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Supplementary Information: Role of Quantum-size Effects on the Dehydrogenation of CH_4 on $3d$ TM_n Clusters: DFT Calculations Combined with Data Mining

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Contents

1 Introduction

A large number of calculations and analyses were performed along the present study, however, several results and analyses, which can be considered as complementary to the main manuscript were not reported within the manuscript due to the space limitation. Thus, the present electronic supporting information file contains complementary results and analyses, which are a plus for the present manuscript, however, they are not essential to the manuscript. Additionally, we report all the results (numbers) employed to build up the figures, which is important for comparison.

2 Flowchart: Designing of the Adsorption Trial Configurations

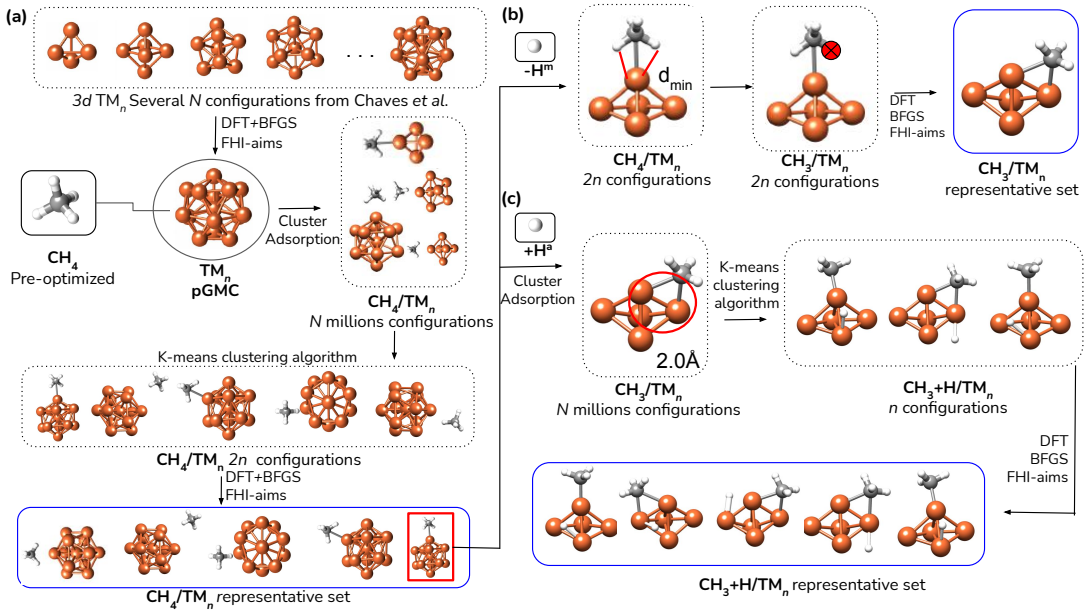

Figure S1: Schematic representation of the designing principles and methodology employed in this present work, namely: (a) adsorption with random orientation (AwRO) using the cluster adsorption algorithm and Euclidean similarity metrics for generating the CH_4/TM_n systems. (b) selective abstraction of one H atom to yield the CH_3/TM_n systems. (c) AwRO of one H atom on the CH_3/TM_n systems to yield the $(CH_3+H)/TM_n$ co-adsorbed configurations.

3 Structural Analysis: Effective Coordination Concept Framework

To characterize the changes the TM_n clusters induced by the adsorption of the CH_4, CH_3, H, CH_3+H systems, we employed the effective coordination concept,[??] which yields the weighted bond length, d^i_{av}, and the effective coordination number, ECN^i, for each atom i in the cluster. In this approach, the d^i_{av} values are calculated using a self-consistent approach based on weighted obtained via an exponential function, as following:

$$d_{av}^{i,new} = \frac{\sum_i d_{ij} exp\left[1-\left(\frac{d_{ij}}{d_{av}^{i,old}}\right)^6\right]}{\sum_j exp\left[1-\left(\frac{d_{ij}}{d_{av}^{i,old}}\right)^6\right]}, \quad (1)$$

where d_{av}^{i} is obtained self-consistently. In the QTNano implementation, the shortest distance between the i and j atoms are employed as the starting value for the $d_{av}^{i,old}$ parameters. The convergence is achieved once the condition, $\mid d_{av}^{i,new} - d_{av}^{i,old} \mid < 10^{-4}$ Å is reached for every atom i within the molecular system. For complicated systems with lower symmetry, the self-consistent scheme reaches its convergence in less than 10 steps.

Once the d_{av}^{i} results are calculated, the ECN^{i} results can be obtained using the following equation,

$$\mathrm{ECN}^{i} = \sum_{j} exp\left[1 - \left(\frac{d_{ij}}{d_{av}^{i}}\right)^{6}\right] . \tag{2}$$

Thus, the average values, i.e., ECN$_{av}$ and d_{av}, are obtained as follows:

$$\mathrm{ECN}_{av} = \frac{1}{N}\sum_{i}\mathrm{ECN}_{i} , \tag{3}$$

and

$$d_{av} = \frac{1}{N}\sum_{i} d_{av}^{i} , \tag{4}$$

where N is the number of atoms in the system. This framework is suitable for transition-metal clusters with lower symmetry as it takes into account tiny differences among the bond lengths. Thus, it can capture small changes in the cluster structure upon the adsorption of molecular systems. At the present work, it was applied for the TM clusters in gas-phase and frozen structures without the adsorbate molecules, i.e., CH_4, CH_3, H, and CH_3+H.

4 Energetic Analyses: Relative Total Energy, Binding Energy, and Adsorption Energies

The energetic analyses is a key point in our investigation, and hence, we provided below the definition for several energetic quantities:

1. Relative total energy, ΔE_{tot}^{i}, calculated with respect to the lowest energy configuration,

$$\Delta E_{tot}^{i} = E_{tot}^{i} - E_{tot}^{lowest} , \tag{5}$$

where E_{tot}^{i} is the total energy of the configuration i, while E_{tot}^{lowest} is the total energy of the lowest energy configuration. Thus, this quantity is useful as only the relative energies (small numbers) are represented and not large total energy values.

2. Binding energy per atom, E_b, using the free-atoms as reference, e.g.,

$$E_b^{\mathrm{TM}_n} = (E_{tot}^{\mathrm{TM}_n} - nE_{tot}^{\mathrm{TM}\ free\ atom})/n\,, \tag{6}$$

$$E_b^{\mathrm{CH_4}} = (E_{tot}^{\mathrm{CH_4}} - E_{tot}^{\mathrm{C}\ free\ atom} - 4E_{tot}^{\mathrm{H}\ free\ atom})/5\,, \tag{7}$$

$$E_b^{\mathrm{CH_3}} = (E_{tot}^{\mathrm{CH_3}} - E_{tot}^{\mathrm{C}\ free\ atom} - 3E_{tot}^{\mathrm{H}\ free\ atom})/4\,, \tag{8}$$

where E_{tot}^{Mol} (Mol = TM_n, $\mathrm{CH_4}$, and $\mathrm{CH_3}$) is the total energy of the molecules in gas-phase, while $E_{tot}^{X\ free\ atom}$ (X = TM, C, and H) is the total energy of the free-atoms obtained from spin-polarized calculations.

3. To evaluate the strength of the $\mathrm{CH_4}$-TM_n, $\mathrm{CH_3}$-TM_n, H-TM_n, and ($\mathrm{CH_3}$+H)-TM_n interactions, we calculated the adsorption energy, E_{ad}, using the following equations:

$$E_{ad}^{\mathrm{CH_4/TM}_n} = E_{tot}^{\mathrm{CH_4/TM}_n} - E_{tot}^{\mathrm{TM}_n\ lowest} - E_{tot}^{\mathrm{CH_4}\ lowest}\,, \tag{9}$$

$$E_{ad}^{\mathrm{CH_3/TM}_n} = E_{tot}^{\mathrm{CH_3/TM}_n} - E_{tot}^{\mathrm{TM}_n\ lowest} - E_{tot}^{\mathrm{CH_3}\ lowest}\,, \tag{10}$$

$$E_{ad}^{\mathrm{(CH_3+H)/TM}_n} = E_{tot}^{\mathrm{(CH_3+H)/TM}_n} - E_{tot}^{\mathrm{TM}_n\ lowest} - E_{tot}^{\mathrm{CH_3}\ lowest} - E_{tot}^{\mathrm{H}\ lowest}\,, \tag{11}$$

$$E_{ad}^{\mathrm{H/TM}_n} = E_{tot}^{\mathrm{H/TM}_n} - E_{tot}^{\mathrm{TM}_n\ lowest} - E_{tot}^{\mathrm{H}\ lowest}\,, \tag{12}$$

where $E_{tot}^{\mathrm{CH_4/TM}_n}$, $E_{tot}^{\mathrm{CH_3/TM}_n}$, $E_{tot}^{\mathrm{(CH_3+H)/TM}_n}$, and $E_{tot}^{\mathrm{H/TM}_n}$ are the total energies in the lowest energy configurations of the $\mathrm{CH_4/TM}_n$, $\mathrm{CH_3/TM}_n$, ($\mathrm{CH_3}$+H)/TM_n, and H/TM_n systems, respectively. The $E_{tot}^{\mathrm{TM}_n\ lowest}$, $E_{tot}^{\mathrm{CH_4}\ lowest}$, $E_{tot}^{\mathrm{CH_3}\ lowest}$, and $E_{tot}^{\mathrm{H}\ lowest}$ are the total energies of those systems in gas-phase.

5 Gas-phase Molecules

The $\mathrm{CH_4}$ molecule has a T_d symmetry with an sp^3 hybridization, which can slightly change upon adsorption, i.e., there is an elongation of the C – H bond length along with change in the

HCH bond angle. Thus, those parameters can be used as indicators to identify the activation of CH_4. In contrast, the CH_3 radical has one unpaired electron located near the C atom (C p_z-state) upon removing one H atom. Thus, the energy difference between the CH_3 planar (sp^2) to T_d (sp^3-like) is -0.3 eV. Furthermore, the HCH changes from the T_d symmetry to 120°, i.e., a feature of sp^2 hybridization. Thus, upon the adsorption of CH_3 on the TM_n clusters, the CH_3-TM_n interaction tends to restore the sp^3 hybridization. Thus, from those calculations for the molecules in gas-phase, we can conclude the activation and bond C–H breaking involves change in the sp^3 hybridization, which helps to explain the high energy cost for this process.

Properties	CH_4	CH_3	$CH_{3,\mathrm{frozen}}$	CH_3^-	CH_3^+	H
Hybridization	sp^3	$sp^2 - p$	sp^3	sp^3	sp^2	s
$d_{av}^{\mathrm{C-H}}$ (Å)	1.10	1.09	1.10	1.12	1.10	
$\theta_{av}^{\mathrm{HCH}}$ (°)	109.47	120.00	109.47	104.50	120.00	
E_b (eV/atom)	−3.65	−3.36	−3.29	−3.31	−1.15	
IP (eV)	12.45	9.99				
EA (eV)	−2.18	−0.20				
ϵ_{homo} (eV)	−9.41	−5.39	−6.18	3.12	−19.90	−7.58
ϵ_{lumo} (eV)	0.38	−2.88	−3.44	5.61	−14.79	2.30
E_g (eV)	9.79	2.51	2.75	2.48	5.40	9.88
m_{tot} (μ_B)	0	1	1	0	0	1

Figure S2: Structural, energetic, and electronic properties for CH_4 and CH_3 molecules, and H atom: average C–H bond length ($d_{av}^{\mathrm{C-H}}$), average HCH bond angle ($\theta_{av}^{\mathrm{HCH}}$), binding energy per atom (E_b), ionization potential (IP), electron affinity (AE), HOMO energy ($\varepsilon_{\mathrm{homo}}$), LUMO energy ($\varepsilon_{\mathrm{lumo}}$), LUMO-HOMO energy separation (E_g) and total magnetic moment (m_{tot}).

6 Gas-phase Clusters

6.1 Computational Details

Below, we summarize complementary computational details for the gas-phase clusters:

1. The initial TM_n configurations where obtained from Chaves *et al.*,[?] which were re-optimized with the DFT-PBE framework and using the light-tier2 basis set with a total energy and atomic forces criteria cutoff of 2.5×10^{-5} eV and 2.5×10^{-3} eV Å^{-1}, while for obtain the equilibrium geometry we used a cutoff of 2.5×10^{-2} eV Å^{-1}.

2. All representative configurations were selected by the clustering *k-means* algorithm by using energy as criteria to reduce the initial set from i configurations to $n/2$, where n represent the number of atoms in the TM_n clusters.

3. Therefore, we re-normalized the putative global minima configurations (pGMCs) with a more accurate criteria for both electronic self-consistency and relaxation, where the convergence for the first one was achieved once the total energy and atomic forces criteria were smaller than 2.5×10^{-6} eV and 2.5×10^{-5} eV Å^{-1}, respectively. The equilibrium geometry was obtained once the atomic forces on every atom were smaller than 2.5×10^{-4} eV Å^{-1}.

4. The properties for the representative set of the TM_n clusters are given on Tables S1-S12, while the refined pGMCS are show on Figure S3 along with their most relevant properties, Table S13.

6.2 Data (Tables): Structural, Electronic, and Energetic Properties

Table S1: Structural, energetic, and electronic properties for the TM_4 clusters: cluster label configuration (i), relative total energy per atom (ΔE_{tot}), binding energy per atom (E_b), total magnetic moment (m_{tot}), HOMO energy $(\varepsilon_{\text{homo}})$, LUMO energy $(\varepsilon_{\text{lumo}})$, LUMO-HOMO energy band gap, (E_g), average effective coordination number (ECN_{av}), average weighted bond lengths (d_{av}), average radius (R_{av}), and total number of bonds (N_b).

TM_4	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	4	2398	−1.53	14	−3.82	−3.72	0.10	1.13	2.09	3.35	3
	pGMC	0	−2.12	14	−3.89	−3.44	0.45	2.66	2.29	1.36	6
Co	3	2385	−1.57	10	−3.96	−3.96	0.00	1.05	2.00	3.26	3
	pGMC	0	−2.17	10	−4.17	−3.66	0.52	2.09	2.15	1.41	6
Ni	1	56	−2.13	4	−3.97	−3.61	0.36	2.50	2.24	1.93	5
	pGMC	0	−2.14	4	−3.42	−3.15	0.28	2.93	2.27	1.28	6
Cu	2	775	−1.42	2	−4.43	−3.88	0.55	2.00	2.31	1.64	4
	pGMC	0	−1.61	0	−4.46	−3.45	1.00	2.47	2.35	2.09	5

Table S2: Structural, energetic, and electronic properties for the TM_5 clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_5	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	4	1024	−2.23	16	−3.69	−3.12	0.57	2.67	2.26	2.32	7
	3	55	−2.42	18	−4.02	−3.64	0.38	3.26	2.35	1.73	9
	pGMC	0	−2.44	16	−3.76	−3.58	0.18	3.38	2.33	1.85	9
Co	7	1432	−2.18	11	−3.74	−3.52	0.22	1.99	2.12	1.89	5
	4	556	−2.36	11	−3.85	−3.33	0.52	2.74	2.21	2.05	8
	pGMC	0	−2.47	13	−4.21	−3.72	0.49	2.85	2.23	1.57	9
Ni	3	579	−2.29	6	−4.14	−4.01	0.13	2.78	2.25	2.27	7
	1	66	−2.39	4	−3.54	−3.24	0.29	3.57	2.30	1.94	9
	pGMC	66	−2.39	4	−3.54	−3.24	0.29	3.57	2.30	1.94	9
Cu	2	294	−1.68	1	−4.35	−3.88	0.47	2.40	2.33	2.41	6
	1	285	−1.69	1	−3.96	−3.75	0.21	3.53	2.40	1.98	9
	pGMC	0	−1.74	1	−4.28	−3.84	0.44	2.79	2.36	2.40	7

Table S3: Structural, energetic, and electronic properties for the TM_6 clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_6	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	7	3050	−2.20	18	−3.42	−3.22	0.20	2.90	2.30	2.44	9
	5	1100	−2.53	20	−4.00	−3.49	0.51	3.00	2.31	1.70	9
	pGMC	0	−2.71	20	−3.92	−3.46	0.46	3.81	2.36	1.81	13
Co	7	2336	−2.43	12	−3.93	−3.61	0.32	2.91	2.22	2.40	9
	4	1062	−2.64	14	−3.81	−3.57	0.24	3.27	2.24	1.89	11
	pGMC	0	−2.82	14	−3.98	−3.45	0.53	4.00	2.27	1.61	12
Ni	6	734	−2.51	6	−3.84	−3.59	0.24	3.32	2.28	2.03	10
	3	412	−2.56	6	−3.80	−3.61	0.18	3.92	2.32	2.09	12
	pGMC	0	−2.63	8	−3.99	−3.82	0.18	3.97	2.32	1.64	12
Cu	7	1432	−1.70	2	−4.46	−4.03	0.43	2.67	2.35	3.22	8
	2	294	−1.89	0	−4.24	−3.78	0.46	3.94	2.41	2.13	12
	pGMC	0	−1.94	0	−4.68	−2.85	1.83	2.99	2.35	2.52	9

Table S4: Structural, energetic, and electronic properties for the TM_7 clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap, E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_7	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	7	3689	−2.34	22	−3.71	−3.33	0.38	2.86	2.28	3.61	11
	6	2834	−2.46	22	−3.34	−3.12	0.22	3.32	2.32	2.46	12
	4	937	−2.73	22	−3.46	−3.14	0.32	4.13	2.36	2.37	15
	pGMC	0	−2.87	22	−3.70	−3.32	0.38	4.17	2.36	1.94	16
Co	14	2751	−2.47	15	−3.89	−3.74	0.15	2.98	2.23	2.97	11
	5	211	−2.83	15	−3.73	−3.49	0.25	4.45	2.31	1.95	16
	1	115	−2.85	15	−3.59	−3.41	0.18	4.24	2.29	1.89	16
	pGMC	0	−2.86	15	−3.72	−3.39	0.33	4.28	2.29	2.21	15
Ni	11	656	−2.66	8	−3.99	−3.83	0.16	3.71	2.29	2.20	13
	4	315	−2.71	8	−4.01	−3.62	0.38	3.75	2.30	1.97	15
	3	181	−2.73	6	−3.83	−3.67	0.17	4.53	2.35	2.00	16
	pGMC	0	−2.76	8	−3.97	−3.81	0.16	4.28	2.32	2.23	15
Cu	11	1526	−1.84	1	−4.36	−3.97	0.39	3.14	2.37	3.62	11
	8	994	−1.92	1	−3.65	−3.51	0.14	3.42	2.39	2.42	12
	2	331	−2.01	1	−4.43	−4.17	0.27	4.18	2.41	2.26	15
	pGMC	0	−2.06	1	−4.05	−3.69	0.36	4.55	2.42	2.01	16

Table S5: Structural, energetic, and electronic properties for the TM_8 clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_8	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	11	5074	−2.32	24	−3.62	−3.34	0.29	3.18	2.33	3.05	13
	10	3829	−2.47	24	−3.47	−3.29	0.17	3.39	2.32	3.37	14
	5	411	−2.90	24	−3.51	−3.15	0.36	4.40	2.36	2.42	19
	pGMC	0	−2.95	24	−3.59	−3.07	0.52	4.19	2.34	2.03	19
Co	18	3181	−2.57	16	−3.86	−3.66	0.20	3.11	2.22	2.89	13
	13	845	−2.86	16	−3.62	−3.21	0.41	3.07	2.20	1.98	18
	2	175	−2.95	16	−3.62	−3.24	0.38	4.53	2.31	2.17	19
	pGMC	0	−2.97	16	−3.57	−3.27	0.29	4.53	2.30	2.08	20
Ni	17	1870	−2.62	8	−4.09	−3.89	0.21	3.49	2.28	3.17	14
	14	926	−2.74	8	−3.87	−3.64	0.23	3.96	2.30	2.55	16
	10	450	−2.80	8	−3.85	−3.75	0.11	4.10	2.31	2.15	18
	pGMC	0	−2.85	8	−3.82	−3.67	0.15	4.54	2.34	2.15	19
Cu	11	1196	−2.01	0	−4.43	−3.25	1.19	3.36	2.36	2.27	14
	10	1187	−2.01	0	−4.35	−3.53	0.82	3.47	2.38	3.41	14
	9	880	−2.05	0	−4.46	−3.96	0.50	4.23	2.41	2.73	18
	pGMC	0	−2.16	0	−4.60	−3.09	1.51	4.50	2.42	2.14	20

Table S6: Structural, energetic, and electronic properties for the TM_9 clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_9	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	15	1165	−2.87	28	−3.59	−3.28	0.31	4.39	2.36	2.94	21
	10	642	−2.93	26	−3.44	−3.17	0.27	4.35	2.34	2.46	22
	6	555	−2.94	28	−3.65	−3.21	0.44	4.46	2.38	2.97	23
	4	90	−2.99	26	−3.46	−2.89	0.57	4.37	2.35	2.10	21
	pGMC	0	−3.00	26	−3.37	−2.96	0.41	4.91	2.38	2.33	23
Co	20	3738	−2.62	17	−3.69	−3.44	0.25	3.50	2.26	3.86	16
	18	1532	−2.86	19	−3.81	−3.51	0.30	4.33	2.30	2.82	20
	5	453	−2.98	17	−3.47	−3.20	0.28	4.66	2.30	2.27	21
	4	418	−2.99	19	−3.74	−3.54	0.19	4.64	2.32	2.87	23
	pGMC	0	−3.03	17	−3.49	−3.32	0.17	4.63	2.30	2.22	21
Ni	19	2427	−2.68	8	−4.12	−3.88	0.24	3.53	2.28	3.91	16
	16	830	−2.86	10	−4.07	−3.89	0.19	4.39	2.32	3.10	20
	10	487	−2.90	8	−3.86	−3.64	0.22	4.58	2.34	2.80	23
	8	337	−2.91	8	−3.72	−3.68	0.04	4.66	2.33	2.29	21
	pGMC	0	−2.95	8	−3.85	−3.75	0.11	4.61	2.34	2.25	21
Cu	17	1119	−2.04	1	−4.27	−4.02	0.25	3.52	2.38	3.31	16
	12	586	−2.10	1	−3.70	−3.48	0.22	4.66	2.42	2.36	21
	6	226	−2.14	1	−3.61	−3.29	0.31	4.83	2.43	2.56	22
	2	79	−2.15	1	−3.59	−3.24	0.35	4.87	2.43	2.55	22
	pGMC	0	−2.16	1	−4.19	−3.91	0.28	4.68	2.43	2.94	23

Table S7: Structural, energetic, and electronic properties for the TM_{10} clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_{10}	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	22	2320	−2.83	28	−3.33	−3.14	0.19	4.69	2.38	2.61	24
	7	315	−3.03	30	−3.69	−3.18	0.51	4.96	2.38	2.96	26
	3	271	−3.03	30	−3.59	−3.18	0.41	4.94	2.39	2.99	26
	2	113	−3.05	28	−3.58	−3.03	0.56	4.50	2.34	2.38	24
	pGMC	0	−3.06	30	−3.29	−3.19	0.10	5.24	2.40	2.42	27
Co	15	594	−3.03	20	−3.61	−3.46	0.16	5.13	2.33	2.97	26
	7	431	−3.05	18	−3.46	−3.22	0.24	4.89	2.31	2.34	25
	2	257	−3.07	18	−3.45	−3.24	0.21	4.77	2.31	2.41	24
	1	251	−3.07	20	−3.74	−3.46	0.29	5.17	2.33	2.82	26
	pGMC	0	−3.09	20	−3.67	−3.47	0.20	4.96	2.32	2.63	25
Ni	25	2915	−2.74	10	−4.08	−3.96	0.11	3.79	2.30	3.39	19
	24	1686	−2.86	8	−3.98	−3.89	0.10	3.38	2.24	2.58	17
	15	770	−2.95	10	−3.97	−3.88	0.09	4.95	2.35	2.71	25
	8	582	−2.97	10	−4.00	−3.89	0.10	4.74	2.34	2.94	24
	pGMC	0	−3.03	8	−3.92	−3.53	0.40	4.71	2.32	2.52	24
Cu	18	986	−2.14	0	−3.60	−3.20	0.40	4.75	2.44	2.65	24
	17	973	−2.14	0	−4.59	−3.37	1.22	3.79	2.39	3.60	19
	16	923	−2.15	2	−3.87	−3.52	0.35	4.97	2.44	2.44	25
	10	487	−2.19	2	−3.92	−3.57	0.36	5.33	2.45	2.49	27
	pGMC	0	−2.24	0	−4.18	−3.15	1.02	5.16	2.44	3.00	26

Table S8: Structural, energetic, and electronic properties for the TM_{11} clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_{11}	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	19	915	−3.04	34	−3.61	−3.26	0.35	4.54	2.38	2.81	29
	11	775	−3.05	34	−3.66	−3.31	0.35	4.64	2.38	2.88	30
	4	558	−3.07	34	−3.57	−3.21	0.36	4.92	2.40	2.48	31
	2	501	−3.07	32	−3.40	−3.20	0.20	5.25	2.40	3.05	30
	1	197	−3.10	34	−3.65	−3.33	0.32	5.22	2.41	2.97	30
	pGMC	0	−3.12	34	−3.45	−3.17	0.27	5.41	2.42	2.51	31
Co	17	338	−3.09	21	−3.62	−3.46	0.16	4.87	2.32	3.13	27
	7	198	−3.10	21	−3.59	−3.39	0.19	5.10	2.33	2.74	31
	6	182	−3.10	21	−3.62	−3.48	0.15	5.05	2.33	2.73	28
	3	82	−3.11	19	−3.43	−3.21	0.22	5.17	2.32	2.45	29
	2	81	−3.11	23	−3.83	−3.66	0.17	5.01	2.33	2.92	28
	pGMC	0	−3.12	23	−3.82	−3.59	0.23	5.09	2.33	3.07	29
Ni	33	3462	−2.75	12	−4.23	−4.14	0.09	3.80	2.30	3.57	21
	15	526	−3.02	10	−3.87	−3.70	0.18	5.31	2.36	3.01	30
	6	319	−3.04	10	−3.95	−3.81	0.14	5.01	2.35	2.94	28
	2	161	−3.05	8	−3.81	−3.59	0.22	4.94	2.34	2.43	28
	1	96	−3.06	8	−3.78	−3.67	0.12	5.23	2.35	2.46	29
	pGMC	0	−3.06	8	−3.81	−3.67	0.15	5.33	2.35	2.41	30
Cu	30	1445	−2.13	1	−4.34	−4.11	0.22	3.81	2.39	3.74	21
	24	664	−2.21	1	−3.90	−3.64	0.26	5.01	2.44	2.92	28
	10	345	−2.23	1	−3.89	−3.63	0.26	5.33	2.45	3.08	30
	8	312	−2.24	1	−3.98	−3.73	0.25	5.13	2.44	2.94	30
	2	150	−2.25	1	−3.94	−3.68	0.27	5.33	2.45	3.23	30
	pGMC	0	−2.27	1	−4.20	−3.96	0.23	5.36	2.45	3.10	30

Table S9: Structural, energetic, and electronic properties for the TM_{12} clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_{12}	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	22	1448	−3.06	36	−3.61	−3.32	0.29	4.92	2.39	3.64	33
	15	1108	−3.09	36	−3.61	−3.30	0.31	4.81	2.39	3.16	33
	10	963	−3.10	34	−3.44	−3.16	0.29	5.48	2.41	2.88	34
	4	675	−3.13	38	−3.60	−3.36	0.24	5.20	2.41	2.74	34
	1	396	−3.15	36	−3.52	−3.21	0.31	5.48	2.42	2.98	34
	pGMC	0	−3.18	36	−3.32	−3.17	0.16	5.54	2.42	2.47	36
Co	30	1460	−3.06	30	−4.03	−3.90	0.13	5.00	2.36	2.25	30
	14	402	−3.15	22	−3.52	−3.31	0.21	5.58	2.35	2.84	34
	4	199	−3.16	24	−3.74	−3.54	0.20	5.60	2.36	2.82	34
	3	189	−3.16	24	−3.69	−3.59	0.10	5.14	2.33	3.17	33
	1	148	−3.17	24	−3.70	−3.50	0.19	5.30	2.34	2.99	35
	pGMC	0	−3.18	24	−3.69	−3.49	0.20	5.25	2.33	2.74	32
Ni	13	556	−3.06	10	−3.90	−3.77	0.13	5.11	2.35	3.12	32
	11	495	−3.07	12	−4.07	−3.88	0.19	5.09	2.35	3.35	31
	8	400	−3.07	12	−3.99	−3.85	0.14	5.55	2.37	2.97	34
	5	348	−3.08	8	−3.75	−3.60	0.14	5.88	2.38	2.38	36
	1	32	−3.11	10	−3.78	−3.70	0.08	5.54	2.36	2.84	34
	pGMC	0	−3.11	10	−3.93	−3.81	0.12	5.47	2.36	2.87	34
Cu	26	2184	−2.14	0	−4.40	−3.86	0.54	3.81	2.38	4.45	23
	24	1699	−2.18	4	−4.27	−4.10	0.17	4.97	2.44	2.35	30
	11	513	−2.28	0	−3.93	−3.67	0.26	5.59	2.46	3.00	34
	9	493	−2.28	0	−4.42	−3.39	1.03	5.15	2.43	3.49	32
	2	188	−2.31	0	−4.30	−3.65	0.65	5.33	2.45	3.14	35
	pGMC	0	−2.32	0	−4.40	−3.59	0.81	5.54	2.45	3.09	34

Table S10: Structural, energetic, and electronic properties for the TM_{13} clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_{13}	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	18	1519	−3.15	38	−3.50	−3.24	0.25	5.22	2.40	3.23	38
	17	1519	−3.15	40	−3.62	−3.35	0.27	5.23	2.41	3.20	37
	13	1356	−3.16	38	−3.52	−3.30	0.22	5.66	2.42	2.99	38
	7	1065	−3.19	40	−3.54	−3.43	0.11	5.63	2.43	3.04	38
	4	972	−3.19	40	−3.56	−3.39	0.17	5.37	2.41	2.63	38
	2	878	−3.20	40	−3.47	−3.25	0.22	5.61	2.42	3.21	39
	pGMC	0	−3.27	44	−3.69	−3.41	0.28	6.31	2.48	2.42	42
Co	32	1029	−3.19	31	−3.81	−3.66	0.15	6.38	2.42	2.34	42
	25	967	−3.20	25	−3.71	−3.54	0.18	5.16	2.33	3.22	34
	15	756	−3.21	25	−3.69	−3.53	0.17	5.28	2.33	3.27	35
	9	711	−3.22	25	−3.66	−3.54	0.12	5.24	2.33	3.20	38
	6	577	−3.23	25	−3.62	−3.44	0.18	5.71	2.35	3.03	38
	1	546	−3.23	27	−3.79	−3.65	0.14	5.34	2.34	3.16	39
	pGMC	0	−3.27	27	−3.89	−3.58	0.31	5.45	2.34	2.77	36
Ni	37	673	−3.11	12	−3.94	−3.88	0.06	5.46	2.37	2.67	36
	25	535	−3.12	12	−3.95	−3.88	0.07	5.00	2.34	3.23	34
	24	528	−3.12	12	−4.01	−3.87	0.14	5.31	2.35	3.32	37
	10	340	−3.13	10	−3.84	−3.77	0.07	5.37	2.35	3.78	35
	6	308	−3.13	8	−3.74	−3.64	0.10	6.38	2.41	2.36	42
	2	165	−3.15	12	−3.97	−3.86	0.11	5.68	2.37	3.10	38
	pGMC	0	−3.16	10	−3.84	−3.72	0.11	5.67	2.37	3.02	38
Cu	49	1473	−2.24	1	−4.00	−3.82	0.17	4.89	2.42	3.40	33
	34	791	−2.29	1	−4.15	−3.91	0.24	5.45	2.45	2.79	36
	25	591	−2.31	1	−4.12	−3.92	0.20	5.46	2.45	2.90	36
	16	505	−2.31	1	−3.76	−3.56	0.21	5.69	2.46	3.12	38
	7	318	−2.33	1	−3.88	−3.65	0.22	5.65	2.46	3.30	38
	5	269	−2.33	1	−3.91	−3.68	0.23	5.67	2.46	3.12	38
	pGMC	0	−2.35	1	−4.03	−3.83	0.20	5.70	2.46	3.08	38

Table S11: Structural, energetic, and electronic properties for the TM_{14} clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_{14}	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	31	1762	−3.18	42	−3.59	−3.34	0.25	5.13	2.39	3.47	40
	29	1704	−3.18	42	−3.60	−3.42	0.18	5.47	2.42	3.19	41
	9	1005	−3.23	42	−3.52	−3.36	0.17	5.75	2.43	3.41	42
	8	994	−3.23	42	−3.56	−3.35	0.21	5.73	2.43	3.07	42
	6	697	−3.25	42	−3.48	−3.20	0.28	5.82	2.43	3.17	43
	5	569	−3.26	44	−3.68	−3.59	0.09	5.79	2.44	2.59	46
	pGMC	0	−3.30	46	−3.80	−3.54	0.26	5.93	2.46	2.62	46
Co	48	992	−3.22	28	−3.68	−3.50	0.19	5.27	2.33	3.28	40
	12	401	−3.26	28	−3.71	−3.58	0.13	5.61	2.36	2.96	40
	5	296	−3.27	28	−3.74	−3.60	0.14	5.37	2.34	3.29	40
	4	293	−3.27	26	−3.59	−3.38	0.21	5.81	2.36	3.07	42
	3	197	−3.28	28	−3.85	−3.68	0.17	5.52	2.34	3.08	41
	2	189	−3.28	28	−3.78	−3.66	0.12	5.81	2.36	3.07	42
	pGMC	0	−3.29	28	−3.72	−3.56	0.15	5.45	2.33	3.23	39
Ni	20	617	−3.16	12	−3.87	−3.76	0.11	5.48	2.36	2.74	40
	18	584	−3.16	10	−3.81	−3.68	0.12	5.52	2.36	2.94	41
	12	419	−3.17	12	−3.99	−3.87	0.12	5.45	2.36	3.15	40
	5	280	−3.18	12	−3.95	−3.92	0.03	5.38	2.35	3.32	38
	4	263	−3.18	14	−4.08	−3.94	0.14	5.82	2.38	3.11	42
	3	243	−3.18	12	−4.00	−3.87	0.13	5.83	2.37	3.52	42
	pGMC	0	−3.20	12	−3.93	−3.85	0.08	5.79	2.37	3.12	42
Cu	44	3677	−2.15	2	−4.00	−3.89	70.11	5.09	2.74	7.47	39
	24	990	−2.34	0	−4.21	−3.60	0.61	5.33	2.44	3.81	39
	20	942	−2.34	2	−3.92	−3.78	0.14	5.98	2.47	3.19	43
	9	735	−2.36	0	−4.18	−3.74	0.45	5.46	2.45	3.78	40
	6	711	−2.36	0	−3.98	−3.61	0.38	5.81	2.46	3.13	42
	4	457	−2.38	0	−4.13	−3.73	0.40	5.77	2.46	3.10	42
	pGMC	0	−2.41	0	−4.45	−3.45	1.00	5.80	2.46	3.21	42

Table S12: Structural, energetic, and electronic properties for the TM_{15} clusters: cluster label configuration i, relative total energy per atom ΔE_{tot}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_{15}	i	ΔE_{tot} (meV)	E_b (eV)	m_{tot} (μ_B)	$\varepsilon_{\text{homo}}$ (eV)	$\varepsilon_{\text{lumo}}$ (eV)	E_g (eV)	ECN_{av} (NNN)	d_{av} (Å)	R_{av} (Å)	N_b
Fe	32	2924	−3.21	44	−3.50	−3.24	0.26	5.36	2.41	3.65	46
	30	2795	−3.22	44	−3.49	−3.33	0.16	5.54	2.41	3.39	44
	23	2566	−3.23	44	−3.56	−3.41	0.15	5.41	2.40	3.21	45
	18	2269	−3.25	44	−3.43	−3.28	0.15	5.89	2.43	3.42	46
	12	1938	−3.27	44	−3.49	−3.23	0.25	5.85	2.43	3.17	46
	5	1603	−3.29	48	−3.78	−3.56	0.22	5.75	2.43	2.84	46
	3	1101	−3.33	48	−3.68	−3.37	0.31	6.35	2.47	3.10	49
	pGMC	0	−3.40	48	−3.76	−3.38	0.38	6.15	2.45	2.56	50
Co	44	1907	−3.20	29	−3.73	−3.57	0.16	5.15	2.32	3.73	40
	32	787	−3.28	29	−3.70	−3.56	0.14	5.61	2.35	3.33	45
	23	567	−3.29	29	−3.63	−3.50	0.13	5.38	2.33	3.48	42
	7	356	−3.31	27	−3.52	−3.41	0.11	5.97	2.36	3.45	46
	4	294	−3.31	29	−3.67	−3.52	0.15	5.54	2.34	3.27	43
	2	258	−3.31	29 ¨	−3.68	−3.54	0.14	5.80	2.35	3.23	44
	1	239	−3.31	29	−3.73	−3.55	0.18	5.78	2.36	3.08	46
	pGMC	0	−3.33	29	−3.69	−3.55	0.13	5.65	2.34	3.21	43
Ni	41	1524	−3.14	14	−4.00	−3.91	0.09	5.44	2.36	3.66	44
	28	881	−3.19	14	−4.00	−3.88	0.12	5.50	2.36	3.39	43
	15	659	−3.20	12	−3.86	−3.80	0.07	5.56	2.36	3.15	45
	12	622	−3.20	12	−3.99	−3.88	0.10	5.66	2.36	3.22	43
	3	369	−3.22	14	−4.13	−3.98	0.15	5.42	2.35	3.31	42
	2	366	−3.22	12	−3.84	−3.74	0.11	6.07	2.39	2.98	47
	1	269	−3.23	12	−3.95	−3.82	0.13	6.41	2.40	3.07	49
	pGMC	0	−3.25	12	−3.94	−3.86	0.08	5.93	2.37	3.48	46
Cu	42	4030	−2.14	1	−4.41	−4.21	0.20	3.70	2.38	5.21	28
	37	1161	−2.34	1	−3.88	−3.68	0.20	5.53	2.45	4.12	42
	36	1054	−2.34	1	−3.77	−3.53	0.23	5.52	2.45	3.76	43
	12	433	−2.38	1	−3.85	−3.63	0.23	5.63	2.45	3.37	43
	6	373	−2.39	3	−4.15	−3.94	0.21	6.41	2.49	3.15	49
	3	282	−2.39	1	−4.07	−3.85	0.22	6.02	2.47	3.09	47
	1	76	−2.41	1	−4.14	−3.96	0.18	5.45	5.45	3.40	41
	pGMC	0	−2.41	1	−3.92	−3.72	0.20	5.91	2.46	3.53	46

Table S13: Structural, energetic, and electronic properties for the TM_n pGMCs: cluster TM atom, number of atoms n, DFT total energy E_{tot}^{DFT}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\mathrm{homo}}$, LUMO energy $\varepsilon_{\mathrm{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_n	n	E_{tot}^{DFT}	E_b	m_{tot}	$\varepsilon_{\mathrm{homo}}$	$\varepsilon_{\mathrm{lumo}}$	E_g	ECN_{av}	d_{av}	N_b	R_{av}
Fe	4	−139056.94713412	−2.14	14	−3.89	−3.45	0.44	2.68	2.29	6	1.36
	5	−173822.74929841	−2.45	16	−3.76	−3.58	0.18	3.38	2.33	9	1.85
	6	−208588.95737418	−2.73	20	−3.93	−3.46	0.47	3.81	2.36	13	1.81
	7	−243354.93417974	−2.89	22	−3.69	−3.31	0.38	4.18	2.36	16	1.95
	8	−278120.64233925	−2.98	24	−3.58	−3.06	0.52	4.20	2.35	19	2.04
	9	−312886.25380373	−3.04	26	−3.35	−2.96	0.39	4.92	2.38	23	2.33
	10	−347652.08303398	−3.11	30	−3.30	−3.19	0.11	5.24	2.41	27	2.42
	11	−382418.08138225	−3.18	34	−3.44	−3.17	0.27	5.41	2.42	31	2.51
	12	−417184.21187631	−3.25	36	−3.31	−3.17	0.14	5.56	2.42	36	2.47
	13	−451950.79109570	−3.35	44	−3.67	−3.38	0.29	6.31	2.49	42	2.42
	14	−486716.83079646	−3.39	46	−3.80	−3.53	0.27	5.94	2.46	46	2.63
	15	−521483.89000220	−3.49	48	−3.74	−3.39	0.36	6.16	2.46	50	2.56
Co	4	−152283.86395647	−2.18	10	−4.17	−3.65	0.52	2.09	2.15	6	1.41
	5	−190356.33035948	−2.48	13	−4.21	−3.72	0.49	2.85	2.23	9	1.56
	6	−228429.70423114	−2.83	14	−3.99	−3.45	0.53	4.00	2.27	12	1.61
	7	−266501.70128176	−2.88	15	−3.73	−3.39	0.34	4.28	2.30	15	2.21
	8	−304574.23102333	−2.99	16	−3.56	−3.26	0.30	4.52	2.31	20	2.10
	9	−342646.67579777	−3.06	17	−3.49	−3.32	0.17	4.63	2.31	21	2.22
	10	−380719.19900567	−3.13	20	−3.67	−3.46	0.21	4.95	2.32	25	2.63
	11	−418791.50158599	−3.17	23	−3.81	−3.58	0.23	5.09	2.33	29	3.07
	12	−456864.28559254	−3.24	24	−3.70	−3.49	0.21	5.24	2.33	33	2.77
	13	−494937.57991850	−3.33	27	−3.89	−3.58	0.31	5.45	2.34	36	2.77
	14	−533010.06879742	−3.36	28	−3.72	−3.56	0.15	5.45	2.33	39	3.22
	15	−571082.90344263	−3.41	29	−3.69	−3.55	0.14	5.65	2.35	43	3.21
Ni	4	−166251.05667065	−2.13	4	−3.42	−3.15	0.28	2.93	2.27	6	1.28
	5	−207815.18483697	−2.40	6	−3.92	−3.59	0.33	3.20	2.28	8	1.64
	6	−249379.59333081	−2.63	8	−3.99	−3.82	0.18	3.97	2.32	12	1.64
	7	−290943.82217984	−2.76	8	−3.97	−3.81	0.16	4.28	2.32	15	2.22
	8	−332508.09774503	−2.87	8	−3.83	−3.68	0.15	4.54	2.34	19	2.14
	9	−374072.57727050	−2.98	8	−3.84	−3.74	0.10	4.62	2.34	21	2.24
	10	−415637.16738596	−3.08	8	−3.93	−3.53	0.40	4.70	2.31	24	2.52
	11	−457201.47501850	−3.13	8	−3.81	−3.66	0.15	5.32	2.35	30	2.41
	12	−498765.85856452	−3.18	10	−3.92	−3.82	0.10	5.48	2.36	34	2.86
	13	−540330.43313167	−3.24	10	−3.84	−3.73	0.11	5.67	2.36	38	3.01
	14	−581895.06227970	−3.29	12	−3.93	−3.86	0.07	5.80	2.37	42	3.10
	15	−623459.75109867	−3.35	12	−3.94	−3.84	0.10	5.93	2.37	46	3.45
Cu	4	−180976.89556019	−1.60	0	−4.45	−3.46	1.00	2.48	2.35	5	2.08
	5	−226221.82570163	−1.74	1	−4.28	−3.84	0.44	2.79	2.35	7	2.39
	6	−271467.41068803	−1.95	0	−4.68	−2.84	1.84	2.99	2.35	9	2.51
	7	−316712.82679610	−2.07	1	−4.05	−3.69	0.36	4.55	2.42	16	2.01

Continued on next page

Table S13: Structural, energetic, and electronic properties for the TM_n pGMCs: cluster TM atom, number of atoms n, DFT total energy E_{tot}^{DFT}, binding energy per atom E_b, total magnetic moment m_{tot}, HOMO energy $\varepsilon_{\mathrm{homo}}$, LUMO energy $\varepsilon_{\mathrm{lumo}}$, LUMO-HOMO energy band gap E_g, average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b.

TM_n	n	E_{tot}^{DFT}	E_b	m_{tot}	$\varepsilon_{\mathrm{homo}}$	$\varepsilon_{\mathrm{lumo}}$	E_g	ECN_{av}	d_{av}	N_b	R_{av}
	8	−361958.39404104	−2.18	0	−4.60	−3.07	1.54	4.49	2.42	20	2.14
	9	−407203.24432142	−2.18	1	−4.18	−3.90	0.28	4.67	2.43	23	2.93
	10	−452448.95152443	−2.27	0	−4.17	−3.15	1.02	5.16	2.44	26	2.99
	11	−497694.20238829	−2.31	1	−4.20	−3.96	0.23	5.36	2.44	30	3.08
	12	−542939.92656457	−2.37	0	−4.40	−3.60	0.80	5.54	2.45	34	3.07
	13	−588185.36659674	−2.41	1	−4.03	−3.83	0.20	5.69	2.45	38	3.06
	14	−633431.30246182	−2.47	0	−4.44	−3.48	0.96	5.81	2.45	42	3.19
	15	−678676.52150806	−2.48	1	−3.93	−3.73	0.20	5.89	2.45	46	3.51

6.3 Analysis: Structural, Electronic, and Energetic Properties

The lowest energy configurations, i.e., the putative global minimum configurations, are shown in Figure S3. The most important remarks are the following:

- The Fe_n cluster are closed packed structures which evolve from tetrahedral to icosahedron motifs with the increasing of n.
- The Co_n preferentially form layered-like hexagonal structures.
- Ni_n clusters present both tetrahedral and icosahedron motifs.
- For small values of n, Cu_n are planar while from n up to 7 the formation of polytetrahedron geometry is preferred.

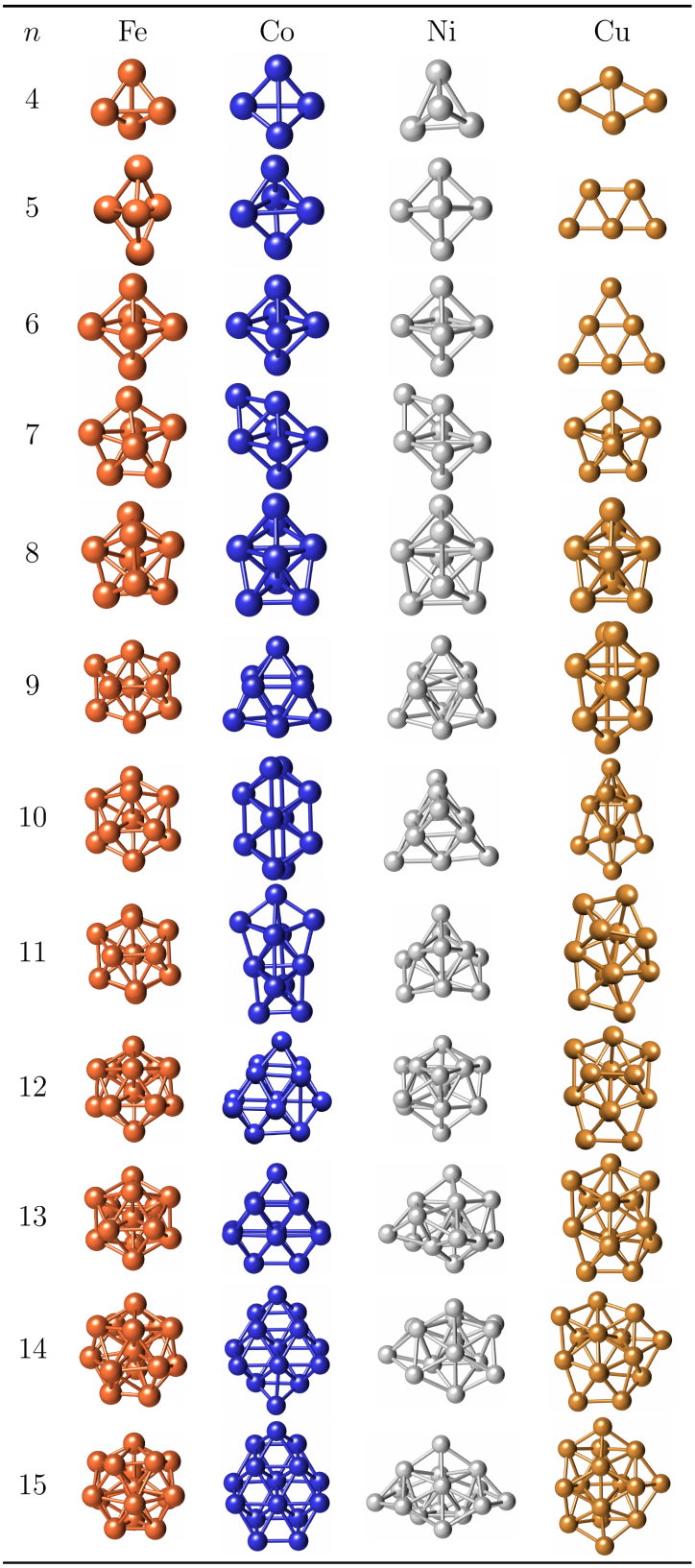

Figure S3: Putative global minimum configurations (pGMCs) for the TM_n clusters, where $n = 4 - 15$ and TM = Fe, Co, Ni, Cu.

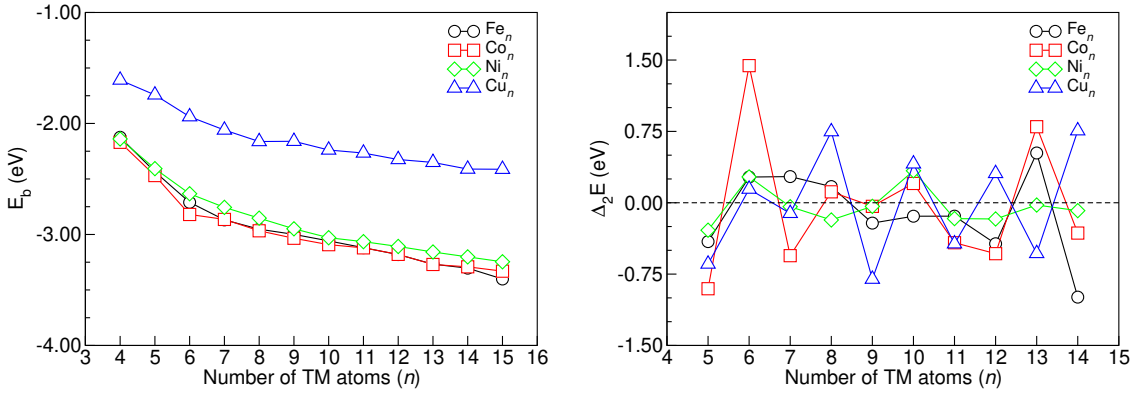

Figure S4: Binding energy and stability function for the pGMCs of the TM_n clusters from $n = 4 - 15$, where TM= Fe, Co, Ni, Cu.

- The increase of the number of TM atoms increase the E_b magnitude in absolute value.
- The occupation of the anti-bonding d-states leads to the decrease of the E_b magnitude, which explain the following trend $|E_b|$: Fe_n ($[Ar]3d^6 4s^2$) > Co_n ($[Ar]3d^7 4s^2$)> Ni_n ($[Ar]3d^9 4s^1$) > Cu_n ($[Ar]3d^{10} 4s^1$).
- The most stable Fe_n cluster are Fe_8 at small regime, i.e., $n < 9$ and Fe_{13} for a larger regime $n > 9$.
- For Co_n both Co_6 and Co_{13} present the larger values of stability function, and hence, are considered magic clusters. The larger differences between $\Delta_2 E$ are observed for these systems.
- Ni_6 and Ni_{10} present only slightly greater values of the stability function in comparison with their neighboring clusters.
- For Cu_n clusters, the most stable clusters are Cu_8 and Cu_{14}. However, a different pattern is observed, i.e., clusters with an even number of atoms are more stable than their odd neighboring ones. In conjunction, they lead to zig-zag behaviour of $\Delta_2 E$.

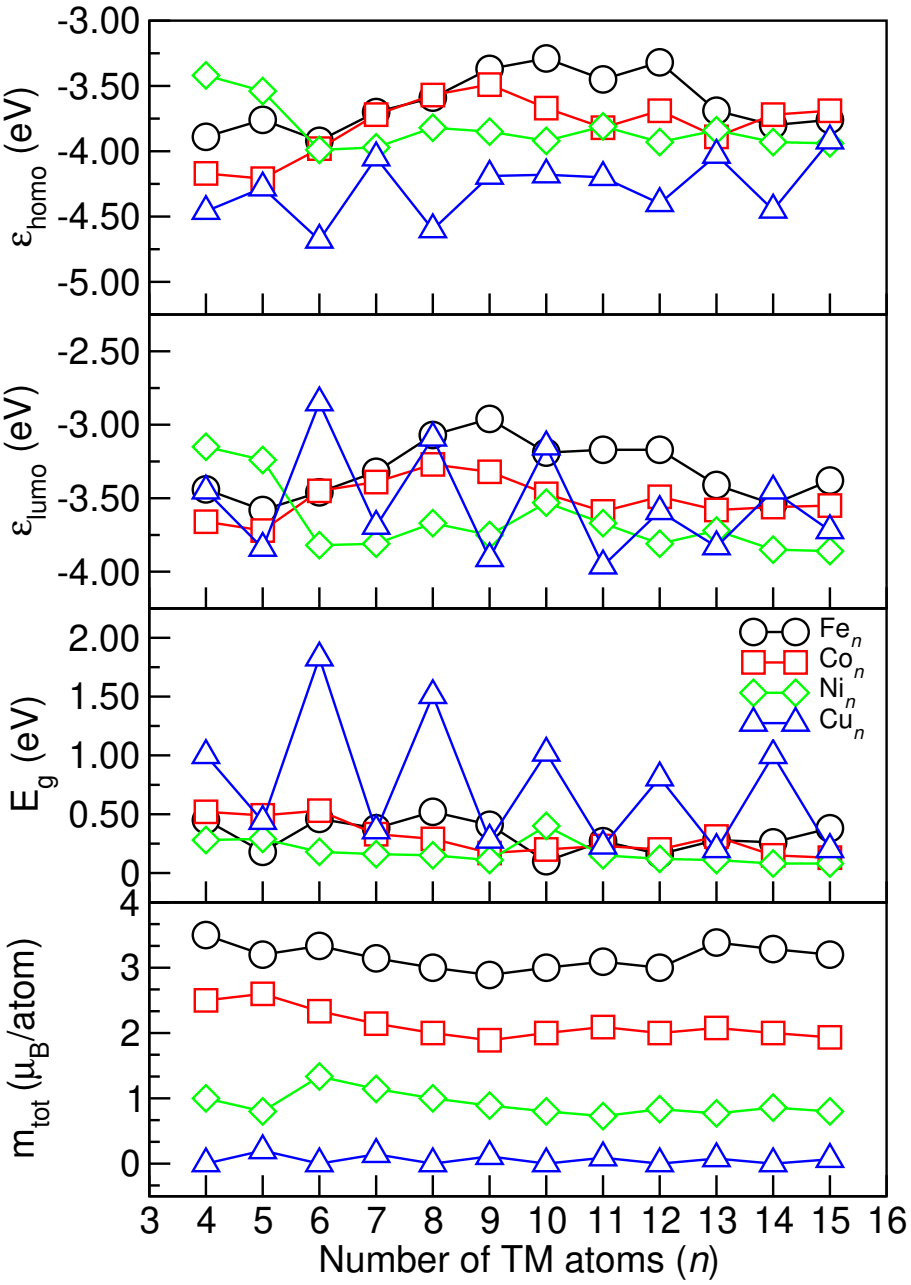

Figure S5: Energetic and electronic properties: HOMO energy $\varepsilon_{\text{homo}}$, LUMO energy $\varepsilon_{\text{lumo}}$, LUMO-HOMO energy band gap E_g, and total magnetic moment per atom m_{tot} for the pGMCs of TM_n clusters from $n = 4 - 15$ where TM = Fe, Co, Ni, Cu.

- The magnitude of HOMO states (ε_{homo}) increases with the gradual occupation of the d-states, which is evinced from $n = 6$, where the larger magnitude is observed for Cu_n, followed by Ni_n, Co_n and Fe_n. For $n = 4$ and $n = 5$ a different trend is observed where Ni present the lowest magnitude of ε_{homo}. For Cu_n the zig-zag pattern is also observed where for even clusters ε_{homo} is greater in comparison with the odd neighbours clusters. Although, from Cu_9 to Cu_{11} is almost the same.

- The same behaviour of ε_{homo} is observed for the LUMO (ε_{lumo}). Thus, for odd Cu_n clusters ε_{homo} are in higher magnitude in comparison with the even clusters, as expected.

- In conjunction, both HOMO and LUMO drives the magnitude of the energy gap, E_g, which reveals that in general Ni_n clusters are more reactive in comparison with Co_n and Fe_{n0}, with few exceptions $n = 5$ (Co_5) and $n = 10$ (Co_{10}). Thus, the reactivity of Fe_n and Co_n should be carefully examined, since there is not a plain trend. Furthermore, as expected the odd Cu_n clusters present lower values of E_g, and hence, are more reactivity, than the stable even Cu_n.

- As expected, m_{tot} increases with n for Fe_n, Co_n and Ni_n clusters, where the slope is less pronounced from the first towards the latter one. In addition, the even Cu_n present a zero total magnetic moment in reason of the complete filled d-states, while for odd Cu_n clusters m_{tot}=1.

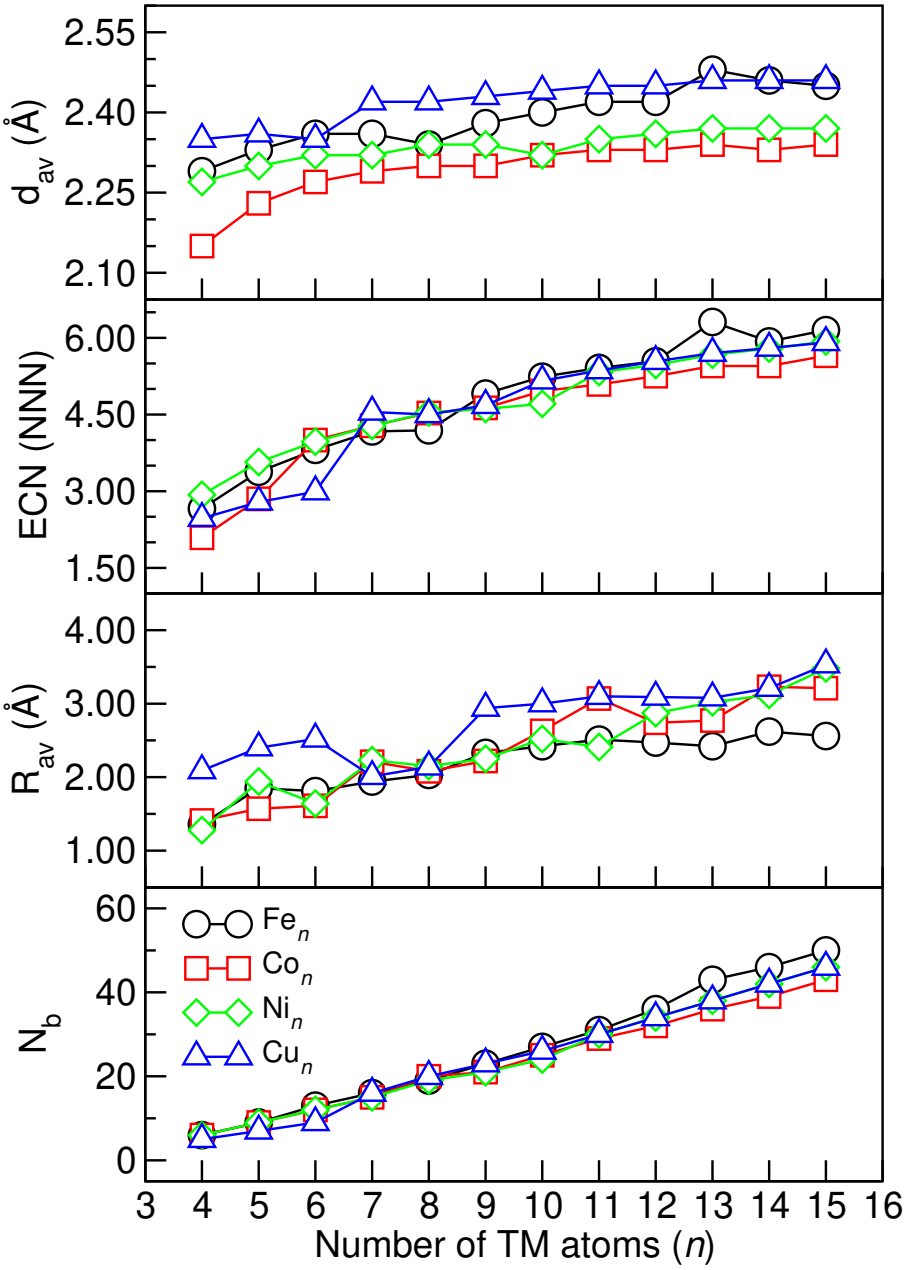

Figure S6: Structural properties: average effective coordination number ECN_{av}, average weighted bond lengths d_{av}, average radius R_{av}, and total number of bonds N_b for the pGMCs of TM_n clusters from $n = 4 - 15$ where TM = Fe, Co, Ni, Cu.

The d_{av} results follow an almost linear increasing with n, with the largest variation for Co_n, while the largest magnitude is observed for Cu_n, which evinces the more compact structures for Fe_n, Co_n and Ni_n. The ECN also increases with the number of transition atoms, which in conjunction with d_{av} explain the increase the average radius, which is related as well to the linear increasing of N_b.

7 CH_4 Adsorption on TM_n Clusters

Table S14: Structural, energetic, and electronic properties for the CH_4/Fe_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	2	−0.35	14	−3.66	−3.28	0.38	2.00	2.36	109.48	9.65	1.28
	1	−0.35	14	−3.66	−3.28	0.38	2.00	2.37	109.48	9.67	1.29
	1	−0.35	14	−3.66	−3.28	0.37	1.99	2.36	109.48	9.67	1.29
	1	−0.35	14	−3.66	−3.28	0.38	2.00	2.37	109.48	9.62	1.28
	1	−0.35	14	−3.66	−3.28	0.38	2.00	2.37	109.48	9.62	1.29
	1	−0.35	14	−3.66	−3.29	0.37	1.99	2.36	109.48	9.68	1.29
	0	−0.35	14	−3.66	−3.28	0.38	2.00	2.36	109.48	9.67	1.29
	0	−0.35	14	−3.66	−3.28	0.37	1.99	2.36	109.48	9.69	1.30
5	368	−0.09	16	−3.61	−3.40	0.21	2.38	2.80	109.47	−0.67	−0.03
	359	−0.10	16	−3.60	−3.24	0.37	2.05	2.39	109.46	−2.44	−0.20
	14	−0.44	16	−3.60	−3.10	0.50	2.00	2.36	109.48	0.19	−0.04
	1	−0.45	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.16	−0.03
	1	−0.45	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.16	−0.03
	1	−0.45	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.18	−0.02
	1	−0.45	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.18	−0.02
	1	−0.45	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.19	−0.02
	0	−0.46	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.19	−0.02
	0	−0.46	16	−3.60	−3.10	0.51	1.99	2.35	109.48	0.16	−0.03
6	333	0.22	18	−3.47	−3.33	0.14	1.91	2.25	109.46	−0.87	−1.11
	109	0.22	18	−3.33	−3.00	0.33	1.97	2.33	109.48	−2.81	−1.18
	109	−0.00	18	−3.33	−3.00	0.33	1.97	2.33	109.48	−2.80	−1.18
	5	−0.00	20	−3.94	−3.47	0.47	3.55	3.89	109.47	0.00	−0.02
	5	−0.11	20	−3.94	−3.47	0.47	3.54	3.80	109.47	−0.01	−0.02
	5	−0.11	20	−3.81	−3.33	0.47	2.36	2.81	109.48	−0.11	0.00
	2	−0.11	20	−3.81	−3.33	0.48	2.35	2.79	109.48	−0.12	0.00
	2	−0.11	20	−3.80	−3.33	0.47	2.33	2.78	109.48	−0.11	0.00
	1	−0.11	20	−3.81	−3.33	0.47	2.35	2.79	109.48	−0.11	0.01
	0	−0.11	20	−3.94	−3.47	0.46	3.62	4.05	109.47	0.01	−0.02
	0	−0.11	20	−3.81	−3.33	0.48	2.34	2.79	109.48	−0.10	0.00
7	226	0.072	20	−3.24	−3.05	0.19	1.96	2.31	109.49	2.91	−0.30
	226	0.072	20	−3.24	−3.05	0.19	1.96	2.31	109.48	2.90	−0.30
	39	−0.12	22	−3.70	−3.32	0.38	3.59	3.84	109.47	0.11	0.00
	28	−0.13	22	−3.70	−3.32	0.38	3.58	4.02	109.47	−0.02	−0.02
	6	−0.15	22	−3.54	−3.17	0.38	2.14	2.57	109.48	0.96	0.18
	4	−0.15	22	−3.53	−3.16	0.37	2.21	2.64	109.48	0.45	0.10
	4	−0.15	22	−3.53	−3.16	0.37	2.23	2.64	109.48	0.27	0.08

Table S14 continued from previous page

	2	−0.15	22	−3.52	−3.15	0.37	2.15	2.57	109.48	0.52	0.14
	1	−0.15	22	−3.53	−3.16	0.37	2.16	2.58	109.48	0.54	0.12
	1	−0.15	22	−3.52	−3.15	0.37	2.16	2.58	109.48	0.56	0.14
	1	−0.15	22	−3.54	−3.17	0.37	2.23	2.65	109.48	0.44	0.08
	1	−0.15	22	−3.53	−3.16	0.37	2.16	2.58	109.48	0.54	0.12
	0	−0.15	22	−3.53	−3.16	0.37	2.15	2.57	109.48	0.57	0.13
	0	−0.15	22	−3.53	−3.16	0.37	2.14	2.56	109.48	0.56	0.13
8	36	−0.12	24	−3.58	−3.06	0.52	3.54	3.94	109.47	0.06	0.00
	26	−0.14	24	−3.59	−3.07	0.53	3.52	3.97	109.47	−0.03	−0.02
	24	−0.14	24	−3.51	−3.00	0.51	2.41	2.86	109.47	0.18	0.05
	24	−0.14	24	−3.51	−3.00	0.51	2.41	2.86	109.47	0.18	0.04
	24	−0.14	24	−3.51	−3.00	0.51	2.43	2.88	109.47	0.13	0.04
	21	−0.14	24	−3.44	−2.94	0.50	2.23	2.65	109.48	0.47	0.09
	21	−0.14	24	−3.43	−2.94	0.50	2.22	2.64	109.48	0.49	0.10
	21	−0.14	24	−3.44	−2.94	0.50	2.23	2.66	109.48	0.48	0.10
	21	−0.14	24	−3.44	−2.94	0.50	2.26	2.68	109.48	0.42	0.09
	20	−0.14	24	−3.43	−2.94	0.50	2.22	2.64	109.48	0.47	0.10
	20	−0.14	24	−3.43	−2.93	0.50	2.21	2.63	109.48	0.50	0.11
	5	−0.16	24	−3.47	−2.96	0.51	2.28	2.71	109.48	0.12	0.04
	4	−0.16	24	−3.46	−2.95	0.51	2.25	2.67	109.48	0.13	0.04
	3	−0.16	24	−3.46	−2.95	0.51	2.26	2.68	109.48	0.12	0.04
	3	−0.16	24	−3.47	−2.96	0.51	2.27	2.70	109.48	0.11	0.04
	0	−0.16	24	−3.42	−2.91	0.51	2.13	2.53	109.48	0.34	0.13
9	78	−0.15	26	−3.38	−2.98	0.40	3.04	3.57	109.47	0.08	0.00
	78	−0.15	26	−3.38	−2.98	0.40	3.08	3.58	109.47	0.10	0.01
	77	−0.15	26	−3.38	−2.98	0.40	2.93	3.53	109.47	0.10	0.00
	46	−0.18	26	−3.24	−2.87	0.37	2.16	2.57	109.48	0.22	0.07
	44	−0.18	26	−3.24	−2.87	0.37	2.15	2.56	109.48	0.21	0.06
	18	−0.21	26	−3.25	−2.88	0.37	2.14	2.57	109.48	0.04	0.08
	17	−0.21	26	−3.25	−2.88	0.37	2.13	2.56	109.48	0.03	0.09
	17	−0.21	26	−3.25	−2.88	0.37	2.13	2.56	109.48	0.03	0.09
	16	−0.21	26	−3.25	−2.88	0.37	2.14	2.57	109.48	0.05	0.09
	16	−0.21	26	−3.25	−2.88	0.37	2.13	2.56	109.48	0.03	0.09
	16	−0.21	26	−3.25	−2.88	0.37	2.14	2.57	109.48	0.04	0.09
	15	−0.21	26	−3.21	−2.77	0.45	2.06	2.44	109.48	−0.12	0.05
	14	−0.21	26	−3.21	−2.77	0.45	2.06	2.44	109.48	−0.10	0.06
	2	−0.23	26	−3.22	−2.76	0.46	2.01	2.42	109.48	−0.17	0.03
	1	−0.23	26	−3.22	−2.76	0.46	2.01	2.42	109.48	−0.20	0.03
	1	−0.23	26	−3.22	−2.76	0.46	2.01	2.42	109.48	−0.16	0.03
	1	−0.23	26	−3.22	−2.76	0.46	2.01	2.42	109.48	−0.15	0.04
	0	−0.23	26	−3.22	−2.76	0.46	2.02	2.42	109.48	−0.20	0.03
10	168	−0.12	32	−3.48	−3.27	0.21	2.48	3.18	109.47	−0.95	0.57
	168	−0.13	32	−3.48	−3.29	0.19	2.58	3.26	109.47	−0.88	0.57
	168	−0.13	32	−3.48	−3.31	0.18	2.61	3.31	109.47	−0.85	0.58
	167	−0.13	32	−3.48	−3.30	0.19	2.59	3.27	109.47	−0.88	0.58
	133	−0.16	30	−3.32	−3.21	0.11	3.59	3.87	109.47	0.06	0.00
	68	−0.23	30	−3.21	−3.09	0.12	2.16	2.56	109.48	0.18	0.09
	67	−0.23	30	−3.21	−3.09	0.12	2.16	2.56	109.48	0.18	0.09
	67	−0.23	30	−3.20	−3.09	0.12	2.09	2.53	109.48	0.15	0.09

Table S14 continued from previous page

	48	−0.25	30	−3.19	−2.94	0.25	2.05	2.43	109.48	0.06	0.05
	48	−0.25	30	−3.19	−2.94	0.25	2.05	2.43	109.48	0.06	0.05
	41	−0.26	30	−3.18	−2.93	0.26	2.02	2.42	109.48	−0.01	0.03
	41	−0.26	30	−3.18	−2.93	0.25	2.02	2.41	109.48	0.00	0.04
	41	−0.26	30	−3.18	−2.93	0.26	2.02	2.41	109.48	−0.01	0.03
	40	−0.26	30	−3.19	−2.93	0.26	2.02	2.42	109.48	0.00	0.03
	40	−0.26	30	−3.19	−2.93	0.26	2.02	2.42	109.48	0.00	0.04
	40	−0.26	30	−3.19	−2.93	0.26	2.02	2.41	109.48	0.00	0.03
	40	−0.26	30	−3.18	−2.93	0.25	2.02	2.41	109.48	−0.01	0.03
	2	−0.30	30	−3.19	−2.94	0.26	2.05	2.43	109.48	0.18	0.08
	0	−0.30	30	−3.19	−2.93	0.26	2.05	2.43	109.48	0.17	0.08
	0	−0.30	30	−3.19	−2.94	0.26	2.06	2.44	109.48	0.17	0.08
11	151	−0.14	32	−3.12	−3.03	0.09	2.03	2.41	109.48	−0.85	−0.70
	107	−0.18	34	−3.24	−3.09	0.14	2.08	2.48	109.48	−0.17	−0.02
	103	−0.18	34	−3.46	−3.18	0.27	3.51	4.01	109.47	0.00	−0.02
	96	−0.19	34	−3.24	−3.10	0.15	2.08	2.46	109.48	−0.18	−0.01
	95	−0.19	34	−3.24	−3.09	0.15	2.08	2.46	109.48	−0.17	0.00
	95	−0.19	34	−3.24	−3.09	0.15	2.07	2.45	109.48	−0.20	−0.01
	94	−0.19	34	−3.24	−3.10	0.15	2.07	2.45	109.48	−0.20	−0.01
	54	−0.23	34	−3.28	−2.99	0.29	2.03	2.41	109.48	0.04	0.06
	53	−0.23	34	−3.28	−2.99	0.29	2.04	2.41	109.48	0.04	0.06
	53	−0.23	34	−3.28	−2.99	0.29	2.04	2.41	109.48	0.04	0.06
	47	−0.24	34	−3.35	−3.09	0.26	2.11	2.52	109.48	0.31	0.10
	47	−0.24	34	−3.35	−3.09	0.26	2.11	2.52	109.48	0.32	0.10
	47	−0.24	34	−3.35	−3.09	0.26	2.13	2.53	109.48	0.32	0.10
	47	−0.24	34	−3.35	−3.09	0.26	2.12	2.53	109.48	0.30	0.10
	46	−0.24	34	−3.35	−3.09	0.26	2.12	2.53	109.48	0.32	0.10
	46	−0.24	34	−3.35	−3.09	0.26	2.11	2.53	109.48	0.32	0.10
	46	−0.24	34	−3.35	−3.09	0.26	2.12	2.53	109.48	0.31	0.10
	3	−0.28	34	−3.29	−3.02	0.28	2.07	2.45	109.48	0.25	0.12
	3	−0.28	34	−3.29	−3.02	0.28	2.07	2.45	109.48	0.24	0.12
	1	−0.28	34	−3.28	−3.01	0.27	2.03	2.43	109.48	0.28	0.13
	0	−0.28	34	−3.28	−3.01	0.27	2.03	2.42	109.48	0.28	0.13
	0	−0.29	34	−3.28	−3.01	0.27	2.03	2.42	109.48	0.28	0.12
12	207	−0.14	36	−3.31	−3.17	0.14	3.44	3.84	109.47	0.02	−0.01
	155	−0.20	36	−3.32	−3.18	0.13	3.49	3.90	109.47	0.07	−0.01
	121	−0.23	36	−3.19	−3.10	0.09	2.13	2.53	109.48	0.76	0.15
	120	−0.23	36	−3.19	−3.10	0.09	2.12	2.53	109.48	0.77	0.15
	78	−0.27	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.46	0.09
	78	−0.27	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.44	0.08
	77	−0.27	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.43	0.08
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.46	0.09
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.40	0.08
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.46	0.09
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.44	0.08
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.42	0.08
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.43	0.08
	77	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.40	0.08
	76	−0.28	36	−3.22	−3.06	0.16	2.02	2.40	109.48	0.43	0.08

Table S14 continued from previous page

	53	−0.30	36	−3.21	−3.08	0.14	2.04	2.44	109.48	0.42	0.10
	52	−0.30	36	−3.22	−3.08	0.14	2.03	2.43	109.48	0.37	0.10
	47	−0.30	36	−3.22	−3.08	0.14	2.05	2.43	109.48	0.35	0.09
	46	−0.31	36	−3.22	−3.08	0.14	2.04	2.42	109.48	0.37	0.09
	46	−0.31	36	−3.22	−3.08	0.14	2.05	2.42	109.48	0.35	0.09
	45	−0.31	36	−3.22	−3.08	0.14	2.05	2.43	109.48	0.33	0.09
	45	−0.31	36	−3.22	−3.08	0.14	2.05	2.43	109.48	0.33	0.09
	45	−0.31	36	−3.22	−3.08	0.14	2.05	2.43	109.48	0.34	0.09
	0	−0.35	38	−3.32	−3.13	0.18	2.05	2.43	109.48	2.22	0.86
13	19	−0.26	42	−3.44	−3.25	0.19	2.02	2.38	109.48	−1.89	−0.68
	18	−0.26	42	−3.44	−3.25	0.19	2.01	2.38	109.48	−1.90	−0.69
	17	−0.26	42	−3.45	−3.26	0.19	2.01	2.38	109.48	−1.94	−0.69
	16	−0.26	42	−3.45	−3.25	0.19	2.02	2.38	109.48	−1.96	−0.69
	16	−0.26	42	−3.45	−3.25	0.20	2.02	2.38	109.48	−1.98	−0.70
	15	−0.26	42	−3.45	−3.26	0.19	2.01	2.38	109.48	−1.97	−0.69
	9	−0.27	44	−3.52	−3.34	0.19	2.06	2.43	109.48	−0.75	−0.10
	7	−0.27	44	−3.53	−3.33	0.19	2.06	2.44	109.48	−0.71	−0.08
	7	−0.27	44	−3.52	−3.33	0.19	2.06	2.43	109.48	−0.73	−0.09
	6	−0.27	44	−3.53	−3.33	0.19	2.06	2.43	109.48	−0.73	−0.09
	6	−0.27	44	−3.52	−3.33	0.19	2.06	2.43	109.48	−0.73	−0.09
	5	−0.27	44	−3.52	−3.34	0.19	2.06	2.43	109.48	−0.73	−0.09
	5	−0.27	44	−3.53	−3.33	0.19	2.06	2.44	109.48	−0.72	−0.09
	5	−0.27	44	−3.53	−3.33	0.19	2.06	2.43	109.48	−0.72	−0.09
	5	−0.27	44	−3.53	−3.34	0.19	2.06	2.44	109.48	−0.71	−0.08
	4	−0.27	44	−3.52	−3.33	0.19	2.06	2.43	109.48	−0.72	−0.09
	3	−0.28	44	−3.52	−3.34	0.19	2.06	2.44	109.48	−0.71	−0.08
	3	−0.28	44	−3.53	−3.34	0.19	2.06	2.43	109.48	−0.72	−0.08
	3	−0.28	44	−3.53	−3.34	0.19	2.06	2.43	109.48	−0.73	−0.08
	3	−0.28	44	−3.52	−3.33	0.19	2.06	2.43	109.48	−0.71	−0.08
	2	−0.28	44	−3.52	−3.33	0.19	2.06	2.43	109.48	−0.72	−0.08
	2	−0.28	44	−3.52	−3.34	0.19	2.06	2.43	109.48	−0.72	−0.08
	2	−0.28	44	−3.53	−3.34	0.19	2.06	2.43	109.48	−0.73	−0.08
	2	−0.28	44	−3.53	−3.34	0.19	2.06	2.43	109.48	−0.72	−0.08
	1	−0.28	44	−3.52	−3.34	0.19	2.06	2.43	109.48	−0.71	−0.08
	0	−0.28	44	−3.53	−3.33	0.19	2.06	2.43	109.48	−0.73	−0.08
14	201	−0.12	46	−3.73	−3.46	0.27	2.41	2.86	109.47	0.10	0.02
	200	−0.12	46	−3.73	−3.46	0.27	2.42	2.87	109.47	0.10	0.02
	188	−0.13	46	−3.72	−3.46	0.26	2.44	2.89	109.47	0.07	0.01
	164	−0.16	44	−3.56	−3.29	0.27	1.96	2.32	109.49	−1.17	−0.64
	121	−0.20	44	−3.55	−3.24	0.32	2.01	2.38	109.48	−0.87	−0.57
	90	−0.23	44	−3.55	−3.21	0.34	1.99	2.34	109.48	−0.61	−0.53
	88	−0.23	44	−3.56	−3.21	0.34	1.99	2.34	109.48	−0.62	−0.52
	73	−0.25	46	−3.60	−3.43	0.17	2.08	2.47	109.48	−0.09	0.00
	73	−0.25	46	−3.60	−3.43	0.16	2.07	2.47	109.48	−0.09	0.00
	71	−0.25	44	−3.53	−3.36	0.17	2.01	2.39	109.48	−0.84	−0.49
	66	−0.26	44	−3.52	−3.38	0.15	2.04	2.41	109.48	−0.43	−0.46
	64	−0.26	44	−3.52	−3.38	0.14	2.04	2.41	109.48	−0.42	−0.46
	62	−0.26	44	−3.52	−3.38	0.15	2.04	2.41	109.48	−0.44	−0.46
	57	−0.26	46	−3.57	−3.44	0.13	2.07	2.42	109.47	−0.22	−0.04

Table S14 continued from previous page

	56	−0.26	46	−3.57	−3.44	0.13	2.07	2.42	109.47	−0.20	−0.04
	56	−0.27	46	−3.57	−3.44	0.13	2.06	2.41	109.47	−0.21	−0.04
	55	−0.27	46	−3.57	−3.44	0.13	2.07	2.42	109.47	−0.20	−0.04
	54	−0.27	46	−3.57	−3.44	0.13	2.07	2.42	109.47	−0.20	−0.04
	53	−0.27	46	−3.57	−3.44	0.13	2.07	2.43	109.47	−0.21	−0.04
	53	−0.27	46	−3.57	−3.44	0.13	2.06	2.42	109.47	−0.22	−0.04
	53	−0.27	46	−3.57	−3.44	0.13	2.07	2.43	109.47	−0.21	−0.04
	20	−0.30	44	−3.56	−3.29	0.27	1.99	2.36	109.48	−0.30	−0.51
	18	−0.30	44	−3.56	−3.29	0.27	1.99	2.36	109.48	−0.30	−0.51
	18	−0.30	44	−3.56	−3.29	0.27	1.99	2.36	109.48	−0.30	−0.51
	4	−0.32	44	−3.55	−3.35	0.19	2.02	2.38	109.48	−0.46	−0.50
	3	−0.32	44	−3.55	−3.35	0.19	2.02	2.38	109.48	−0.46	−0.49
	2	−0.32	44	−3.55	−3.35	0.19	2.02	2.38	109.48	−0.46	−0.49
	0	−0.32	44	−3.54	−3.35	0.19	2.02	2.38	109.48	−0.47	−0.49
15	166	−0.14	48	−3.66	−3.27	0.38	2.30	2.76	109.48	0.12	0.02
	154	−0.16	48	−3.65	−3.24	0.41	2.22	2.66	109.48	−0.03	0.00
	153	−0.16	48	−3.65	−3.24	0.41	2.22	2.66	109.48	−0.05	0.00
	153	−0.16	48	−3.65	−3.24	0.41	2.22	2.66	109.48	−0.02	0.01
	153	−0.16	48	−3.65	−3.24	0.41	2.21	2.64	109.48	−0.04	0.00
	153	−0.16	48	−3.65	−3.24	0.41	2.22	2.66	109.48	−0.05	0.00
	152	−0.16	48	−3.65	−3.24	0.41	2.23	2.66	109.48	−0.03	0.00
	152	−0.16	48	−3.65	−3.24	0.41	2.22	2.66	109.48	−0.03	0.00
	150	−0.16	48	−3.65	−3.24	0.41	2.21	2.65	109.48	−0.04	0.00
	149	−0.16	48	−3.65	−3.24	0.41	2.22	2.66	109.48	−0.03	0.00
	4	−0.30	48	−3.68	−3.24	0.44	2.07	2.45	109.48	−0.06	0.00
	3	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.06	0.01
	3	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.05	0.01
	2	−0.31	48	−3.68	−3.24	0.44	2.07	2.45	109.48	−0.06	0.01
	2	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.06	0.01
	2	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.07	0.00
	2	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.05	0.01
	2	−0.31	48	−3.69	−3.24	0.44	2.08	2.46	109.48	−0.09	0.00
	1	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.06	0.01
	1	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.07	0.01
	1	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.06	0.00
	1	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.05	0.01
	1	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.06	0.01
	1	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.07	0.00
	1	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.08	0.00
	1	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.06	0.01
	1	−0.31	48	−3.68	−3.24	0.44	2.08	2.46	109.48	−0.06	0.01
	0	−0.31	48	−3.68	−3.24	0.44	2.07	2.45	109.48	−0.07	0.00
	0	−0.31	48	−3.68	−3.24	0.44	2.07	2.45	109.48	−0.07	0.00
	0	−0.31	48	−3.68	−3.24	0.44	2.07	2.46	109.48	−0.06	0.01

Table S15: Structural, energetic, and electronic properties for the CH_4/Co_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	104	−0.16	10	−3.74	−3.52	0.21	2.06	2.43	109.47	4.72	0.76
	104	−0.16	10	−3.74	−3.52	0.22	2.04	2.43	109.47	4.53	0.73
	103	−0.16	10	−3.74	−3.52	0.22	2.05	2.44	109.47	4.50	0.73
	103	−0.16	10	−3.73	−3.52	0.21	2.06	2.43	109.47	4.83	0.77
	103	−0.16	10	−3.74	−3.52	0.21	2.06	2.43	109.47	4.72	0.76
	1	−0.26	8	−3.21	−2.90	0.31	1.77	2.11	109.46	13.27	1.42
	0	−0.26	8	−3.21	−2.90	0.31	1.77	2.11	109.46	13.21	1.41
	0	−0.26	8	−3.21	−2.90	0.31	1.77	2.11	109.46	13.23	1.42
5	305	−0.10	13	−4.06	−3.59	0.47	2.42	2.87	109.47	1.39	0.22
	115	−0.29	11	−3.44	−3.15	0.29	1.88	2.20	109.45	11.65	0.94
	2	−0.41	11	−3.53	−3.21	0.32	1.84	2.12	109.41	8.25	0.49
	1	−0.41	11	−3.53	−3.21	0.32	1.84	2.12	109.41	8.26	0.49
	1	−0.41	11	−3.53	−3.21	0.32	1.84	2.12	109.41	8.24	0.49
	1	−0.41	11	−3.53	−3.21	0.32	1.84	2.12	109.41	8.26	0.49
	0	−0.41	11	−3.53	−3.21	0.32	1.84	2.11	109.41	8.27	0.50
	0	−0.41	11	−3.53	−3.21	0.32	1.84	2.12	109.41	8.28	0.49
	0	−0.41	11	−3.53	−3.21	0.32	1.85	2.12	109.41	8.24	0.49
	0	−0.41	11	−3.53	−3.21	0.32	1.84	2.12	109.41	8.29	0.50
6	2	−0.16	14	−3.83	−3.27	0.56	2.15	2.56	109.48	0.03	0.09
	1	−0.16	14	−3.83	−3.27	0.56	2.16	2.57	109.48	0.03	0.08
	1	−0.16	14	−3.82	−3.27	0.56	2.14	2.55	109.48	0.03	0.09
	1	−0.16	14	−3.83	−3.27	0.56	2.16	2.57	109.48	0.03	0.08
	1	−0.16	14	−3.83	−3.27	0.56	2.16	2.57	109.48	0.03	0.08
	1	−0.16	14	−3.83	−3.27	0.56	2.15	2.56	109.48	0.03	0.09
	0	−0.16	14	−3.83	−3.27	0.56	2.16	2.57	109.48	0.03	0.08
	0	−0.16	14	−3.83	−3.27	0.56	2.15	2.56	109.48	0.03	0.09
	0	−0.16	14	−3.83	−3.27	0.56	2.17	2.58	109.48	0.03	0.08
	0	−0.16	14	−3.83	−3.27	0.56	2.15	2.56	109.48	0.03	0.08
	0	−0.16	14	−3.83	−3.27	0.56	2.16	2.57	109.48	0.03	0.08
	0	−0.16	14	−3.83	−3.27	0.56	2.15	2.55	109.48	0.03	0.08
7	139	−0.17	15	−3.62	−3.28	0.34	2.19	2.61	109.48	−0.07	0.11
	138	−0.17	15	−3.63	−3.28	0.35	2.23	2.65	109.48	−0.21	0.04
	138	−0.17	15	−3.62	−3.27	0.35	2.19	2.60	109.48	−0.06	0.11
	137	−0.17	15	−3.63	−3.28	0.35	2.21	2.63	109.48	−0.21	0.05
	136	−0.17	15	−3.63	−3.28	0.35	2.21	2.63	109.48	−0.21	0.05
	135	−0.17	15	−3.63	−3.28	0.35	2.21	2.63	109.48	−0.21	0.05
	89	−0.22	15	−3.55	−3.21	0.34	2.05	2.43	109.48	−0.07	0.11
	88	−0.22	15	−3.56	−3.22	0.34	2.05	2.42	109.48	−0.06	0.11
	88	−0.22	15	−3.56	−3.21	0.34	2.05	2.42	109.48	−0.04	0.11

Table S15 continued from previous page

	87	−0.22	15	−3.56	−3.21	0.35	2.05	2.43	109.48	−0.04	0.12
	86	−0.22	15	−3.56	−3.21	0.35	2.05	2.43	109.48	−0.05	0.11
	2	−0.30	15	−3.58	−3.23	0.35	1.93	2.31	109.47	−0.11	0.06
	1	−0.30	15	−3.58	−3.23	0.35	1.93	2.31	109.47	−0.11	0.06
	0	−0.30	15	−3.58	−3.23	0.35	1.93	2.30	109.47	−0.07	0.07
8	134	−0.17	16	−3.47	−3.14	0.33	2.27	2.70	109.48	−0.48	0.00
	129	−0.17	16	−3.44	−3.12	0.32	2.13	2.53	109.48	−1.22	−0.04
	129	−0.17	16	−3.44	−3.12	0.32	2.12	2.52	109.48	−1.22	−0.05
	128	−0.17	16	−3.44	−3.12	0.32	2.12	2.53	109.48	−1.22	−0.05
	128	−0.17	16	−3.44	−3.12	0.32	2.12	2.52	109.48	−1.22	−0.05
	128	−0.17	16	−3.44	−3.12	0.32	2.12	2.53	109.48	−1.22	−0.05
	128	−0.17	16	−3.44	−3.12	0.32	2.12	2.53	109.48	−1.21	−0.05
	128	−0.17	16	−3.44	−3.12	0.32	2.12	2.52	109.48	−1.22	−0.04
	2	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.33	0.02
	1	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.33	0.02
	1	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.32	0.02
	1	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.32	0.02
	1	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.31	0.02
	1	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.31	0.02
	0	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.31	0.02
	0	−0.30	16	−3.41	−3.11	0.30	1.95	2.32	109.47	−0.31	0.02
9	118	−0.22	17	−3.37	−3.19	0.19	2.10	2.51	109.48	0.06	0.10
	118	−0.22	17	−3.37	−3.19	0.19	2.10	2.51	109.48	0.06	0.10
	118	−0.22	17	−3.37	−3.19	0.19	2.10	2.51	109.48	0.06	0.10
	118	−0.22	17	−3.37	−3.19	0.19	2.10	2.51	109.48	0.06	0.10
	117	−0.22	17	−3.37	−3.19	0.19	2.10	2.51	109.48	0.06	0.10
	117	−0.22	17	−3.39	−3.18	0.21	2.08	2.49	109.48	−0.20	0.06
	117	−0.22	17	−3.39	−3.18	0.21	2.08	2.49	109.48	−0.20	0.06
	117	−0.22	17	−3.39	−3.18	0.21	2.08	2.49	109.48	−0.21	0.06
	117	−0.22	17	−3.39	−3.18	0.21	2.09	2.49	109.48	−0.21	0.06
	116	−0.22	17	−3.39	−3.18	0.21	2.08	2.49	109.48	−0.20	0.06
	2	−0.34	17	−3.34	−3.16	0.18	1.93	2.28	109.47	−0.23	0.03
	1	−0.34	17	−3.34	−3.15	0.18	1.93	2.28	109.47	−0.23	0.03
	1	−0.34	17	−3.34	−3.16	0.18	1.93	2.28	109.48	−0.23	0.03
	1	−0.34	17	−3.34	−3.16	0.18	1.93	2.28	109.47	−0.23	0.03
	1	−0.34	17	−3.34	−3.16	0.18	1.93	2.28	109.47	−0.24	0.03
	1	−0.34	17	−3.34	−3.16	0.18	1.94	2.28	109.47	−0.23	0.03
	0	−0.34	17	−3.34	−3.16	0.18	1.93	2.28	109.47	−0.24	0.03
	0	−0.34	17	−3.34	−3.16	0.18	1.93	2.28	109.47	−0.24	0.03
10	87	−0.15	20	−3.69	−3.48	0.21	3.52	3.93	109.47	−0.72	−0.01
	87	−0.15	20	−3.68	−3.48	0.21	3.50	3.93	109.47	−0.73	−0.01
	71	−0.27	20	−3.53	−3.31	0.22	1.98	2.35	109.48	−0.87	0.08
	70	−0.27	20	−3.52	−3.31	0.21	1.98	2.35	109.48	−0.85	0.08
	70	−0.27	20	−3.53	−3.31	0.22	1.98	2.35	109.48	−0.87	0.08
	70	−0.27	20	−3.53	−3.31	0.22	1.98	2.35	109.48	−0.87	0.08
	68	−0.27	20	−3.53	−3.31	0.22	1.98	2.35	109.48	−0.87	0.08
	67	−0.27	20	−3.53	−3.31	0.22	1.98	2.35	109.48	−0.88	0.08
	67	−0.27	20	−3.53	−3.31	0.21	1.98	2.34	109.48	−0.86	0.08
	67	−0.27	20	−3.53	−3.31	0.21	1.98	2.35	109.48	−0.87	0.08

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	2	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.83	0.03
	2	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.83	0.03
	2	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.82	0.03
	2	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.83	0.03
	1	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.83	0.03
	1	−0.34	20	−3.57	−3.35	0.22	2.00	2.37	109.48	−0.83	0.03
	1	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.83	0.03
	1	−0.34	20	−3.57	−3.35	0.22	1.99	2.37	109.48	−0.82	0.03
	0	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.82	0.03
	0	−0.34	20	−3.57	−3.35	0.22	1.99	2.36	109.48	−0.83	0.03
11	132	−0.15	23	−3.90	−3.68	0.22	3.49	3.86	109.47	−0.21	0.02
	131	−0.15	23	−3.61	−3.42	0.18	2.12	2.50	109.47	−0.15	0.05
	130	−0.15	23	−3.60	−3.42	0.18	2.11	2.50	109.48	−0.15	0.05
	88	−0.19	23	−3.68	−3.44	0.24	2.06	2.45	109.48	−0.12	0.06
	87	−0.19	21	−3.35	−3.21	0.14	1.91	2.24	109.45	0.10	−0.15
	42	−0.24	23	−3.80	−3.57	0.23	2.15	2.56	109.48	−0.30	0.12
	29	−0.25	23	−3.67	−3.44	0.23	2.02	2.38	109.47	−0.15	0.06
	29	−0.25	23	−3.67	−3.44	0.23	2.02	2.38	109.46	−0.16	0.05
	28	−0.25	23	−3.67	−3.44	0.23	2.02	2.38	109.46	−0.15	0.06
	28	−0.25	23	−3.67	−3.44	0.23	2.02	2.38	109.46	−0.16	0.05
	27	−0.25	23	−3.67	−3.44	0.23	2.02	2.37	109.46	−0.16	0.05
	11	−0.27	23	−3.69	−3.49	0.20	2.10	2.50	109.48	−0.17	0.07
	10	−0.27	23	−3.69	−3.49	0.20	2.10	2.50	109.48	−0.16	0.06
	10	−0.27	23	−3.69	−3.49	0.20	2.10	2.50	109.48	−0.17	0.06
	10	−0.27	23	−3.69	−3.49	0.20	2.10	2.50	109.48	−0.17	0.06
	2	−0.27	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
	1	−0.28	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
	1	−0.28	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
	0	−0.28	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
	0	−0.28	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
	0	−0.28	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
	0	−0.28	23	−3.69	−3.46	0.24	2.02	2.40	109.48	−0.14	−0.02
12	199	−0.18	24	−3.72	−3.52	0.20	3.16	3.67	109.47	0.06	0.00
	197	−0.18	24	−3.72	−3.51	0.20	3.39	3.81	109.47	0.04	0.00
	195	−0.18	24	−3.65	−3.45	0.20	2.42	2.96	109.47	−0.02	0.00
	195	−0.18	24	−3.65	−3.45	0.20	2.44	2.94	109.47	−0.02	0.00
	194	−0.18	24	−3.65	−3.44	0.20	2.43	2.91	109.47	−0.03	0.00
	194	−0.18	24	−3.65	−3.44	0.20	2.44	2.92	109.47	−0.03	0.00
	194	−0.18	24	−3.65	−3.45	0.20	2.46	2.95	109.47	−0.01	0.00
	159	−0.22	24	−3.58	−3.37	0.21	2.03	2.46	109.48	−0.17	0.04
	159	−0.22	24	−3.58	−3.37	0.21	2.03	2.46	109.48	−0.16	0.04
	158	−0.22	24	−3.58	−3.37	0.21	2.03	2.46	109.48	−0.15	0.04
	158	−0.22	24	−3.58	−3.37	0.21	2.03	2.46	109.48	−0.16	0.04
	158	−0.22	24	−3.58	−3.37	0.21	2.03	2.46	109.48	−0.17	0.04
	158	−0.22	24	−3.58	−3.37	0.21	2.03	2.46	109.48	−0.15	0.04
	157	−0.22	24	−3.58	−3.37	0.21	2.03	2.45	109.48	−0.16	0.04
	88	−0.29	24	−3.58	−3.36	0.21	1.99	2.36	109.48	−0.32	0.01
	83	−0.30	24	−3.58	−3.36	0.22	1.97	2.34	109.48	−0.36	0.00
	82	−0.30	24	−3.61	−3.42	0.18	2.01	2.41	109.48	0.43	0.18

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	82	−0.30	24	−3.58	−3.36	0.21	1.96	2.34	109.48	−0.38	0.00
	81	−0.30	24	−3.58	−3.36	0.21	1.97	2.34	109.48	−0.38	0.00
	81	−0.30	24	−3.58	−3.36	0.21	1.97	2.34	109.48	−0.37	0.00
	80	−0.30	24	−3.58	−3.36	0.22	1.96	2.34	109.48	−0.38	0.00
	19	−0.36	24	−3.60	−3.39	0.21	1.93	2.33	109.48	−0.15	0.03
	17	−0.36	24	−3.60	−3.39	0.21	1.93	2.33	109.48	−0.14	0.03
	0	−0.38	24	−3.59	−3.38	0.21	1.93	2.29	109.48	−0.30	0.01
13	51	−0.20	27	−3.85	−3.53	0.32	2.40	2.91	109.47	−0.04	0.01
	51	−0.20	27	−3.85	−3.53	0.32	2.39	2.88	109.47	−0.05	0.01
	51	−0.20	27	−3.85	−3.53	0.32	2.37	2.89	109.47	−0.05	0.01
	51	−0.20	27	−3.85	−3.53	0.32	2.37	2.88	109.47	−0.04	0.01
	51	−0.20	27	−3.85	−3.53	0.32	2.36	2.89	109.47	−0.05	0.01
	50	−0.20	27	−3.85	−3.53	0.32	2.36	2.86	109.47	−0.04	0.01
	50	−0.20	27	−3.85	−3.53	0.32	2.38	2.89	109.47	−0.05	0.01
	49	−0.20	27	−3.85	−3.53	0.32	2.37	2.88	109.47	−0.04	0.01
	49	−0.20	27	−3.85	−3.53	0.32	2.36	2.86	109.47	−0.04	0.01
	49	−0.20	27	−3.85	−3.53	0.32	2.38	2.87	109.47	−0.04	0.01
	17	−0.24	27	−3.69	−3.43	0.26	2.03	2.41	109.48	−0.02	0.05
	15	−0.24	27	−3.69	−3.43	0.26	2.03	2.41	109.48	−0.01	0.05
	15	−0.24	27	−3.69	−3.43	0.26	2.03	2.41	109.48	−0.02	0.05
	15	−0.24	27	−3.69	−3.43	0.26	2.03	2.41	109.48	−0.02	0.05
	14	−0.24	27	−3.69	−3.43	0.26	2.03	2.41	109.48	−0.02	0.05
	14	−0.24	27	−3.69	−3.43	0.26	2.03	2.41	109.48	−0.02	0.05
	3	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.10	0.04
	2	−0.25	27	−3.73	−3.48	0.26	2.01	2.43	109.48	−0.09	0.05
	2	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.05
	1	−0.25	27	−3.73	−3.48	0.26	2.02	2.43	109.48	−0.09	0.05
	1	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.05
	1	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.05
	1	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.05
	1	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.05
	0	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.04
	0	−0.25	27	−3.73	−3.48	0.25	2.02	2.43	109.48	−0.09	0.05
14	132	−0.19	28	−3.71	−3.55	0.17	2.64	3.37	109.47	−0.04	−0.02
	131	−0.19	28	−3.74	−3.58	0.15	3.43	3.84	109.47	0.01	−0.01
	131	−0.19	28	−3.71	−3.55	0.16	2.67	3.40	109.47	−0.01	−0.02
	117	−0.21	28	−3.67	−3.52	0.16	2.34	2.87	109.47	−0.08	0.00
	116	−0.21	28	−3.67	−3.52	0.16	2.36	2.88	109.47	−0.08	0.00
	98	−0.22	28	−3.58	−3.44	0.14	2.04	2.44	109.48	−0.03	0.03
	92	−0.23	28	−3.58	−3.44	0.15	2.03	2.42	109.48	−0.01	0.03
	91	−0.23	28	−3.60	−3.46	0.15	2.04	2.45	109.48	−0.14	0.03
	91	−0.23	28	−3.60	−3.45	0.15	2.04	2.45	109.48	−0.14	0.03
	91	−0.23	28	−3.61	−3.46	0.15	2.04	2.44	109.48	−0.14	0.03
	88	−0.23	28	−3.61	−3.46	0.15	2.02	2.44	109.48	−0.22	0.04
	87	−0.24	28	−3.61	−3.46	0.15	2.02	2.44	109.48	−0.23	0.03
	86	−0.24	28	−3.61	−3.46	0.15	2.02	2.44	109.48	−0.22	0.03
	75	−0.25	28	−3.59	−3.43	0.17	2.00	2.37	109.46	0.13	0.06
	73	−0.25	28	−3.59	−3.43	0.17	2.01	2.37	109.47	0.12	0.06
	70	−0.25	28	−3.59	−3.43	0.17	1.99	2.37	109.46	0.15	0.06

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	36	−0.29	28	−3.62	−3.48	0.15	2.06	2.45	109.48	−0.05	0.07
	28	−0.30	28	−3.65	−3.47	0.18	2.03	2.45	109.48	−0.01	0.12
	26	−0.30	28	−3.64	−3.47	0.18	2.03	2.45	109.48	−0.02	0.12
	26	−0.30	28	−3.65	−3.47	0.18	2.03	2.45	109.48	−0.01	0.13
	25	−0.30	28	−3.65	−3.47	0.18	2.03	2.46	109.48	−0.03	0.12
	3	−0.32	28	−3.58	−3.42	0.16	1.98	2.35	109.48	0.13	0.09
	3	−0.32	28	−3.58	−3.42	0.16	1.98	2.35	109.48	0.14	0.10
	3	−0.32	28	−3.58	−3.43	0.16	1.98	2.35	109.48	0.14	0.09
	2	−0.32	28	−3.58	−3.42	0.16	1.98	2.35	109.48	0.14	0.09
	1	−0.32	28	−3.58	−3.43	0.16	1.98	2.35	109.48	0.14	0.09
	1	−0.32	28	−3.58	−3.42	0.16	1.98	2.35	109.48	0.14	0.09
	0	−0.32	28	−3.58	−3.42	0.16	1.98	2.35	109.48	0.14	0.09
15	157	−0.20	29	−3.70	−3.57	0.14	2.98	3.67	109.47	0.01	0.00
	154	−0.21	29	−3.70	−3.56	0.14	2.92	3.62	109.47	0.01	0.00
	154	−0.21	29	−3.70	−3.56	0.14	2.94	3.64	109.47	0.01	0.00
	153	−0.21	29	−3.70	−3.56	0.14	2.92	3.61	109.47	0.00	−0.01
	153	−0.21	29	−3.70	−3.56	0.14	2.98	3.65	109.47	0.01	0.00
	150	−0.21	29	−3.56	−3.43	0.13	2.06	2.45	109.48	−0.33	0.00
	139	−0.22	29	−3.57	−3.43	0.14	1.97	2.41	109.48	−0.09	0.02
	136	−0.23	29	−3.57	−3.43	0.14	1.97	2.39	109.48	−0.08	0.02
	136	−0.23	29	−3.57	−3.43	0.14	1.97	2.40	109.48	−0.08	0.02
	135	−0.23	29	−3.57	−3.43	0.14	1.97	2.40	109.48	−0.09	0.02
	134	−0.23	29	−3.57	−3.43	0.14	1.97	2.40	109.48	−0.08	0.02
	133	−0.23	29	−3.57	−3.43	0.14	1.97	2.39	109.48	−0.08	0.02
	124	−0.24	29	−3.57	−3.43	0.14	2.02	2.44	109.48	−0.13	0.04
	123	−0.24	29	−3.57	−3.44	0.14	2.03	2.44	109.48	−0.14	0.04
	122	−0.24	29	−3.57	−3.44	0.14	2.02	2.44	109.48	−0.15	0.04
	105	−0.26	29	−3.57	−3.44	0.14	2.02	2.41	109.48	−0.05	0.04
	57	−0.30	29	−3.62	−3.46	0.16	2.01	2.48	109.48	−0.04	0.04
	55	−0.31	29	−3.62	−3.46	0.16	2.00	2.48	109.48	−0.02	0.04
	54	−0.31	29	−3.62	−3.46	0.16	2.01	2.47	109.48	−0.02	0.04
	53	−0.31	29	−3.62	−3.46	0.16	2.00	2.48	109.48	−0.02	0.04
	51	−0.31	29	−3.62	−3.45	0.17	1.98	2.43	109.48	−0.06	0.04
	51	−0.31	29	−3.62	−3.46	0.16	2.01	2.48	109.48	−0.02	0.04
	51	−0.31	29	−3.62	−3.45	0.17	1.98	2.43	109.48	−0.05	0.04
	51	−0.31	29	−3.62	−3.45	0.17	1.98	2.43	109.48	−0.05	0.04
	50	−0.31	29	−3.62	−3.45	0.17	1.98	2.42	109.48	−0.05	0.04
	49	−0.31	29	−3.62	−3.45	0.17	1.98	2.43	109.48	−0.05	0.04
	49	−0.31	29	−3.62	−3.45	0.17	1.98	2.42	109.48	−0.05	0.04
	4	−0.36	29	−3.54	−3.42	0.12	1.97	2.33	109.48	−0.04	0.03
	1	−0.36	29	−3.54	−3.42	0.13	1.95	2.32	109.48	−0.07	0.02
	0	−0.36	29	−3.54	−3.42	0.13	1.95	2.32	109.48	−0.05	0.02

Table S16: Structural, energetic, and electronic properties for the CH_4/Ni_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	2	−0.58	4	−3.23	−3.02	0.21	1.78	2.09	109.48	0.74	0.29
	2	−0.58	4	−3.23	−3.02	0.21	1.78	2.09	109.48	0.75	0.28
	2	−0.58	4	−3.23	−3.01	0.21	1.77	2.09	109.48	0.74	0.27
	2	−0.58	4	−3.23	−3.02	0.21	1.78	2.09	109.48	0.74	0.27
	1	−0.58	4	−3.23	−3.02	0.21	1.78	2.09	109.48	0.75	0.28
	1	−0.58	4	−3.23	−3.02	0.21	1.78	2.09	109.48	0.75	0.28
	0	−0.58	4	−3.23	−3.01	0.22	1.78	2.09	109.48	0.72	0.27
	0	−0.58	4	−3.23	−3.02	0.21	1.78	2.09	109.48	0.75	0.28
5	237	−0.34	6	−3.58	−3.51	0.07	1.77	2.15	109.48	4.99	0.70
	119	−0.46	6	−3.68	−3.44	0.24	1.89	2.23	109.47	−0.19	0.06
	119	−0.46	6	−3.68	−3.44	0.24	1.89	2.23	109.47	−0.20	0.07
	119	−0.46	6	−3.68	−3.44	0.24	1.89	2.23	109.47	−0.19	0.06
	118	−0.46	6	−3.68	−3.44	0.24	1.89	2.23	109.47	−0.19	0.07
	23	−0.56	4	−3.48	−3.22	0.26	1.76	2.07	109.49	11.82	1.30
	22	−0.56	4	−3.48	−3.22	0.26	1.76	2.07	109.49	11.82	1.29
	22	−0.56	4	−3.48	−3.22	0.26	1.76	2.07	109.48	11.83	1.29
	22	−0.56	4	−3.48	−3.22	0.26	1.76	2.07	109.49	11.82	1.29
	0	−0.58	4	−3.37	−3.16	0.21	1.78	2.09	109.48	11.40	1.12
6	133	−0.12	8	−3.96	−3.78	0.17	2.35	3.21	109.47	0.04	0.00
	132	−0.12	8	−3.94	−3.77	0.17	2.34	3.14	109.47	−0.04	−0.01
	85	−0.16	8	−3.87	−3.68	0.19	1.96	2.40	109.48	−0.04	0.17
	83	−0.17	8	−3.86	−3.68	0.19	1.98	2.40	109.48	−0.02	0.15
	83	−0.17	8	−3.87	−3.68	0.19	1.97	2.40	109.48	−0.06	0.16
	82	−0.17	8	−3.87	−3.68	0.19	1.96	2.39	109.48	−0.03	0.18
	3	−0.25	6	−3.65	−3.46	0.19	1.79	2.11	109.49	0.55	0.13
	2	−0.25	6	−3.65	−3.46	0.19	1.79	2.11	109.49	0.55	0.13
	2	−0.25	6	−3.65	−3.46	0.19	1.79	2.11	109.49	0.55	0.13
	2	−0.25	6	−3.65	−3.46	0.19	1.79	2.11	109.49	0.55	0.13
	1	−0.25	6	−3.65	−3.46	0.19	1.79	2.11	109.49	0.55	0.13
	0	−0.25	6	−3.66	−3.46	0.19	1.79	2.11	109.49	0.55	0.14
7	208	−0.12	8	−3.92	−3.75	0.17	2.32	3.14	109.47	−0.02	0.00
	147	−0.18	8	−3.80	−3.64	0.16	2.03	2.42	109.48	−0.18	0.13
	147	−0.18	8	−3.81	−3.64	0.16	2.03	2.41	109.48	−0.18	0.13
	147	−0.18	8	−3.81	−3.64	0.16	2.03	2.41	109.48	−0.17	0.12
	146	−0.18	8	−3.81	−3.64	0.16	2.03	2.42	109.48	−0.17	0.12
	126	−0.20	8	−3.83	−3.63	0.20	1.98	2.36	109.48	−0.33	0.08
	126	−0.20	8	−3.83	−3.63	0.19	1.98	2.36	109.48	−0.33	0.08
	122	−0.21	8	−3.87	−3.71	0.16	2.03	2.43	109.48	−0.06	0.13
	121	−0.21	8	−3.87	−3.71	0.16	2.03	2.43	109.48	−0.06	0.13

Table S16 continued from previous page

	121	−0.21	8	−3.87	−3.71	0.16	2.03	2.43	109.48	−0.06	0.13
	120	−0.21	8	−3.87	−3.71	0.16	2.03	2.43	109.48	−0.06	0.13
	117	−0.21	6	−3.64	−3.48	0.16	1.80	2.12	109.48	−0.45	0.24
	0	−0.33	8	−3.70	−3.53	0.17	1.80	2.14	109.48	−0.20	0.27
	0	−0.33	8	−3.70	−3.53	0.17	1.80	2.14	109.48	−0.20	0.27
8	223	−0.18	8	−3.74	−3.58	0.15	2.04	2.46	109.48	−0.58	0.10
	143	−0.26	8	−3.72	−3.54	0.18	1.98	2.36	109.48	−0.01	0.04
	142	−0.26	8	−3.72	−3.54	0.18	1.98	2.36	109.48	−0.02	0.04
	142	−0.26	8	−3.72	−3.54	0.18	1.98	2.36	109.48	0.01	0.04
	141	−0.27	8	−3.72	−3.54	0.18	1.98	2.36	109.48	−0.01	0.04
	2	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.43	0.13
	1	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.42	0.13
	1	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.43	0.14
	1	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.43	0.13
	1	−0.41	8	−3.70	−3.53	0.16	1.81	2.14	109.48	0.43	0.14
	1	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.42	0.13
	1	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.41	0.13
	1	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.43	0.13
	0	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.42	0.14
	0	−0.41	8	−3.70	−3.53	0.16	1.81	2.14	109.48	0.43	0.13
	0	−0.41	8	−3.69	−3.53	0.16	1.81	2.14	109.48	0.44	0.14
9	95	−0.28	8	−3.74	−3.62	0.13	1.94	2.35	109.48	−0.29	0.09
	95	−0.28	8	−3.74	−3.62	0.13	1.94	2.35	109.48	−0.28	0.08
	94	−0.28	8	−3.75	−3.62	0.13	1.95	2.36	109.48	−0.28	0.09
	94	−0.28	8	−3.75	−3.62	0.13	1.94	2.35	109.48	−0.28	0.08
	94	−0.28	8	−3.75	−3.62	0.13	1.94	2.35	109.48	−0.29	0.09
	94	−0.28	8	−3.74	−3.62	0.13	1.95	2.35	109.48	−0.29	0.08
	94	−0.28	8	−3.74	−3.62	0.13	1.94	2.35	109.48	−0.29	0.09
	94	−0.28	8	−3.74	−3.62	0.13	1.95	2.35	109.48	−0.29	0.08
	94	−0.28	8	−3.75	−3.62	0.13	1.94	2.35	109.48	−0.29	0.08
	93	−0.28	8	−3.74	−3.62	0.13	1.94	2.35	109.48	−0.28	0.08
	93	−0.28	8	−3.74	−3.62	0.13	1.94	2.35	109.48	−0.28	0.09
	93	−0.28	8	−3.75	−3.62	0.13	1.94	2.35	109.48	−0.28	0.09
	2	−0.37	8	−3.68	−3.56	0.12	1.83	2.18	109.49	0.41	0.14
	2	−0.37	8	−3.68	−3.56	0.12	1.83	2.17	109.49	0.40	0.14
	1	−0.37	8	−3.68	−3.56	0.12	1.83	2.17	109.49	0.42	0.14
	1	−0.37	8	−3.68	−3.56	0.12	1.83	2.17	109.49	0.42	0.14
	1	−0.37	8	−3.68	−3.56	0.12	1.83	2.17	109.49	0.40	0.14
	0	−0.37	8	−3.68	−3.57	0.12	1.83	2.18	109.49	0.39	0.14
10	112	−0.20	8	−3.95	−3.55	0.40	3.32	3.73	109.47	0.01	0.00
	112	−0.20	8	−3.95	−3.55	0.40	3.34	3.74	109.47	0.01	0.00
	111	−0.20	8	−3.95	−3.55	0.40	3.33	3.74	109.47	0.01	0.00
	111	−0.20	8	−3.95	−3.55	0.40	3.35	3.75	109.47	0.01	0.00
	111	−0.20	8	−3.95	−3.55	0.40	3.34	3.75	109.47	0.01	0.00
	111	−0.20	8	−3.95	−3.55	0.40	3.34	3.74	109.47	0.00	0.00
	111	−0.20	8	−3.95	−3.55	0.40	3.33	3.74	109.47	0.01	0.00
	2	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.39	0.12
	2	−0.31	8	−3.75	−3.37	0.38	1.90	2.28	109.48	0.39	0.12
	1	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.39	0.12

Table S16 continued from previous page

	1	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.38	0.12
	1	−0.31	8	−3.75	−3.37	0.38	1.90	2.28	109.48	0.39	0.12
	1	−0.31	8	−3.75	−3.37	0.38	1.90	2.28	109.48	0.39	0.12
	1	−0.31	8	−3.75	−3.37	0.38	1.90	2.28	109.48	0.39	0.12
	1	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.39	0.12
	1	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.39	0.12
	0	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.39	0.12
	0	−0.31	8	−3.75	−3.37	0.38	1.91	2.28	109.48	0.39	0.12
	0	−0.31	8	−3.75	−3.37	0.38	1.90	2.28	109.48	0.39	0.12
	0	−0.31	8	−3.75	−3.37	0.38	1.90	2.27	109.48	0.39	0.12
11	219	−0.18	8	−3.79	−3.65	0.14	2.39	3.19	109.47	−0.01	0.00
	219	−0.18	8	−3.79	−3.65	0.14	2.37	3.19	109.47	0.02	0.00
	60	−0.34	8	−3.68	−3.50	0.19	1.86	2.31	109.48	−0.20	0.15
	60	−0.34	8	−3.68	−3.50	0.19	1.86	2.31	109.48	−0.20	0.15
	60	−0.34	8	−3.68	−3.50	0.19	1.86	2.30	109.48	−0.20	0.15
	2	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	2	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	2	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.03	0.17
	2	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.03	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.03	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.03	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.03	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	1	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	0	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.18
	0	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.04	0.17
	0	−0.40	8	−3.61	−3.49	0.11	1.84	2.18	109.48	0.03	0.17
12	321	−0.18	10	−3.90	−3.79	0.11	2.32	3.17	109.47	0.09	0.00
	316	−0.18	8	−3.74	−3.61	0.12	2.49	3.27	109.47	0.71	−0.16
	316	−0.19	10	−3.90	−3.79	0.11	2.32	3.15	109.47	0.02	0.00
	202	−0.30	10	−3.75	−3.68	0.06	1.92	2.36	109.48	0.10	0.09
	202	−0.30	10	−3.75	−3.68	0.06	1.92	2.36	109.48	0.09	0.09
	201	−0.30	10	−3.75	−3.68	0.06	1.91	2.36	109.48	0.09	0.09
	174	−0.33	8	−3.58	−3.45	0.13	1.89	2.24	109.48	0.84	−0.03
	170	−0.33	10	−3.70	−3.59	0.11	1.91	2.26	109.48	0.63	0.12
	170	−0.33	10	−3.70	−3.59	0.11	1.91	2.26	109.48	0.64	0.12
	133	−0.37	8	−3.61	−3.48	0.13	1.86	2.28	109.48	0.84	−0.03
	133	−0.37	8	−3.61	−3.48	0.13	1.85	2.27	109.48	0.84	−0.03
	125	−0.38	10	−3.72	−3.63	0.09	1.88	2.24	109.48	0.54	0.17
	123	−0.38	10	−3.72	−3.64	0.08	1.88	2.24	109.48	0.53	0.17
	122	−0.38	10	−3.72	−3.64	0.08	1.88	2.24	109.48	0.52	0.17
	112	−0.39	10	−3.71	−3.63	0.07	1.84	2.19	109.48	0.36	0.16
	111	−0.39	10	−3.71	−3.63	0.07	1.84	2.18	109.48	0.36	0.16
	111	−0.39	10	−3.71	−3.63	0.07	1.84	2.18	109.48	0.36	0.16

Table S16 continued from previous page

	111	−0.39	10	−3.71	−3.63	0.07	1.84	2.19	109.48	0.36	0.16
	110	−0.39	10	−3.71	−3.63	0.07	1.84	2.18	109.48	0.36	0.16
	110	−0.39	10	−3.71	−3.63	0.07	1.84	2.18	109.48	0.36	0.16
	0	−0.50	8	−3.64	−3.50	0.14	1.79	2.12	109.48	0.97	0.03
	0	−0.50	8	−3.64	−3.50	0.14	1.80	2.12	109.48	0.97	0.03
	0	−0.50	8	−3.64	−3.50	0.14	1.79	2.12	109.48	0.97	0.03
	0	−0.50	8	−3.64	−3.50	0.14	1.79	2.12	109.48	0.97	0.03
13	249	−0.19	10	−3.84	−3.73	0.11	2.67	3.39	109.47	0.06	0.00
	247	−0.19	10	−3.84	−3.73	0.11	2.59	3.40	109.47	0.03	−0.01
	245	−0.19	10	−3.85	−3.74	0.11	3.26	3.64	109.47	0.03	−0.01
	245	−0.19	10	−3.85	−3.74	0.11	2.51	3.35	109.47	0.15	0.02
	138	−0.30	10	−3.73	−3.59	0.13	2.00	2.40	109.48	0.24	0.14
	138	−0.30	10	−3.73	−3.59	0.13	2.00	2.40	109.48	0.24	0.15
	138	−0.30	10	−3.73	−3.59	0.13	2.00	2.40	109.48	0.23	0.15
	94	−0.34	10	−3.67	−3.55	0.13	1.88	2.24	109.48	0.17	0.11
	94	−0.34	10	−3.67	−3.55	0.13	1.88	2.24	109.48	0.16	0.10
	72	−0.37	10	−3.71	−3.62	0.09	1.83	2.17	109.47	0.15	0.14
	71	−0.37	10	−3.71	−3.62	0.09	1.83	2.17	109.47	0.15	0.13
	71	−0.37	10	−3.71	−3.62	0.09	1.83	2.17	109.47	0.14	0.14
	61	−0.38	10	−3.72	−3.63	0.09	1.81	2.14	109.48	0.39	0.18
	61	−0.38	10	−3.72	−3.63	0.09	1.81	2.14	109.48	0.38	0.18
	61	−0.38	10	−3.72	−3.63	0.09	1.81	2.14	109.48	0.39	0.18
	61	−0.38	10	−3.72	−3.63	0.09	1.81	2.14	109.48	0.40	0.18
	46	−0.39	10	−3.72	−3.60	0.12	1.86	2.23	109.48	0.41	0.13
	46	−0.39	10	−3.72	−3.60	0.12	1.86	2.23	109.48	0.40	0.13
	46	−0.39	10	−3.72	−3.60	0.12	1.86	2.23	109.48	0.41	0.13
	45	−0.39	10	−3.72	−3.60	0.12	1.86	2.23	109.48	0.41	0.13
	1	−0.44	10	−3.73	−3.62	0.11	1.83	2.20	109.48	0.24	0.11
	1	−0.44	10	−3.73	−3.62	0.11	1.83	2.20	109.48	0.24	0.11
	1	−0.44	10	−3.73	−3.62	0.11	1.83	2.20	109.48	0.24	0.11
	1	−0.44	10	−3.73	−3.62	0.11	1.83	2.21	109.48	0.24	0.12
	1	−0.44	10	−3.73	−3.62	0.11	1.83	2.20	109.48	0.24	0.12
	0	−0.44	10	−3.73	−3.62	0.11	1.83	2.20	109.48	0.24	0.11
14	175	−0.19	12	−3.90	−3.82	0.07	2.23	3.11	109.47	0.03	0.00
	171	−0.19	12	−3.93	−3.86	0.07	2.68	3.42	109.47	0.04	−0.03
	171	−0.19	12	−3.93	−3.86	0.07	2.63	3.39	109.47	0.06	0.00
	167	−0.20	12	−3.90	−3.83	0.07	2.28	3.12	109.47	−0.03	−0.02
	164	−0.20	12	−3.93	−3.85	0.07	2.51	3.30	109.47	0.03	−0.02
	162	−0.20	12	−3.94	−3.86	0.07	3.21	3.69	109.47	0.01	−0.01
	116	−0.25	12	−3.80	−3.73	0.07	1.94	2.35	109.48	0.20	0.05
	115	−0.25	12	−3.80	−3.73	0.07	1.94	2.35	109.48	0.19	0.05
	109	−0.26	12	−3.78	−3.70	0.08	1.87	2.22	109.48	0.35	0.14
	108	−0.26	12	−3.80	−3.67	0.12	2.00	2.38	109.48	0.47	0.16
	108	−0.26	12	−3.78	−3.70	0.08	1.87	2.23	109.48	0.35	0.14
	108	−0.26	12	−3.80	−3.67	0.12	2.00	2.38	109.48	0.47	0.16
	108	−0.26	12	−3.80	−3.67	0.12	2.00	2.38	109.48	0.47	0.16
	83	−0.28	12	−3.82	−3.73	0.09	2.01	2.41	109.48	0.06	0.06
	22	−0.34	12	−3.82	−3.74	0.07	1.90	2.29	109.48	0.22	0.07
	21	−0.34	12	−3.82	−3.74	0.07	1.90	2.29	109.48	0.22	0.07

Table S16 continued from previous page

	16	−0.35	12	−3.78	−3.70	0.08	1.83	2.18	109.48	0.25	0.12
	16	−0.35	12	−3.78	−3.70	0.08	1.83	2.18	109.48	0.24	0.12
	14	−0.35	12	−3.83	−3.74	0.09	1.90	2.28	109.48	0.31	0.08
	14	−0.35	12	−3.83	−3.74	0.09	1.90	2.28	109.48	0.31	0.08
	14	−0.35	12	−3.83	−3.74	0.09	1.91	2.29	109.48	0.31	0.08
	13	−0.35	12	−3.83	−3.74	0.08	1.90	2.28	109.48	0.32	0.08
	1	−0.36	12	−3.76	−3.68	0.09	1.89	2.24	109.48	0.11	0.08
	1	−0.36	12	−3.83	−3.73	0.10	1.87	2.28	109.48	0.22	0.09
	1	−0.36	12	−3.83	−3.73	0.10	1.87	2.28	109.48	0.22	0.09
	1	−0.36	12	−3.76	−3.67	0.09	1.89	2.24	109.48	0.11	0.08
	1	−0.36	12	−3.83	−3.73	0.10	1.88	2.28	109.48	0.22	0.09
	0	−0.36	12	−3.83	−3.73	0.09	1.87	2.27	109.48	0.22	0.10
15	168	−0.20	12	−3.95	−3.85	0.10	3.28	3.53	109.47	0.04	−0.01
	167	−0.20	12	−3.95	−3.85	0.10	3.29	3.59	109.47	0.02	−0.01
	164	−0.20	12	−3.94	−3.83	0.10	2.45	3.29	109.47	−0.01	0.00
	50	−0.32	12	−3.79	−3.67	0.11	1.98	2.36	109.48	−0.06	0.10
	50	−0.32	12	−3.79	−3.67	0.11	1.98	2.37	109.48	−0.05	0.11
	23	−0.34	12	−3.84	−3.71	0.12	1.90	2.29	109.48	0.13	0.07
	23	−0.34	12	−3.83	−3.71	0.12	1.89	2.28	109.48	0.13	0.07
	23	−0.34	12	−3.84	−3.71	0.12	1.89	2.28	109.48	0.13	0.07
	22	−0.34	12	−3.83	−3.71	0.12	1.89	2.28	109.48	0.13	0.07
	22	−0.34	12	−3.78	−3.70	0.08	1.91	2.27	109.48	0.15	0.06
	22	−0.34	12	−3.82	−3.74	0.08	1.91	2.31	109.48	0.13	0.07
	22	−0.34	12	−3.82	−3.74	0.08	1.92	2.32	109.48	0.13	0.07
	22	−0.34	12	−3.84	−3.71	0.13	1.89	2.28	109.48	0.13	0.07
	22	−0.34	12	−3.78	−3.70	0.08	1.91	2.27	109.48	0.15	0.07
	22	−0.34	12	−3.78	−3.69	0.08	1.91	2.26	109.48	0.16	0.07
	21	−0.35	12	−3.78	−3.69	0.08	1.91	2.27	109.48	0.15	0.06
	21	−0.35	12	−3.78	−3.70	0.08	1.91	2.27	109.48	0.15	0.06
	21	−0.35	12	−3.82	−3.74	0.08	1.91	2.32	109.48	0.13	0.07
	21	−0.35	12	−3.82	−3.74	0.08	1.91	2.31	109.48	0.13	0.07
	21	−0.35	12	−3.82	−3.74	0.08	1.91	2.31	109.48	0.13	0.07
	21	−0.35	12	−3.82	−3.74	0.08	1.91	2.31	109.48	0.13	0.07
	2	−0.36	12	−3.77	−3.68	0.09	1.82	2.18	109.48	0.21	0.14
	2	−0.36	12	−3.77	−3.68	0.09	1.82	2.17	109.48	0.21	0.14
	1	−0.36	12	−3.76	−3.68	0.08	1.81	2.17	109.48	0.21	0.14
	1	−0.37	12	−3.76	−3.68	0.08	1.82	2.18	109.48	0.21	0.14
	1	−0.37	12	−3.77	−3.68	0.09	1.82	2.17	109.48	0.21	0.14
	1	−0.37	12	−3.76	−3.68	0.08	1.82	2.17	109.48	0.21	0.14
	0	−0.37	12	−3.76	−3.68	0.08	1.82	2.17	109.48	0.21	0.14
	0	−0.37	12	−3.77	−3.68	0.09	1.82	2.18	109.48	0.21	0.14
	0	−0.37	12	−3.77	−3.68	0.09	1.82	2.18	109.48	0.20	0.14

Table S17: Structural, energetic, and electronic properties for the CH_4/Cu_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	2	−0.16	0	−4.23	−2.60	1.63	1.91	2.27	109.49	−0.15	0.07
	1	−0.16	0	−4.23	−2.61	1.63	1.91	2.27	109.49	−0.11	0.05
	1	−0.16	0	−4.23	−2.60	1.63	1.91	2.27	109.49	−0.12	0.05
	0	−0.16	0	−4.23	−2.61	1.63	1.91	2.27	109.48	−0.10	0.04
	0	−0.16	0	−4.23	−2.61	1.63	1.92	2.27	109.49	−0.09	0.03
	0	−0.26	0	−4.23	−2.60	1.63	1.91	2.27	109.49	−0.11	0.05
	0	−0.26	0	−4.23	−2.61	1.63	1.92	2.27	109.49	−0.11	0.05
	0	−0.26	0	−4.23	−2.61	1.63	1.91	2.27	109.49	−0.13	0.06
5	74	−0.09	1	−4.25	−3.81	0.43	3.22	3.48	109.47	0.01	−0.02
	18	−0.14	1	−4.29	−3.85	0.44	3.34	3.76	109.47	−0.01	−0.04
	17	−0.14	1	−4.29	−3.85	0.44	3.36	3.78	109.47	−0.01	−0.04
	17	−0.14	1	−4.29	−3.85	0.44	3.33	3.74	109.47	−0.01	−0.05
	17	−0.14	1	−4.29	−3.85	0.44	3.36	3.77	109.47	−0.01	−0.04
	17	−0.14	1	−4.29	−3.85	0.44	3.34	3.75	109.47	−0.01	−0.04
	17	−0.14	1	−4.29	−3.85	0.44	3.34	3.75	109.47	−0.01	−0.04
	15	−0.14	1	−4.29	−3.85	0.44	3.33	3.73	109.47	−0.01	−0.04
	1	−0.16	1	−3.91	−3.49	0.42	2.03	2.42	109.48	−0.07	0.18
	0	−0.16	1	−3.91	−3.49	0.42	2.03	2.41	109.48	−0.06	0.18
6	16	−0.15	0	−4.36	−2.46	1.91	2.05	2.44	109.48	0.12	0.21
	12	−0.15	0	−4.36	−2.45	1.91	2.03	2.42	109.48	0.13	0.20
	12	−0.15	0	−4.36	−2.45	1.91	2.04	2.43	109.48	0.12	0.20
	11	−0.15	0	−4.36	−2.45	1.91	2.04	2.43	109.48	0.12	0.20
	11	−0.15	0	−4.36	−2.45	1.91	2.04	2.43	109.48	0.12	0.20
	11	−0.15	0	−4.36	−2.45	1.91	2.04	2.43	109.48	0.12	0.20
	11	−0.15	0	−4.36	−2.45	1.91	2.03	2.43	109.48	0.13	0.20
	3	−0.16	0	−4.69	−2.86	1.83	3.40	3.80	109.47	−0.01	−0.04
	2	−0.16	0	−4.69	−2.86	1.83	3.40	3.80	109.47	−0.01	−0.04
	2	−0.16	0	−4.69	−2.86	1.83	3.38	3.79	109.47	−0.01	−0.03
	1	−0.16	0	−4.69	−2.86	1.83	3.39	3.79	109.47	−0.02	−0.04
	0	−0.16	0	−4.69	−2.86	1.83	3.38	3.79	109.47	−0.02	−0.04
7	61	−0.11	1	−3.98	−3.65	0.33	2.34	3.39	109.47	−0.02	−0.01
	59	−0.11	1	−3.95	−3.62	0.33	2.28	3.31	109.47	−0.01	−0.02
	58	−0.11	1	−3.96	−3.63	0.33	2.30	3.34	109.47	−0.02	−0.02
	57	−0.11	1	−3.96	−3.63	0.33	2.31	3.33	109.47	−0.01	−0.01
	48	−0.12	1	−4.04	−3.69	0.35	3.46	3.87	109.47	0.00	−0.02
	2	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.11	0.11
	2	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.12	0.12
	2	−0.17	1	−3.87	−3.51	0.36	2.05	2.45	109.48	0.12	0.12
	1	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.12	0.12

Table S17 continued from previous page

	1	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.12	0.12
	1	−0.17	1	−3.87	−3.51	0.36	2.05	2.45	109.48	0.12	0.12
	1	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.11	0.12
	1	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.12	0.12
	0	−0.17	1	−3.87	−3.51	0.36	2.05	2.44	109.48	0.12	0.11
8	75	−0.11	0	−4.56	−3.05	1.51	2.38	3.33	109.47	−0.02	0.00
	74	−0.11	0	−4.56	−3.05	1.51	2.34	3.32	109.47	−0.03	0.00
	74	−0.11	0	−4.55	−3.05	1.51	2.36	3.31	109.47	−0.03	−0.01
	74	−0.11	0	−4.55	−3.05	1.50	2.34	3.31	109.47	−0.01	−0.01
	74	−0.11	0	−4.55	−3.05	1.51	2.37	3.29	109.47	−0.02	−0.01
	73	−0.11	0	−4.55	−3.05	1.50	2.36	3.24	109.47	−0.01	−0.01
	73	−0.11	0	−4.56	−3.05	1.51	2.38	3.30	109.47	−0.02	0.00
	2	−0.18	0	−4.38	−2.81	1.57	2.04	2.44	109.48	0.15	0.09
	2	−0.18	0	−4.38	−2.81	1.57	2.04	2.44	109.48	0.15	0.09
	2	−0.18	0	−4.38	−2.80	1.58	2.04	2.44	109.48	0.14	0.09
	2	−0.18	0	−4.38	−2.81	1.57	2.04	2.43	109.48	0.14	0.09
	1	−0.18	0	−4.38	−2.81	1.58	2.04	2.43	109.48	0.14	0.09
	1	−0.18	0	−4.38	−2.81	1.58	2.05	2.44	109.48	0.15	0.09
	1	−0.18	0	−4.38	−2.80	1.58	2.04	2.43	109.48	0.15	0.09
	1	−0.18	0	−4.38	−2.80	1.58	2.04	2.43	109.48	0.14	0.09
	0	−0.18	0	−4.38	−2.81	1.57	2.04	2.44	109.48	0.16	0.09
9	206	−0.03	1	−3.59	−3.27	0.32	3.31	3.69	109.47	3.32	0.19
	157	−0.02	1	−4.25	−3.97	0.28	3.38	3.69	109.47	3.15	0.06
	156	−0.02	1	−4.25	−3.97	0.28	3.39	3.65	109.47	3.16	0.06
	156	−0.02	1	−4.25	−3.97	0.28	3.38	3.67	109.47	3.17	0.06
	144	−0.03	1	−4.23	−3.94	0.29	3.38	3.83	109.47	2.88	0.04
	84	−0.10	1	−4.11	−3.83	0.28	2.41	3.26	109.47	0.03	−0.01
	81	−0.10	1	−3.82	−3.55	0.27	2.08	2.48	109.48	0.17	0.10
	80	−0.10	1	−3.83	−3.56	0.27	2.08	2.48	109.48	0.18	0.09
	80	−0.10	1	−3.83	−3.56	0.27	2.09	2.49	109.48	0.16	0.10
	70	−0.10	1	−4.16	−3.87	0.28	2.32	3.28	109.47	0.00	−0.01
	63	−0.11	1	−4.15	−3.87	0.28	2.29	3.23	109.47	−0.02	−0.01
	63	−0.11	1	−4.15	−3.87	0.28	2.34	3.20	109.47	−0.04	−0.01
	62	−0.11	1	−4.15	−3.87	0.28	2.31	3.23	109.47	−0.03	−0.02
	61	−0.12	1	−4.15	−3.86	0.28	2.30	3.19	109.47	−0.03	−0.01
	8	−0.17	1	−4.02	−3.74	0.28	2.09	2.50	109.48	0.09	0.12
	7	−0.17	1	−4.02	−3.74	0.28	2.08	2.49	109.48	0.10	0.12
	0	−0.18	1	−4.02	−3.73	0.28	2.05	2.44	109.48	0.14	0.10
	0	−0.18	1	−4.01	−3.73	0.28	2.05	2.44	109.48	0.14	0.10
10	130	−0.07	0	−4.14	−3.15	0.99	2.47	3.56	109.47	0.00	−0.01
	54	−0.15	0	−4.13	−3.11	1.02	2.20	3.11	109.47	0.01	−0.02
	53	−0.15	0	−4.13	−3.11	1.02	2.19	3.07	109.47	0.01	−0.02
	51	−0.15	0	−4.13	−3.11	1.02	2.21	3.07	109.47	0.02	−0.01
	51	−0.15	0	−4.13	−3.11	1.02	2.22	3.07	109.47	0.03	−0.01
	51	−0.15	0	−4.13	−3.11	1.02	2.21	3.07	109.47	0.02	−0.01
	50	−0.15	0	−4.13	−3.11	1.02	2.20	3.06	109.47	0.02	−0.01
	48	−0.16	0	−4.13	−3.11	1.02	2.17	3.06	109.47	−0.01	−0.01
	47	−0.16	0	−4.13	−3.11	1.02	2.19	3.09	109.47	0.00	−0.02
	46	−0.16	0	−4.13	−3.11	1.02	2.17	3.06	109.47	−0.01	−0.01

Table S17 continued from previous page

	46	−0.16	0	−4.10	−3.09	1.02	2.35	2.81	109.47	0.01	0.00
	46	−0.16	0	−4.13	−3.11	1.02	2.17	3.07	109.47	0.00	−0.01
	2	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.04	0.04
	1	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.04	0.02
	1	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.04	0.03
	1	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.03	0.03
	1	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.04	0.03
	1	−0.20	0	−4.02	−3.00	1.02	2.03	2.41	109.48	0.05	0.04
	0	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.04	0.02
	0	−0.20	0	−4.02	−3.00	1.02	2.02	2.41	109.48	0.04	0.03
11	114	−0.09	1	−4.16	−3.93	0.23	2.41	3.27	109.47	−0.04	−0.02
	113	−0.09	1	−4.03	−3.81	0.22	2.25	2.73	109.47	0.05	−0.01
	69	−0.14	1	−4.19	−3.96	0.23	3.29	3.63	109.47	0.08	−0.02
	69	−0.14	1	−4.20	−3.97	0.23	3.35	3.74	109.47	0.06	−0.02
	62	−0.14	1	−4.20	−3.97	0.23	3.38	3.73	109.47	0.04	−0.02
	60	−0.15	1	−4.20	−3.97	0.23	3.40	3.76	109.47	0.04	−0.02
	59	−0.15	1	−4.20	−3.97	0.23	3.33	3.79	109.47	0.01	−0.02
	57	−0.15	1	−4.19	−3.96	0.23	3.29	3.70	109.47	0.01	−0.02
	56	−0.15	1	−4.19	−3.96	0.23	3.30	3.73	109.47	0.01	−0.02
	56	−0.15	1	−4.19	−3.96	0.23	3.30	3.69	109.47	0.01	−0.02
	37	−0.17	1	−4.11	−3.88	0.23	2.18	2.62	109.48	0.23	0.05
	37	−0.17	1	−4.11	−3.88	0.23	2.19	2.63	109.48	0.24	0.04
	24	−0.18	1	−3.95	−3.71	0.23	2.02	2.41	109.48	0.05	0.04
	20	−0.19	1	−4.04	−3.82	0.22	2.12	2.53	109.48	0.24	0.09
	20	−0.19	1	−4.04	−3.82	0.22	2.11	2.52	109.48	0.25	0.09
	19	−0.19	1	−4.04	−3.82	0.22	2.12	2.53	109.48	0.24	0.09
	19	−0.19	1	−4.04	−3.81	0.22	2.11	2.52	109.48	0.25	0.09
	1	−0.20	1	−4.04	−3.81	0.23	2.05	2.45	109.48	0.25	0.07
	1	−0.20	1	−4.04	−3.81	0.23	2.05	2.45	109.48	0.26	0.07
	1	−0.21	1	−4.04	−3.81	0.23	2.05	2.45	109.48	0.26	0.07
	0	−0.21	1	−4.04	−3.81	0.23	2.05	2.45	109.48	0.25	0.06
	0	−0.21	1	−4.04	−3.81	0.23	2.05	2.45	109.48	0.25	0.07
12	112	−0.14	0	−4.39	−3.59	0.81	3.32	3.73	109.47	−0.06	−0.02
	112	−0.14	0	−4.39	−3.59	0.81	3.35	3.76	109.47	−0.04	−0.02
	92	−0.16	0	−4.40	−3.60	0.80	3.37	3.68	109.47	0.10	−0.01
	60	−0.19	0	−4.29	−3.51	0.78	2.10	2.55	109.48	0.31	0.08
	60	−0.19	0	−4.29	−3.51	0.78	2.11	2.54	109.48	0.31	0.09
	59	−0.19	0	−4.28	−3.50	0.78	2.11	2.53	109.48	0.31	0.09
	59	−0.19	0	−4.29	−3.51	0.78	2.10	2.54	109.48	0.32	0.08
	59	−0.19	0	−4.29	−3.51	0.78	2.10	2.54	109.48	0.32	0.08
	59	−0.19	0	−4.17	−3.32	0.86	2.02	2.41	109.48	0.05	0.03
	58	−0.19	0	−4.17	−3.32	0.85	2.02	2.42	109.48	0.05	0.03
	58	−0.19	0	−4.17	−3.32	0.85	2.01	2.41	109.48	0.05	0.04
	57	−0.19	0	−4.17	−3.32	0.86	2.01	2.41	109.48	0.05	0.03
	56	−0.19	0	−4.25	−3.37	0.88	2.11	2.53	109.48	0.12	0.07
	55	−0.19	0	−4.25	−3.37	0.88	2.11	2.52	109.48	0.12	0.07
	1	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.33	0.09
	1	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.32	0.10
	1	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.33	0.09

Table S17 continued from previous page

	1	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.33	0.10
	1	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.33	0.10
	1	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.33	0.10
	0	−0.25	0	−4.30	−3.42	0.88	2.05	2.46	109.48	0.33	0.10
	0	−0.25	0	−4.30	−3.42	0.88	2.07	2.46	109.48	0.33	0.10
	0	−0.25	0	−4.30	−3.42	0.88	2.06	2.46	109.48	0.33	0.10
	0	−0.25	0	−4.30	−3.42	0.88	2.06	2.45	109.48	0.34	0.10
13	131	−0.10	1	−3.90	−3.70	0.20	2.27	2.77	109.47	0.02	−0.02
	120	−0.11	1	−3.84	−3.66	0.18	2.33	2.80	109.47	0.11	0.02
	120	−0.11	1	−3.84	−3.66	0.18	2.35	2.80	109.47	0.12	0.02
	120	−0.11	1	−3.83	−3.63	0.20	2.11	2.52	109.48	−0.02	0.06
	116	−0.12	1	−3.95	−3.74	0.21	2.26	2.76	109.47	0.00	0.00
	114	−0.12	1	−3.95	−3.74	0.21	2.28	2.74	109.47	0.01	0.00
	113	−0.12	1	−3.95	−3.74	0.21	2.26	2.74	109.47	−0.01	0.00
	82	−0.15	1	−4.00	−3.80	0.20	2.22	3.06	109.47	0.01	−0.01
	79	−0.16	1	−3.99	−3.79	0.20	2.23	3.10	109.47	0.03	0.00
	75	−0.16	1	−3.97	−3.77	0.20	2.34	2.81	109.47	0.05	0.01
	74	−0.16	1	−3.97	−3.77	0.20	2.30	2.79	109.47	0.05	0.01
	74	−0.16	1	−3.97	−3.77	0.20	2.34	2.80	109.47	0.05	0.01
	73	−0.16	1	−4.03	−3.83	0.20	3.30	3.56	109.47	0.13	−0.02
	73	−0.16	1	−3.97	−3.77	0.20	2.33	2.79	109.47	0.06	0.01
	60	−0.17	1	−3.79	−3.59	0.21	2.03	2.43	109.48	0.17	0.05
	60	−0.17	1	−3.79	−3.58	0.21	2.03	2.43	109.48	0.17	0.05
	60	−0.17	1	−3.79	−3.59	0.21	2.03	2.43	109.48	0.17	0.05
	59	−0.17	1	−3.79	−3.59	0.21	2.03	2.43	109.48	0.17	0.05
	2	−0.23	1	−3.92	−3.71	0.21	2.06	2.48	109.48	0.44	0.08
	1	−0.23	1	−3.92	−3.71	0.21	2.06	2.48	109.48	0.44	0.08
	1	−0.23	1	−3.92	−3.71	0.21	2.06	2.49	109.48	0.46	0.08
	1	−0.23	1	−3.92	−3.71	0.21	2.07	2.48	109.48	0.44	0.08
	1	−0.23	1	−3.92	−3.71	0.21	2.07	2.48	109.48	0.44	0.08
	1	−0.23	1	−3.91	−3.71	0.21	2.06	2.48	109.48	0.45	0.08
	0	−0.23	1	−3.92	−3.71	0.21	2.06	2.48	109.48	0.44	0.08
	0	−0.23	1	−3.92	−3.71	0.21	2.06	2.48	109.48	0.46	0.08
14	91	−0.15	0	−4.44	−3.46	0.98	3.21	3.61	109.47	−0.03	−0.02
	91	−0.15	0	−4.42	−3.45	0.97	2.42	3.23	109.47	−0.05	−0.02
	90	−0.15	0	−4.43	−3.45	0.98	3.32	3.74	109.47	−0.03	−0.02
	90	−0.15	0	−4.42	−3.45	0.97	2.42	3.21	109.47	−0.04	−0.02
	90	−0.16	0	−4.42	−3.45	0.97	2.40	3.19	109.47	−0.01	−0.01
	90	−0.16	0	−4.42	−3.46	0.96	2.40	3.20	109.47	0.01	−0.01
	89	−0.16	0	−4.42	−3.46	0.96	2.39	3.19	109.47	0.02	−0.01
	89	−0.16	0	−4.42	−3.46	0.96	2.40	3.20	109.47	0.01	−0.01
	82	−0.16	0	−4.44	−3.46	0.98	3.26	3.60	109.47	0.09	−0.01
	38	−0.21	0	−4.30	−3.35	0.95	2.05	2.47	109.48	0.29	0.06
	37	−0.21	0	−4.30	−3.35	0.95	2.06	2.47	109.48	0.27	0.07
	37	−0.21	0	−4.30	−3.35	0.95	2.05	2.46	109.48	0.29	0.07
	36	−0.21	0	−4.30	−3.35	0.95	2.05	2.46	109.48	0.29	0.06
	36	−0.21	0	−4.30	−3.36	0.95	2.05	2.46	109.48	0.28	0.06
	2	−0.24	0	−4.33	−3.36	0.97	2.06	2.47	109.48	0.59	0.09
	2	−0.24	0	−4.33	−3.36	0.96	2.06	2.47	109.48	0.59	0.09

Table S17 continued from previous page

	1	−0.24	0	−4.33	−3.36	0.96	2.05	2.47	109.48	0.59	0.09
	1	−0.24	0	−4.33	−3.36	0.97	2.05	2.47	109.48	0.59	0.09
	1	−0.24	0	−4.33	−3.36	0.96	2.05	2.47	109.48	0.59	0.09
	1	−0.24	0	−4.33	−3.36	0.96	2.06	2.47	109.48	0.60	0.09
	1	−0.24	0	−4.33	−3.36	0.97	2.07	2.47	109.48	0.59	0.09
	1	−0.24	0	−4.33	−3.36	0.96	2.05	2.47	109.48	0.60	0.09
	1	−0.24	0	−4.33	−3.36	0.97	2.06	2.48	109.48	0.59	0.09
	1	−0.24	0	−4.33	−3.36	0.97	2.05	2.47	109.48	0.59	0.09
	0	−0.24	0	−4.33	−3.36	0.97	2.06	2.47	109.48	0.59	0.09
	0	−0.24	0	−4.33	−3.36	0.97	2.07	2.47	109.48	0.58	0.09
	0	−0.24	0	−4.33	−3.36	0.96	2.07	2.47	109.48	0.58	0.09
	0	−0.24	0	−4.33	−3.36	0.97	2.06	2.47	109.48	0.59	0.09
15	109	−0.16	1	−3.76	−3.57	0.20	2.17	2.62	109.48	0.42	0.08
	108	−0.16	1	−3.76	−3.56	0.20	2.13	2.62	109.48	0.44	0.08
	106	−0.16	1	−3.77	−3.57	0.19	2.14	2.63	109.48	0.40	0.08
	106	−0.16	1	−3.77	−3.57	0.19	2.13	2.63	109.48	0.40	0.08
	105	−0.16	1	−3.77	−3.57	0.19	2.11	2.62	109.48	0.42	0.08
	105	−0.16	1	−3.77	−3.57	0.19	2.14	2.63	109.48	0.41	0.08
	105	−0.16	1	−3.76	−3.57	0.19	2.11	2.62	109.48	0.45	0.08
	102	−0.16	1	−3.92	−3.73	0.20	2.82	3.43	109.47	0.16	−0.01
	101	−0.16	1	−3.91	−3.71	0.20	2.32	3.14	109.47	0.01	−0.01
	100	−0.16	1	−3.91	−3.71	0.20	2.31	3.01	109.47	0.00	−0.01
	100	−0.16	1	−3.91	−3.71	0.20	2.33	3.16	109.47	0.01	−0.02
	100	−0.16	1	−3.91	−3.72	0.20	2.32	3.16	109.47	−0.01	−0.02
	99	−0.17	1	−3.91	−3.72	0.20	2.31	3.15	109.47	−0.02	−0.01
	99	−0.17	1	−3.91	−3.71	0.20	2.31	3.15	109.47	−0.03	−0.01
	98	−0.17	1	−3.91	−3.72	0.20	2.32	3.16	109.47	−0.01	−0.01
	97	−0.17	1	−3.92	−3.72	0.20	2.34	3.18	109.47	−0.03	−0.01
	77	−0.19	1	−3.74	−3.54	0.19	2.04	2.43	109.48	−0.02	0.01
	77	−0.19	1	−3.73	−3.54	0.19	2.03	2.42	109.48	−0.03	0.01
	76	−0.19	1	−3.73	−3.54	0.19	2.04	2.43	109.48	−0.03	0.01
	47	−0.22	1	−3.80	−3.60	0.20	2.05	2.46	109.48	0.26	0.06
	47	−0.22	1	−3.80	−3.60	0.20	2.05	2.46	109.48	0.24	0.06
	47	−0.22	1	−3.80	−3.60	0.20	2.05	2.46	109.48	0.24	0.06
	46	−0.22	1	−3.80	−3.60	0.20	2.05	2.46	109.48	0.23	0.05
	46	−0.22	1	−3.80	−3.60	0.20	2.05	2.46	109.48	0.25	0.06
	46	−0.22	1	−3.80	−3.60	0.20	2.04	2.46	109.48	0.25	0.05
	46	−0.22	1	−3.80	−3.60	0.20	2.04	2.46	109.48	0.27	0.05
	2	−0.26	1	−3.82	−3.63	0.20	2.06	2.46	109.48	0.63	0.09
	1	−0.26	1	−3.82	−3.63	0.20	2.05	2.46	109.48	0.65	0.09
	1	−0.26	1	−3.82	−3.63	0.20	2.06	2.46	109.48	0.63	0.09
	0	−0.26	1	−3.82	−3.63	0.20	2.06	2.46	109.48	0.63	0.09

- In general, the interaction of CH_4 molecule occurs between most preferable two, and one H atoms and the TM atom that directly interacts with the CH_4 at top sites.
- For few systems, the CH_4 molecule is oriented with an "umbrella" configurations, where three H atoms are pointed directly to the TM atom.
- The cluster size n does not have any influence of the nature of CH_4 – TM interaction.

- However, for few cases we can also observed that the H – TM interactions are also assisted by the C atom, in particular for Ni_n clusters.

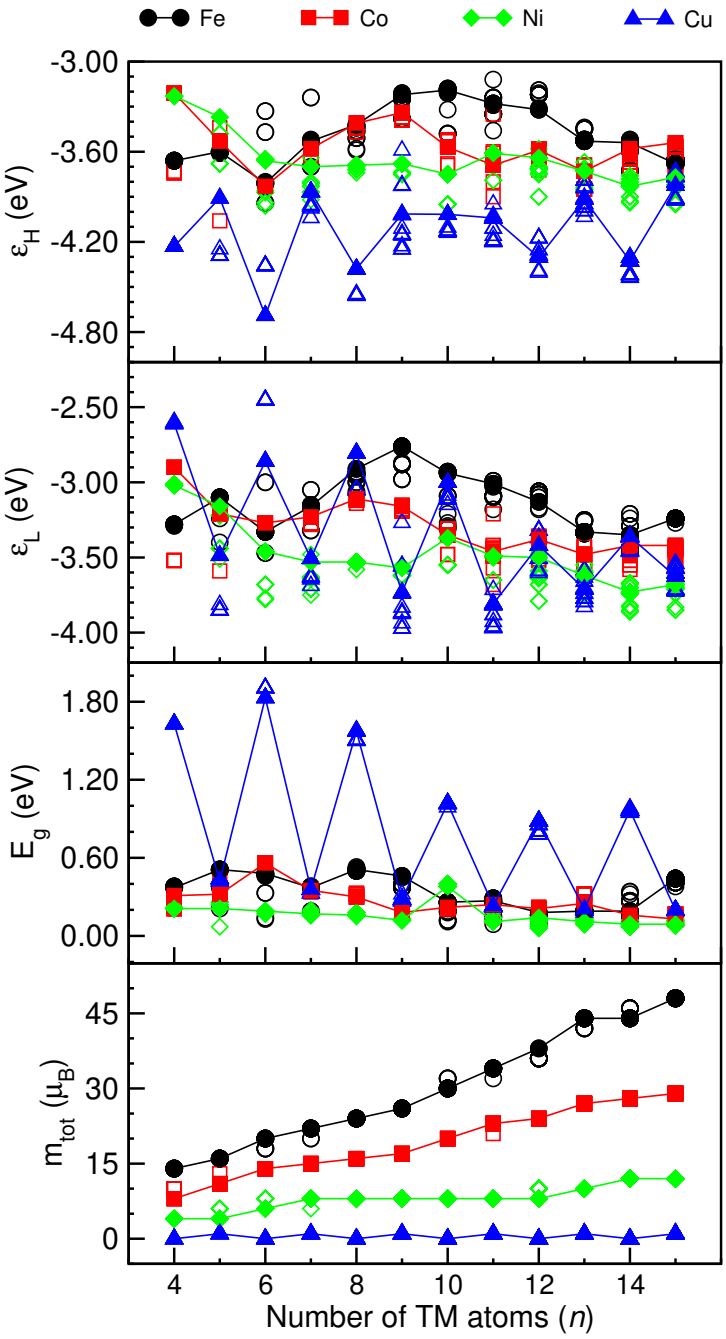

Figure S7: Energetic and electronic properties for CH_4 adsorption on TM_n clusters where $n = 4 - 15$ and TM = Fe, Co, Ni, Cu.

- In general, the magnitude of ε_{homo} for the CH_4/TM_n systems follows the same tendency of the TM_n clusters, where it increases with the occupation of d-states for almost all values of n, i.e., $n > 6$. However, for all configurations the largest magnitude of ε_{homo} is observed for Cu_n clusters.

- The trend of ε_{lumo} also follows the d-states occupation, as previously observed for $n > 6$. However, the zig-zag behaviour for Cu_n, can be explained due to the unpaired electron for

odd systems, which shifts the ε_{lumo} towards higher negative values while for even clusters, due to the complete d-states occupation, the ε_{lumo} assumes lower magnitudes.

- For almost all values of n, E_g assumes the lowest value for CH_4/Ni_n systems, and increases its magnitude from Co_n to Fe_n. Thus, as expected for Cu_n clusters with an even number of atoms present the largest magnitude of E_g.

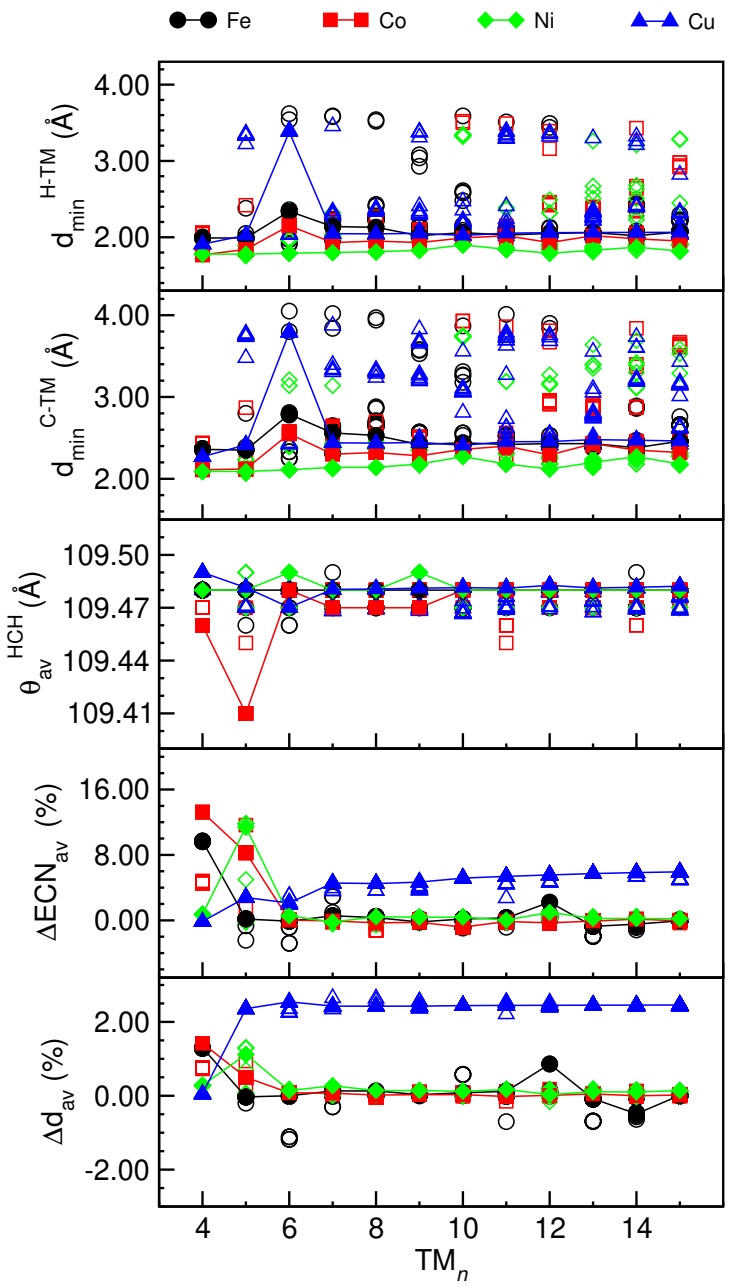

Figure S8: Structural properties for CH_4 adsorption on TM_n clusters where $n = 4 - 15$ and TM = Fe, Co, Ni, Cu.

- The magnitude of ${d_{min}}^{\mathrm{H-TM}}$ is smaller than ${d_{min}}^{\mathrm{C-TM}}$, which evinces that CH_4 preferable interacts via H rather than C atom. However, for few configurations the oppositely is observed.

- d_{min}^{H-TM} and d_{min}^{C-TM} increase from Ni_n towards Cu_n clusters, with the only exception for Cu_6, where the CH_4 molecule is at larger distances from the cluster, and oriented with an umbrella conformation, except for Co_5.
- Thus, those parameters along with the average HCH bong angle are not affected with n.
- The small variance of ΔECN_{av} and Δd_{av} indicate small deformation on the cluster upon adsorption. The largest changes occurs for n = 4 and 5 and for Cu_n clusters.

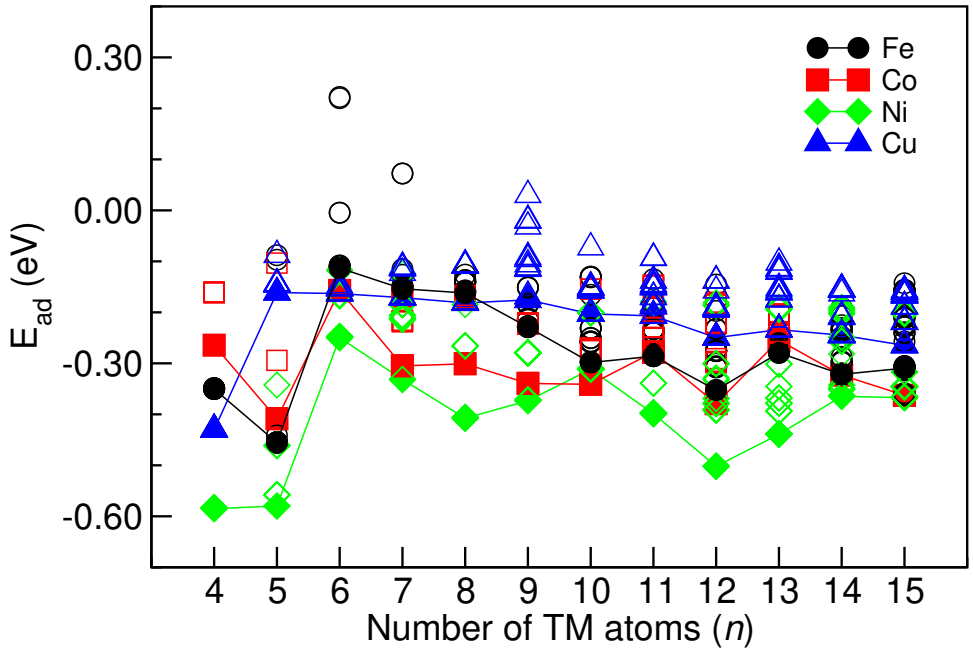

Figure S9: Adsorption energy for the CH_4/TM_n systems where $n = 4 - 15$ and TM = Fe, Co, Ni, and Cu.

- Except for Ni_{10} the magnitude of E_{ad} is higher for all values of n, while for $n > 8$ the smallest value of E_{ad} is observed for Cu_n.
- Among the TM, Ni_n present largest variation of E_{ad} range, where $Ni_{4/5}$ and Ni_{12} present the largest values of E_{ad}.
- Furthermore, the remaining clusters present small variations, where for Fe_n, the energy plateau starts from $n > 9$, while for Co_n and Cu_n, from $n > 6$ and $n > 5$, respectively.
- Small size clusters ($n < 9$), a different behaviour is observed, where Cu_n present adsorbs stronger CH_4 than Fe_n clusters. For $n = 4$, E_{ad} is about -0.45 eV, the second larger values after Ni_4, i.e., -0.60 eV.

8 CH_3 Adsorption on TM_n Clusters

Table S18: Structural, energetic, and electronic properties for the CH_3/Fe_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	8	−2.43	13	−3.87	−3.50	0.37	2.55	1.98	109.01	1.57	0.20

Table S18 continued from previous page

	1	−2.44	13	−3.82	−3.45	0.36	2.56	1.98	109.18	1.30	0.08
	1	−2.44	13	−3.82	−3.46	0.36	2.56	1.98	109.16	1.29	0.09
	0	−2.44	13	−3.82	−3.45	0.36	2.56	1.98	109.17	1.30	0.08
	0	−2.44	13	−3.82	−3.46	0.36	2.55	1.98	109.17	1.37	0.10
	0	−2.44	13	−3.82	−3.45	0.36	2.56	1.98	109.19	1.30	0.08
	0	−2.44	13	−3.82	−3.46	0.36	2.56	1.98	109.17	1.38	0.10
	0	−2.44	13	−3.81	−3.45	0.36	2.56	1.98	109.17	1.21	0.07
5	234	−2.35	15	−3.81	−3.74	0.08	2.55	1.97	109.13	4.81	0.60
	151	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.21	−0.35	1.13
	151	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.21	−0.35	1.13
	150	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.20	−0.34	1.13
	150	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.21	−0.35	1.13
	149	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.21	−0.37	1.13
	149	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.22	−0.33	1.13
	149	−2.44	17	−4.09	−3.81	0.27	2.56	1.98	109.21	−0.30	1.13
	4	−2.58	17	−3.78	−3.42	0.36	2.27	2.15	106.42	1.53	0.65
	0	−2.59	17	−3.77	−3.40	0.37	2.21	2.12	106.42	1.49	0.65
6	245	−2.11	19	−4.05	−3.65	0.39	2.52	1.96	110.17	0.44	−0.05
	245	−2.11	19	−4.05	−3.65	0.39	2.52	1.96	110.19	0.43	−0.06
	244	−2.11	19	−4.04	−3.65	0.39	2.52	1.96	110.18	0.44	−0.06
	244	−2.11	19	−4.05	−3.65	0.39	2.52	1.96	110.20	0.45	−0.06
	243	−2.11	19	−4.04	−3.65	0.39	2.52	1.96	110.17	0.45	−0.05
	210	−2.15	19	−3.93	−3.46	0.47	2.53	1.96	110.11	−2.38	−0.42
	207	−2.15	19	−3.93	−3.46	0.47	2.53	1.95	110.13	−2.39	−0.43
	62	−2.30	19	−4.02	−3.53	0.48	2.55	1.96	109.67	−0.71	−0.14
	61	−2.30	19	−4.02	−3.53	0.48	2.55	1.96	109.69	−0.70	−0.13
	61	−2.30	19	−4.02	−3.53	0.48	2.54	1.96	109.68	−0.71	−0.13
	2	−2.36	19	−3.65	−3.29	0.37	2.19	2.08	106.73	−0.70	−0.35
	0	−2.36	19	−3.66	−3.29	0.37	2.19	2.08	106.75	−0.71	−0.35
7	343	−1.99	23	−4.00	−3.72	0.29	2.57	1.99	108.33	3.74	1.59
	342	−1.99	23	−4.02	−3.68	0.34	2.59	1.99	108.31	3.50	1.60
	341	−1.99	23	−4.02	−3.68	0.34	2.59	1.99	108.28	3.49	1.60
	341	−1.99	23	−4.02	−3.68	0.34	2.59	1.99	108.30	3.50	1.59
	340	−1.99	23	−4.00	−3.71	0.29	2.58	1.99	108.32	3.76	1.59
	340	−1.99	23	−4.00	−3.72	0.29	2.58	1.99	108.31	3.79	1.60
	337	−1.99	23	−4.02	−3.77	0.24	2.59	2.00	108.27	2.30	1.43
	337	−1.99	23	−4.01	−3.77	0.24	2.59	1.99	108.27	2.32	1.44
	336	−1.99	23	−4.01	−3.77	0.24	2.59	1.99	108.27	2.35	1.44
	258	−2.07	23	−3.81	−3.44	0.37	2.21	2.12	105.93	2.03	1.07
	169	−2.16	21	−3.80	−3.43	0.37	2.44	1.95	109.90	−0.70	−0.08
	98	−2.23	21	−3.47	−3.06	0.42	2.16	2.06	106.57	−0.36	−0.21
	15	−2.31	21	−3.72	−3.37	0.36	2.54	1.96	109.90	0.86	0.09
	0	−2.33	21	−3.74	−3.34	0.40	2.53	1.96	109.85	0.65	0.05
8	295	−1.94	25	−3.68	−3.61	0.06	2.59	1.99	108.38	1.43	1.36
	218	−2.02	25	−3.86	−3.48	0.39	2.54	1.98	108.59	1.60	1.32
	217	−2.02	25	−3.87	−3.48	0.39	2.54	1.98	108.59	1.77	1.34
	216	−2.02	25	−3.86	−3.48	0.38	2.54	1.98	108.58	1.66	1.33
	216	−2.02	25	−3.87	−3.48	0.39	2.54	1.98	108.59	1.68	1.32

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	204	−2.03	25	−3.48	−3.27	0.21	2.16	2.09	105.80	−0.05	0.87
	173	−2.06	25	−3.91	−3.50	0.41	2.57	1.98	108.61	1.44	1.32
	94	−2.14	25	−3.54	−3.02	0.52	2.30	2.16	106.17	3.80	1.12
	47	−2.19	23	−3.74	−3.17	0.57	2.49	1.94	110.20	−0.12	−0.09
	47	−2.19	23	−3.74	−3.17	0.57	2.49	1.94	110.21	−0.15	−0.10
	46	−2.19	23	−3.74	−3.17	0.57	2.49	1.94	110.17	−0.14	−0.10
	46	−2.19	23	−3.74	−3.17	0.57	2.49	1.94	110.19	−0.14	−0.10
	31	−2.21	23	−3.73	−3.23	0.50	2.50	1.94	109.95	−0.25	−0.11
	30	−2.21	23	−3.73	−3.23	0.50	2.50	1.94	109.92	−0.23	−0.11
	1	−2.24	23	−3.68	−3.14	0.54	2.53	1.95	109.98	−0.40	−0.14
	0	−2.24	23	−3.68	−3.14	0.54	2.53	1.95	109.97	−0.40	−0.14
9	317	−2.05	25	−3.36	−2.98	0.39	2.56	1.98	109.48	−2.04	−0.38
	316	−2.05	25	−3.36	−2.98	0.39	2.56	1.98	109.49	−2.09	−0.39
	316	−2.05	25	−3.36	−2.98	0.39	2.56	1.98	109.47	−2.03	−0.38
	262	−2.11	27	−3.62	−3.47	0.15	2.54	1.99	108.38	0.41	0.75
	189	−2.18	27	−3.66	−3.37	0.28	2.54	1.98	109.01	−1.07	0.60
	189	−2.18	27	−3.66	−3.37	0.28	2.54	1.98	109.01	−1.07	0.60
	189	−2.18	27	−3.66	−3.38	0.28	2.54	1.98	109.00	−1.07	0.60
	188	−2.18	25	−3.48	−3.16	0.32	2.50	1.95	109.99	−0.58	−0.10
	188	−2.18	27	−3.66	−3.37	0.29	2.54	1.98	109.00	−1.07	0.60
	188	−2.18	27	−3.66	−3.38	0.28	2.54	1.98	109.01	−1.06	0.60
	188	−2.18	27	−3.66	−3.37	0.28	2.54	1.98	109.01	−1.08	0.60
	3	−2.37	27	−3.72	−3.42	0.30	2.56	1.98	108.80	−0.27	0.65
	2	−2.37	27	−3.72	−3.41	0.30	2.56	1.98	108.78	−0.27	0.65
	2	−2.37	27	−3.72	−3.41	0.30	2.56	1.98	108.78	−0.30	0.64
	2	−2.37	27	−3.72	−3.41	0.30	2.55	1.98	108.78	−0.27	0.65
	1	−2.37	27	−3.72	−3.42	0.30	2.56	1.98	108.80	−0.28	0.65
	1	−2.37	27	−3.72	−3.42	0.30	2.56	1.98	108.79	−0.26	0.65
	0	−2.37	27	−3.72	−3.41	0.30	2.56	1.98	108.79	−0.28	0.65
10	594	−2.00	31	−3.47	−3.31	0.16	2.57	1.99	109.39	−3.36	0.14
	592	−2.00	31	−3.47	−3.31	0.16	2.58	1.99	109.38	−3.36	0.14
	592	−2.00	31	−3.47	−3.31	0.16	2.57	1.99	109.38	−3.34	0.14
	319	−2.27	31	−3.42	−3.28	0.14	2.20	2.02	107.14	−1.57	0.38
	319	−2.27	31	−3.42	−3.28	0.14	2.19	2.02	107.12	−1.57	0.38
	254	−2.34	29	−3.54	−3.21	0.33	2.51	1.98	108.90	−0.43	−0.15
	254	−2.34	29	−3.54	−3.21	0.33	2.51	1.98	108.91	−0.44	−0.15
	252	−2.34	29	−3.54	−3.21	0.33	2.51	1.98	108.89	−0.45	−0.15
	103	−2.49	29	−3.55	−3.25	0.30	2.54	1.98	108.92	−0.05	−0.09
	102	−2.49	29	−3.55	−3.25	0.30	2.55	1.98	108.93	−0.05	−0.08
	102	−2.49	29	−3.55	−3.25	0.30	2.55	1.98	108.91	−0.06	−0.09
	102	−2.49	29	−3.55	−3.25	0.30	2.54	1.98	108.93	−0.06	−0.09
	102	−2.49	29	−3.55	−3.25	0.30	2.55	1.98	108.92	−0.05	−0.08
	101	−2.49	29	−3.55	−3.25	0.30	2.54	1.98	108.92	−0.06	−0.09
	101	−2.49	29	−3.55	−3.25	0.30	2.55	1.98	108.93	−0.05	−0.09
	100	−2.49	29	−3.55	−3.25	0.30	2.54	1.98	108.92	−0.06	−0.09
	100	−2.49	29	−3.55	−3.25	0.30	2.55	1.98	108.93	−0.05	−0.09
	2	−2.59	29	−3.54	−3.20	0.33	2.57	1.98	108.91	−0.01	−0.12
	2	−2.59	29	−3.54	−3.20	0.33	2.57	1.98	108.92	−0.01	−0.12
	0	−2.59	29	−3.54	−3.20	0.33	2.56	1.98	108.94	−0.01	−0.12

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11	235	−2.25	33	−3.53	−3.34	0.19	2.52	1.98	108.95	−0.59	−0.15
	235	−2.25	33	−3.53	−3.33	0.19	2.52	1.98	108.94	−0.55	−0.15
	234	−2.25	33	−3.52	−3.33	0.19	2.49	1.98	108.96	−0.69	−0.17
	234	−2.25	33	−3.52	−3.34	0.19	2.50	1.98	108.96	−0.56	−0.15
	234	−2.25	33	−3.53	−3.34	0.19	2.51	1.98	108.95	−0.56	−0.15
	233	−2.25	33	−3.53	−3.33	0.19	2.52	1.98	108.98	−0.53	−0.14
	233	−2.25	33	−3.52	−3.33	0.19	2.50	1.98	108.94	−0.65	−0.16
	107	−2.37	33	−3.55	−3.36	0.19	2.53	1.98	109.14	0.10	−0.12
	106	−2.37	33	−3.55	−3.38	0.17	2.54	1.98	109.11	0.51	−0.04
	106	−2.38	33	−3.55	−3.35	0.20	2.53	1.98	109.11	−0.03	−0.13
	106	−2.38	33	−3.55	−3.35	0.20	2.53	1.98	109.14	−0.04	−0.13
	104	−2.38	33	−3.55	−3.34	0.20	2.54	1.98	109.11	−0.05	−0.13
	2	−2.48	33	−3.47	−3.31	0.16	2.56	1.98	109.22	0.44	−0.03
	1	−2.48	33	−3.47	−3.23	0.25	2.55	1.98	109.22	0.01	−0.12
	1	−2.48	33	−3.47	−3.23	0.25	2.54	1.98	109.25	0.01	−0.11
	1	−2.48	33	−3.48	−3.31	0.16	2.56	1.98	109.23	0.44	−0.03
	1	−2.48	33	−3.47	−3.31	0.16	2.55	1.98	109.23	0.43	−0.03
	1	−2.48	33	−3.48	−3.31	0.17	2.56	1.98	109.21	0.43	−0.03
	1	−2.48	33	−3.47	−3.23	0.25	2.55	1.98	109.25	0.01	−0.11
	1	−2.48	33	−3.48	−3.31	0.17	2.57	1.98	109.19	0.42	−0.03
	0	−2.48	33	−3.47	−3.23	0.25	2.55	1.98	109.22	0.00	−0.12
	0	−2.48	33	−3.47	−3.23	0.25	2.55	1.98	109.23	−0.01	−0.12
12	191	−2.39	37	−3.63	−3.24	0.39	2.45	1.98	108.71	2.23	0.79
	191	−2.39	37	−3.63	−3.25	0.39	2.47	1.98	108.70	2.30	0.80
	141	−2.44	35	−3.33	−3.18	0.15	2.21	2.14	106.00	−0.84	−0.27
	40	−2.54	37	−3.57	−3.33	0.24	2.56	1.98	109.32	2.58	0.74
	40	−2.54	37	−3.57	−3.33	0.24	2.56	1.98	109.30	2.57	0.73
	39	−2.54	37	−3.56	−3.33	0.23	2.56	1.98	109.32	2.55	0.73
	39	−2.54	37	−3.56	−3.33	0.24	2.56	1.98	109.31	2.56	0.73
	39	−2.54	37	−3.57	−3.33	0.24	2.56	1.98	109.31	2.57	0.74
	38	−2.54	37	−3.57	−3.33	0.24	2.56	1.98	109.31	2.58	0.74
	38	−2.54	37	−3.57	−3.33	0.24	2.56	1.98	109.32	2.58	0.74
	38	−2.54	37	−3.57	−3.33	0.24	2.55	1.98	109.32	2.57	0.73
	37	−2.54	37	−3.57	−3.33	0.23	2.56	1.98	109.32	2.58	0.74
	37	−2.54	37	−3.57	−3.33	0.24	2.55	1.98	109.33	2.57	0.73
	37	−2.54	37	−3.56	−3.33	0.23	2.56	1.98	109.32	2.57	0.74
	24	−2.56	37	−3.56	−3.15	0.41	2.10	2.08	105.53	1.02	0.56
	2	−2.58	37	−3.58	−3.32	0.26	2.55	1.98	109.24	2.45	0.74
	2	−2.58	37	−3.58	−3.32	0.26	2.55	1.98	109.24	2.43	0.74
	1	−2.58	37	−3.59	−3.32	0.26	2.55	1.98	109.24	2.43	0.74
	1	−2.58	37	−3.58	−3.32	0.26	2.55	1.98	109.24	2.43	0.74
	1	−2.58	37	−3.58	−3.32	0.26	2.55	1.98	109.25	2.44	0.74
	1	−2.58	37	−3.58	−3.32	0.26	2.56	1.98	109.24	2.43	0.73
	1	−2.58	37	−3.58	−3.32	0.26	2.55	1.98	109.25	2.43	0.73
	1	−2.58	37	−3.59	−3.32	0.26	2.55	1.98	109.23	2.42	0.73
	0	−2.58	37	−3.58	−3.32	0.26	2.55	1.98	109.24	2.44	0.74
13	80	−2.35	41	−3.65	−3.54	0.11	2.53	1.98	109.49	−2.24	−0.83
	78	−2.35	41	−3.65	−3.54	0.11	2.53	1.98	109.47	−2.18	−0.82
	78	−2.35	41	−3.65	−3.54	0.11	2.53	1.98	109.47	−2.23	−0.83

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	77	−2.35	41	−3.65	−3.54	0.11	2.53	1.98	109.46	−2.17	−0.82
	55	−2.37	41	−3.65	−3.51	0.14	2.56	1.98	109.49	−2.53	−0.86
	54	−2.37	41	−3.65	−3.51	0.14	2.56	1.98	109.50	−2.54	−0.86
	7	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.71	0.30	−0.08
	7	−2.42	43	−3.71	−3.56	0.16	2.57	1.99	109.69	0.29	−0.08
	5	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.70	0.31	−0.08
	5	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.69	0.30	−0.08
	5	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.70	0.30	−0.08
	5	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.68	0.30	−0.08
	5	−2.42	43	−3.71	−3.55	0.16	2.56	1.99	109.69	0.31	−0.08
	5	−2.42	43	−3.71	−3.56	0.15	2.57	1.99	109.68	0.31	−0.08
	5	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.68	0.31	−0.08
	4	−2.42	43	−3.71	−3.56	0.15	2.57	1.99	109.70	0.30	−0.08
	4	−2.42	43	−3.71	−3.55	0.16	2.56	1.99	109.69	0.30	−0.08
	4	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.69	0.30	−0.08
	3	−2.42	43	−3.71	−3.55	0.15	2.57	1.99	109.70	0.31	−0.08
	3	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.69	0.30	−0.08
	3	−2.42	43	−3.71	−3.56	0.16	2.57	1.99	109.69	0.29	−0.08
	3	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.70	0.31	−0.08
	3	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.69	0.30	−0.08
	2	−2.42	43	−3.71	−3.55	0.16	2.57	1.99	109.68	0.30	−0.08
	2	−2.42	43	−3.71	−3.56	0.15	2.57	1.99	109.68	0.30	−0.08
	0	−2.42	43	−3.71	−3.55	0.16	2.56	1.99	109.68	0.30	−0.08
14	109	−2.29	43	−3.73	−3.63	0.10	2.53	1.95	109.47	−0.83	−0.61
	104	−2.30	43	−3.72	−3.62	0.11	2.55	1.98	109.80	−0.35	−0.65
	104	−2.30	43	−3.72	−3.62	0.10	2.55	1.98	109.78	−0.34	−0.65
	102	−2.30	43	−3.72	−3.62	0.10	2.55	1.98	109.78	−0.33	−0.65
	101	−2.30	43	−3.72	−3.62	0.10	2.55	1.98	109.77	−0.32	−0.64
	67	−2.33	45	−3.82	−3.61	0.21	2.55	1.96	109.60	−0.31	−0.13
	66	−2.34	45	−3.82	−3.61	0.21	2.54	1.96	109.60	−0.33	−0.13
	47	−2.35	43	−3.73	−3.62	0.10	2.55	1.98	109.35	−0.19	−0.59
	47	−2.35	43	−3.73	−3.62	0.10	2.55	1.98	109.36	−0.19	−0.60
	45	−2.36	43	−3.73	−3.62	0.11	2.55	1.98	109.35	−0.20	−0.59
	39	−2.36	45	−3.84	−3.57	0.27	2.56	1.99	109.88	−0.28	−0.16
	37	−2.36	45	−3.84	−3.57	0.27	2.55	1.99	109.88	−0.29	−0.16
	37	−2.36	45	−3.84	−3.57	0.27	2.56	1.99	109.88	−0.27	−0.16
	37	−2.36	45	−3.84	−3.57	0.27	2.56	1.99	109.88	−0.28	−0.16
	36	−2.37	45	−3.84	−3.57	0.27	2.56	1.99	109.88	−0.29	−0.16
	36	−2.37	45	−3.84	−3.57	0.27	2.56	1.99	109.88	−0.28	−0.16
	35	−2.37	45	−3.84	−3.57	0.27	2.56	1.99	109.88	−0.28	−0.16
	35	−2.37	43	−3.74	−3.64	0.10	2.53	1.97	109.48	−0.33	−0.61
	35	−2.37	45	−3.84	−3.57	0.27	2.55	1.99	109.88	−0.29	−0.16
	34	−2.37	43	−3.74	−3.64	0.10	2.53	1.97	109.49	−0.33	−0.61
	33	−2.37	43	−3.74	−3.64	0.10	2.53	1.97	109.48	−0.33	−0.61
	30	−2.37	43	−3.73	−3.61	0.12	2.55	1.97	109.58	−0.90	−0.65
	11	−2.39	43	−3.73	−3.63	0.11	2.55	1.97	109.27	−0.61	−0.64
	8	−2.39	43	−3.73	−3.63	0.11	2.55	1.97	109.29	−0.61	−0.63
	2	−2.40	45	−3.84	−3.59	0.25	2.53	1.98	109.59	−0.20	−0.11
	2	−2.40	45	−3.84	−3.59	0.25	2.54	1.98	109.60	−0.20	−0.11
	2	−2.40	45	−3.84	−3.60	0.25	2.54	1.98	109.59	−0.19	−0.11

Table S18 continued from previous page

	0	−2.40	45	−3.81	−3.61	0.20	2.55	1.97	109.85	−0.19	−0.13
15	164	−2.21	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.17	−0.11
	163	−2.21	47	−3.80	−3.71	0.09	2.56	1.99	110.03	−0.16	−0.10
	162	−2.21	47	−3.80	−3.71	0.09	2.56	1.99	110.04	−0.16	−0.11
	162	−2.21	47	−3.80	−3.71	0.09	2.56	1.99	110.02	−0.18	−0.11
	162	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.00	−0.18	−0.11
	161	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.02	−0.18	−0.11
	161	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.18	−0.11
	161	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.17	−0.11
	161	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.18	−0.11
	161	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.00	−0.18	−0.11
	160	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.00	−0.17	−0.11
	160	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.00	−0.19	−0.11
	160	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.17	−0.11
	160	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.02	−0.18	−0.11
	160	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.18	−0.11
	159	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.02	−0.16	−0.11
	159	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.18	−0.11
	158	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.00	−0.18	−0.11
	158	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.01	−0.18	−0.11
	157	−2.22	47	−3.80	−3.71	0.09	2.56	1.99	110.02	−0.18	−0.11
	18	−2.36	47	−3.82	−3.52	0.30	2.55	1.97	109.74	−0.05	−0.08
	6	−2.37	47	−3.83	−3.52	0.31	2.54	1.97	109.70	−0.18	−0.11
	5	−2.37	47	−3.83	−3.53	0.31	2.53	1.97	109.68	−0.15	−0.11
	5	−2.37	47	−3.83	−3.52	0.31	2.54	1.97	109.74	−0.15	−0.11
	5	−2.37	47	−3.83	−3.52	0.31	2.53	1.97	109.69	−0.15	−0.11
	5	−2.37	47	−3.83	−3.52	0.31	2.54	1.97	109.71	−0.18	−0.11
	5	−2.37	47	−3.83	−3.52	0.31	2.54	1.97	109.70	−0.16	−0.11
	5	−2.37	47	−3.83	−3.52	0.30	2.53	1.97	109.70	−0.12	−0.10
	3	−2.37	47	−3.83	−3.52	0.30	2.53	1.97	109.70	−0.12	−0.11
	0	−2.38	47	−3.83	−3.52	0.31	2.54	1.97	109.70	−0.15	−0.11

Table S19: Structural, energetic, and electronic properties for the CH_3/Co_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	1	−2.60	9	−4.14	−3.68	0.45	2.46	1.93	109.66	1.73	0.15
	1	−2.60	9	−4.13	−3.68	0.45	2.46	1.93	109.68	1.63	0.13
	1	−2.60	9	−4.13	−3.69	0.45	2.46	1.93	109.66	1.79	0.16
	1	−2.60	9	−4.14	−3.69	0.45	2.46	1.93	109.68	1.84	0.17
	0	−2.60	9	−4.14	−3.69	0.45	2.46	1.93	109.69	1.79	0.16
	0	−2.60	9	−4.13	−3.69	0.45	2.46	1.93	109.68	1.76	0.16

Table S19 continued from previous page

	0	−2.60	9	−4.13	−3.69	0.45	2.46	1.93	109.68	1.81	0.17
	0	−2.60	9	−4.13	−3.69	0.44	2.46	1.93	109.70	1.79	0.17
5	242	−2.31	12	−4.29	−3.80	0.49	2.51	1.94	110.48	−2.44	−0.37
	66	−2.49	12	−3.99	−3.79	0.20	2.52	1.94	109.96	9.75	0.97
	66	−2.49	12	−3.99	−3.79	0.20	2.52	1.94	109.97	9.73	0.97
	65	−2.49	12	−3.99	−3.79	0.20	2.52	1.94	109.99	9.74	0.96
	65	−2.49	12	−3.99	−3.79	0.20	2.52	1.94	109.96	9.74	0.97
	65	−2.49	12	−3.99	−3.79	0.20	2.52	1.94	109.97	9.74	0.96
	64	−2.49	12	−3.99	−3.79	0.20	2.52	1.94	109.98	9.76	0.97
	33	−2.52	12	−4.08	−3.68	0.40	2.52	1.94	109.97	10.19	1.27
	0	−2.55	12	−4.23	−3.79	0.44	2.49	1.94	109.91	2.60	0.38
	0	−2.55	12	−4.23	−3.79	0.44	2.49	1.94	109.92	2.63	0.38
6	3	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.61	0.01	−0.04
	2	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.61	0.01	−0.04
	1	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.62	0.01	−0.04
	1	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.63	0.01	−0.04
	1	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.62	0.01	−0.04
	1	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.62	0.01	−0.04
	1	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.62	0.01	−0.03
	1	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.63	0.01	−0.04
	0	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.63	0.01	−0.04
	0	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.62	0.01	−0.04
	0	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.63	0.01	−0.04
	0	−2.34	13	−4.07	−3.56	0.51	2.51	1.94	110.61	0.01	−0.04
7	104	−2.25	14	−3.72	−3.44	0.28	2.50	1.94	110.41	−0.25	−0.02
	103	−2.25	14	−3.74	−3.43	0.31	2.51	1.94	110.42	−0.64	−0.07
	102	−2.26	14	−3.74	−3.43	0.31	2.50	1.94	110.42	−0.61	−0.07
	102	−2.26	14	−3.71	−3.44	0.27	2.50	1.94	110.39	−0.24	−0.02
	101	−2.26	14	−3.74	−3.43	0.31	2.51	1.94	110.43	−0.66	−0.08
	101	−2.26	14	−3.74	−3.43	0.31	2.51	1.94	110.43	−0.65	−0.08
	100	−2.26	16	−4.26	−3.96	0.31	2.54	1.95	109.08	−0.07	0.61
	99	−2.26	16	−4.26	−3.95	0.31	2.54	1.95	109.09	−0.07	0.61
	46	−2.31	14	−3.71	−3.34	0.37	1.99	1.95	106.82	0.04	−0.12
	1	−2.36	14	−3.83	−3.45	0.38	2.47	1.93	110.53	−0.21	−0.03
	0	−2.36	14	−3.83	−3.45	0.38	2.47	1.93	110.51	−0.19	−0.02
	0	−2.36	14	−3.83	−3.45	0.38	2.47	1.93	110.52	−0.20	−0.02
	0	−2.36	14	−3.83	−3.45	0.38	2.46	1.93	110.52	−0.20	−0.02
	0	−2.36	14	−3.83	−3.45	0.38	2.45	1.93	110.50	−0.20	−0.02
8	357	−2.02	17	−3.68	−3.63	0.06	2.52	1.97	109.27	−1.13	0.38
	357	−2.02	17	−3.68	−3.63	0.06	2.52	1.97	109.26	−1.15	0.38
	357	−2.02	17	−3.68	−3.62	0.06	2.52	1.97	109.26	−1.17	0.38
	357	−2.02	17	−3.68	−3.62	0.06	2.52	1.97	109.24	−1.17	0.38
	356	−2.02	17	−3.68	−3.62	0.06	2.52	1.97	109.25	−1.14	0.38
	356	−2.02	17	−3.69	−3.62	0.06	2.52	1.97	109.26	−1.16	0.38
	355	−2.02	17	−3.69	−3.63	0.06	2.52	1.97	109.26	−1.17	0.38
	192	−2.19	15	−3.56	−3.27	0.29	2.43	1.94	110.41	−0.58	−0.11
	2	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.27	−0.92	0.29
	1	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.24	−0.96	0.28

Table S19 continued from previous page

	1	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.26	−0.96	0.28
	1	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.25	−0.94	0.28
	1	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.26	−0.95	0.28
	1	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.26	−0.94	0.28
	1	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.26	−0.95	0.28
	0	−2.38	17	−3.94	−3.66	0.28	2.51	1.94	109.26	−0.94	0.28
9	162	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.31	−0.35	0.34
	162	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.31	−0.36	0.33
	161	−2.25	18	−3.73	−3.55	0.18	2.51	1.96	109.33	−0.35	0.33
	161	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.32	−0.35	0.33
	161	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.32	−0.35	0.33
	161	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.32	−0.36	0.33
	161	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.32	−0.34	0.34
	161	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.30	−0.36	0.33
	161	−2.25	18	−3.74	−3.55	0.18	2.51	1.96	109.33	−0.35	0.34
	160	−2.25	18	−3.74	−3.55	0.19	2.51	1.96	109.32	−0.35	0.33
	1	−2.41	18	−3.87	−3.70	0.17	2.53	1.94	109.21	0.20	0.41
	1	−2.41	18	−3.87	−3.70	0.17	2.53	1.94	109.19	0.20	0.41
	1	−2.41	18	−3.87	−3.70	0.17	2.54	1.94	109.22	0.20	0.41
	0	−2.41	18	−3.87	−3.70	0.17	2.54	1.94	109.22	0.20	0.41
	0	−2.41	18	−3.87	−3.70	0.17	2.53	1.94	109.22	0.20	0.41
	0	−2.41	18	−3.87	−3.70	0.17	2.53	1.94	109.22	0.20	0.41
	0	−2.41	18	−3.87	−3.70	0.17	2.53	1.94	109.23	0.19	0.41
	0	−2.41	18	−3.87	−3.70	0.17	2.53	1.94	109.21	0.20	0.41
10	371	−2.14	19	−3.54	−3.38	0.16	1.87	2.01	106.37	−1.07	0.09
	218	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.56	−0.58	0.22
	218	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.54	−0.59	0.22
	218	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.55	−0.59	0.22
	217	−2.30	21	−3.90	−3.69	0.21	2.49	1.95	109.55	−0.61	0.22
	217	−2.30	21	−3.90	−3.70	0.21	2.49	1.95	109.55	−0.59	0.22
	216	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.56	−0.58	0.22
	216	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.58	−0.59	0.22
	216	−2.30	21	−3.90	−3.69	0.21	2.50	1.95	109.54	−0.59	0.22
	216	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.56	−0.58	0.22
	215	−2.30	21	−3.90	−3.70	0.21	2.50	1.95	109.57	−0.57	0.22
	122	−2.39	21	−3.91	−3.84	0.07	2.51	1.94	109.85	−0.13	0.47
	121	−2.39	21	−3.91	−3.84	0.06	2.51	1.94	109.86	−0.13	0.47
	10	−2.50	19	−3.75	−3.56	0.20	2.49	1.93	110.01	0.11	−0.02
	3	−2.51	19	−3.76	−3.55	0.21	2.47	1.93	110.05	0.09	−0.01
	3	−2.51	19	−3.76	−3.55	0.21	2.47	1.93	110.05	0.09	−0.01
	3	−2.51	19	−3.76	−3.55	0.21	2.47	1.93	110.05	0.07	−0.01
	3	−2.51	19	−3.76	−3.55	0.20	2.47	1.93	110.06	0.09	−0.01
	2	−2.51	19	−3.76	−3.55	0.21	2.47	1.93	110.06	0.07	−0.01
	0	−2.51	19	−3.76	−3.55	0.21	2.47	1.93	110.05	0.07	−0.01
11	425	−2.16	24	−3.99	−3.78	0.21	2.52	1.96	109.95	−1.84	0.00
	257	−2.16	22	−3.82	−3.66	0.16	2.42	1.93	110.03	−0.02	−0.05
	192	−2.16	22	−3.90	−3.64	0.26	2.46	1.93	110.09	0.17	−0.05
	192	−2.16	22	−3.90	−3.64	0.26	2.46	1.93	110.08	0.18	−0.05
	191	−2.33	22	−3.89	−3.64	0.26	2.46	1.93	110.08	0.16	−0.05

Table S19 continued from previous page

	191	−2.40	22	−3.89	−3.63	0.26	2.46	1.93	110.07	0.17	−0.05
	191	−2.40	22	−3.89	−3.63	0.26	2.47	1.93	110.07	0.16	−0.05
	28	−2.40	22	−3.88	−3.59	0.29	2.49	1.94	110.03	0.11	−0.01
	28	−2.40	22	−3.88	−3.59	0.29	2.49	1.93	110.01	0.12	−0.01
	27	−2.40	22	−3.88	−3.59	0.29	2.50	1.94	110.02	0.11	0.00
	25	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.45	−0.61	−0.08
	25	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.44	−0.61	−0.08
	25	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.42	−0.62	−0.08
	25	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.44	−0.61	−0.08
	25	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.44	−0.61	−0.08
	25	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.44	−0.61	−0.08
	24	−2.56	22	−3.71	−3.54	0.17	1.86	1.99	106.44	−0.61	−0.08
	1	−2.56	22	−3.74	−3.55	0.18	1.89	2.00	106.73	−0.90	−0.01
	0	−2.56	22	−3.74	−3.55	0.19	1.89	2.00	106.75	−0.89	−0.01
	3	−2.56	19	−3.76	−3.55	0.20	2.47	1.93	110.06	0.09	−0.01
	2	−2.59	19	−3.76	−3.55	0.21	2.47	1.93	110.06	0.07	−0.01
	0	−2.59	19	−3.76	−3.55	0.21	2.47	1.93	110.05	0.07	−0.01
12	435	−2.12	25	−3.71	−3.55	0.16	2.15	2.12	105.84	−0.94	0.21
	312	−2.24	25	−3.80	−3.63	0.17	2.04	2.07	106.56	−0.53	0.20
	311	−2.24	25	−3.80	−3.63	0.17	2.04	2.07	106.55	−0.53	0.21
	311	−2.24	25	−3.80	−3.63	0.17	2.04	2.07	106.55	−0.53	0.20
	311	−2.24	25	−3.80	−3.63	0.17	2.04	2.08	106.56	−0.52	0.20
	226	−2.33	25	−3.80	−3.66	0.14	2.05	2.00	106.74	−0.80	0.18
	223	−2.33	25	−3.76	−3.59	0.17	2.06	2.06	106.27	−0.05	0.34
	200	−2.35	25	−3.95	−3.73	0.22	2.50	1.95	109.88	−0.09	0.37
	135	−2.42	25	−3.92	−3.81	0.11	2.47	1.94	109.72	−0.57	0.18
	134	−2.42	25	−3.92	−3.81	0.11	2.47	1.94	109.73	−0.53	0.18
	78	−2.48	25	−3.97	−3.80	0.17	2.48	1.93	109.97	−0.44	0.18
	78	−2.48	25	−3.97	−3.80	0.17	2.48	1.93	109.98	−0.45	0.18
	78	−2.48	25	−3.97	−3.80	0.17	2.48	1.93	109.99	−0.45	0.18
	78	−2.48	25	−3.97	−3.80	0.17	2.48	1.93	109.99	−0.46	0.18
	77	−2.48	25	−3.97	−3.80	0.17	2.48	1.93	109.99	−0.50	0.18
	77	−2.48	25	−3.97	−3.80	0.17	2.48	1.93	109.98	−0.45	0.18
	67	−2.49	23	−3.75	−3.54	0.21	2.48	1.94	110.06	0.30	0.03
	66	−2.49	23	−3.75	−3.54	0.21	2.49	1.94	110.09	0.26	0.03
	66	−2.49	23	−3.75	−3.54	0.21	2.49	1.94	110.06	0.26	0.03
	65	−2.49	23	−3.75	−3.54	0.21	2.48	1.94	110.06	0.29	0.03
	65	−2.49	23	−3.75	−3.54	0.21	2.48	1.94	110.06	0.27	0.03
	28	−2.53	23	−3.76	−3.55	0.21	2.47	1.94	110.03	0.47	0.08
	28	−2.53	23	−3.76	−3.55	0.21	2.47	1.94	110.03	0.47	0.08
	0	−2.55	25	−3.96	−3.78	0.18	2.47	1.94	109.46	−1.42	−0.01
13	634	−2.15	26	−3.80	−3.65	0.15	2.05	2.09	106.38	−0.50	0.06
	634	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.39	−0.50	0.05
	634	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.41	−0.51	0.05
	633	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.41	−0.51	0.05
	633	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.37	−0.50	0.05
	633	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.38	−0.50	0.05
	633	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.40	−0.50	0.06
	632	−2.15	26	−3.80	−3.65	0.15	2.05	2.09	106.39	−0.50	0.05

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	632	−2.15	26	−3.80	−3.65	0.15	2.05	2.10	106.39	−0.50	0.05
	526	−2.25	26	−3.83	−3.62	0.20	2.02	1.99	106.69	−0.23	0.06
	336	−2.44	26	−3.98	−3.61	0.37	2.49	1.94	109.98	0.09	−0.03
	336	−2.44	26	−3.97	−3.61	0.36	2.48	1.94	109.92	0.11	−0.03
	336	−2.44	26	−3.98	−3.61	0.37	2.48	1.94	110.02	0.07	−0.02
	335	−2.45	26	−3.98	−3.61	0.37	2.49	1.94	109.95	0.08	−0.03
	335	−2.45	26	−3.98	−3.61	0.37	2.49	1.94	109.96	0.10	−0.03
	335	−2.45	26	−3.98	−3.61	0.37	2.49	1.94	109.99	0.08	−0.02
	335	−2.45	26	−3.98	−3.61	0.37	2.49	1.94	109.99	0.08	−0.02
	334	−2.45	26	−3.97	−3.61	0.37	2.49	1.94	109.99	0.08	−0.03
	256	−2.52	26	−4.00	−3.65	0.35	2.50	1.93	110.11	0.23	0.01
	255	−2.53	26	−4.00	−3.65	0.35	2.50	1.93	110.11	0.23	0.01
	255	−2.53	26	−4.00	−3.65	0.35	2.50	1.93	110.12	0.23	0.01
	255	−2.53	26	−4.00	−3.65	0.35	2.50	1.93	110.11	0.22	0.01
	255	−2.53	26	−4.00	−3.65	0.35	2.50	1.93	110.12	0.23	0.01
	255	−2.53	26	−4.00	−3.65	0.35	2.50	1.93	110.10	0.23	0.01
	1	−2.78	26	−3.82	−3.53	0.28	2.14	2.07	106.89	0.22	0.00
	0	−2.78	26	−3.82	−3.53	0.28	2.13	2.07	106.88	0.22	0.00
14	469	−2.30	29	−3.93	−3.66	0.27	2.03	2.02	106.51	−0.61	−0.03
	469	−2.30	29	−3.93	−3.66	0.27	2.03	2.02	106.51	−0.60	−0.03
	450	−2.32	29	−3.89	−3.79	0.10	2.51	1.94	109.78	0.70	0.15
	449	−2.32	29	−3.89	−3.79	0.10	2.51	1.94	109.80	0.70	0.15
	449	−2.32	29	−3.89	−3.79	0.10	2.51	1.94	109.80	0.70	0.16
	383	−2.38	29	−3.91	−3.76	0.16	2.46	1.95	109.77	0.82	0.39
	383	−2.38	29	−3.92	−3.76	0.16	2.46	1.95	109.78	0.82	0.39
	382	−2.38	29	−3.91	−3.76	0.16	2.46	1.95	109.79	0.83	0.39
	382	−2.38	29	−3.91	−3.76	0.16	2.46	1.95	109.78	0.83	0.39
	382	−2.38	29	−3.91	−3.76	0.16	2.46	1.95	109.77	0.82	0.39
	381	−2.39	29	−3.91	−3.76	0.16	2.46	1.95	109.75	0.83	0.39
	355	−2.41	29	−3.93	−3.70	0.23	2.01	2.01	106.47	0.04	0.05
	354	−2.41	29	−3.93	−3.70	0.23	2.01	2.01	106.46	0.04	0.05
	294	−2.47	29	−4.03	−3.83	0.20	2.50	1.94	109.82	0.54	0.10
	294	−2.47	29	−4.03	−3.83	0.20	2.51	1.94	109.83	0.53	0.10
	293	−2.47	29	−4.03	−3.83	0.20	2.50	1.94	109.81	0.55	0.10
	293	−2.47	29	−4.03	−3.83	0.20	2.51	1.94	109.82	0.54	0.10
	293	−2.47	29	−4.03	−3.83	0.20	2.51	1.94	109.82	0.54	0.10
	292	−2.47	29	−4.03	−3.83	0.20	2.51	1.94	109.83	0.54	0.10
	291	−2.48	29	−4.03	−3.83	0.20	2.51	1.94	109.82	0.54	0.10
	238	−2.53	27	−3.79	−3.61	0.18	2.49	1.93	110.11	0.22	−0.20
	236	−2.53	27	−3.79	−3.61	0.18	2.48	1.93	110.12	0.23	−0.20
	2	−2.76	27	−3.67	−3.52	0.15	2.10	2.06	106.79	0.21	−0.21
	2	−2.76	27	−3.67	−3.52	0.15	2.11	2.06	106.79	0.20	−0.21
	1	−2.77	27	−3.67	−3.52	0.15	2.09	2.06	106.77	0.20	−0.21
	1	−2.77	27	−3.67	−3.52	0.15	2.11	2.06	106.80	0.21	−0.21
	0	−2.77	27	−3.67	−3.52	0.15	2.10	2.06	106.78	0.20	−0.22
	0	−2.77	27	−3.67	−3.52	0.15	2.10	2.06	106.78	0.20	−0.22
15	467	−2.23	30	−3.75	−3.58	0.17	2.04	2.06	105.51	−1.08	0.16
	467	−2.23	30	−3.75	−3.58	0.17	2.04	2.06	105.49	−1.08	0.16
	465	−2.23	30	−3.75	−3.58	0.17	2.04	2.06	105.53	−1.06	0.16

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463	−2.23	30	−3.75	−3.58	0.17	2.04	2.06	105.53	−1.07	0.16
321	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.86	−0.57	0.21
321	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.86	−0.58	0.21
321	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.86	−0.55	0.21
320	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.83	−0.56	0.21
320	−2.37	30	−3.89	−3.68	0.20	2.46	1.95	109.87	−0.57	0.20
320	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.87	−0.56	0.21
320	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.84	−0.57	0.21
320	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.87	−0.57	0.21
319	−2.37	30	−3.89	−3.69	0.20	2.46	1.95	109.85	−0.56	0.21
319	−2.37	30	−3.89	−3.68	0.20	2.46	1.95	109.86	−0.57	0.20
318	−2.37	30	−3.89	−3.68	0.20	2.46	1.95	109.84	−0.56	0.21
315	−2.38	30	−3.79	−3.63	0.16	1.97	2.01	106.31	−0.70	0.16
314	−2.38	30	−3.79	−3.63	0.16	1.97	2.02	106.30	−0.70	0.16
310	−2.38	30	−3.80	−3.74	0.06	2.45	1.93	110.05	−0.30	0.15
261	−2.43	28	−3.73	−3.59	0.13	2.51	1.93	110.17	0.04	−0.04
261	−2.43	28	−3.73	−3.60	0.13	2.51	1.93	110.16	0.04	−0.05
156	−2.54	30	−3.88	−3.65	0.23	2.05	2.04	105.90	−0.25	0.12
156	−2.54	30	−3.88	−3.65	0.23	2.05	2.04	105.92	−0.24	0.11
156	−2.54	30	−3.88	−3.65	0.23	2.05	2.03	105.92	−0.24	0.12
156	−2.54	30	−3.88	−3.65	0.23	2.05	2.03	105.92	−0.25	0.12
155	−2.54	30	−3.88	−3.64	0.23	2.05	2.04	105.92	−0.24	0.12
154	−2.54	30	−3.88	−3.65	0.23	2.05	2.03	105.92	−0.24	0.11
142	−2.55	30	−4.02	−3.79	0.24	2.51	1.94	109.92	−0.15	0.15
139	−2.55	30	−4.02	−3.79	0.23	2.51	1.94	109.92	−0.14	0.15
138	−2.55	30	−4.02	−3.79	0.24	2.51	1.94	109.94	−0.15	0.15
0	−2.69	28	−3.63	−3.52	0.11	2.08	2.06	106.70	0.01	−0.05

Table S20: Structural, energetic, and electronic properties for the CH_3/Ni_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	2	−2.80	3	−3.81	−3.26	0.55	2.48	1.91	108.92	0.17	0.02
	2	−2.80	3	−3.80	−3.26	0.54	2.48	1.91	108.92	0.14	0.02
	2	−2.80	3	−3.80	−3.26	0.54	2.48	1.91	108.90	0.18	0.02
	2	−2.80	3	−3.80	−3.26	0.54	2.48	1.91	108.90	0.13	0.02
	2	−2.80	3	−3.80	−3.26	0.54	2.48	1.91	108.91	0.15	0.02
	1	−2.80	3	−3.80	−3.26	0.54	2.48	1.91	108.89	0.13	0.01
	0	−2.81	3	−3.80	−3.26	0.54	2.48	1.91	108.92	0.12	0.01
	0	−2.81	3	−3.80	−3.26	0.54	2.48	1.91	108.92	0.15	0.01
5	280	−2.29	7	−4.45	−4.31	0.14	2.47	1.92	109.73	−1.31	−0.22
	280	−2.29	7	−4.45	−4.32	0.13	2.47	1.92	109.70	−1.29	−0.23

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	278	−2.29	7	−4.45	−4.32	0.14	2.46	1.92	109.72	−1.29	−0.23
	278	−2.29	7	−4.45	−4.32	0.14	2.47	1.92	109.73	−1.30	−0.22
	38	−2.53	5	−4.14	−3.65	0.49	2.48	1.90	109.81	1.85	0.31
	38	−2.53	5	−4.14	−3.66	0.49	2.48	1.90	109.87	1.28	0.22
	38	−2.53	5	−4.14	−3.66	0.49	2.48	1.90	109.85	1.44	0.25
	37	−2.53	5	−4.14	−3.65	0.49	2.48	1.90	109.84	1.69	0.29
	36	−2.53	5	−4.14	−3.65	0.49	2.48	1.90	109.83	1.71	0.29
	0	−2.57	5	−3.90	−3.70	0.20	2.45	1.91	109.61	10.67	1.08
6	2	−2.39	7	−4.07	−3.86	0.21	2.44	1.91	110.20	−0.74	0.06
	2	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.20	−0.77	0.06
	1	−2.39	7	−4.06	−3.85	0.21	2.44	1.91	110.18	−0.80	0.06
	1	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.18	−0.79	0.06
	1	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.20	−0.75	0.06
	1	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.20	−0.76	0.06
	1	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.19	−0.75	0.06
	1	−2.39	7	−4.07	−3.86	0.21	2.44	1.91	110.21	−0.77	0.06
	1	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.21	−0.77	0.06
	0	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.19	−0.77	0.06
	0	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.20	−0.75	0.06
	0	−2.39	7	−4.06	−3.86	0.21	2.44	1.91	110.21	−0.75	0.06
7	209	−2.20	9	−4.34	−4.18	0.16	2.50	1.92	109.94	0.02	0.21
	209	−2.20	9	−4.34	−4.18	0.16	2.50	1.92	109.94	0.02	0.21
	106	−2.31	7	−4.10	−3.90	0.20	2.46	1.92	110.36	−0.37	0.02
	105	−2.31	7	−4.10	−3.90	0.20	2.46	1.92	110.38	−0.38	0.03
	104	−2.31	7	−4.10	−3.90	0.20	2.47	1.92	110.38	−0.37	0.02
	104	−2.31	7	−4.10	−3.90	0.20	2.47	1.92	110.38	−0.36	0.02
	3	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.26	−0.14	0.13
	2	−2.41	7	−4.10	−3.90	0.20	2.43	1.90	110.26	−0.13	0.13
	2	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.27	−0.14	0.12
	1	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.27	−0.14	0.13
	1	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.27	−0.14	0.13
	1	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.25	−0.14	0.12
	1	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.27	−0.14	0.13
	0	−2.41	7	−4.10	−3.90	0.20	2.44	1.90	110.27	−0.14	0.13
8	39	−2.23	9	−4.16	−4.01	0.16	2.47	1.91	109.63	0.32	0.22
	38	−2.23	9	−4.17	−4.01	0.16	2.47	1.91	109.66	0.31	0.22
	38	−2.23	9	−4.17	−4.01	0.16	2.47	1.91	109.67	0.31	0.23
	38	−2.23	9	−4.16	−4.01	0.16	2.47	1.91	109.66	0.32	0.22
	38	−2.23	9	−4.17	−4.01	0.16	2.47	1.91	109.67	0.32	0.22
	38	−2.23	9	−4.17	−4.01	0.16	2.47	1.91	109.67	0.31	0.22
	38	−2.23	9	−4.16	−4.01	0.16	2.47	1.91	109.67	0.33	0.22
	38	−2.23	9	−4.16	−4.01	0.16	2.47	1.91	109.65	0.33	0.22
	37	−2.23	9	−4.16	−4.01	0.16	2.47	1.91	109.66	0.33	0.22
	37	−2.23	9	−4.16	−4.01	0.16	2.47	1.91	109.66	0.33	0.22
	37	−2.23	9	−4.17	−4.01	0.16	2.47	1.91	109.67	0.31	0.22
	1	−2.26	7	−3.91	−3.77	0.14	2.42	1.90	110.38	−1.42	−0.10
	1	−2.27	7	−3.91	−3.77	0.14	2.42	1.90	110.38	−1.39	−0.10
	0	−2.27	7	−3.91	−3.77	0.14	2.43	1.90	110.38	−1.38	−0.10
	0	−2.27	7	−3.91	−3.76	0.15	2.43	1.90	110.37	−1.43	−0.10

Table S20 continued from previous page

	0	−2.27	7	−3.91	−3.76	0.14	2.43	1.90	110.38	−1.42	−0.10
9	67	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.48	−0.12	−0.02
	67	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.47	−0.14	−0.02
	67	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.45	−0.13	−0.02
	66	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.48	−0.14	−0.02
	66	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.48	−0.13	−0.02
	66	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.48	−0.14	−0.02
	66	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.47	−0.13	−0.02
	65	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.46	−0.13	−0.02
	65	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.47	−0.13	−0.02
	65	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.48	−0.14	−0.02
	65	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.47	−0.13	−0.02
	64	−2.21	7	−3.97	−3.82	0.15	2.37	1.90	110.46	−0.14	−0.02
	2	−2.27	9	−4.14	−4.03	0.11	2.49	1.91	109.75	−0.02	0.16
	1	−2.27	9	−4.14	−4.03	0.11	2.48	1.91	109.74	−0.02	0.16
	0	−2.27	9	−4.14	−4.03	0.11	2.48	1.91	109.75	−0.02	0.17
	0	−2.27	9	−4.14	−4.03	0.11	2.48	1.91	109.75	−0.02	0.17
	0	−2.27	9	−4.14	−4.03	0.11	2.48	1.91	109.75	−0.02	0.17
	0	−2.27	9	−4.14	−4.03	0.11	2.48	1.91	109.74	−0.01	0.17
10	350	−1.86	7	−3.55	−3.47	0.09	1.98	1.94	105.87	−0.12	0.60
	349	−1.86	7	−3.55	−3.47	0.09	1.97	1.94	105.90	−0.12	0.60
	259	−1.95	7	−3.99	−3.68	0.31	2.45	1.91	109.90	−0.59	0.09
	242	−1.97	9	−4.23	−4.06	0.17	2.48	1.91	109.90	0.23	0.31
	241	−1.97	9	−4.23	−4.06	0.17	2.48	1.91	109.89	0.22	0.31
	241	−1.97	9	−4.22	−4.06	0.17	2.48	1.91	109.91	0.24	0.31
	241	−1.97	9	−4.22	−4.05	0.17	2.48	1.91	109.90	0.22	0.32
	241	−1.97	9	−4.22	−4.05	0.17	2.48	1.91	109.89	0.22	0.31
	241	−1.97	9	−4.23	−4.06	0.17	2.48	1.91	109.90	0.24	0.32
	241	−1.97	9	−4.23	−4.06	0.17	2.48	1.91	109.89	0.24	0.31
	241	−1.97	9	−4.22	−4.05	0.17	2.48	1.91	109.91	0.22	0.32
	241	−1.97	9	−4.23	−4.06	0.17	2.48	1.91	109.91	0.23	0.31
	241	−1.97	9	−4.23	−4.05	0.17	2.48	1.91	109.90	0.21	0.31
	241	−1.97	9	−4.22	−4.04	0.18	2.48	1.91	109.90	0.21	0.31
	240	−1.97	9	−4.22	−4.04	0.18	2.48	1.91	109.89	0.20	0.31
	240	−1.97	9	−4.23	−4.05	0.17	2.48	1.91	109.90	0.22	0.31
	237	−1.97	9	−4.16	−3.94	0.22	2.44	1.92	109.98	0.40	0.35
	96	−2.11	9	−3.83	−3.71	0.11	1.93	1.98	106.44	−0.44	0.30
	1	−2.21	7	−3.65	−3.54	0.11	1.80	1.93	107.21	−0.42	0.45
	0	−2.21	7	−3.65	−3.54	0.11	1.80	1.93	107.27	−0.41	0.46
11	661	−1.91	9	−3.86	−3.79	0.07	2.53	1.96	109.10	−2.60	0.12
	612	−1.95	9	−3.82	−3.71	0.11	1.86	2.05	105.90	−2.68	0.19
	477	−2.09	9	−3.92	−3.84	0.08	2.48	1.93	109.23	−1.13	0.28
	476	−2.09	9	−3.92	−3.84	0.08	2.47	1.93	109.21	−1.15	0.28
	476	−2.09	9	−3.92	−3.84	0.08	2.48	1.92	109.24	−1.14	0.28
	2	−2.56	9	−3.91	−3.74	0.18	1.79	1.98	105.44	−0.32	0.35
	2	−2.56	9	−3.91	−3.74	0.18	1.79	1.98	105.44	−0.31	0.35
	1	−2.56	9	−3.91	−3.74	0.18	1.79	1.98	105.46	−0.31	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.44	−0.31	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.45	−0.32	0.35

Table S20 continued from previous page

	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.46	−0.31	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.46	−0.31	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.44	−0.30	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.46	−0.31	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.48	−0.32	0.35
	1	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.46	−0.32	0.35
	0	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.47	−0.31	0.35
	0	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.45	−0.31	0.35
	0	−2.57	9	−3.91	−3.74	0.18	1.79	1.98	105.46	−0.32	0.35
	0	−2.57	9	−3.91	−3.73	0.18	1.79	1.98	105.44	−0.31	0.35
	0	−2.57	9	−3.91	−3.73	0.18	1.79	1.98	105.44	−0.32	0.35
	0	−2.57	7	−3.65	−3.54	0.11	1.80	1.93	107.27	−0.41	0.46
12	548	−2.29	11	−4.09	−4.00	0.09	2.44	1.91	109.63	0.32	0.22
	547	−2.29	11	−4.09	−4.00	0.09	2.44	1.91	109.62	0.33	0.22
	547	−2.29	11	−4.09	−4.00	0.09	2.44	1.91	109.66	0.32	0.22
	547	−2.29	11	−4.09	−4.00	0.09	2.44	1.91	109.67	0.32	0.22
	547	−2.29	11	−4.09	−4.00	0.09	2.44	1.91	109.64	0.33	0.22
	547	−2.29	11	−4.09	−4.00	0.09	2.44	1.91	109.65	0.33	0.22
	524	−2.31	9	−3.93	−3.84	0.09	2.43	1.91	109.65	0.46	0.06
	524	−2.31	9	−3.93	−3.84	0.09	2.43	1.91	109.67	0.47	0.06
	524	−2.31	9	−3.93	−3.84	0.09	2.42	1.91	109.64	0.46	0.06
	524	−2.31	9	−3.93	−3.84	0.09	2.42	1.91	109.66	0.46	0.06
	523	−2.31	9	−3.93	−3.84	0.09	2.43	1.91	109.65	0.46	0.06
	476	−2.36	9	−3.78	−3.66	0.12	1.87	1.96	106.43	−0.18	0.16
	476	−2.36	9	−3.78	−3.66	0.13	1.87	1.96	106.43	−0.18	0.16
	289	−2.54	9	−3.96	−3.82	0.14	2.43	1.90	109.88	0.53	−0.04
	289	−2.54	9	−3.96	−3.82	0.14	2.43	1.90	109.83	0.56	−0.04
	289	−2.54	9	−3.96	−3.82	0.14	2.43	1.90	109.83	0.55	−0.03
	288	−2.55	9	−3.96	−3.82	0.14	2.43	1.90	109.85	0.56	−0.03
	187	−2.65	9	−3.83	−3.71	0.12	1.93	2.03	106.26	0.51	−0.02
	114	−2.72	9	−3.82	−3.72	0.11	1.84	1.97	105.82	0.58	0.03
	113	−2.72	9	−3.82	−3.72	0.11	1.84	1.97	105.81	0.58	0.03
	47	−2.79	9	−3.79	−3.71	0.08	1.82	1.96	105.90	0.41	0.11
	1	−2.83	9	−3.82	−3.66	0.16	1.77	1.95	105.93	0.05	0.17
	1	−2.83	9	−3.82	−3.66	0.16	1.77	1.95	105.92	0.04	0.17
	0	−2.83	9	−3.82	−3.66	0.16	1.77	1.95	105.93	0.05	0.17
13	719	−1.98	11	−3.96	−3.86	0.09	2.50	1.95	109.69	−2.92	−0.02
	719	−1.98	11	−3.96	−3.86	0.09	2.50	1.95	109.70	−2.95	−0.02
	718	−1.98	11	−3.96	−3.86	0.09	2.50	1.95	109.69	−2.92	−0.02
	416	−2.28	9	−3.78	−3.66	0.12	1.98	1.94	107.50	−0.89	0.05
	374	−2.32	11	−4.00	−3.90	0.10	2.42	1.91	109.88	−0.25	0.17
	373	−2.32	11	−4.00	−3.90	0.10	2.42	1.91	109.90	−0.27	0.18
	373	−2.32	11	−4.00	−3.90	0.10	2.42	1.91	109.87	−0.24	0.17
	373	−2.32	11	−4.00	−3.90	0.10	2.42	1.91	109.89	−0.27	0.19
	373	−2.32	11	−4.00	−3.90	0.10	2.42	1.91	109.91	−0.27	0.18
	372	−2.32	11	−4.00	−3.90	0.10	2.42	1.91	109.85	−0.25	0.18
	9	−2.69	11	−3.94	−3.79	0.15	1.79	1.96	106.11	0.14	0.29
	8	−2.69	11	−3.94	−3.79	0.15	1.79	1.96	106.10	0.13	0.29
	8	−2.69	11	−3.94	−3.79	0.15	1.79	1.96	106.10	0.14	0.29

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	8	−2.69	11	−3.94	−3.79	0.15	1.79	1.96	106.11	0.13	0.29
	7	−2.69	11	−3.94	−3.79	0.15	1.79	1.96	106.11	0.13	0.29
	2	−2.69	11	−3.88	−3.79	0.09	1.87	1.98	106.03	0.19	0.16
	2	−2.69	11	−3.88	−3.79	0.09	1.87	1.98	106.04	0.19	0.16
	1	−2.69	11	−3.88	−3.79	0.09	1.87	1.98	106.04	0.19	0.16
	1	−2.69	11	−3.89	−3.79	0.09	1.87	1.98	106.04	0.19	0.16
	1	−2.69	11	−3.89	−3.79	0.09	1.87	1.98	106.04	0.19	0.16
	1	−2.69	11	−3.89	−3.79	0.09	1.87	1.98	106.05	0.19	0.16
	1	−2.69	11	−3.89	−3.79	0.09	1.86	1.98	106.02	0.19	0.16
	1	−2.69	11	−3.88	−3.79	0.09	1.87	1.98	106.05	0.19	0.16
	1	−2.69	11	−3.89	−3.79	0.09	1.87	1.98	106.04	0.19	0.16
	0	−2.70	11	−3.88	−3.79	0.09	1.87	1.98	106.06	0.19	0.16
	0	−2.70	11	−3.88	−3.79	0.09	1.86	1.98	106.04	0.19	0.16
14	757	−1.93	13	−4.03	−3.94	0.09	2.53	1.96	109.42	−2.77	−0.02
	712	−1.97	13	−4.07	−3.96	0.11	2.53	1.96	109.39	−0.91	0.11
	707	−1.98	13	−4.07	−3.96	0.11	2.50	1.95	109.33	−0.90	0.10
	687	−2.00	13	−4.01	−3.94	0.07	2.54	1.96	109.73	−2.42	−0.02
	493	−2.19	13	−4.15	−4.07	0.08	2.47	1.91	109.86	0.01	0.10
	492	−2.19	13	−4.15	−4.07	0.08	2.47	1.91	109.85	0.02	0.11
	469	−2.21	11	−3.86	−3.76	0.10	2.00	1.95	107.49	−0.76	0.05
	410	−2.27	11	−3.98	−3.84	0.15	2.49	1.91	110.23	0.11	0.00
	372	−2.31	11	−3.86	−3.74	0.12	2.01	2.05	105.57	0.10	0.14
	108	−2.58	11	−3.90	−3.75	0.14	1.94	1.99	106.95	−0.43	−0.01
	108	−2.58	11	−3.90	−3.76	0.14	1.94	1.99	106.95	−0.43	0.00
	96	−2.59	11	−3.85	−3.76	0.08	1.78	1.93	106.37	0.13	0.12
	96	−2.59	11	−3.84	−3.76	0.08	1.78	1.93	106.39	0.13	0.12
	94	−2.59	11	−3.85	−3.76	0.08	1.78	1.93	106.38	0.12	0.12
	75	−2.61	11	−3.91	−3.78	0.12	1.87	1.98	106.52	−0.18	0.04
	75	−2.61	11	−3.91	−3.78	0.12	1.87	1.98	106.54	−0.17	0.04
	75	−2.61	11	−3.91	−3.78	0.12	1.87	1.98	106.54	−0.18	0.04
	75	−2.61	11	−3.91	−3.78	0.12	1.87	1.98	106.53	−0.16	0.04
	21	−2.66	11	−3.89	−3.76	0.13	1.91	1.99	106.54	0.06	−0.02
	20	−2.66	11	−3.89	−3.76	0.13	1.91	1.99	106.55	0.07	−0.02
	2	−2.68	11	−3.90	−3.79	0.12	1.83	1.97	106.40	0.11	0.05
	2	−2.68	11	−3.90	−3.79	0.12	1.83	1.97	106.42	0.10	0.05
	1	−2.68	11	−3.91	−3.77	0.14	1.84	1.98	106.18	0.28	0.03
	1	−2.68	11	−3.91	−3.77	0.14	1.85	1.98	106.20	0.27	0.03
	1	−2.68	11	−3.91	−3.77	0.14	1.85	1.98	106.19	0.27	0.03
	0	−2.68	11	−3.91	−3.77	0.14	1.85	1.98	106.20	0.27	0.03
	0	−2.68	11	−3.91	−3.77	0.14	1.85	1.98	106.21	0.27	0.03
	0	−2.68	11	−3.91	−3.77	0.14	1.84	1.98	106.19	0.28	0.03
15	606	−2.04	11	−3.87	−3.77	0.10	1.92	1.93	107.92	−0.31	0.21
	494	−2.15	13	−4.11	−4.00	0.11	2.48	1.92	109.78	−0.05	0.11
	494	−2.15	13	−4.11	−4.00	0.11	2.48	1.92	109.77	−0.05	0.11
	493	−2.15	13	−4.11	−4.00	0.11	2.48	1.92	109.77	−0.05	0.11
	493	−2.15	13	−4.11	−4.00	0.11	2.48	1.92	109.77	−0.06	0.11
	493	−2.15	13	−4.11	−4.00	0.11	2.48	1.92	109.78	−0.05	0.11
	493	−2.15	13	−4.11	−4.00	0.11	2.48	1.92	109.77	−0.06	0.11
	488	−2.16	13	−4.14	−4.05	0.09	2.46	1.91	109.96	0.19	0.13

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488	−2.16	13	−4.14	−4.05	0.09	2.46	1.91	109.96	0.18	0.13
488	−2.16	13	−4.15	−4.06	0.09	2.50	1.91	109.96	0.22	0.13
488	−2.16	13	−4.14	−4.05	0.09	2.46	1.91	109.96	0.19	0.13
488	−2.16	13	−4.14	−4.05	0.09	2.47	1.91	109.97	0.20	0.13
462	−2.18	13	−4.12	−4.07	0.05	2.49	1.91	109.97	−0.12	0.14
462	−2.18	13	−4.12	−4.07	0.05	2.49	1.91	109.97	−0.13	0.14
462	−2.18	13	−4.12	−4.07	0.05	2.49	1.91	109.97	−0.13	0.14
462	−2.18	13	−4.13	−4.07	0.06	2.49	1.91	109.99	−0.12	0.14
461	−2.18	13	−4.13	−4.07	0.06	2.49	1.91	109.98	−0.12	0.14
461	−2.18	13	−4.12	−4.07	0.05	2.49	1.91	109.97	−0.12	0.14
461	−2.18	13	−4.13	−4.07	0.06	2.49	1.91	109.99	−0.12	0.14
461	−2.18	13	−4.12	−4.07	0.05	2.49	1.91	109.97	−0.14	0.14
461	−2.19	13	−4.12	−4.07	0.05	2.49	1.91	109.96	−0.13	0.13
450	−2.20	11	−3.91	−3.82	0.10	1.98	1.94	107.40	−0.81	0.02
449	−2.20	11	−3.91	−3.82	0.10	1.98	1.94	107.37	−0.81	0.02
111	−2.53	13	−3.99	−3.90	0.09	1.85	1.99	106.03	−0.17	0.16
110	−2.54	13	−3.99	−3.90	0.09	1.85	1.99	106.04	−0.17	0.16
40	−2.61	11	−3.90	−3.81	0.09	1.84	1.99	106.35	−0.22	0.02
1	−2.64	11	−3.92	−3.81	0.11	1.86	1.99	106.53	−0.50	−0.01
0	−2.65	11	−3.92	−3.81	0.11	1.85	1.99	106.54	−0.50	−0.01
0	−2.65	11	−3.92	−3.81	0.11	1.86	1.99	106.55	−0.50	−0.01
0	−2.65	11	−3.92	−3.81	0.11	1.85	1.99	106.54	−0.50	−0.01

Table S21: Structural, energetic, and electronic properties for the CH_3/Cu_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	1	−2.03	1	−4.55	−4.12	0.43	2.39	1.92	109.38	−0.10	0.02
	1	−2.03	1	−4.55	−4.12	0.43	2.39	1.92	109.39	−0.10	0.02
	1	−2.03	1	−4.56	−4.12	0.43	2.39	1.92	109.39	−0.11	0.02
	0	−2.03	1	−4.55	−4.12	0.43	2.39	1.92	109.37	−0.10	0.02
	0	−2.03	1	−4.55	−4.12	0.43	2.39	1.92	109.38	−0.09	0.01
	0	−2.03	1	−4.55	−4.12	0.43	2.38	1.92	109.39	−0.12	0.01
	0	−2.03	1	−4.55	−4.12	0.43	2.39	1.92	109.37	−0.10	0.00
	0	−2.03	1	−4.55	−4.12	0.43	2.39	1.92	109.37	−0.10	0.01
5	570	−2.40	0	−4.39	−3.16	1.23	2.03	2.05	106.46	−2.81	−0.14
	569	−2.40	0	−4.39	−3.16	1.23	2.02	2.05	106.43	−2.76	−0.14
	569	−2.40	0	−4.39	−3.16	1.23	2.02	2.05	106.47	−2.75	−0.13
	569	−2.40	0	−4.39	−3.16	1.23	2.02	2.05	106.46	−2.71	−0.13
	569	−2.40	0	−4.39	−3.16	1.23	2.02	2.05	106.46	−2.79	−0.14
	1	−2.97	0	−4.55	−2.68	1.87	1.99	2.03	105.87	−0.14	−0.20
	1	−2.97	0	−4.55	−2.68	1.87	1.99	2.03	105.87	−0.14	−0.19

Table S21 continued from previous page

	0	−2.97	0	−4.55	−2.68	1.87	1.99	2.03	105.86	−0.14	−0.20
	0	−2.97	0	−4.55	−2.68	1.87	1.99	2.03	105.88	−0.14	−0.19
	0	−2.97	0	−4.55	−2.68	1.87	1.99	2.03	105.89	−0.14	−0.19
6	9	−1.60	1	−4.58	−4.24	0.34	2.48	1.92	111.05	0.20	0.42
	9	−1.61	1	−4.57	−4.23	0.34	2.48	1.92	111.04	0.20	0.43
	9	−1.61	1	−4.57	−4.24	0.34	2.48	1.92	111.05	0.19	0.42
	8	−1.61	1	−4.56	−4.22	0.34	2.48	1.92	111.03	0.20	0.43
	8	−1.61	1	−4.58	−4.24	0.34	2.48	1.92	111.05	0.19	0.42
	8	−1.61	1	−4.56	−4.22	0.34	2.48	1.92	111.03	0.20	0.43
	5	−1.61	1	−4.56	−4.22	0.34	2.48	1.92	111.03	0.20	0.43
	0	−1.61	1	−4.02	−3.57	0.45	2.02	2.06	106.87	−1.65	0.24
	0	−1.61	1	−4.02	−3.57	0.45	2.02	2.06	106.85	−1.62	0.25
	0	−1.61	1	−4.02	−3.57	0.45	2.02	2.06	106.84	−1.65	0.24
	0	−1.61	1	−4.02	−3.57	0.45	2.02	2.06	106.85	−1.63	0.25
	0	−1.61	1	−4.02	−3.57	0.45	2.02	2.06	106.88	−1.65	0.26
7	768	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.60	−0.15	0.31
	767	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.60	−0.14	0.31
	767	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.59	−0.14	0.31
	767	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.61	−0.15	0.31
	767	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.59	−0.14	0.31
	767	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.61	−0.14	0.31
	766	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.60	−0.15	0.31
	766	−1.68	2	−4.25	−4.04	0.21	2.49	1.92	110.61	−0.14	0.31
	519	−1.93	0	−4.35	−3.57	0.78	2.40	1.92	109.97	−3.16	0.18
	517	−1.93	0	−4.35	−3.57	0.78	2.40	1.92	109.93	−3.16	0.19
	3	−2.45	0	−4.78	−2.86	1.92	2.51	1.93	110.17	−0.22	0.16
	2	−2.45	0	−4.78	−2.86	1.92	2.51	1.93	110.19	−0.23	0.17
	2	−2.45	0	−4.78	−2.86	1.92	2.51	1.93	110.16	−0.22	0.16
	0	−2.45	0	−4.78	−2.86	1.92	2.51	1.93	110.17	−0.22	0.17
8	197	−1.53	1	−4.12	−3.76	0.35	2.51	1.94	110.60	−0.34	0.37
	196	−1.53	1	−4.12	−3.77	0.35	2.51	1.94	110.61	−0.35	0.38
	196	−1.53	1	−4.12	−3.77	0.35	2.51	1.94	110.61	−0.34	0.38
	196	−1.53	1	−4.12	−3.77	0.35	2.51	1.94	110.61	−0.36	0.37
	196	−1.53	1	−4.12	−3.77	0.35	2.51	1.94	110.61	−0.34	0.38
	195	−1.53	1	−4.12	−3.77	0.35	2.51	1.94	110.61	−0.34	0.37
	195	−1.53	1	−4.12	−3.77	0.35	2.51	1.94	110.61	−0.34	0.38
	1	−1.73	1	−4.31	−4.03	0.28	2.49	1.92	110.45	0.53	0.32
	1	−1.73	1	−4.32	−4.04	0.28	2.49	1.92	110.46	0.55	0.32
	1	−1.73	1	−4.32	−4.04	0.28	2.49	1.92	110.45	0.53	0.32
	1	−1.73	1	−4.31	−4.03	0.28	2.49	1.92	110.44	0.54	0.32
	1	−1.73	1	−4.31	−4.04	0.28	2.49	1.92	110.44	0.54	0.32
	1	−1.73	1	−4.31	−4.03	0.28	2.49	1.92	110.46	0.53	0.33
	0	−1.73	1	−4.32	−4.04	0.28	2.49	1.92	110.47	0.53	0.32
	0	−1.73	1	−4.31	−4.04	0.28	2.49	1.92	110.45	0.54	0.32
	0	−1.73	1	−4.31	−4.03	0.28	2.49	1.92	110.43	0.53	0.32
9	1012	−1.51	2	−4.21	−3.99	0.22	2.49	1.94	110.72	−0.30	0.31
	971	−1.55	2	−4.20	−3.95	0.25	2.50	1.94	110.71	−0.30	0.29
	969	−1.55	2	−4.20	−3.95	0.25	2.50	1.94	110.70	−0.30	0.29

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	856	−1.67	2	−4.26	−4.00	0.26	2.49	1.93	110.54	0.03	0.26
	855	−1.67	2	−4.26	−4.00	0.26	2.49	1.93	110.54	0.03	0.27
	184	−2.34	0	−4.54	−3.38	1.16	2.02	2.00	106.60	−0.88	−0.18
	184	−2.34	0	−4.54	−3.38	1.15	2.01	2.00	106.57	−0.89	−0.17
	125	−2.40	0	−4.32	−3.35	0.96	1.95	2.01	106.65	−2.57	−0.19
	124	−2.40	0	−4.18	−3.09	1.09	2.07	2.07	105.83	−0.24	−0.09
	101	−2.42	0	−4.77	−3.38	1.38	2.49	1.91	110.03	−0.29	−0.01
	100	−2.42	0	−4.77	−3.38	1.38	2.49	1.91	110.03	−0.29	−0.01
	100	−2.42	0	−4.77	−3.38	1.38	2.49	1.91	110.02	−0.29	−0.01
	100	−2.42	0	−4.77	−3.38	1.39	2.49	1.91	110.03	−0.29	−0.01
	46	−2.48	0	−4.80	−3.22	1.58	2.49	1.91	110.08	0.00	0.00
	45	−2.48	0	−4.80	−3.22	1.58	2.48	1.91	110.11	0.00	0.01
	4	−2.52	0	−4.20	−2.98	1.22	1.93	2.03	106.60	1.25	0.20
	1	−2.52	0	−4.19	−3.01	1.18	1.99	1.98	106.97	1.70	0.25
	0	−2.52	0	−4.19	−3.01	1.18	1.99	1.98	106.98	1.70	0.25
10	165	−1.82	1	−3.79	−3.51	0.28	2.48	1.93	109.59	−0.19	0.16
	165	−1.82	1	−3.79	−3.51	0.28	2.48	1.93	109.60	−0.17	0.15
	165	−1.82	1	−3.79	−3.51	0.28	2.48	1.93	109.60	−0.19	0.15
	164	−1.82	1	−3.79	−3.51	0.28	2.48	1.93	109.64	−0.17	0.15
	164	−1.82	1	−3.78	−3.50	0.28	2.48	1.93	109.64	−0.13	0.15
	164	−1.82	1	−3.79	−3.51	0.28	2.48	1.93	109.56	−0.17	0.16
	75	−1.91	1	−3.84	−3.58	0.26	1.93	2.05	106.10	−1.66	0.04
	74	−1.91	1	−3.84	−3.58	0.26	1.93	2.05	106.09	−1.65	0.04
	69	−1.92	1	−3.79	−3.54	0.24	1.95	2.00	106.43	−1.01	0.11
	69	−1.92	1	−3.79	−3.54	0.24	1.95	2.00	106.43	−1.02	0.11
	68	−1.92	1	−3.79	−3.54	0.24	1.95	2.00	106.42	−1.02	0.11
	68	−1.92	1	−3.79	−3.54	0.24	1.95	2.00	106.42	−1.02	0.11
	3	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.95	−0.52	−0.07
	2	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.94	−0.53	−0.07
	1	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.93	−0.53	−0.08
	1	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.95	−0.51	−0.07
	1	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.94	−0.52	−0.07
	0	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.93	−0.52	−0.07
	0	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.95	−0.51	−0.07
	0	−1.99	1	−3.82	−3.53	0.28	1.97	2.03	105.94	−0.50	−0.07
11	617	−1.89	2	−4.16	−4.07	0.09	2.00	2.04	106.82	−0.96	0.03
	615	−1.89	2	−4.17	−4.07	0.09	2.00	2.04	106.83	−0.99	0.03
	486	−2.02	0	−4.39	−3.70	0.69	2.41	1.94	110.19	0.03	0.19
	483	−2.02	0	−4.39	−3.69	0.69	2.42	1.94	110.15	0.04	0.19
	427	−2.08	0	−4.28	−3.45	0.83	2.01	2.00	107.23	−1.06	0.10
	426	−2.08	0	−4.28	−3.45	0.84	2.01	2.00	107.22	−1.05	0.11
	261	−2.24	0	−4.26	−3.38	0.88	1.96	2.02	107.16	−7.66	−0.35
	130	−2.37	0	−4.35	−3.56	0.79	1.94	1.98	106.59	−1.04	−0.01
	129	−2.37	0	−4.35	−3.56	0.79	1.94	1.98	106.57	−1.04	−0.01
	129	−2.37	0	−4.35	−3.56	0.79	1.94	1.98	106.58	−1.04	−0.01
	128	−2.37	0	−4.35	−3.56	0.79	1.94	1.98	106.57	−1.03	0.00
	128	−2.37	0	−4.35	−3.56	0.78	1.94	1.98	106.59	−1.03	0.00
	127	−2.38	0	−4.35	−3.56	0.79	1.94	1.98	106.58	−1.03	0.00
	1	−2.50	0	−4.38	−3.23	1.15	1.96	2.01	106.29	−0.85	−0.06

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	1	−2.50	0	−4.38	−3.23	1.15	1.96	2.01	106.31	−0.82	−0.05
	1	−2.50	0	−4.38	−3.23	1.14	1.96	2.01	106.29	−0.84	−0.05
	1	−2.50	0	−4.38	−3.23	1.14	1.96	2.01	106.30	−0.84	−0.05
	0	−2.50	0	−4.38	−3.23	1.14	1.96	2.01	106.28	−0.84	−0.05
	0	−2.50	0	−4.38	−3.23	1.14	1.96	2.01	106.28	−0.85	−0.05
	0	−2.50	0	−4.38	−3.23	1.14	1.95	2.01	106.27	−0.83	−0.05
	0	−2.50	0	−4.38	−3.23	1.15	1.95	2.01	106.28	−0.83	−0.05
	0	−2.50	0	−4.38	−3.23	1.15	1.96	2.01	106.29	−0.85	−0.05
12	438	−1.58	1	−4.30	−4.05	0.25	2.48	1.94	110.80	0.08	0.20
	438	−1.58	1	−4.30	−4.05	0.25	2.48	1.94	110.79	0.08	0.21
	437	−1.58	1	−4.29	−4.04	0.25	2.48	1.94	110.77	0.08	0.20
	436	−1.58	1	−4.29	−4.04	0.25	2.48	1.94	110.77	0.08	0.21
	330	−1.69	1	−4.02	−3.81	0.21	2.02	2.04	106.77	−2.22	0.00
	150	−1.87	1	−3.83	−3.60	0.23	2.05	2.10	105.49	−1.25	0.09
	129	−1.89	1	−4.04	−3.77	0.27	1.96	2.02	107.38	−2.35	0.14
	119	−1.90	1	−4.09	−3.84	0.26	2.03	1.98	107.66	−1.83	0.17
	119	−1.90	1	−4.09	−3.84	0.26	2.03	1.98	107.67	−1.86	0.16
	119	−1.90	1	−4.10	−3.84	0.26	2.03	1.98	107.68	−1.85	0.16
	118	−1.90	1	−4.09	−3.84	0.26	2.03	1.98	107.68	−1.87	0.16
	118	−1.90	1	−4.10	−3.84	0.26	2.03	1.98	107.68	−1.85	0.17
	118	−1.90	1	−4.09	−3.84	0.26	2.03	1.98	107.68	−1.85	0.17
	118	−1.90	1	−4.10	−3.84	0.26	2.03	1.98	107.69	−1.86	0.16
	118	−1.90	1	−4.09	−3.84	0.26	2.03	1.98	107.67	−1.86	0.17
	118	−1.90	1	−4.09	−3.84	0.26	2.03	1.98	107.68	−1.84	0.16
	116	−1.90	1	−4.36	−4.16	0.20	2.47	1.92	110.18	0.41	0.13
	115	−1.90	1	−4.36	−4.16	0.20	2.48	1.92	110.17	0.40	0.13
	115	−1.90	1	−4.36	−4.16	0.20	2.48	1.92	110.18	0.40	0.13
	115	−1.90	1	−4.37	−4.17	0.20	2.48	1.92	110.14	0.38	0.12
	115	−1.90	1	−4.36	−4.16	0.20	2.48	1.92	110.15	0.40	0.12
	45	−1.97	1	−4.26	−4.05	0.21	1.99	1.99	106.76	−0.25	0.09
	1	−2.02	1	−3.83	−3.57	0.25	2.00	2.01	106.74	−0.06	0.05
	0	−2.02	1	−3.83	−3.57	0.25	2.00	2.01	106.76	−0.06	0.05
13	661	−1.65	2	−4.08	−3.95	0.14	2.49	1.93	110.30	0.03	0.16
	661	−1.65	2	−4.08	−3.95	0.13	2.49	1.93	110.31	0.02	0.16
	660	−1.65	2	−4.08	−3.96	0.13	2.49	1.93	110.30	0.01	0.16
	162	−2.15	0	−4.32	−3.53	0.79	2.44	1.92	109.84	−0.04	0.07
	162	−2.15	0	−4.32	−3.53	0.79	2.44	1.92	109.86	−0.03	0.07
	161	−2.15	0	−4.32	−3.54	0.79	2.44	1.92	109.84	−0.02	0.07
	125	−2.18	0	−4.20	−3.43	0.77	1.97	2.00	106.83	−1.23	0.08
	125	−2.18	0	−4.20	−3.43	0.77	1.97	2.00	106.84	−1.24	0.08
	125	−2.18	0	−4.20	−3.43	0.77	1.97	2.00	106.84	−1.22	0.08
	125	−2.18	0	−4.20	−3.43	0.77	1.97	2.00	106.85	−1.23	0.08
	125	−2.19	0	−4.20	−3.43	0.77	1.97	2.00	106.86	−1.24	0.08
	125	−2.19	0	−4.20	−3.43	0.77	1.97	2.00	106.82	−1.22	0.08
	124	−2.19	0	−4.20	−3.43	0.77	1.97	2.00	106.86	−1.23	0.08
	124	−2.19	0	−4.20	−3.43	0.77	1.97	2.00	106.83	−1.22	0.08
	80	−2.23	0	−4.55	−3.50	1.05	2.52	1.94	109.49	0.14	0.07
	79	−2.23	0	−4.55	−3.50	1.05	2.52	1.94	109.49	0.14	0.07
	48	−2.26	0	−4.30	−3.64	0.66	2.01	1.98	106.79	−0.45	0.04

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	48	−2.26	0	−4.30	−3.64	0.66	2.01	1.98	106.76	−0.45	0.04
	47	−2.26	0	−4.30	−3.64	0.66	2.01	1.98	106.79	−0.47	0.04
	47	−2.26	0	−4.30	−3.64	0.66	2.01	1.98	106.76	−0.44	0.04
	46	−2.26	0	−4.30	−3.64	0.66	2.01	1.98	106.77	−0.45	0.04
	46	−2.26	0	−4.54	−3.67	0.87	2.47	1.92	109.95	0.05	0.04
	45	−2.26	0	−4.54	−3.67	0.88	2.47	1.92	109.96	0.03	0.04
	45	−2.26	0	−4.54	−3.67	0.87	2.48	1.92	109.95	0.06	0.04
	45	−2.27	0	−4.54	−3.67	0.88	2.47	1.92	109.95	0.03	0.04
	0	−2.31	0	−4.39	−3.34	1.05	2.05	2.06	106.00	−4.56	−0.46
14	124	−1.68	1	−4.28	−4.08	0.20	2.49	1.93	110.41	0.48	0.16
	124	−1.68	1	−4.28	−4.08	0.20	2.49	1.93	110.40	0.49	0.16
	124	−1.68	1	−4.28	−4.08	0.20	2.49	1.93	110.39	0.49	0.16
	123	−1.68	1	−4.28	−4.07	0.20	2.50	1.93	110.40	0.48	0.16
	123	−1.68	1	−4.28	−4.08	0.20	2.49	1.93	110.39	0.49	0.16
	114	−1.69	1	−4.23	−4.04	0.20	2.43	1.93	110.26	0.46	0.17
	114	−1.69	1	−4.23	−4.03	0.20	2.43	1.93	110.24	0.45	0.17
	114	−1.69	1	−4.22	−4.03	0.20	2.42	1.93	110.22	0.46	0.17
	113	−1.69	1	−4.22	−4.02	0.20	2.42	1.93	110.20	0.45	0.16
	112	−1.69	1	−4.20	−4.00	0.20	2.43	1.94	110.17	0.42	0.16
	102	−1.70	1	−4.10	−3.89	0.22	2.49	1.92	110.39	0.48	0.16
	68	−1.73	1	−3.94	−3.75	0.19	2.03	1.98	107.70	−1.23	0.05
	68	−1.73	1	−3.93	−3.74	0.19	2.03	1.98	107.68	−1.21	0.05
	68	−1.73	1	−3.94	−3.75	0.19	2.03	1.98	107.70	−1.25	0.05
	68	−1.73	1	−3.93	−3.75	0.19	2.03	1.98	107.69	−1.21	0.05
	68	−1.73	1	−3.94	−3.75	0.19	2.03	1.98	107.70	−1.25	0.05
	68	−1.73	1	−3.93	−3.75	0.19	2.03	1.98	107.70	−1.24	0.05
	68	−1.73	1	−3.94	−3.75	0.19	2.03	1.98	107.70	−1.23	0.05
	67	−1.73	1	−3.94	−3.75	0.19	2.03	1.98	107.72	−1.25	0.05
	67	−1.73	1	−3.93	−3.75	0.19	2.03	1.98	107.67	−1.24	0.05
	19	−1.78	1	−3.93	−3.76	0.17	1.96	2.07	106.90	−1.70	−0.01
	18	−1.78	1	−3.94	−3.76	0.17	1.96	2.07	106.91	−1.70	−0.01
	18	−1.78	1	−3.93	−3.76	0.17	1.96	2.07	106.88	−1.70	−0.01
	18	−1.78	1	−3.93	−3.76	0.17	1.96	2.07	106.89	−1.66	−0.01
	1	−1.80	1	−3.95	−3.78	0.17	1.97	2.00	107.11	−0.85	0.07
	1	−1.80	1	−3.95	−3.78	0.17	1.97	2.00	107.10	−0.87	0.06
	0	−1.80	1	−3.96	−3.78	0.17	1.97	2.00	107.10	−0.86	0.06
	0	−1.80	1	−3.96	−3.78	0.17	1.97	2.00	107.11	−0.88	0.06
15	590	−1.71	0	−4.22	−3.82	0.40	2.54	1.96	109.28	−1.72	−0.01
	588	−1.71	0	−4.22	−3.82	0.40	2.54	1.96	109.28	−1.75	−0.02
	588	−1.71	0	−4.22	−3.82	0.40	2.54	1.96	109.28	−1.76	−0.01
	365	−1.93	0	−4.30	−3.89	0.41	2.51	1.93	109.41	0.02	0.10
	365	−1.94	0	−4.30	−3.89	0.41	2.51	1.93	109.40	0.03	0.09
	365	−1.94	0	−4.30	−3.89	0.42	2.51	1.93	109.43	0.05	0.09
	365	−1.94	0	−4.30	−3.89	0.41	2.51	1.93	109.42	0.03	0.09
	365	−1.94	0	−4.30	−3.89	0.41	2.51	1.93	109.41	0.04	0.10
	364	−1.94	0	−4.30	−3.89	0.41	2.51	1.93	109.41	0.02	0.10
	364	−1.94	0	−4.30	−3.89	0.41	2.51	1.93	109.41	0.05	0.10
	361	−1.94	0	−4.31	−3.90	0.41	2.51	1.94	109.69	0.76	0.13
	361	−1.94	0	−4.30	−3.90	0.41	2.51	1.94	109.68	0.77	0.13

Table S21 continued from previous page

361	−1.94	0	−4.31	−3.90	0.41	2.51	1.94	109.68	0.77	0.13
360	−1.94	0	−4.30	−3.90	0.41	2.51	1.94	109.68	0.76	0.13
334	−1.97	0	−4.27	−3.62	0.65	2.02	2.00	106.99	−2.91	−0.18
100	−2.20	0	−4.26	−3.46	0.80	2.01	1.97	107.28	−0.19	0.09
99	−2.20	0	−4.26	−3.46	0.80	2.02	1.97	107.32	−0.20	0.08
98	−2.20	0	−4.26	−3.46	0.80	2.02	1.97	107.33	−0.20	0.09
94	−2.21	0	−4.44	−3.49	0.95	2.48	1.93	109.95	0.37	0.11
93	−2.21	0	−4.44	−3.49	0.95	2.48	1.93	109.94	0.37	0.11
93	−2.21	0	−4.44	−3.48	0.95	2.48	1.93	109.93	0.37	0.10
92	−2.21	0	−4.44	−3.48	0.95	2.48	1.93	109.93	0.37	0.10
92	−2.21	0	−4.44	−3.49	0.95	2.48	1.93	109.94	0.36	0.10
92	−2.21	0	−4.44	−3.49	0.95	2.48	1.93	109.95	0.36	0.10
92	−2.21	0	−4.44	−3.49	0.95	2.48	1.93	109.94	0.37	0.10
91	−2.21	0	−4.44	−3.49	0.95	2.48	1.93	109.95	0.37	0.11
91	−2.21	0	−4.44	−3.48	0.95	2.48	1.93	109.95	0.37	0.10
1	−2.30	0	−4.51	−3.60	0.91	2.48	1.92	109.80	0.62	0.13
0	−2.30	0	−4.51	−3.59	0.92	2.48	1.92	109.83	0.59	0.13
0	−2.30	0	−4.51	−3.59	0.92	2.48	1.92	109.82	0.60	0.13

- For each TM_n cluster the site preference present a particular behaviour, despite that CH_3 – TM interaction occurs between the C and the TM atom that CH_3 is directly bonded.
- CH_3 binds to the TM atom via bridge or top coordination.

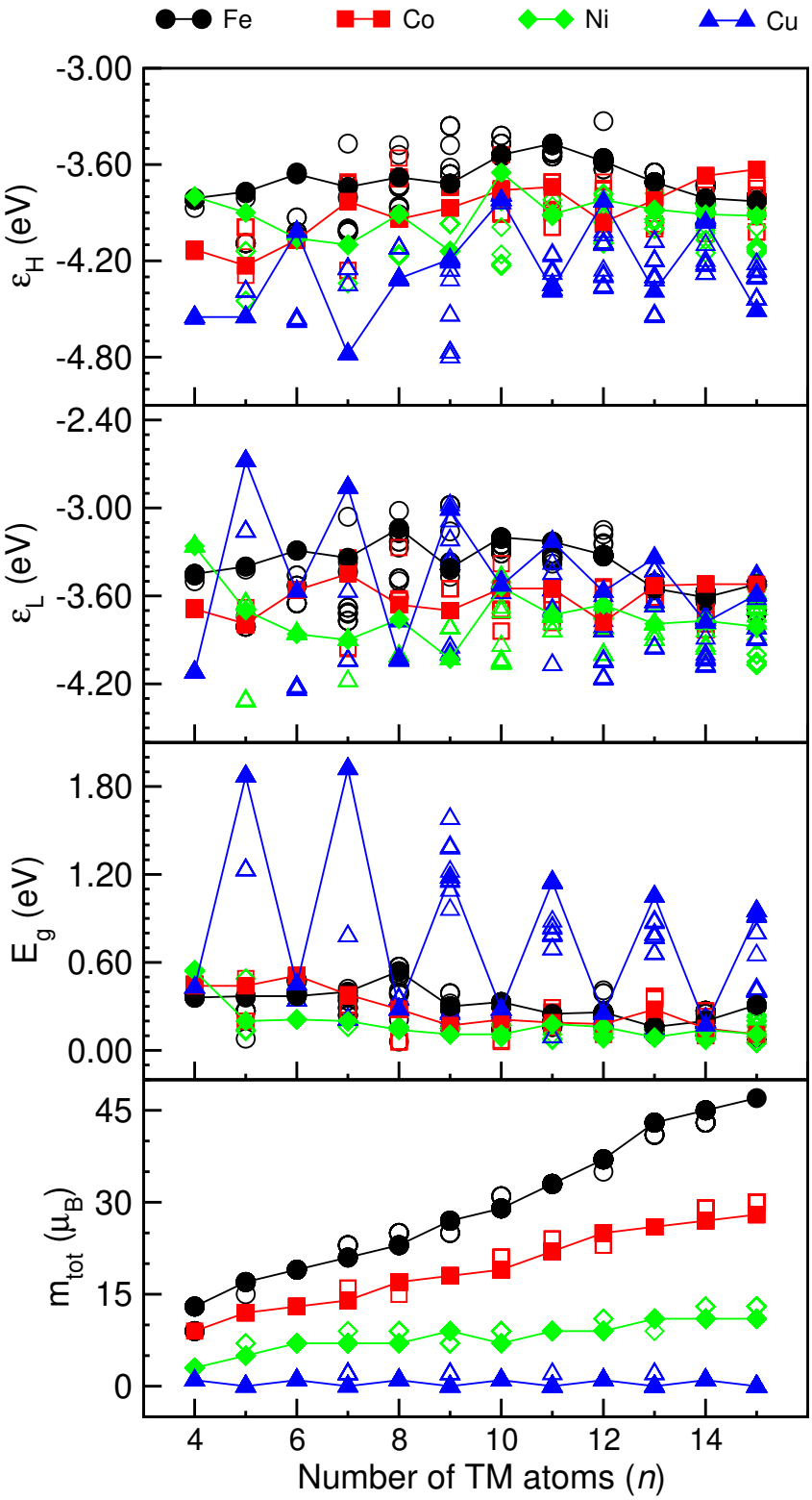

Figure S10: Energetic and electronic properties for CH_3/TM_n systems from from $n = 4 - 15$ where TM = Fe, Co, Ni, Cu.

- For CH_3/TM_n systems the magnitude of ε_{homo} increases with the d-states, where Cu_n/Fe_n presents the highest/smallest values, with few exceptions cases, i.e., Cu_6. The same behavior is observed for ε_{lumo}.

- Conversely the magnitude of E_g decreases with the occupation of the d-states from Fe_n to Ni_n, where in general Cu_n presents the highest magnitude.

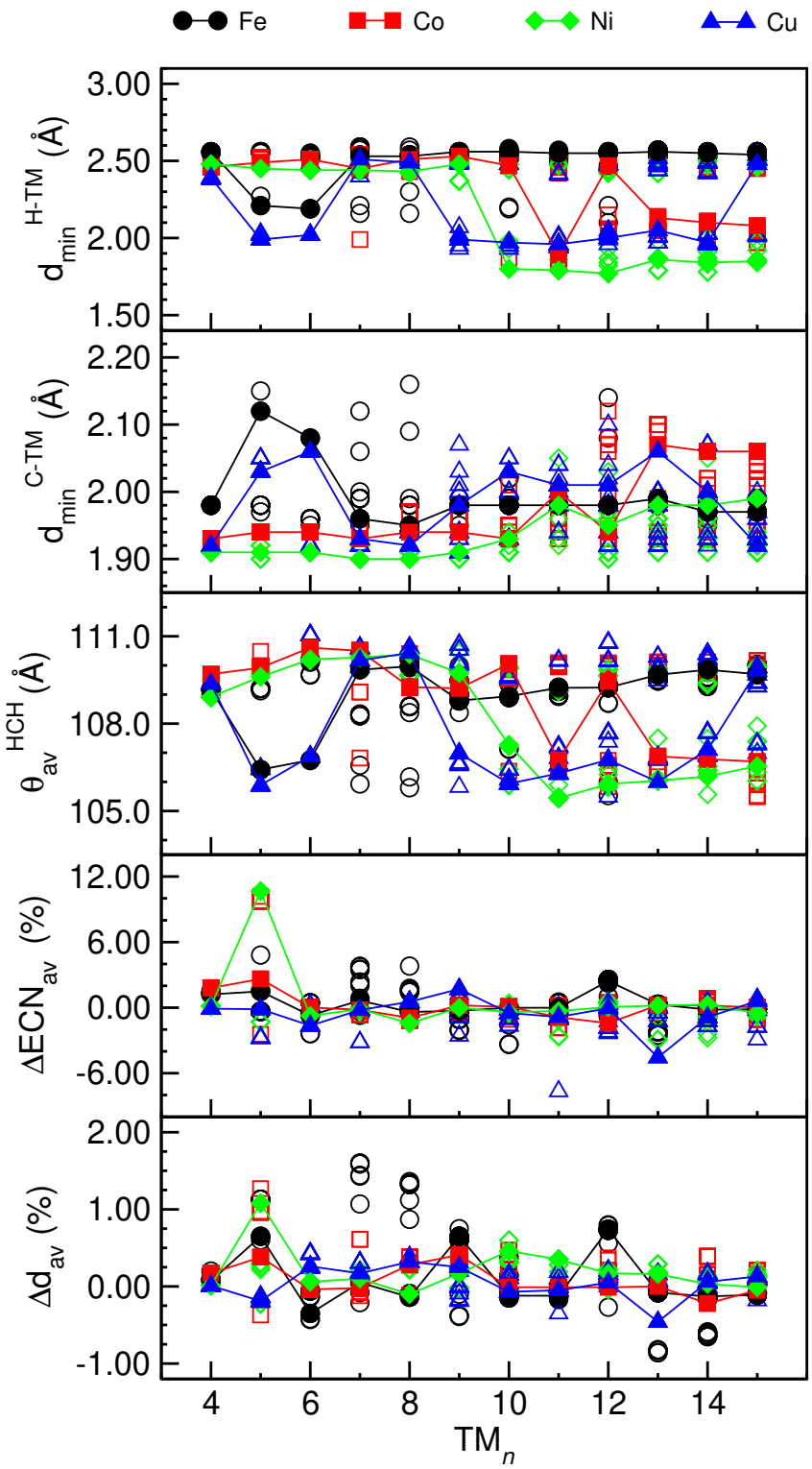

Figure S11: Structural properties for CH_3/TM_n systems from from $n = 4 - 15$ where TM = Fe, Co, Ni, Cu.

- For Ni_n and Co_n clusters we have a similar behaviour of $d_{min}{}^{H-TM}$, i.e., it is almost constant from $n = 4$ to $n = 9(10)$ (Co), for the higher values of n it decreases about

0.75 Å, except for Co_{14}. Oppositely, $d_{min}{}^{H-TM}$ has the same behavior on Cu_n and Fe_n clusters for n = 4 to n = 8. However, from Cu_9 the magnitude decreases but remains almost the same for the remaining sizes.

- The tendencies of $d_{min}{}^{C-TM}$ magnitude is opposite than the one observed for $d_{min}{}^{C-TM}$. Where Ni_n clusters present the smallest values. Thus, the magnitude of $d_{min}{}^{C-TM}$ is smaller than $d_{min}{}^{H-TM}$, characterizing the CH_3-TM interaction via C atom, however, the exception occurs for $Cu_{5/6}$ and $Fe_{5/6}$, which those parameters have almost the same magnitude.

- The average HCH bond angle follows the same behaviour than $d_{min}{}^{H-TM}$, and capture the exceptions, explaining the tendencies observed on the structural properties.

- The largest structural deformations are observed for Co_6 and Ni_6 clusters.

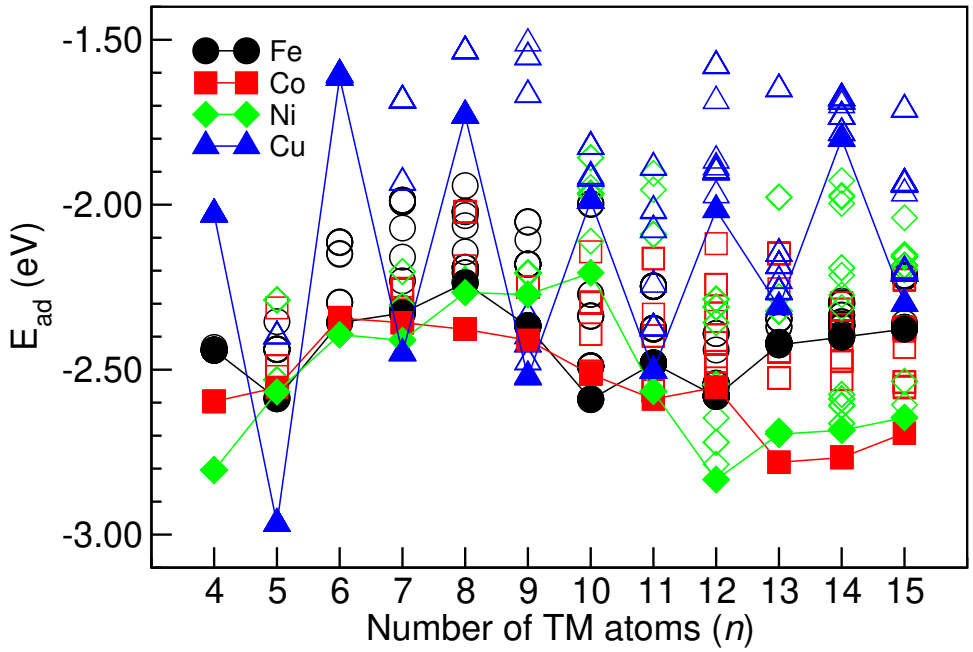

Figure S12: Adsorption energy for the CH_3/TM_n systems where $n = 4-15$ and TM = Fe, Co, Ni, and Cu.

- The adsorption of CH_3 on TM_n has a greater magnitude than CH_4, which the first one has a phyisisorption nature, i.e., below -0.5 eV in magnitude, while the last one, presents $|E_{ad}|$ at least three times greater, characterizing a chemisorption nature process.

- Different from CH_4, the adsorption of CH_3 does not present a clear behaviour with the increasing of n.

- The highest value of E_ad is observed for Cu_5 while Ni_4 and Ni_{13} have the second highest magnitudes. For $n > 12$, there is competition between Ni and Co clusters.

- For Cu_n, the E_{ad} has the highest values for $n = 5$, 7 and 9. Thus, the adsorption of CH_3 is less favorable on Cu_n with an even n.

9 CH_3+H Adsorption on TM_n Clusters

Table S22: Structural, energetic, and electronic properties for the (CH_3+H)/Fe_n system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{H^a-TM}$), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	$d_{min}^{H^a-TM}$ (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	1	−5.62	12	−3.84	−3.47	0.37	2.53	1.99	3.62	108.49	11.39	0.10
	1	−5.62	12	−3.84	−3.47	0.37	2.53	1.99	3.62	108.53	11.38	0.11
	0	−5.62	12	−3.84	−3.47	0.37	2.53	1.99	3.62	108.50	11.38	0.11
	0	−5.62	12	−3.84	−3.47	0.37	2.53	1.99	3.62	108.54	11.37	0.10
5	320	−5.37	16	−4.10	−3.65	0.45	2.54	1.98	3.41	109.45	1.54	0.70
	269	−5.42	16	−4.12	−3.66	0.46	2.54	1.98	3.44	109.48	1.65	0.69
	0	−5.69	16	−3.81	−3.42	0.40	2.22	2.10	3.64	106.05	−1.33	−0.15
	0	−5.69	16	−3.81	−3.42	0.39	2.21	2.09	3.64	106.05	−1.35	−0.14
6	435	−4.96	18	−3.85	−3.49	0.36	2.53	1.97	2.78	109.61	−0.10	−0.25
	434	−4.96	18	−3.85	−3.49	0.36	2.54	1.97	2.79	109.59	−0.10	−0.24
	258	−5.13	18	−3.56	−2.96	0.60	2.18	2.08	3.15	106.92	−0.15	−0.57
	1	−5.39	18	−3.71	−3.36	0.35	2.17	2.09	3.79	106.24	−1.43	−0.84
	1	−5.39	18	−3.71	−3.36	0.35	2.17	2.08	3.79	106.27	−1.34	−0.82
	0	−5.39	18	−3.71	−3.36	0.35	2.17	2.09	3.79	106.24	−1.31	−0.82
7	821	−4.36	22	−4.16	−3.83	0.33	2.58	1.99	5.23	108.33	3.97	1.66
	158	−5.02	20	−3.75	−3.07	0.68	2.50	1.96	2.83	110.18	−0.42	−0.45
	96	−5.08	22	−3.72	−3.32	0.40	2.22	2.13	3.79	106.14	0.12	0.67
	94	−5.08	22	−3.72	−3.31	0.40	2.19	2.13	3.79	106.11	0.06	0.67
	93	−5.08	22	−3.72	−3.32	0.41	2.20	2.13	3.79	106.12	0.06	0.67
	86	−5.09	22	−3.82	−3.41	0.41	2.18	2.09	3.66	106.32	1.09	0.99
	0	−5.18	20	−3.54	−2.76	0.78	2.17	2.05	3.79	106.15	−0.88	−0.68
8	850	−4.51	24	−3.66	−3.32	0.34	2.52	1.97	2.62	109.51	0.27	0.88
	799	−4.56	24	−3.78	−3.42	0.37	2.49	1.96	2.77	109.98	2.39	1.17
	553	−4.80	22	−3.63	−3.04	0.59	2.50	1.95	2.96	110.07	0.25	−0.24
	341	−5.01	24	−3.58	−2.99	0.59	2.21	2.08	3.75	105.93	−0.20	0.58
	226	−5.13	24	−3.67	−3.21	0.46	2.18	2.04	3.70	106.17	0.25	0.68
	183	−5.17	24	−3.64	−3.16	0.48	2.13	2.04	3.68	106.23	2.46	0.85
	52	−5.30	24	−3.59	−3.04	0.55	2.29	2.11	3.71	105.98	3.53	1.02
	0	−5.36	24	−3.62	−3.09	0.54	2.12	2.07	3.70	106.27	3.03	1.00
9	457	−4.88	26	−3.68	−3.51	0.18	2.49	1.96	2.73	109.56	−1.04	0.34
	440	−4.90	26	−3.58	−3.19	0.39	2.51	1.99	3.09	108.89	−1.17	0.38
	439	−4.90	26	−3.57	−3.19	0.38	2.51	1.99	3.09	108.90	−1.17	0.38
	435	−4.91	26	−3.58	−3.18	0.39	2.51	1.99	3.13	108.84	−1.26	0.37
	374	−4.97	26	−3.75	−3.40	0.35	2.50	1.96	2.97	109.63	−0.63	0.43

Table S22 continued from previous page

	92	−5.25	26	−3.44	−3.03	0.41	2.19	2.08	3.64	106.37	−0.86	0.58
	5	−5.34	26	−3.47	−3.10	0.37	2.12	2.02	3.69	106.65	−2.76	−0.09
	0	−5.34	26	−3.48	−3.16	0.32	2.14	2.02	3.71	106.72	−2.96	−0.05
10	766	−4.85	30	−3.49	−3.29	0.19	2.54	1.99	3.74	109.50	−5.10	−0.24
	765	−4.85	30	−3.48	−3.29	0.19	2.54	1.99	3.74	109.51	−5.06	−0.23
	764	−4.85	30	−3.49	−3.29	0.19	2.54	1.99	3.73	109.51	−5.12	−0.24
	544	−5.07	32	−3.59	−3.40	0.19	2.15	2.04	4.83	106.75	−2.21	0.81
	365	−5.25	30	−3.69	−3.40	0.30	2.52	1.98	3.04	109.33	−2.76	0.02
	365	−5.25	30	−3.69	−3.40	0.30	2.52	1.98	3.05	109.34	−2.72	0.02
	4	−5.61	30	−3.49	−3.19	0.30	2.11	2.07	3.65	106.00	−0.82	0.36
	2	−5.62	30	−3.49	−3.19	0.30	2.11	2.07	3.65	105.96	−0.80	0.36
	0	−5.62	30	−3.49	−3.19	0.30	2.11	2.07	3.65	105.98	−0.80	0.35
11	422	−4.96	34	−3.65	−3.52	0.13	2.53	1.99	3.90	108.83	−0.62	0.35
	421	−4.96	34	−3.65	−3.52	0.13	2.54	1.99	3.87	108.84	−0.57	0.36
	420	−4.96	34	−3.65	−3.52	0.13	2.53	1.99	3.89	108.86	−0.57	0.36
	244	−5.14	34	−3.59	−3.46	0.12	2.54	1.98	2.98	109.34	−0.57	0.40
	210	−5.17	34	−3.58	−3.45	0.13	2.51	1.98	2.94	109.58	−0.25	0.43
	158	−5.22	32	−3.66	−3.31	0.36	2.52	1.98	4.27	109.06	−1.07	−0.40
	156	−5.23	32	−3.66	−3.30	0.36	2.52	1.98	4.21	109.09	−1.12	−0.42
	156	−5.23	32	−3.66	−3.31	0.36	2.52	1.98	4.27	109.04	−1.18	−0.41
	131	−5.25	32	−3.59	−3.30	0.29	2.54	1.98	3.15	109.37	−1.90	−0.45
	2	−5.38	32	−3.40	−3.02	0.38	2.09	2.12	3.63	105.60	−3.24	−0.61
	0	−5.38	32	−3.40	−3.03	0.38	2.10	2.12	3.63	105.60	−3.24	−0.61
12	244	−5.25	38	−3.66	−3.42	0.24	2.50	1.98	2.97	109.55	2.03	1.18
	121	−5.37	38	−3.66	−3.36	0.30	2.09	2.07	3.39	105.77	0.47	0.98
	120	−5.37	38	−3.66	−3.36	0.31	2.09	2.07	3.38	105.76	0.48	0.98
	72	−5.42	36	−3.54	−3.13	0.41	2.10	2.12	3.31	105.66	−1.05	0.08
	30	−5.46	36	−3.61	−3.33	0.28	2.53	1.97	4.15	109.31	0.96	0.36
	27	−5.46	36	−3.61	−3.33	0.28	2.53	1.97	4.16	109.33	1.02	0.36
	0	−5.49	36	−3.42	−3.16	0.26	2.02	2.09	3.76	105.30	0.21	0.31
13	169	−5.16	42	−3.74	−3.47	0.27	2.50	1.98	3.00	109.89	−1.95	−0.41
	163	−5.17	42	−3.74	−3.47	0.27	2.49	1.98	3.04	109.82	−2.01	−0.42
	163	−5.17	42	−3.73	−3.46	0.27	2.49	1.98	3.06	109.77	−2.00	−0.43
	135	−5.20	42	−3.75	−3.43	0.32	2.52	1.98	3.10	109.71	−1.66	−0.37
	134	−5.20	42	−3.75	−3.43	0.32	2.53	1.98	3.12	109.68	−1.65	−0.37
	48	−5.28	42	−3.64	−3.31	0.33	2.08	2.08	3.57	105.96	−1.88	−0.37
	47	−5.28	42	−3.64	−3.32	0.33	2.08	2.08	3.57	105.97	−1.92	−0.38
	5	−5.33	42	−3.79	−3.52	0.27	2.53	1.98	4.16	109.83	−0.92	−0.36
	4	−5.33	42	−3.79	−3.53	0.26	2.52	1.98	4.18	109.83	−0.95	−0.35
	4	−5.33	42	−3.79	−3.53	0.26	2.52	1.98	4.20	109.79	−0.96	−0.36
	3	−5.33	42	−3.79	−3.53	0.26	2.52	1.98	4.19	109.83	−0.95	−0.36
	1	−5.33	42	−3.79	−3.53	0.26	2.53	1.98	4.17	109.81	−0.93	−0.36
	0	−5.33	42	−3.79	−3.53	0.26	2.52	1.98	4.18	109.83	−0.94	−0.36
14	453	−5.14	44	−3.81	−3.51	0.30	2.54	1.98	2.94	110.08	−0.17	−0.27
	452	−5.14	44	−3.81	−3.51	0.30	2.54	1.98	2.95	110.08	−0.18	−0.26
	419	−5.18	44	−3.81	−3.52	0.28	2.51	1.99	2.96	110.06	−0.74	−0.33
	419	−5.18	44	−3.84	−3.53	0.31	2.51	1.98	3.07	109.65	−0.59	−0.21
	419	−5.18	44	−3.82	−3.51	0.31	2.52	1.98	2.88	110.19	−0.73	−0.33

Table S22 continued from previous page

	417	−5.18	44	−3.82	−3.51	0.31	2.53	1.98	2.89	110.17	−0.71	−0.34
	417	−5.18	44	−3.82	−3.51	0.31	2.52	1.98	2.89	110.19	−0.75	−0.34
	415	−5.18	44	−3.84	−3.55	0.29	2.50	1.97	3.00	109.75	−0.90	−0.30
	368	−5.23	44	−3.85	−3.66	0.19	2.54	1.97	3.88	109.88	−0.52	−0.24
	274	−5.32	44	−3.85	−3.48	0.37	2.53	1.97	3.04	109.57	−0.39	−0.24
	2	−5.59	44	−3.72	−3.40	0.33	2.07	2.06	3.63	105.97	−0.66	−0.29
	0	−5.60	44	−3.72	−3.40	0.33	2.07	2.06	3.63	105.97	−0.66	−0.29
	0	−5.60	42	−3.79	−3.53	0.26	2.52	1.98	4.18	109.83	−0.94	−0.36
15	150	−5.02	48	−3.89	−3.56	0.33	2.54	1.99	2.87	109.90	−0.21	0.16
	150	−5.02	48	−3.89	−3.56	0.33	2.54	1.99	2.88	109.90	−0.21	0.16
	149	−5.02	48	−3.88	−3.56	0.33	2.54	1.99	2.89	109.86	−0.21	0.15
	149	−5.02	48	−3.89	−3.56	0.33	2.54	1.99	2.89	109.88	−0.22	0.16
	149	−5.02	48	−3.89	−3.56	0.33	2.54	1.99	2.88	109.89	−0.23	0.16
	149	−5.02	48	−3.89	−3.56	0.33	2.54	1.99	2.87	109.88	−0.21	0.16
	149	−5.02	48	−3.88	−3.56	0.33	2.54	1.99	2.90	109.87	−0.21	0.16
	148	−5.02	48	−3.88	−3.55	0.33	2.53	1.99	2.90	109.86	−0.22	0.16
	147	−5.02	48	−3.89	−3.56	0.33	2.54	1.99	2.89	109.87	−0.21	0.16
	146	−5.02	48	−3.88	−3.56	0.33	2.53	1.99	2.89	109.86	−0.22	0.15
	113	−5.05	46	−3.85	−3.53	0.31	2.51	1.97	4.02	109.84	−0.77	−0.29
	113	−5.05	46	−3.85	−3.54	0.31	2.52	1.97	4.03	109.81	−0.78	−0.30
	109	−5.06	46	−3.85	−3.53	0.31	2.52	1.96	4.03	109.83	−0.77	−0.29
	108	−5.06	46	−3.85	−3.54	0.31	2.52	1.97	4.02	109.84	−0.78	−0.29
	0	−5.17	46	−3.85	−3.59	0.26	2.52	1.97	4.08	109.73	−0.32	−0.21

Table S23: Structural, energetic, and electronic properties for the $(CH_3+H)/Co_n$ system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{H^a-TM}$), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	$d_{min}^{H^a-TM}$ (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	8	−5.43	8	−3.88	−3.33	0.55	2.49	1.96	3.45	108.66	14.42	2.07
	2	−5.44	8	−3.90	−3.33	0.56	2.47	1.95	3.44	108.73	14.48	2.09
	1	−5.44	8	−3.88	−3.32	0.56	2.47	1.95	3.45	108.68	14.38	2.06
	0	−5.44	8	−3.93	−3.34	0.59	2.46	1.95	3.43	108.83	14.47	2.14
5	82	−5.18	11	−4.14	−3.81	0.33	2.50	1.95	3.00	109.86	8.70	0.78
	82	−5.18	11	−4.14	−3.81	0.33	2.50	1.95	3.00	109.86	8.70	0.78
	80	−5.18	11	−4.14	−3.80	0.33	2.50	1.95	2.98	109.87	8.72	0.79
	43	−5.22	11	−3.82	−3.55	0.27	2.12	2.05	3.70	106.22	−1.11	0.13
	0	−5.26	11	−4.01	−3.70	0.31	2.50	1.94	3.59	109.25	4.19	0.50
6	52	−4.75	12	−3.83	−3.44	0.38	2.46	1.95	3.34	109.45	−0.32	0.26
	52	−4.76	12	−3.83	−3.45	0.38	2.45	1.95	3.34	109.47	−0.31	0.26
	50	−4.76	12	−3.84	−3.45	0.39	2.46	1.95	3.32	109.56	−0.31	0.26

Table S23 continued from previous page

	49	−4.76	12	−3.84	−3.46	0.39	2.46	1.95	3.30	109.60	−0.29	0.25
	0	−4.81	12	−3.66	−3.30	0.36	2.04	2.04	3.72	106.01	−1.20	0.33
	0	−4.81	12	−3.66	−3.30	0.37	2.04	2.04	3.72	106.05	−1.22	0.33
7	588	−4.69	13	−3.81	−3.50	0.31	2.48	1.96	2.57	109.87	−0.95	−0.15
	587	−4.69	13	−3.81	−3.50	0.31	2.47	1.96	2.56	109.94	−0.95	−0.15
	481	−4.80	15	−3.91	−3.55	0.35	2.32	1.93	3.60	108.97	−1.00	0.51
	464	−4.82	15	−3.93	−3.57	0.36	2.40	1.93	3.59	109.35	−1.08	0.51
	286	−5.00	13	−3.60	−3.42	0.18	2.41	1.93	3.41	109.96	−1.05	0.07
	182	−5.10	15	−4.12	−3.77	0.34	2.43	1.94	3.52	108.54	−1.11	0.47
	0	−5.28	15	−3.93	−3.60	0.33	2.04	1.98	3.48	106.45	−0.88	0.36
8	548	−4.87	16	−3.86	−3.65	0.21	2.42	1.93	4.27	110.58	−1.70	−0.06
	411	−5.00	16	−3.74	−3.56	0.18	2.03	2.02	4.82	106.65	−1.20	0.04
	411	−5.00	16	−3.74	−3.56	0.18	2.03	2.02	4.82	106.62	−1.22	0.04
	410	−5.00	16	−3.74	−3.56	0.18	2.03	2.02	4.84	106.64	−1.29	0.04
	296	−5.12	16	−3.66	−3.42	0.24	1.92	2.01	3.71	106.18	−0.95	0.42
	0	−5.41	16	−3.73	−3.44	0.29	2.09	2.00	3.60	106.82	−2.77	0.07
9	231	−4.94	19	−3.88	−3.66	0.22	2.08	2.00	3.36	106.41	−3.93	0.23
	231	−4.94	19	−3.88	−3.66	0.22	2.08	2.00	3.36	106.40	−3.87	0.23
	231	−4.94	19	−3.88	−3.66	0.22	2.08	2.00	3.35	106.38	−3.92	0.23
	29	−5.14	17	−3.61	−3.35	0.26	1.96	2.04	3.65	105.64	−2.37	0.14
	27	−5.14	17	−3.61	−3.35	0.26	1.96	2.04	3.65	105.64	−2.30	0.16
	27	−5.14	17	−3.61	−3.35	0.26	1.96	2.04	3.65	105.66	−2.31	0.14
	2	−5.17	17	−3.78	−3.62	0.16	2.47	1.94	3.54	108.67	−0.56	0.38
	0	−5.17	17	−3.78	−3.63	0.15	2.46	1.94	3.53	108.70	−0.57	0.37
10	706	−4.90	20	−3.85	−3.68	0.17	2.49	1.96	3.50	108.91	−0.61	0.37
	698	−4.90	20	−3.70	−3.52	0.18	2.11	2.03	3.59	106.34	−1.29	0.27
	697	−4.90	20	−3.81	−3.56	0.25	1.82	2.02	3.29	106.10	−1.82	0.35
	508	−5.09	20	−3.88	−3.71	0.16	2.46	1.94	3.21	110.08	−0.66	0.37
	299	−5.30	20	−3.84	−3.58	0.25	2.43	1.95	3.59	109.08	0.00	0.39
	297	−5.31	20	−3.84	−3.59	0.25	2.42	1.95	3.58	109.14	0.00	0.39
	239	−5.36	20	−3.77	−3.61	0.16	1.91	2.02	3.38	105.97	−0.45	0.44
	238	−5.36	20	−3.78	−3.61	0.16	1.91	2.02	3.38	105.99	−0.46	0.43
	169	−5.43	20	−3.79	−3.53	0.26	2.03	2.02	3.64	106.15	−1.17	0.26
	0	−5.60	18	−3.56	−3.35	0.20	1.83	1.97	3.67	106.22	0.04	0.03
11	513	−4.99	23	−3.88	−3.71	0.17	2.52	1.96	4.71	109.86	−2.15	−0.03
	421	−5.08	21	−3.83	−3.60	0.23	2.49	1.93	2.79	110.68	−0.38	−0.06
	420	−5.08	21	−3.83	−3.60	0.23	2.49	1.93	2.81	110.68	−0.40	−0.08
	348	−5.15	23	−3.94	−3.74	0.20	2.45	1.94	3.61	109.00	−0.29	0.27
	304	−5.19	21	−3.63	−3.50	0.13	1.86	1.98	3.41	106.60	−1.06	−0.11
	304	−5.20	21	−3.63	−3.49	0.13	1.86	1.98	3.41	106.59	−1.04	−0.11
	266	−5.23	21	−3.67	−3.51	0.16	1.97	2.01	3.40	107.17	−1.09	−0.04
	265	−5.23	21	−3.67	−3.51	0.16	1.96	2.01	3.41	107.16	−1.11	−0.05
	120	−5.38	21	−3.75	−3.55	0.20	1.93	2.04	3.61	105.58	−0.73	−0.15
	0	−5.50	21	−3.70	−3.54	0.16	1.91	2.02	3.61	105.61	−0.31	0.08
12	724	−5.16	24	−3.78	−3.62	0.17	2.06	1.99	3.40	106.92	−0.87	0.16
	688	−5.20	26	−3.92	−3.73	0.18	2.06	2.05	3.59	106.32	−1.26	0.42

Table S23 continued from previous page

	688	−5.20	26	−3.92	−3.73	0.18	2.06	2.04	3.59	106.36	−1.30	0.42
	563	−5.33	24	−3.95	−3.78	0.18	2.49	1.94	3.89	110.21	−0.20	0.22
	563	−5.33	24	−3.95	−3.78	0.17	2.49	1.94	3.88	110.21	−0.19	0.22
	562	−5.33	24	−3.95	−3.78	0.18	2.49	1.94	3.88	110.21	−0.21	0.22
	473	−5.42	24	−3.74	−3.63	0.12	1.97	1.99	3.67	105.79	−0.05	0.26
	454	−5.44	24	−3.99	−3.71	0.28	2.49	1.95	4.04	109.79	0.36	0.33
	449	−5.44	24	−3.81	−3.60	0.21	2.02	2.02	3.64	106.12	0.10	0.32
	447	−5.44	24	−3.81	−3.66	0.15	2.00	2.00	3.65	106.06	0.23	0.27
	268	−5.62	24	−3.82	−3.65	0.17	1.94	2.02	3.57	106.19	−0.55	0.12
	0	−5.89	24	−3.85	−3.71	0.15	1.98	2.00	3.56	106.48	−0.14	0.18
13	570	−5.06	27	−3.90	−3.67	0.22	2.02	2.01	3.48	106.49	−0.57	0.23
	514	−5.11	25	−3.75	−3.63	0.12	2.48	1.94	3.38	109.73	−0.08	0.07
	267	−5.36	25	−3.85	−3.54	0.31	2.13	2.07	3.18	106.58	−0.09	−0.07
	265	−5.36	25	−3.85	−3.54	0.31	2.13	2.07	3.19	106.57	−0.07	−0.07
	265	−5.36	25	−3.85	−3.54	0.31	2.13	2.07	3.18	106.58	−0.08	−0.07
	264	−5.36	25	−3.85	−3.54	0.31	2.13	2.07	3.20	106.54	−0.06	−0.06
	115	−5.51	27	−3.94	−3.67	0.27	2.00	1.98	3.68	105.92	−0.03	0.24
	114	−5.51	27	−3.94	−3.66	0.27	2.00	1.98	3.68	105.96	−0.04	0.24
	113	−5.51	27	−3.94	−3.67	0.27	2.00	1.98	3.68	105.91	−0.03	0.24
	112	−5.52	27	−3.94	−3.66	0.27	2.00	1.98	3.68	105.96	−0.05	0.23
	2	−5.63	25	−3.70	−3.54	0.16	1.98	2.03	3.56	106.78	0.37	0.06
	0	−5.63	25	−3.70	−3.54	0.16	1.98	2.03	3.56	106.78	0.37	0.06
	0	−5.63	25	−3.70	−3.54	0.16	1.98	2.03	3.56	106.78	0.38	0.06
14	641	−4.92	28	−3.87	−3.73	0.15	2.44	1.96	2.75	109.83	−0.87	0.30
	536	−5.02	26	−3.77	−3.62	0.15	2.46	1.92	2.84	110.48	0.12	0.04
	438	−5.12	28	−3.90	−3.74	0.16	2.45	1.95	3.42	109.15	0.17	0.31
	379	−5.18	28	−3.84	−3.68	0.17	2.38	1.94	3.50	108.95	−0.36	0.25
	339	−5.22	28	−3.93	−3.73	0.20	2.01	2.01	4.64	106.36	0.03	0.19
	311	−5.25	28	−3.84	−3.70	0.14	2.03	2.00	3.63	106.27	−0.31	0.40
	253	−5.30	28	−3.81	−3.67	0.13	2.05	2.04	3.64	106.40	0.49	0.57
	249	−5.31	28	−3.82	−3.65	0.17	2.03	2.04	3.65	106.27	−0.13	0.48
	186	−5.37	28	−3.82	−3.64	0.18	1.99	1.98	3.66	106.03	−0.13	0.23
	102	−5.46	26	−3.71	−3.56	0.15	1.90	2.01	3.55	106.35	0.38	0.05
	100	−5.46	26	−3.70	−3.56	0.15	1.90	2.01	3.55	106.31	0.38	0.05
	100	−5.46	26	−3.71	−3.56	0.15	1.90	2.01	3.55	106.34	0.38	0.05
	1	−5.56	28	−3.76	−3.63	0.13	1.98	1.98	3.69	105.88	−0.15	0.18
	0	−5.56	28	−3.76	−3.63	0.13	1.98	1.98	3.69	105.94	−0.19	0.18
15	644	−5.00	29	−3.72	−3.56	0.16	1.99	2.07	3.04	105.40	−0.44	0.32
	642	−5.00	29	−3.72	−3.56	0.16	1.99	2.07	3.04	105.40	−0.45	0.32
	620	−5.03	31	−3.88	−3.74	0.13	2.04	2.01	3.32	106.86	−0.57	0.42
	547	−5.10	29	−3.86	−3.72	0.14	2.48	1.94	3.41	109.43	−0.46	0.19
	429	−5.22	29	−3.72	−3.61	0.11	2.04	2.03	3.14	106.56	−0.48	0.26
	428	−5.22	29	−3.72	−3.61	0.10	2.04	2.03	3.14	106.55	−0.48	0.26
	382	−5.26	29	−3.91	−3.72	0.19	2.51	1.94	3.98	109.87	−0.02	0.14
	345	−5.30	29	−3.77	−3.58	0.18	2.06	2.02	3.56	106.31	−1.34	0.09
	344	−5.30	29	−3.77	−3.58	0.18	2.06	2.02	3.56	106.33	−1.34	0.09
	267	−5.38	29	−3.78	−3.64	0.14	2.02	2.03	3.64	106.35	−0.40	0.24
	267	−5.38	29	−3.78	−3.64	0.14	2.02	2.03	3.64	106.35	−0.38	0.24

Table S23 continued from previous page

	141	−5.50	29	−3.80	−3.64	0.16	1.99	2.05	3.67	105.85	0.16	0.17
	141	−5.51	29	−3.80	−3.64	0.16	1.97	2.04	3.67	105.76	0.15	0.18
	18	−5.63	29	−3.91	−3.70	0.21	1.94	2.01	3.56	105.51	−0.03	0.15
	0	−5.65	29	−3.75	−3.61	0.14	1.93	2.00	3.60	106.30	−0.24	0.11

Table S24: Structural, energetic, and electronic properties for the $(CH_3+H)/Ni_n$ system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{H^a-TM}$), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	$d_{min}^{H^a-TM}$ (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	381	−5.01	2	−3.83	−3.46	0.36	2.44	1.90	2.31	109.90	0.36	0.37
	380	−5.01	2	−3.83	−3.46	0.36	2.43	1.90	2.31	109.91	0.39	0.37
	2	−5.39	2	−4.04	−3.56	0.48	2.47	1.91	2.72	109.37	0.20	1.09
	0	−5.39	2	−4.04	−3.56	0.48	2.47	1.91	2.72	109.38	0.21	1.11
5	109	−5.25	6	−4.39	−3.90	0.49	2.44	1.91	3.48	109.84	4.14	1.16
	108	−5.25	6	−4.39	−3.90	0.49	2.44	1.91	3.47	109.86	4.09	1.16
	2	−5.35	4	−3.96	−3.71	0.25	2.43	1.92	2.74	109.67	11.01	1.41
	0	−5.35	4	−3.96	−3.71	0.25	2.43	1.92	2.73	109.67	11.01	1.43
	0	−5.36	4	−3.96	−3.71	0.25	2.43	1.92	2.73	109.69	11.01	1.41
6	30	−5.17	6	−4.11	−3.88	0.23	2.38	1.91	3.10	110.34	−0.92	0.58
	28	−5.17	6	−4.11	−3.88	0.23	2.37	1.91	3.11	110.32	−0.94	0.55
	28	−5.17	6	−4.11	−3.88	0.23	2.37	1.91	3.13	110.28	−0.97	0.56
	28	−5.17	6	−4.11	−3.87	0.23	2.37	1.91	3.11	110.33	−0.97	0.56
	2	−5.19	6	−4.00	−3.81	0.19	2.40	1.91	3.43	109.84	−0.41	0.64
	0	−5.20	6	−4.01	−3.82	0.19	2.40	1.91	3.43	109.90	−0.38	0.62
7	364	−4.87	6	−4.08	−3.90	0.18	2.45	1.92	2.44	110.51	−0.53	0.32
	357	−4.88	6	−4.08	−3.90	0.18	2.45	1.92	2.46	110.48	−0.55	0.31
	160	−5.07	8	−4.27	−4.17	0.10	2.43	1.90	3.46	109.35	−0.43	0.56
	95	−5.14	6	−4.03	−3.86	0.17	2.41	1.90	4.57	110.18	−0.67	0.24
	91	−5.14	6	−4.03	−3.85	0.17	2.41	1.90	4.56	110.18	−0.70	0.23
	3	−5.23	6	−3.84	−3.64	0.20	1.81	1.99	3.76	105.80	−5.12	0.40
	0	−5.23	6	−3.87	−3.77	0.10	2.40	1.90	3.61	109.32	−1.40	0.46
8	785	−4.75	8	−4.20	−4.01	0.19	2.47	1.90	2.68	110.64	0.05	0.30
	616	−4.92	6	−3.99	−3.77	0.22	2.37	1.90	4.13	110.43	−0.33	0.11
	332	−5.20	6	−3.82	−3.66	0.16	1.85	1.99	3.52	106.27	−2.48	0.32
	332	−5.21	6	−3.82	−3.66	0.16	1.85	1.99	3.52	106.28	−2.52	0.30
	29	−5.51	8	−4.06	−3.89	0.17	1.94	1.99	3.59	105.72	−0.86	0.18
	29	−5.51	8	−4.06	−3.89	0.17	1.94	1.99	3.59	105.73	−0.88	0.17
	28	−5.51	8	−4.06	−3.89	0.17	1.94	1.99	3.59	105.68	−0.88	0.18
	0	−5.54	8	−4.07	−3.83	0.24	1.82	2.02	3.59	105.92	−0.48	0.29

Table S24 continued from previous page

9	675	−4.73	8	−4.15	−3.97	0.18	2.44	1.92	3.59	110.29	−0.11	0.23
	516	−4.89	6	−3.97	−3.79	0.18	2.38	1.91	2.99	109.96	−0.72	0.23
	512	−4.89	6	−3.97	−3.79	0.18	2.38	1.91	2.96	110.05	−0.73	0.22
	458	−4.94	6	−3.98	−3.82	0.17	2.43	1.89	4.67	110.72	0.25	0.19
	325	−5.08	6	−3.96	−3.81	0.16	1.86	1.94	3.54	106.04	0.29	0.32
	325	−5.08	6	−3.96	−3.81	0.16	1.86	1.94	3.54	106.05	0.31	0.32
	248	−5.15	8	−4.09	−3.99	0.09	2.42	1.91	3.55	108.87	−0.24	0.46
	246	−5.16	8	−4.09	−3.99	0.10	2.41	1.91	3.56	108.80	−0.26	0.46
	0	−5.40	8	−4.00	−3.87	0.13	1.93	1.96	3.58	105.96	−0.35	0.34
10	949	−4.40	8	−4.27	−3.99	0.27	1.89	1.92	5.44	107.33	0.36	0.70
	671	−4.68	6	−3.65	−3.55	0.10	1.80	1.93	4.50	107.07	−0.36	0.87
	307	−5.04	8	−4.10	−3.85	0.25	2.41	1.90	3.46	108.98	−0.37	0.47
	1	−5.35	8	−3.93	−3.70	0.23	1.90	1.93	3.57	106.07	−2.01	0.36
	1	−5.35	8	−3.93	−3.70	0.23	1.90	1.93	3.57	106.13	−1.95	0.37
	1	−5.35	8	−3.93	−3.69	0.24	1.90	1.93	3.57	106.12	−2.01	0.35
	0	−5.35	8	−3.94	−3.72	0.22	1.88	1.94	3.56	105.96	−0.92	0.41
11	816	−4.61	6	−3.69	−3.57	0.12	1.78	1.98	3.03	106.38	−2.24	0.59
	622	−4.80	8	−4.02	−3.80	0.22	2.49	1.92	2.53	109.86	−0.80	0.38
	619	−4.80	8	−4.01	−3.80	0.22	2.48	1.92	2.51	109.90	−0.79	0.38
	367	−5.06	8	−3.84	−3.73	0.11	1.77	2.01	3.62	105.49	−2.84	0.38
	3	−5.42	8	−3.81	−3.70	0.11	1.75	1.97	3.58	105.42	−0.70	0.56
	1	−5.42	8	−3.80	−3.70	0.11	1.75	1.97	3.58	105.40	−0.73	0.55
	1	−5.42	8	−3.80	−3.70	0.11	1.75	1.97	3.58	105.42	−0.72	0.56
	1	−5.42	8	−3.80	−3.70	0.11	1.75	1.97	3.58	105.40	−0.74	0.56
	1	−5.42	8	−3.81	−3.70	0.11	1.75	1.97	3.58	105.43	−0.71	0.56
	0	−5.42	8	−3.80	−3.70	0.11	1.75	1.97	3.58	105.45	−0.71	0.56
	0	−5.42	8	−3.80	−3.70	0.11	1.75	1.97	3.58	105.41	−0.72	0.56
12	876	−4.99	8	−3.77	−3.67	0.10	1.90	1.95	3.35	106.62	−25.58	−6.47
	394	−5.47	10	−3.97	−3.90	0.07	1.89	1.94	3.16	106.57	0.62	0.30
	387	−5.47	8	−3.88	−3.79	0.10	1.81	1.93	3.17	106.85	0.30	0.18
	387	−5.47	8	−3.88	−3.79	0.10	1.81	1.93	3.16	106.85	0.30	0.19
	386	−5.48	8	−3.88	−3.79	0.10	1.81	1.93	3.16	106.85	0.32	0.19
	361	−5.50	10	−3.95	−3.93	0.03	1.84	1.96	3.16	106.07	0.46	0.30
	317	−5.54	8	−3.85	−3.71	0.14	1.81	1.93	3.41	106.56	−0.28	0.31
	193	−5.67	8	−3.82	−3.70	0.11	1.79	1.98	3.59	105.69	0.09	0.22
	117	−5.74	8	−3.82	−3.72	0.10	1.83	1.98	3.53	106.21	0.78	0.20
	2	−5.86	10	−4.00	−3.89	0.11	1.87	1.98	3.53	105.89	0.50	0.21
	1	−5.86	10	−4.00	−3.89	0.11	1.87	1.98	3.54	105.91	0.50	0.22
	0	−5.86	10	−4.00	−3.88	0.11	1.87	1.98	3.53	105.90	0.50	0.22
13	884	−4.76	12	−4.14	−4.00	0.14	2.52	1.96	3.84	109.71	−2.86	0.07
	720	−4.92	10	−4.01	−3.90	0.10	2.51	1.96	3.74	109.54	−2.48	−0.03
	449	−5.20	10	−3.89	−3.75	0.14	1.87	1.96	3.63	106.05	−1.30	0.38
	324	−5.32	10	−3.91	−3.82	0.10	1.82	1.96	3.03	106.28	−0.43	0.47
	258	−5.39	10	−3.94	−3.84	0.09	1.80	2.00	3.49	106.29	−1.34	0.21
	253	−5.39	10	−3.92	−3.83	0.09	1.89	1.97	3.48	106.31	−0.63	0.28
	238	−5.41	10	−3.93	−3.85	0.09	1.82	1.93	3.48	106.82	−0.36	0.37
	161	−5.48	10	−3.99	−3.87	0.12	1.89	1.99	3.24	106.17	−0.49	0.15
	47	−5.60	10	−3.95	−3.85	0.09	1.91	1.96	3.39	106.28	−0.83	0.17

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	47	−5.60	10	−3.95	−3.85	0.09	1.91	1.96	3.39	106.29	−0.81	0.17
	3	−5.64	10	−3.90	−3.82	0.08	1.83	1.98	3.56	106.09	−0.48	0.28
	2	−5.64	10	−3.91	−3.82	0.08	1.83	1.98	3.56	106.09	−0.43	0.28
	0	−5.64	10	−3.91	−3.82	0.08	1.83	1.98	3.56	106.09	−0.47	0.28
14	1134	−4.77	12	−3.95	−3.84	0.11	1.88	2.06	3.04	106.49	−2.22	0.22
	956	−4.94	12	−4.07	−3.98	0.09	2.53	1.96	3.73	109.67	−2.19	−0.06
	954	−4.95	12	−4.01	−3.88	0.13	1.81	2.05	2.98	105.88	−1.14	0.26
	628	−5.27	10	−3.89	−3.77	0.13	2.07	1.96	3.43	107.61	−0.39	0.11
	492	−5.41	10	−3.88	−3.80	0.08	1.84	1.93	3.48	107.01	−0.14	0.16
	490	−5.41	10	−3.88	−3.80	0.08	1.84	1.93	3.48	107.04	−0.10	0.16
	475	−5.43	10	−3.90	−3.78	0.12	1.79	1.98	3.56	106.06	−0.21	0.20
	464	−5.44	10	−3.93	−3.83	0.10	1.83	1.95	3.33	106.50	−0.88	0.10
	355	−5.55	10	−3.94	−3.83	0.10	1.86	1.95	3.02	106.59	0.18	0.07
	333	−5.57	10	−3.93	−3.79	0.14	1.96	1.97	3.32	106.69	−0.39	0.04
	298	−5.60	10	−3.95	−3.82	0.13	1.88	1.98	3.15	106.72	−0.14	0.05
	233	−5.67	12	−4.05	−3.93	0.11	1.90	2.02	3.56	106.04	−0.04	0.16
	227	−5.67	12	−4.06	−3.95	0.11	1.90	1.98	3.51	106.40	0.34	0.24
	0	−5.90	10	−3.94	−3.80	0.15	1.94	2.01	3.54	106.48	0.25	−0.03
15	888	−4.94	10	−3.93	−3.80	0.13	2.01	1.94	4.35	108.20	−0.80	0.10
	884	−4.94	12	−4.00	−3.89	0.11	2.00	1.96	6.16	107.22	−1.12	0.19
	458	−5.37	10	−3.94	−3.82	0.12	1.88	1.98	3.13	106.77	−1.10	0.07
	361	−5.47	12	−4.02	−3.94	0.08	1.95	2.03	3.08	106.76	−0.65	0.00
	360	−5.47	12	−4.02	−3.94	0.08	1.95	2.03	3.09	106.72	−0.64	0.01
	288	−5.54	10	−3.91	−3.82	0.09	1.84	1.96	3.34	106.61	−0.92	0.04
	286	−5.54	10	−3.91	−3.82	0.09	1.84	1.95	3.33	106.63	−0.93	0.04
	284	−5.54	10	−3.89	−3.79	0.10	1.85	1.96	3.55	106.56	−0.32	0.14
	52	−5.78	10	−3.97	−3.86	0.10	1.90	1.97	3.51	106.43	−0.15	0.02
	35	−5.79	10	−3.96	−3.85	0.11	1.84	1.99	3.54	106.14	−0.23	0.03
	33	−5.79	10	−3.96	−3.85	0.11	1.83	1.99	3.54	106.14	−0.22	0.03
	31	−5.80	12	−4.00	−3.89	0.11	1.90	1.96	3.50	106.87	0.24	0.27
	30	−5.80	12	−4.00	−3.89	0.11	1.90	1.96	3.50	106.87	0.24	0.27
	30	−5.80	12	−4.00	−3.89	0.11	1.90	1.96	3.50	106.86	0.24	0.27
	0	−5.83	12	−4.02	−3.91	0.11	1.81	1.99	3.55	106.10	0.24	0.25

Table S25: Structural, energetic, and electronic properties for the $(CH_3+H)/Cu_n$ system: relative total energy (ΔE_{tot}), adsorption energy (E_{ad}) total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the molecular hydrogen, H^m, to the nearest TM atom ($d_{min}^{H^m-TM}$), minimum distance of the carbon atom to the nearest TM atom (d_{min}^{C-TM}), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{H^a-TM}$), average HCH bond angle (θ_{av}^{HCH}), changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (Å)	$d_{min}^{H^m-TM}$ (Å)	d_{min}^{C-TM} (Å)	$d_{min}^{H^a-TM}$ (Å)	θ_{av}^{HCH} (°)	ΔECN_{av} (%)	Δd_{av} (%)
4	1	−4.92	0	−4.82	−3.20	1.62	2.51	1.95	2.81	109.64	0.20	0.58
	1	−4.92	0	−4.82	−3.20	1.63	2.49	1.95	2.77	109.70	0.19	0.60

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	0	−4.92	0	−4.82	−3.20	1.62	2.49	1.95	2.78	109.66	0.19	0.55
5	88	−0.38	1	−4.23	−3.72	0.51	2.03	2.02	3.54	106.55	−4.99	0.56
	88	−0.38	1	−4.23	−3.72	0.51	2.03	2.03	3.54	106.55	−4.81	0.54
	62	−0.41	1	−4.24	−3.72	0.52	1.96	2.02	3.52	105.71	−4.24	0.54
	62	−0.41	1	−4.24	−3.72	0.52	1.96	2.02	3.51	105.71	−4.15	0.53
	0	−0.47	1	−4.31	−3.87	0.44	1.97	1.96	3.57	105.72	−2.16	0.36
6	1	−0.54	0	−4.56	−2.90	1.65	1.98	2.00	3.55	105.62	−9.66	0.12
	0	−0.54	0	−4.55	−2.90	1.65	1.98	2.00	3.55	105.66	−9.52	0.15
	0	−0.54	0	−4.53	−2.90	1.64	1.99	2.00	3.55	105.71	−8.35	0.22
	0	−0.54	0	−4.54	−2.90	1.63	1.99	2.00	3.55	105.66	−8.50	0.25
7	475	−4.48	1	−4.11	−3.81	0.30	2.50	1.93	3.59	109.04	−1.21	0.77
	474	−4.48	1	−4.11	−3.81	0.30	2.49	1.93	3.59	109.04	−1.25	0.77
	417	−4.53	1	−4.16	−3.85	0.31	2.48	1.93	3.54	109.47	−2.36	0.70
	376	−4.57	1	−4.19	−3.85	0.34	2.50	1.93	4.04	110.21	−0.86	0.37
	142	−4.81	1	−3.97	−3.56	0.41	1.95	2.01	3.60	105.78	−12.80	−0.47
	1	−4.95	1	−3.87	−3.51	0.36	2.05	2.44	1.11	109.22	0.11	0.11
	0	−4.95	1	−3.87	−3.51	0.36	2.05	2.44	1.11	109.23	0.12	0.12
8	378	0.23	0	−4.40	−3.04	1.36	1.90	2.01	3.63	106.15	−10.09	−0.25
	134	−0.01	0	−4.23	−3.21	1.02	1.96	2.02	3.62	105.87	−1.89	−0.29
	75	−0.07	0	−4.69	−3.13	1.55	2.42	1.91	3.52	108.57	4.95	0.77
	13	−0.13	0	−4.49	−2.97	1.52	1.87	2.00	3.59	106.09	−3.26	0.09
	12	−0.13	0	−4.49	−2.97	1.52	1.88	2.00	3.59	106.11	−3.29	0.09
	3	−0.14	0	−4.49	−3.02	1.48	2.02	1.97	3.58	105.62	0.81	0.44
	2	−0.14	0	−4.49	−3.01	1.48	2.02	1.97	3.58	105.64	0.72	0.43
	0	−0.15	0	−4.49	−3.02	1.48	2.02	1.97	3.58	105.62	0.66	0.44
9	767	0.36	1	−4.07	−3.78	0.29	1.95	2.03	3.27	106.65	−2.24	−0.01
	571	0.16	1	−3.96	−3.65	0.31	1.97	2.00	3.53	106.74	−3.36	0.32
	566	0.16	1	−4.25	−4.01	0.23	1.96	2.03	3.61	106.15	−2.65	0.15
	394	−0.01	1	−4.24	−3.95	0.29	1.93	2.02	3.59	106.19	−0.83	0.32
	363	−0.04	1	−3.76	−3.37	0.39	1.89	2.03	3.58	106.19	−1.50	0.19
	205	−0.20	1	−4.38	−4.11	0.27	2.01	2.01	3.60	105.66	−3.12	0.16
	204	−0.20	1	−4.39	−4.12	0.27	2.00	2.01	3.60	105.62	−2.74	0.19
	14	−0.39	1	−4.07	−3.81	0.26	2.01	2.03	3.58	105.85	4.83	0.75
	0	−0.41	1	−4.09	−3.83	0.26	1.96	2.00	3.56	105.92	4.99	0.71
10	674	0.22	0	−4.12	−3.55	0.57	1.98	2.01	4.88	106.40	−2.28	−0.03
	673	0.22	0	−4.12	−3.55	0.57	1.97	2.01	4.89	106.39	−2.25	−0.02
	499	0.05	0	−4.10	−3.14	0.96	1.89	2.02	3.58	105.94	−8.35	−0.18
	403	−0.05	0	−4.12	−3.12	1.00	1.92	2.01	3.57	105.81	−8.98	−0.24
	403	−0.05	0	−4.12	−3.12	1.00	1.91	2.01	3.57	105.79	−8.94	−0.24
	402	−0.05	0	−4.12	−3.12	0.99	1.91	2.01	3.57	105.80	−8.96	−0.24
	382	−0.07	0	−4.56	−3.18	1.38	2.44	1.92	3.52	109.00	−0.40	0.33
	381	−0.07	0	−4.56	−3.18	1.38	2.45	1.92	3.52	108.97	−0.40	0.33
	378	−0.08	0	−4.56	−3.18	1.38	2.44	1.92	3.52	108.93	−0.41	0.32
	0	−0.45	0	−4.48	−3.19	1.29	1.96	2.00	3.58	105.60	−1.58	0.06
11	752	0.67	1	−4.33	−4.13	0.20	1.99	1.99	4.13	107.34	−1.16	0.47
	277	0.20	1	−3.85	−3.60	0.26	1.94	2.01	3.55	106.69	−7.62	−0.18
	196	0.12	1	−4.24	−4.01	0.23	2.00	2.01	3.57	106.29	−2.10	0.29

Table S25 continued from previous page

	111	0.03	1	−4.31	−4.11	0.20	2.05	1.99	3.30	106.87	−1.84	0.09
	110	0.03	1	−4.31	−4.11	0.20	2.05	1.99	3.30	106.90	−1.86	0.09
	110	0.03	1	−4.31	−4.11	0.20	2.06	1.99	3.30	106.87	−1.94	0.09
	108	0.03	1	−4.07	−3.84	0.23	2.06	1.99	3.37	107.13	−1.47	0.10
	65	−0.01	1	−4.26	−4.06	0.20	2.16	1.96	3.35	107.87	−1.78	0.08
	44	−0.04	1	−4.10	−3.85	0.25	2.02	1.99	4.21	107.49	−4.58	0.04
	43	−0.04	1	−4.10	−3.84	0.25	2.02	1.99	4.23	107.45	−4.49	0.05
	0	−0.08	1	−4.28	−4.05	0.23	1.97	2.01	4.07	107.01	−7.75	−0.23
12	1005	0.84	2	−3.75	−3.65	0.10	2.03	2.10	3.49	105.41	−2.47	0.16
	861	0.69	2	−3.87	−3.77	0.10	2.00	2.00	3.36	106.67	−0.86	0.29
	344	0.18	0	−4.14	−3.45	0.69	2.11	1.96	4.05	107.91	−2.60	0.18
	340	0.17	0	−4.50	−3.75	0.75	2.46	1.92	3.53	109.04	−1.17	0.39
	334	0.17	0	−4.29	−3.57	0.72	2.13	2.02	3.48	106.47	−2.29	0.14
	288	0.12	0	−4.45	−3.42	1.03	2.12	2.04	3.44	106.17	−7.32	−0.38
	8	−0.16	0	−4.51	−3.24	1.26	1.95	2.03	3.74	107.09	−4.15	0.04
	8	−0.16	0	−4.51	−3.24	1.26	1.95	2.03	3.75	107.08	−4.25	0.04
	7	−0.16	0	−4.51	−3.25	1.26	1.95	2.03	3.75	107.07	−4.31	0.02
	7	−0.16	0	−4.51	−3.24	1.26	1.94	2.03	3.74	107.07	−4.13	0.04
	0	−0.17	0	−4.31	−3.51	0.80	2.01	2.01	3.55	106.69	−2.13	0.14
	0	−0.17	0	−4.31	−3.48	0.82	1.97	2.03	3.60	106.62	−2.04	0.15
13	437	−4.44	1	−3.88	−3.62	0.26	1.99	2.06	3.26	106.62	−4.06	0.10
	311	−4.56	1	−4.02	−3.81	0.21	2.07	2.05	3.28	106.84	−1.23	0.09
	309	−4.56	1	−4.02	−3.81	0.21	2.06	2.06	3.29	106.83	−1.27	0.09
	251	−4.62	1	−4.04	−3.86	0.18	1.92	2.02	3.36	106.64	−5.34	−0.33
	250	−4.62	1	−4.04	−3.86	0.18	1.93	2.02	3.36	106.68	−5.31	−0.33
	239	−4.63	1	−3.89	−3.65	0.25	1.99	2.02	3.59	106.22	−2.12	0.06
	236	−4.64	1	−3.89	−3.65	0.25	1.99	2.02	3.59	106.21	−2.03	0.08
	201	−4.67	1	−4.03	−3.82	0.21	2.00	2.00	3.60	106.42	−6.01	−0.32
	93	−4.78	1	−4.06	−3.84	0.22	2.04	1.99	3.59	106.26	−3.58	0.12
	57	−4.82	1	−4.14	−3.93	0.21	2.00	2.00	3.55	106.13	−0.65	0.25
	0	−4.87	1	−3.94	−3.71	0.23	2.02	2.01	3.55	105.99	−5.56	−0.26
14	698	0.66	0	−4.23	−3.92	0.32	2.08	2.01	3.61	106.86	−2.90	0.33
	630	0.59	0	−4.21	−3.69	0.51	1.93	2.01	3.23	107.01	−6.25	−0.41
	627	0.59	0	−4.21	−3.69	0.51	1.93	2.01	3.23	107.02	−6.23	−0.40
	613	0.58	0	−4.08	−3.73	0.35	1.93	2.02	3.26	106.84	−8.25	−0.49
	612	0.58	0	−4.08	−3.73	0.34	1.93	2.02	3.25	106.86	−8.12	−0.49
	603	0.57	0	−4.41	−3.70	0.71	2.50	1.94	4.45	109.73	0.27	0.26
	425	0.39	0	−4.21	−3.56	0.65	1.89	2.05	3.24	106.62	−5.21	−0.03
	419	0.38	0	−4.25	−3.55	0.69	1.92	2.02	3.27	106.40	−4.13	0.07
	418	0.38	0	−4.25	−3.55	0.69	1.92	2.02	3.27	106.43	−4.20	0.07
	410	0.37	0	−4.53	−3.52	1.01	2.48	1.93	3.60	109.16	−0.07	0.28
	410	0.37	0	−4.53	−3.52	1.01	2.48	1.93	3.60	109.16	−0.07	0.28
	410	0.37	0	−4.53	−3.52	1.01	2.49	1.93	3.60	109.16	−0.06	0.29
	410	0.37	0	−4.53	−3.52	1.01	2.48	1.93	3.61	109.14	−0.03	0.29
	0	−0.04	0	−4.56	−3.56	1.00	2.36	1.93	3.57	108.44	−1.12	0.07
15	597	0.63	1	−4.20	−4.05	0.15	2.47	1.97	3.05	109.57	−2.35	0.07
	597	0.63	1	−4.20	−4.05	0.15	2.47	1.97	3.05	109.57	−2.35	0.07
	456	0.49	1	−4.15	−3.95	0.20	2.05	1.98	3.52	107.58	−3.68	−0.04

Table S25 continued from previous page

298	0.33	1	−4.31	−4.13	0.17	2.03	2.02	3.31	107.39	−5.81	−0.22
249	0.28	1	−4.32	−4.16	0.16	2.12	2.06	3.62	106.93	−5.70	−0.11
249	0.28	1	−4.32	−4.16	0.15	2.09	2.06	3.61	106.91	−5.69	−0.11
248	0.28	1	−4.08	−3.88	0.19	2.47	1.93	3.63	109.35	−0.68	0.22
247	0.28	1	−4.08	−3.88	0.19	2.47	1.93	3.63	109.34	−0.65	0.22
246	0.28	1	−4.08	−3.89	0.19	2.47	1.93	3.63	109.34	−0.66	0.21
241	0.27	1	−3.92	−3.73	0.19	1.95	2.03	3.61	106.40	−1.14	0.16
168	0.20	1	−4.04	−3.85	0.19	2.47	1.92	3.60	109.23	0.12	0.23
168	0.20	1	−4.04	−3.85	0.19	2.47	1.92	3.60	109.23	0.11	0.23
165	0.20	1	−4.08	−3.90	0.19	1.94	2.00	3.50	106.59	−5.05	−0.31
134	0.17	1	−4.01	−3.84	0.17	2.46	1.92	3.59	108.93	−1.01	0.13
0	0.03	1	−3.89	−3.69	0.20	2.00	2.01	3.58	106.28	−1.63	0.11

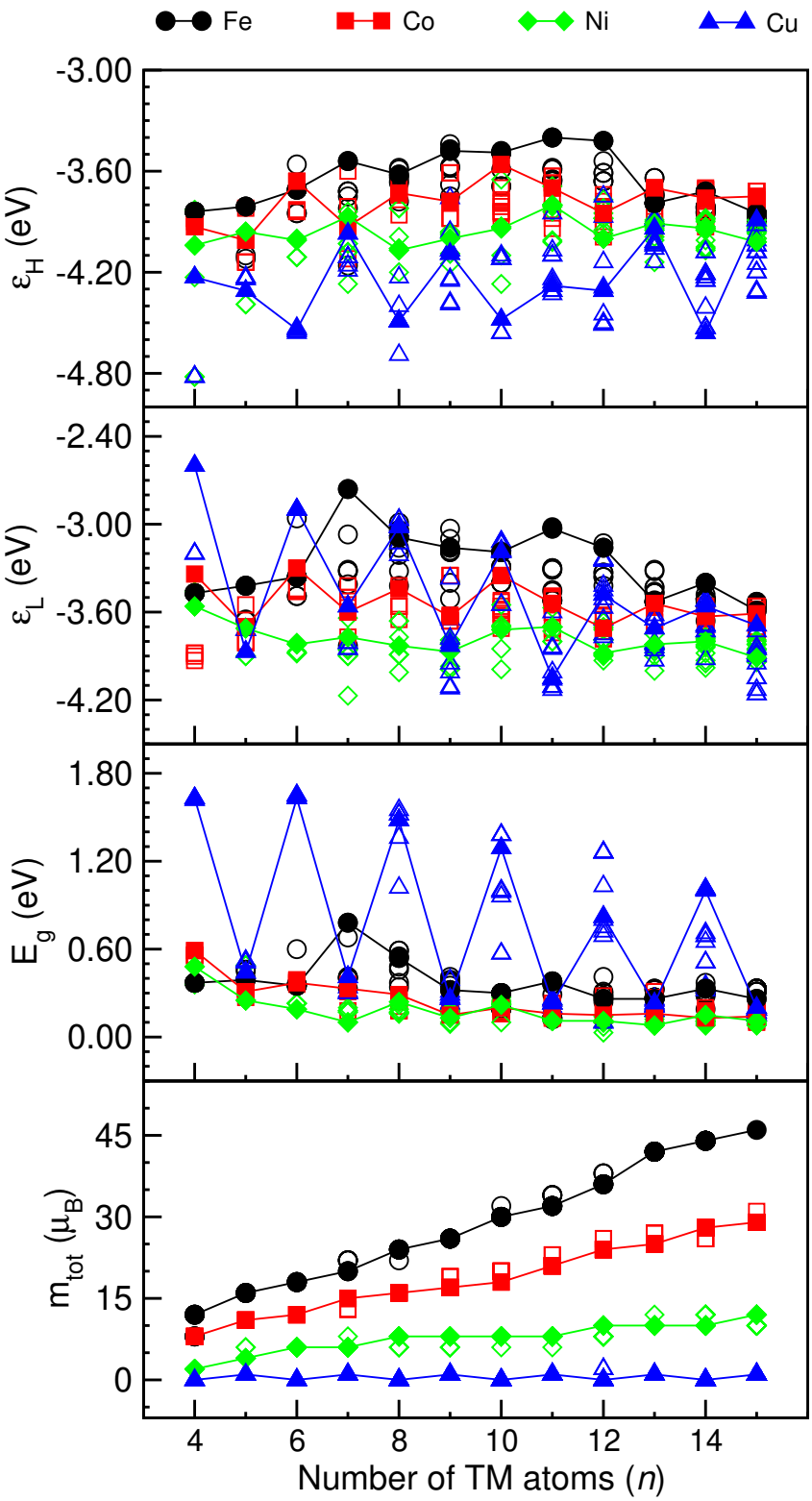

Figure S13: Energetic and electronic properties for $(CH_3+H)/TM_n$ systems from from $n = 4-15$ where TM = Fe, Co, Ni, Cu.

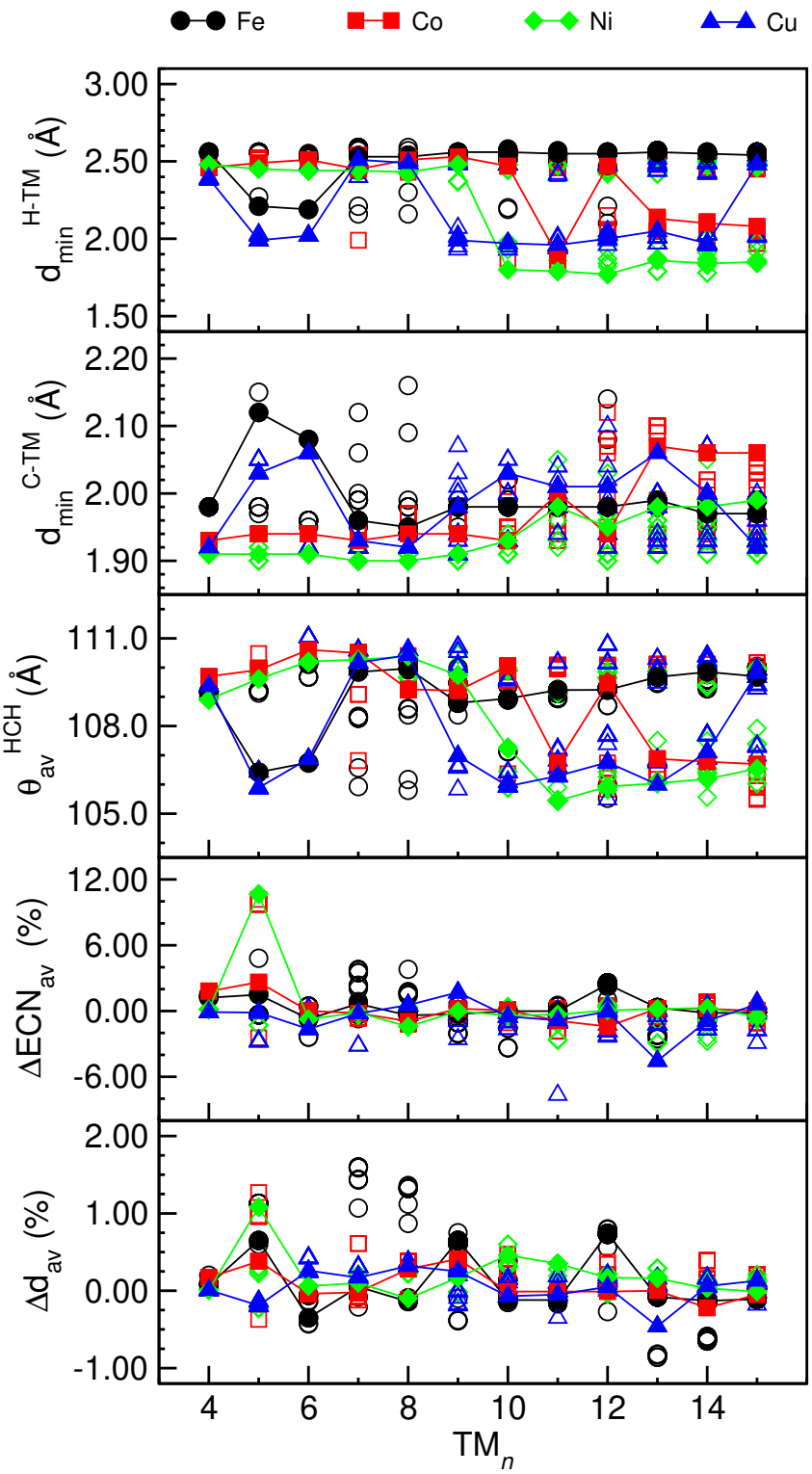

Figure S14: Structural properties for (CH_3+H)/TM_n systems from from $n = 4 - 15$ where TM = Fe, Co, Ni, Cu.

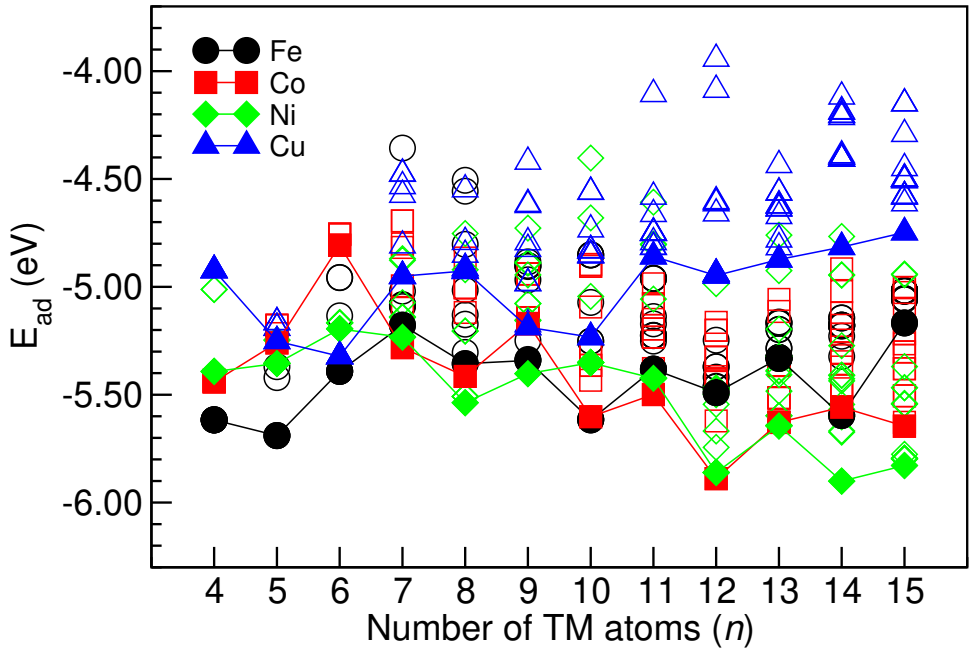

Figure S15: Adsorption energy for the $(CH_3+H)/TM_n$ systems where $n = 4-15$ and TM = Fe, Co, Ni, and Cu.

- The electronic and structural properties follow the same trends of CH_3/TM_n systems.
- The H co-adsorption changes significantly the behaviour of E_{ad} magnitude. For instance, for $n < 7$ the $|E_{ad}|$ is larger for Fe_n, while for $n > 7$, the largest values of are observed for Ni_n, except for Ni_{10}, however those values are close in magnitude with Co_n clusters, with the exception of Ni_9.
- For almost all values of n, $|E_{ad}|$ has the smallest value for Cu_n clusters, where the only exception occurs for Cu_6.

10 H Adsorption on TM_n Clusters

Table S26: Structural, energetic, and electronic properties for the H/Fe_n system: configuration number (i), relative total energy (ΔE_{tot}), total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{H^a-TM}$), average effective coordination number of the atomic hydrogen, ECN_{av}^{H}, changes in the TM_n clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (eV)	$d_{min}^{H^m-TM}$ (Å)	ECN^H (NNN)	ΔECN_{av} (%)	Δd_{av} (%)
4	2	−3.06	13	−3.74	−3.51	0.23	1.72	2.00	2.96	2.30
	2	−3.06	13	−3.74	−3.51	0.23	1.72	2.00	2.96	2.30
	1	−3.06	13	−3.74	−3.51	0.23	1.72	2.00	2.96	2.30
	0	−3.06	13	−3.74	−3.51	0.23	1.72	2.00	2.96	2.30
5	82	−2.99	17	−3.92	−3.61	0.31	1.80	2.94	2.17	0.61

Table S26 continued from previous page

	81	−2.99	17	−3.92	−3.61	0.31	1.80	2.97	2.08	0.61
	0	−3.07	17	−3.98	−3.61	0.37	1.72	2.00	1.00	0.69
	0	−3.07	17	−3.98	−3.61	0.37	1.72	2.00	1.01	0.69
6	2	−2.84	19	−3.75	−3.42	0.33	1.71	2.00	0.15	−0.22
	2	−2.84	19	−3.76	−3.42	0.33	1.71	2.00	0.14	−0.23
	1	−2.84	19	−3.76	−3.42	0.33	1.71	2.00	0.17	−0.22
	1	−2.84	19	−3.75	−3.42	0.33	1.71	2.00	0.15	−0.21
	1	−2.84	19	−3.76	−3.42	0.33	1.71	2.00	0.14	−0.22
	0	−2.84	19	−3.76	−3.42	0.33	1.71	2.00	0.14	−0.22
7	437	−2.38	21	−3.84	−3.44	0.40	1.57	1.00	0.86	0.07
	0	−2.82	21	−3.66	−2.98	0.68	1.79	2.99	−0.67	−0.44
	0	−2.82	21	−3.66	−2.98	0.68	1.79	2.99	−0.63	−0.44
	0	−2.82	21	−3.66	−2.98	0.68	1.79	2.99	−0.61	−0.44
	0	−2.82	21	−3.66	−2.98	0.68	1.79	2.99	−0.70	−0.44
	0	−2.82	21	−3.66	−2.98	0.68	1.79	2.99	−0.64	−0.44
	0	−2.82	21	−3.66	−2.98	0.68	1.79	2.99	−0.66	−0.44
8	328	−2.43	25	−3.69	−3.32	0.37	1.73	1.98	0.90	1.03
	298	−2.46	25	−3.73	−3.22	0.51	1.73	2.00	3.86	1.16
	296	−2.46	25	−3.63	−3.28	0.35	1.83	2.89	2.52	1.19
	189	−2.57	25	−3.68	−3.15	0.52	1.73	2.00	3.62	1.12
	133	−2.62	23	−3.52	−2.85	0.66	1.77	2.53	1.03	−0.02
	133	−2.63	23	−3.51	−2.85	0.66	1.78	2.59	0.97	−0.03
	132	−2.63	23	−3.52	−2.85	0.67	1.77	2.50	1.04	−0.02
	0	−2.76	23	−3.66	−2.89	0.77	1.71	2.00	0.72	−0.05
9	241	−2.54	27	−3.26	−3.09	0.17	1.80	3.00	−0.60	0.59
	240	−2.54	27	−3.26	−3.09	0.17	1.80	3.00	−0.59	0.59
	239	−2.54	27	−3.26	−3.09	0.17	1.80	3.00	−0.58	0.59
	57	−2.72	27	−3.57	−3.21	0.36	1.81	2.76	−1.42	0.46
	56	−2.72	27	−3.57	−3.21	0.36	1.81	2.77	−1.40	0.46
	37	−2.74	27	−3.51	−3.17	0.35	1.74	1.96	−1.15	0.33
	1	−2.78	27	−3.49	−3.16	0.33	1.84	3.00	−1.52	0.16
	0	−2.78	27	−3.49	−3.15	0.33	1.84	3.00	−1.49	0.16
10	65	−2.76	31	−3.52	−3.34	0.19	1.72	1.99	−0.41	0.55
	64	−2.76	31	−3.52	−3.34	0.19	1.72	1.99	−0.40	0.56
	62	−2.76	31	−3.52	−3.34	0.19	1.72	1.99	−0.41	0.56
	62	−2.76	31	−3.52	−3.34	0.18	1.72	1.99	−0.44	0.55
	44	−2.78	31	−3.50	−3.37	0.13	1.73	2.00	−2.03	0.27
	44	−2.78	31	−3.50	−3.37	0.13	1.73	2.00	−2.05	0.27
	44	−2.78	31	−3.50	−3.37	0.13	1.73	2.00	−2.04	0.27
	0	−2.82	31	−3.47	−3.33	0.14	1.82	2.88	−2.97	0.10
	0	−2.82	31	−3.47	−3.33	0.14	1.82	2.88	−2.96	0.11
11	135	−2.75	35	−3.52	−3.42	0.10	1.83	2.91	−0.70	0.46
	54	−2.83	33	−3.48	−3.16	0.32	1.82	2.84	−1.01	−0.38
	31	−2.86	33	−3.43	−3.20	0.22	1.83	2.88	−2.07	−0.41
	30	−2.86	33	−3.43	−3.20	0.22	1.83	2.88	−2.09	−0.41
	30	−2.86	33	−3.43	−3.20	0.22	1.83	2.87	−2.09	−0.41
	30	−2.86	33	−3.43	−3.20	0.22	1.83	2.88	−2.09	−0.40

Table S26 continued from previous page

	30	−2.86	33	−3.42	−3.20	0.22	1.83	2.88	−2.05	−0.40
	29	−2.86	33	−3.43	−3.20	0.22	1.83	2.88	−2.06	−0.40
	29	−2.86	33	−3.43	−3.20	0.22	1.83	2.88	−2.08	−0.40
	2	−2.89	33	−3.44	−3.22	0.23	1.74	2.00	−1.26	−0.31
	0	−2.89	33	−3.44	−3.22	0.23	1.75	2.00	−1.23	−0.30
12	96	−2.84	39	−3.54	−3.41	0.13	1.82	2.99	1.68	1.24
	96	−2.84	39	−3.54	−3.41	0.13	1.81	2.99	1.69	1.24
	95	−2.84	39	−3.54	−3.41	0.13	1.81	2.99	1.67	1.24
	39	−2.90	37	−3.46	−3.29	0.17	1.73	2.00	0.48	0.39
	38	−2.90	37	−3.46	−3.28	0.17	1.73	2.00	0.51	0.39
	10	−2.93	37	−3.45	−3.19	0.26	1.82	2.82	−0.22	0.23
	0	−2.94	37	−3.41	−3.19	0.22	1.83	3.00	0.06	0.34
13	43	−2.82	43	−3.75	−3.43	0.32	1.83	2.84	−0.99	−0.28
	43	−2.82	43	−3.74	−3.43	0.32	1.83	2.83	−0.98	−0.27
	42	−2.82	43	−3.74	−3.43	0.31	1.83	2.85	−0.98	−0.27
	40	−2.82	43	−3.75	−3.43	0.32	1.83	2.83	−0.99	−0.27
	38	−2.83	43	−3.74	−3.43	0.31	1.83	2.85	−0.96	−0.27
	5	−2.86	43	−3.77	−3.46	0.31	1.73	2.00	−0.86	−0.24
	5	−2.86	43	−3.77	−3.46	0.31	1.73	2.00	−0.85	−0.24
	4	−2.86	43	−3.77	−3.45	0.32	1.73	2.00	−0.85	−0.23
	3	−2.86	43	−3.77	−3.46	0.31	1.73	2.00	−0.85	−0.23
	2	−2.86	43	−3.77	−3.46	0.31	1.73	2.00	−0.85	−0.23
	1	−2.86	43	−3.77	−3.45	0.32	1.73	2.00	−0.84	−0.23
	1	−2.86	43	−3.77	−3.45	0.31	1.73	2.00	−0.84	−0.23
	0	−2.86	43	−3.77	−3.45	0.32	1.73	2.00	−0.86	−0.24
14	84	−2.82	45	−3.80	−3.58	0.22	1.74	2.00	−0.45	−0.15
	66	−2.83	45	−3.81	−3.51	0.30	1.83	2.97	−0.96	−0.24
	65	−2.83	45	−3.81	−3.50	0.30	1.83	2.97	−0.94	−0.24
	55	−2.85	45	−3.78	−3.41	0.36	1.81	2.94	−0.16	−0.14
	55	−2.85	45	−3.78	−3.41	0.36	1.81	2.94	−0.14	−0.14
	28	−2.87	45	−3.80	−3.49	0.32	1.82	2.92	−0.31	−0.17
	27	−2.87	45	−3.80	−3.49	0.32	1.82	2.91	−0.31	−0.17
	27	−2.87	45	−3.80	−3.49	0.32	1.82	2.91	−0.30	−0.18
	3	−2.90	45	−3.81	−3.51	0.30	1.71	1.99	−0.17	−0.13
	2	−2.90	45	−3.81	−3.51	0.30	1.71	1.99	−0.15	−0.13
	2	−2.90	45	−3.79	−3.42	0.37	1.80	2.92	−0.33	−0.15
	0	−2.90	45	−3.79	−3.42	0.37	1.80	2.92	−0.33	−0.15
15	149	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.10	0.22
	149	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.10	0.22
	148	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.08	0.22
	148	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.06	0.22
	148	−2.63	49	−3.65	−3.58	0.07	1.82	2.96	−0.06	0.22
	148	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.07	0.22
	147	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.07	0.22
	147	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.07	0.22
	147	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.07	0.22
	144	−2.63	49	−3.65	−3.58	0.07	1.82	2.95	−0.07	0.22
	99	−2.68	47	−3.77	−3.40	0.37	1.73	2.00	−0.64	−0.18

Table S26 continued from previous page

	98	−2.68	47	−3.77	−3.40	0.37	1.73	2.00	−0.67	−0.18
	97	−2.68	47	−3.77	−3.40	0.37	1.73	2.00	−0.64	−0.18
	94	−2.68	47	−3.77	−3.40	0.37	1.73	2.00	−0.66	−0.17
	0	−2.77	47	−3.78	−3.49	0.29	1.73	2.00	−0.20	−0.11

Table S27: Structural, energetic, and electronic properties for the H/Co$_n$ system: configuration number (i), relative total energy (ΔE_{tot}), total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{H^a-TM}$), average effective coordination number of the atomic hydrogen, ECN$_{av}^{H}$, changes in the TM$_n$ clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	ε_{homo} (eV)	ε_{lumo} (eV)	E_g (eV)	$d_{min}^{H^m-TM}$ (Å)	ECN^H (NNN)	ΔECN_{av} (%)	Δd_{av} (%)
4	3	−2.96	9	−3.92	−3.35	0.57	1.69	2.00	8.84	1.13
	2	−2.96	9	−3.92	−3.35	0.57	1.69	2.00	8.93	1.14
	1	−2.96	9	−3.92	−3.35	0.57	1.69	2.00	8.90	1.14
	0	−2.96	9	−3.92	−3.35	0.57	1.69	2.00	8.70	1.13
5	164	−2.64	12	−4.05	−3.78	0.27	1.68	2.00	5.38	0.67
	163	−2.64	12	−4.05	−3.78	0.27	1.68	1.99	5.39	0.68
	161	−2.64	12	−4.05	−3.78	0.28	1.68	1.99	5.34	0.69
	150	−2.65	12	−4.02	−3.78	0.24	1.73	2.32	6.78	0.58
	0	−2.80	12	−4.21	−3.69	0.51	1.70	2.00	1.13	0.22
6	1	−2.56	13	−3.85	−3.46	0.39	1.69	2.00	−0.38	0.08
	1	−2.56	13	−3.85	−3.46	0.39	1.69	2.00	−0.37	0.07
	0	−2.57	13	−3.85	−3.46	0.39	1.69	2.00	−0.38	0.08
	0	−2.57	13	−3.85	−3.46	0.39	1.69	2.00	−0.40	0.07
	0	−2.57	13	−3.85	−3.46	0.39	1.69	2.00	−0.38	0.07
	0	−2.57	13	−3.85	−3.46	0.39	1.69	2.00	−0.39	0.07
7	290	−2.39	16	−3.96	−3.65	0.31	1.82	3.00	−0.11	0.54
	289	−2.39	16	−3.96	−3.65	0.31	1.82	3.00	−0.13	0.54
	52	−2.62	14	−3.79	−3.50	0.29	1.66	1.97	−0.11	−0.15
	52	−2.62	14	−3.79	−3.50	0.29	1.66	1.97	−0.12	−0.16
	52	−2.62	14	−3.79	−3.50	0.29	1.66	1.96	−0.11	−0.15
	50	−2.63	14	−3.79	−3.50	0.29	1.66	1.97	−0.10	−0.16
	0	−2.68	14	−3.55	−3.35	0.20	1.77	3.00	−0.98	−0.16
8	31	−2.60	17	−3.87	−3.70	0.17	1.72	2.00	−2.15	0.04
	31	−2.60	17	−3.87	−3.70	0.17	1.72	2.00	−2.13	0.04
	1	−2.63	17	−3.89	−3.70	0.20	1.72	2.00	−1.92	0.02
	0	−2.63	17	−3.89	−3.69	0.20	1.72	2.00	−1.96	0.03
	0	−2.63	17	−3.89	−3.70	0.20	1.72	2.00	−1.94	0.02
	0	−2.63	17	−3.89	−3.69	0.20	1.72	2.00	−1.96	0.02
9	14	−2.66	18	−3.72	−3.51	0.20	1.71	2.00	−0.88	0.19
	13	−2.66	18	−3.72	−3.51	0.20	1.71	2.00	−0.89	0.19
	13	−2.66	18	−3.72	−3.51	0.20	1.71	2.00	−0.88	0.19
	1	−2.68	16	−3.46	−3.24	0.22	1.76	2.99	−0.14	−0.02
	0	−2.68	16	−3.46	−3.24	0.22	1.76	3.00	−0.15	−0.02

Table S27 continued from previous page

	0	−2.68	16	−3.46	−3.24	0.22	1.76	2.99	−0.14	−0.02
	0	−2.68	16	−3.46	−3.25	0.22	1.76	2.99	−0.14	−0.01
	0	−2.68	16	−3.46	−3.24	0.22	1.76	3.00	−0.14	−0.02
10	190	−2.58	19	−3.61	−3.49	0.12	1.75	2.99	−0.19	0.00
	190	−2.58	19	−3.61	−3.49	0.12	1.75	2.99	−0.19	−0.01
	149	−2.62	21	−3.85	−3.65	0.20	1.74	2.40	−0.97	0.32
	149	−2.62	21	−3.85	−3.65	0.20	1.74	2.39	−0.96	0.32
	148	−2.62	21	−3.85	−3.65	0.20	1.74	2.38	−0.94	0.32
	85	−2.68	21	−3.90	−3.70	0.20	1.70	2.00	−0.93	0.21
	85	−2.68	21	−3.90	−3.70	0.20	1.70	2.00	−0.94	0.21
	2	−2.77	19	−3.58	−3.35	0.23	1.76	2.74	0.28	0.00
	2	−2.77	19	−3.58	−3.35	0.23	1.76	2.74	0.27	0.00
	0	−2.77	19	−3.58	−3.35	0.23	1.77	2.75	0.27	0.01
11	200	−2.62	22	−3.63	−3.51	0.13	1.73	2.43	−0.15	0.03
	199	−2.62	22	−3.63	−3.51	0.13	1.73	2.43	−0.14	0.04
	199	−2.62	22	−3.63	−3.50	0.12	1.73	2.40	−0.13	0.04
	171	−2.65	22	−3.78	−3.58	0.21	1.73	2.50	−0.27	0.05
	169	−2.65	22	−3.78	−3.58	0.21	1.73	2.49	−0.27	0.05
	133	−2.69	22	−3.93	−3.67	0.27	1.66	2.00	−0.40	−0.02
	13	−2.81	22	−3.76	−3.51	0.25	1.75	2.51	−0.53	−0.05
	12	−2.81	22	−3.76	−3.51	0.25	1.75	2.53	−0.56	−0.06
	12	−2.81	22	−3.76	−3.51	0.25	1.75	2.50	−0.51	−0.06
	0	−2.82	22	−3.85	−3.63	0.22	1.69	2.00	−0.06	−0.14
12	437	−2.67	25	−3.89	−3.71	0.18	1.69	1.99	−0.52	0.28
	339	−2.77	25	−3.77	−3.69	0.08	1.80	3.00	−1.08	0.09
	330	−2.78	25	−3.87	−3.72	0.16	1.71	2.00	−1.15	0.04
	329	−2.78	25	−3.87	−3.71	0.16	1.71	2.00	−1.16	0.03
	329	−2.78	25	−3.87	−3.72	0.16	1.71	2.00	−1.18	0.04
	312	−2.79	23	−3.65	−3.50	0.14	1.79	2.94	0.12	0.03
	309	−2.80	25	−3.86	−3.66	0.19	1.78	2.97	−0.62	0.17
	308	−2.80	25	−3.86	−3.66	0.19	1.78	2.97	−0.65	0.16
	307	−2.80	25	−3.86	−3.66	0.20	1.78	2.97	−0.63	0.16
	272	−2.83	23	−3.76	−3.53	0.23	1.69	1.96	−0.11	−0.08
	85	−3.02	23	−3.72	−3.57	0.15	1.69	2.00	0.64	0.05
	0	−3.11	25	−3.88	−3.76	0.12	1.71	2.00	−0.10	0.17
13	399	−2.67	26	−3.70	−3.58	0.12	1.75	2.57	0.02	0.05
	398	−2.67	26	−3.70	−3.58	0.12	1.75	2.57	0.00	0.04
	306	−2.77	26	−3.74	−3.60	0.14	1.80	3.00	0.27	0.06
	304	−2.77	26	−3.74	−3.60	0.14	1.80	3.00	0.27	0.06
	304	−2.77	26	−3.74	−3.60	0.14	1.80	3.00	0.27	0.06
	304	−2.77	26	−3.74	−3.59	0.14	1.80	3.00	0.27	0.06
	304	−2.77	26	−3.74	−3.59	0.14	1.80	3.00	0.27	0.06
	303	−2.77	26	−3.74	−3.59	0.14	1.80	3.00	0.26	0.06
	303	−2.77	26	−3.74	−3.59	0.14	1.80	3.00	0.26	0.06
	3	−3.07	26	−3.93	−3.61	0.31	1.69	2.00	0.20	−0.05
	1	−3.07	26	−3.93	−3.61	0.31	1.69	2.00	0.20	−0.05
	1	−3.07	26	−3.93	−3.62	0.31	1.69	2.00	0.20	−0.05
	0	−3.07	26	−3.92	−3.61	0.31	1.69	2.00	0.21	−0.05

Table S27 continued from previous page

14	252	−2.62	27	−3.72	−3.57	0.15	1.79	2.99	0.14	0.07
	251	−2.62	27	−3.72	−3.57	0.15	1.79	2.99	0.13	0.07
	250	−2.63	27	−3.72	−3.57	0.15	1.79	2.99	0.15	0.08
	250	−2.63	27	−3.72	−3.57	0.15	1.79	2.99	0.15	0.08
	242	−2.63	27	−3.76	−3.62	0.14	1.76	2.68	1.08	0.44
	240	−2.64	29	−3.85	−3.67	0.18	1.77	2.71	0.12	0.34
	217	−2.66	29	−3.90	−3.70	0.19	1.77	2.77	−0.13	0.26
	216	−2.66	29	−3.89	−3.75	0.14	1.76	2.57	−0.40	0.42
	146	−2.73	29	−3.91	−3.76	0.15	1.78	3.00	−0.06	0.44
	81	−2.80	27	−3.71	−3.58	0.13	1.80	2.97	0.02	−0.03
	80	−2.80	27	−3.71	−3.58	0.13	1.80	2.97	0.04	−0.02
	37	−2.84	27	−3.79	−3.61	0.19	1.69	1.99	0.00	0.02
	37	−2.84	27	−3.79	−3.61	0.18	1.69	1.99	−0.04	−0.02
	0	−2.88	29	−3.91	−3.75	0.15	1.70	2.00	0.14	0.26
15	314	−2.56	30	−3.82	−3.72	0.10	1.76	2.87	−0.58	0.29
	312	−2.56	30	−3.82	−3.72	0.10	1.76	2.87	−0.57	0.28
	218	−2.66	30	−3.84	−3.69	0.15	1.76	2.91	−0.46	0.20
	216	−2.66	30	−3.84	−3.69	0.15	1.76	2.91	−0.46	0.20
	196	−2.68	30	−3.77	−3.64	0.14	1.77	2.71	0.02	0.24
	196	−2.68	30	−3.77	−3.64	0.14	1.77	2.71	0.03	0.23
	177	−2.70	30	−3.85	−3.66	0.19	1.79	2.87	0.13	0.19
	175	−2.70	30	−3.85	−3.66	0.19	1.79	2.91	0.12	0.19
	126	−2.75	30	−3.87	−3.70	0.17	1.78	2.96	−0.64	0.19
	126	−2.75	30	−3.87	−3.70	0.17	1.78	2.96	−0.66	0.19
	122	−2.75	28	−3.65	−3.55	0.10	1.76	2.95	−0.30	0.02
	122	−2.75	28	−3.65	−3.55	0.09	1.76	2.95	−0.30	0.02
	115	−2.76	30	−3.86	−3.66	0.20	1.70	2.00	−0.17	0.19
	75	−2.80	30	−3.82	−3.66	0.16	1.79	2.94	0.06	0.22
	0	−2.87	28	−3.74	−3.60	0.13	1.69	2.00	0.05	−0.09

Table S28: Structural, energetic, and electronic properties for the H/Ni$_n$ system: configuration number (i), relative total energy (ΔE_{tot}), total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{\mathrm{H}^a-\mathrm{TM}}$), average effective coordination number of the atomic hydrogen, ECN$_{av}^{H}$, changes in the TM$_n$ clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E$_{ad}$ (eV)	m_{tot} (μ_B)	$\varepsilon_{\mathrm{homo}}$ (eV)	$\varepsilon_{\mathrm{lumo}}$ (eV)	E_g (eV)	$d_{min}^{\mathrm{H}^m-\mathrm{TM}}$ (Å)	ECN$^{\mathrm{H}}$ (NNN)	ΔECN_{av} (%)	Δd_{av} (%)
4	26	−2.89	5	−3.79	−3.64	0.14	1.65	2.00	0.14	0.46
	25	−2.89	5	−3.79	−3.65	0.15	1.65	2.00	0.09	0.51
	0	−2.92	3	−3.89	−3.36	0.54	1.51	1.00	−0.40	0.20
	0	−2.92	3	−3.89	−3.36	0.53	1.51	1.00	−0.49	0.21
5	245	−2.68	5	−3.95	−3.63	0.32	1.65	2.00	−0.36	0.20
	242	−2.68	5	−3.95	−3.63	0.32	1.65	2.00	−0.39	0.22
	1	−2.93	5	−3.84	−3.65	0.19	1.66	1.98	11.95	1.19
	1	−2.93	5	−3.84	−3.65	0.19	1.66	1.99	11.95	1.21

Table S28 continued from previous page

	0	−2.93	5	−3.84	−3.65	0.19	1.66	1.98	11.92	1.21
6	10	−2.80	7	−3.99	−3.81	0.17	1.66	2.00	0.01	0.16
	9	−2.80	7	−3.98	−3.81	0.17	1.66	2.00	0.03	0.16
	9	−2.80	7	−3.98	−3.81	0.17	1.66	2.00	0.07	0.16
	8	−2.80	7	−3.99	−3.81	0.18	1.66	2.00	−0.01	0.13
	0	−2.81	7	−3.95	−3.67	0.28	1.71	3.00	0.11	0.58
	0	−2.81	7	−3.95	−3.67	0.28	1.71	3.00	0.12	0.57
7	207	−2.59	7	−3.92	−3.76	0.16	1.71	2.94	−0.43	0.45
	109	−2.69	7	−3.97	−3.79	0.18	1.61	1.95	−0.16	0.19
	108	−2.69	7	−3.97	−3.79	0.18	1.61	1.95	−0.16	0.19
	108	−2.69	7	−3.97	−3.79	0.18	1.61	1.95	−0.17	0.19
	107	−2.69	7	−3.97	−3.79	0.18	1.61	1.95	−0.16	0.19
	105	−2.70	7	−3.97	−3.79	0.18	1.61	1.95	−0.15	0.19
	0	−2.80	7	−3.94	−3.81	0.13	1.74	3.00	−0.27	0.16
8	326	−2.53	9	−4.05	−4.01	0.04	1.68	2.00	0.50	0.06
	288	−2.57	9	−4.12	−3.98	0.14	1.68	2.00	−0.42	0.07
	232	−2.62	7	−3.85	−3.66	0.19	1.71	3.00	0.42	0.23
	230	−2.63	7	−3.85	−3.67	0.19	1.71	3.00	0.38	0.23
	96	−2.76	7	−3.88	−3.70	0.18	1.73	2.95	−0.68	0.12
	3	−2.85	7	−3.88	−3.72	0.16	1.64	1.99	0.06	0.04
	0	−2.86	7	−3.88	−3.72	0.16	1.64	1.99	0.08	0.04
	0	−2.86	7	−3.87	−3.72	0.15	1.64	1.99	0.13	0.03
9	130	−2.60	7	−3.87	−3.68	0.19	1.64	1.99	1.18	0.05
	130	−2.60	7	−3.87	−3.68	0.20	1.64	1.99	1.18	0.06
	45	−2.68	7	−3.96	−3.81	0.15	1.73	3.00	0.50	0.04
	15	−2.71	7	−3.94	−3.81	0.14	1.73	3.00	0.10	−0.02
	15	−2.71	7	−3.94	−3.80	0.14	1.73	3.00	0.10	−0.03
	1	−2.73	7	−3.89	−3.78	0.12	1.73	2.99	−0.20	0.10
	1	−2.73	7	−3.89	−3.78	0.11	1.73	3.00	−0.19	0.09
	1	−2.73	7	−3.90	−3.78	0.11	1.73	2.99	−0.18	0.09
	0	−2.73	7	−3.89	−3.78	0.11	1.73	2.99	−0.20	0.09
10	475	−2.01	9	−4.26	−4.09	0.17	1.51	1.00	−0.15	0.45
	62	−2.42	7	−3.67	−3.60	0.07	1.69	2.96	0.15	0.46
	62	−2.42	7	−3.67	−3.60	0.07	1.69	2.96	0.15	0.46
	62	−2.42	7	−3.67	−3.60	0.07	1.69	2.96	0.13	0.45
	61	−2.42	7	−3.67	−3.60	0.07	1.69	2.96	0.14	0.46
	1	−2.48	7	−3.73	−3.65	0.08	1.60	2.00	0.28	0.32
	0	−2.48	7	−3.73	−3.65	0.08	1.60	2.00	0.29	0.32
11	165	−2.57	7	−3.73	−3.63	0.10	1.70	2.98	−0.73	0.22
	164	−2.57	7	−3.74	−3.63	0.10	1.70	2.98	−0.71	0.22
	164	−2.57	7	−3.73	−3.63	0.10	1.70	2.98	−0.72	0.22
	164	−2.57	7	−3.74	−3.63	0.10	1.70	2.98	−0.70	0.22
	163	−2.57	7	−3.73	−3.63	0.10	1.70	2.98	−0.69	0.22
	163	−2.57	7	−3.73	−3.63	0.10	1.70	2.98	−0.69	0.22
	163	−2.57	7	−3.73	−3.63	0.11	1.70	2.98	−0.71	0.23
	118	−2.62	7	−3.78	−3.64	0.13	1.69	3.00	−0.33	0.23
	117	−2.62	7	−3.78	−3.64	0.13	1.69	3.00	−0.32	0.24

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	3	−2.73	9	−3.99	−3.84	0.15	1.67	2.00	−0.03	0.21
	0	−2.73	9	−3.99	−3.84	0.15	1.67	2.00	−0.04	0.22
12	364	−2.63	9	−3.86	−3.74	0.11	1.73	2.98	0.37	0.14
	226	−2.77	9	−3.94	−3.82	0.11	1.66	2.00	0.90	0.01
	190	−2.81	9	−3.86	−3.75	0.11	1.72	2.98	0.35	0.12
	169	−2.83	9	−3.92	−3.79	0.14	1.65	2.00	0.80	0.00
	166	−2.83	9	−3.89	−3.73	0.16	1.73	2.91	0.65	0.13
	28	−2.97	9	−3.94	−3.80	0.15	1.65	1.99	0.78	−0.03
	28	−2.97	9	−3.94	−3.80	0.15	1.66	1.99	0.79	−0.03
	2	−3.00	9	−3.89	−3.81	0.07	1.65	1.97	0.66	0.03
	2	−3.00	9	−3.88	−3.81	0.07	1.65	1.97	0.69	0.04
	1	−3.00	9	−3.89	−3.81	0.07	1.65	1.97	0.67	0.04
	0	−3.00	9	−3.89	−3.81	0.07	1.65	1.97	0.68	0.03
13	329	−2.66	9	−3.85	−3.73	0.12	1.69	2.81	−0.41	0.19
	279	−2.71	9	−3.84	−3.75	0.09	1.70	2.98	−0.14	0.16
	258	−2.73	11	−3.94	−3.88	0.06	1.73	2.96	−1.27	0.12
	258	−2.73	11	−3.94	−3.88	0.06	1.73	2.96	−1.18	0.13
	258	−2.73	11	−3.94	−3.88	0.06	1.73	2.96	−1.27	0.12
	176	−2.81	11	−4.03	−3.90	0.13	1.66	1.98	0.16	0.12
	103	−2.88	9	−3.88	−3.77	0.11	1.63	2.00	0.17	0.01
	103	−2.88	9	−3.88	−3.77	0.10	1.63	2.00	0.19	0.02
	24	−2.96	11	−4.02	−3.93	0.09	1.66	2.00	0.08	0.10
	24	−2.96	11	−4.01	−3.92	0.09	1.66	1.99	0.09	0.10
	1	−2.98	9	−3.91	−3.80	0.12	1.64	1.99	0.23	−0.04
	1	−2.98	9	−3.91	−3.80	0.12	1.64	1.99	0.21	−0.05
	0	−2.98	9	−3.92	−3.80	0.12	1.64	1.99	0.21	−0.05
14	361	−2.60	11	−3.92	−3.81	0.12	1.71	2.99	−0.14	0.12
	234	−2.73	11	−3.93	−3.81	0.12	1.71	2.99	−0.18	0.14
	178	−2.78	11	−3.96	−3.81	0.15	1.72	2.99	0.06	0.12
	174	−2.79	11	−3.97	−3.87	0.11	1.62	1.97	0.19	0.03
	127	−2.83	11	−4.00	−3.89	0.11	1.64	1.98	−0.05	−0.05
	114	−2.85	11	−3.93	−3.82	0.11	1.72	2.83	0.27	0.07
	113	−2.85	11	−3.93	−3.82	0.11	1.72	2.82	0.28	0.07
	62	−2.90	11	−4.00	−3.86	0.14	1.64	2.00	−0.28	−0.03
	23	−2.94	11	−3.99	−3.85	0.14	1.64	1.99	0.19	−0.03
	22	−2.94	11	−3.99	−3.85	0.14	1.64	1.99	0.20	−0.02
	4	−2.95	11	−3.98	−3.90	0.08	1.65	2.00	0.28	−0.03
	4	−2.96	11	−3.98	−3.90	0.08	1.65	2.00	0.29	−0.03
	4	−2.96	11	−3.98	−3.90	0.08	1.65	2.00	0.27	−0.04
	0	−2.96	11	−3.98	−3.90	0.08	1.64	2.00	0.40	−0.03
15	193	−2.73	11	−3.96	−3.89	0.07	1.71	2.89	−1.05	0.03
	192	−2.73	11	−3.96	−3.89	0.07	1.71	2.88	−1.05	0.02
	94	−2.83	13	−4.08	−3.97	0.11	1.64	1.97	−0.02	0.11
	93	−2.83	13	−4.08	−3.97	0.11	1.64	1.97	−0.02	0.11
	93	−2.83	13	−4.08	−3.97	0.11	1.64	1.97	−0.02	0.11
	92	−2.83	13	−4.08	−3.97	0.11	1.64	1.97	−0.02	0.11
	85	−2.83	11	−3.94	−3.84	0.09	1.72	2.98	−0.15	0.06
	51	−2.87	11	−3.98	−3.91	0.07	1.64	2.00	0.09	−0.03

Table S28 continued from previous page

	50	−2.87	11	−3.98	−3.91	0.07	1.64	2.00	0.10	−0.03
	50	−2.87	11	−3.98	−3.91	0.07	1.64	1.99	0.10	−0.02
	1	−2.92	11	−4.00	−3.89	0.11	1.64	2.00	−0.45	−0.06
	1	−2.92	11	−4.00	−3.89	0.11	1.64	2.00	−0.44	−0.05
	1	−2.92	11	−4.00	−3.89	0.11	1.64	2.00	−0.43	−0.06
	1	−2.92	11	−4.00	−3.89	0.11	1.64	2.00	−0.46	−0.06
	0	−2.92	11	−4.00	−3.89	0.11	1.64	2.00	−0.44	−0.06

Table S29: Structural, energetic, and electronic properties for the H/Cu$_n$ system: configuration number (i), relative total energy (ΔE_{tot}), total magnetic moment (m_{tot}), HOMO energy (ε_{homo}), LUMO energy (ε_{lumo}), LUMO-HOMO energy gap (E_g), minimum distance of the atomic hydrogen, H^a, to the nearest TM atom ($d_{min}^{\mathrm{H}^a-\mathrm{TM}}$), average effective coordination number of the atomic hydrogen, ECN$_{av}^{H}$, changes in the TM$_n$ clusters due to the adsorption, effective coordination number (ΔECN_{av}), average weighted bond lengths (Δd_{av}).

n	ΔE_{tot} (meV)	E_{ad} (eV)	m_{tot} (μ_B)	$\varepsilon_{\mathrm{homo}}$ (eV)	$\varepsilon_{\mathrm{lumo}}$ (eV)	E_g (eV)	$d_{min}^{\mathrm{H}^m-\mathrm{TM}}$ (Å)	ECN$^{\mathrm{H}}$ (NNN)	ΔECN_{av} (%)	Δd_{av} (%)
4	0	−2.65	1	−4.31	−3.78	0.53	1.65	1.99	0.70	0.26
	0	−2.65	1	−4.31	−3.78	0.53	1.65	1.99	0.70	0.26
	0	−2.65	1	−4.31	−3.79	0.53	1.65	1.99	0.70	0.26
5	1326	−2.07	2	−4.19	−3.95	0.24	1.66	2.00	−0.23	0.38
	1326	−2.07	2	−4.19	−3.95	0.24	1.66	2.00	−0.22	0.40
	1325	−2.07	2	−4.19	−3.95	0.24	1.66	2.00	−0.23	0.38
	0	−3.40	0	−4.71	−2.78	1.93	1.65	2.00	−0.15	−0.08
	0	−3.40	0	−4.71	−2.78	1.93	1.65	2.00	−0.14	−0.11
6	1	−2.20	1	−4.17	−3.77	0.41	1.65	1.99	−0.23	0.41
	1	−2.20	1	−4.17	−3.77	0.41	1.65	2.00	−0.19	0.42
	0	−2.20	1	−4.17	−3.77	0.41	1.65	2.00	−0.23	0.42
	0	−2.20	1	−4.17	−3.77	0.41	1.65	2.00	−0.20	0.41
7	550	−2.12	2	−4.16	−3.85	0.31	1.67	2.00	−0.81	0.20
	548	−2.12	2	−4.16	−3.85	0.31	1.67	2.00	−0.80	0.20
	87	−2.58	0	−4.35	−2.96	1.39	1.73	2.39	−2.90	0.00
	87	−2.58	0	−4.35	−2.96	1.39	1.73	2.37	−2.86	0.01
	0	−2.67	0	−4.46	−2.85	1.60	1.66	1.96	−4.04	0.00
8	149	−1.95	1	−4.31	−3.98	0.32	1.67	2.00	3.79	0.34
	109	−1.99	1	−4.17	−3.87	0.30	1.76	3.00	−0.17	0.22
	109	−1.99	1	−4.17	−3.87	0.30	1.76	2.99	−0.18	0.21
	91	−2.01	1	−3.68	−3.27	0.42	1.65	1.96	4.18	0.52
	91	−2.01	1	−3.68	−3.27	0.42	1.65	1.96	4.24	0.52
	90	−2.01	1	−3.68	−3.27	0.41	1.65	1.96	4.16	0.51
	90	−2.01	1	−3.68	−3.27	0.42	1.65	1.96	4.17	0.52
	0	−2.10	1	−4.05	−3.75	0.30	1.66	2.00	3.29	0.28
9	779	−2.15	2	−4.18	−4.03	0.15	1.66	2.00	2.69	0.16
	353	−2.58	0	−4.49	−3.41	1.08	1.66	2.00	3.24	0.05
	350	−2.58	0	−4.34	−3.70	0.64	1.65	1.91	−0.53	0.26
	350	−2.58	0	−4.34	−3.70	0.64	1.65	1.91	−0.55	0.25
	348	−2.58	0	−4.34	−3.70	0.64	1.65	1.91	−0.58	0.23

Table S29 continued from previous page

	231	−2.70	0	−4.48	−3.41	1.07	1.67	1.99	2.58	−0.01
	175	−2.75	0	−4.28	−3.67	0.61	1.69	2.00	4.83	0.21
	164	−2.77	0	−4.48	−3.23	1.25	1.69	2.00	−1.15	−0.02
	0	−2.93	0	−4.37	−3.37	1.00	1.68	1.99	6.36	0.58
10	165	−2.22	1	−4.08	−3.77	0.31	1.68	2.00	−0.84	0.09
	165	−2.23	1	−4.08	−3.77	0.31	1.68	2.00	−0.86	0.08
	164	−2.23	1	−4.08	−3.77	0.31	1.68	2.00	−0.89	0.09
	104	−2.29	1	−3.91	−3.64	0.27	1.66	2.00	−1.26	0.12
	104	−2.29	1	−3.91	−3.64	0.27	1.66	2.00	−1.22	0.13
	104	−2.29	1	−3.91	−3.64	0.27	1.66	2.00	−1.23	0.12
	3	−2.39	1	−3.93	−3.65	0.28	1.67	2.00	−0.40	−0.07
	1	−2.39	1	−3.93	−3.65	0.28	1.67	2.00	−0.39	−0.06
	0	−2.39	1	−3.93	−3.65	0.28	1.67	2.00	−0.39	−0.07
	0	−2.39	1	−3.93	−3.65	0.28	1.67	2.00	−0.39	−0.05
11	587	−2.32	2	−4.36	−4.16	0.20	1.65	2.00	−0.79	0.05
	586	−2.32	2	−4.36	−4.16	0.19	1.65	2.00	−0.75	0.05
	585	−2.32	2	−4.36	−4.16	0.20	1.65	2.00	−0.88	0.04
	457	−2.45	0	−4.31	−3.66	0.66	1.74	3.00	−1.61	0.05
	278	−2.63	0	−4.58	−3.34	1.24	1.71	2.38	−6.02	−0.27
	132	−2.78	0	−4.45	−3.63	0.82	1.65	1.97	−0.94	0.03
	129	−2.78	0	−4.45	−3.65	0.80	1.65	1.97	−0.99	0.05
	129	−2.78	0	−4.45	−3.65	0.80	1.65	1.97	−0.99	0.03
	100	−2.81	0	−4.47	−3.40	1.08	1.75	2.97	−0.93	0.07
	99	−2.81	0	−4.47	−3.40	1.07	1.75	2.97	−0.95	0.06
	0	−2.91	0	−4.50	−3.37	1.12	1.65	1.99	−0.71	−0.02
12	446	−2.04	1	−3.85	−3.56	0.29	1.64	2.00	−5.85	−0.30
	346	−2.14	1	−3.99	−3.80	0.19	1.75	2.96	−0.99	0.16
	299	−2.19	1	−3.88	−3.63	0.25	1.65	2.00	−0.58	0.06
	197	−2.29	1	−3.90	−3.67	0.23	1.74	3.00	−0.70	0.17
	166	−2.32	1	−4.19	−3.96	0.22	1.75	3.00	−0.70	0.16
	55	−2.43	1	−4.39	−4.18	0.20	1.66	1.99	−0.19	0.10
	54	−2.43	1	−4.39	−4.18	0.20	1.66	1.99	−0.22	0.10
	54	−2.43	1	−4.39	−4.18	0.20	1.66	1.99	−0.22	0.09
	54	−2.43	1	−4.39	−4.18	0.20	1.66	1.99	−0.22	0.11
	53	−2.44	1	−4.38	−4.18	0.20	1.66	1.99	−0.21	0.10
	53	−2.44	1	−4.39	−4.18	0.20	1.66	1.99	−0.22	0.11
	0	−2.49	1	−4.00	−3.73	0.27	1.66	2.00	0.07	0.07
13	831	−2.00	2	−4.03	−3.86	0.16	1.66	2.00	−4.66	−0.40
	710	−2.12	0	−4.11	−3.73	0.38	1.77	2.99	−6.62	−0.50
	658	−2.17	2	−4.04	−3.89	0.16	1.74	2.91	−2.16	0.07
	635	−2.19	0	−4.06	−3.82	0.24	1.75	2.97	−5.03	−0.20
	412	−2.42	0	−4.13	−3.62	0.51	1.66	1.99	−0.64	−0.03
	411	−2.42	0	−4.13	−3.62	0.51	1.66	1.99	−0.64	−0.03
	410	−2.42	0	−4.13	−3.63	0.50	1.66	1.99	−0.64	−0.03
	177	−2.65	0	−4.45	−3.60	0.85	1.68	2.00	−5.10	−0.55
	175	−2.65	0	−4.45	−3.60	0.85	1.68	2.00	−5.08	−0.54
	121	−2.71	0	−4.38	−3.70	0.68	1.67	1.99	−0.70	0.01
	119	−2.71	0	−4.38	−3.71	0.67	1.67	1.99	−0.69	0.02

Table S29 continued from previous page

	3	−2.83	0	−4.35	−3.48	0.87	1.67	2.00	−0.44	0.00
	0	−2.83	0	−4.35	−3.48	0.87	1.67	2.00	−0.42	0.00
14	259	−2.04	1	−4.19	−3.93	0.26	1.66	2.00	−4.82	−0.40
	259	−2.04	1	−4.18	−3.93	0.26	1.66	1.99	−4.81	−0.40
	186	−2.12	1	−3.90	−3.68	0.22	1.66	2.00	−0.03	0.09
	132	−2.17	1	−4.24	−3.99	0.25	1.66	2.00	−0.63	−0.01
	117	−2.19	1	−4.06	−3.87	0.18	1.74	2.92	−1.46	0.07
	116	−2.19	1	−4.06	−3.87	0.18	1.74	2.92	−1.44	0.08
	99	−2.20	1	−4.22	−3.99	0.22	1.75	2.94	−0.01	0.13
	99	−2.21	1	−4.22	−4.00	0.22	1.75	2.94	−0.04	0.11
	98	−2.21	1	−4.22	−4.00	0.22	1.75	2.94	−0.03	0.13
	98	−2.21	1	−4.22	−4.00	0.22	1.75	2.94	−0.02	0.13
	91	−2.21	1	−4.09	−3.86	0.23	1.75	2.99	−6.54	−0.47
	90	−2.21	1	−4.09	−3.86	0.23	1.75	2.99	−6.56	−0.46
	1	−2.30	1	−4.13	−3.96	0.17	1.74	2.99	−0.72	0.10
	0	−2.30	1	−4.13	−3.96	0.17	1.74	2.99	−0.70	0.09
15	582	−2.11	2	−4.07	−4.00	0.07	1.75	2.99	−5.93	−0.44
	320	−2.38	0	−4.07	−3.76	0.31	1.67	2.00	0.03	0.00
	318	−2.38	0	−4.15	−3.76	0.39	1.76	2.99	−2.24	−0.14
	267	−2.43	0	−4.13	−3.72	0.41	1.76	2.98	−3.59	−0.24
	267	−2.43	0	−4.13	−3.72	0.41	1.76	2.98	−3.59	−0.24
	63	−2.63	0	−4.27	−3.70	0.57	1.75	2.95	−3.00	−0.09
	63	−2.63	0	−4.27	−3.70	0.57	1.75	2.94	−2.88	−0.09
	62	−2.63	0	−4.27	−3.70	0.57	1.75	2.94	−2.80	−0.08
	2	−2.69	0	−4.28	−3.71	0.57	1.76	2.99	−1.05	0.02
	1	−2.69	0	−4.28	−3.71	0.57	1.76	2.99	−1.06	0.02
	1	−2.69	0	−4.28	−3.71	0.57	1.76	2.99	−1.06	0.02
	1	−2.69	0	−4.28	−3.71	0.57	1.76	2.99	−1.09	0.02
	0	−2.70	0	−4.28	−3.71	0.57	1.76	2.99	−1.02	0.02
	0	−2.70	0	−4.28	−3.71	0.57	1.76	2.99	−1.04	0.02
	0	−2.70	0	−4.28	−3.71	0.57	1.76	2.99	−1.03	0.02

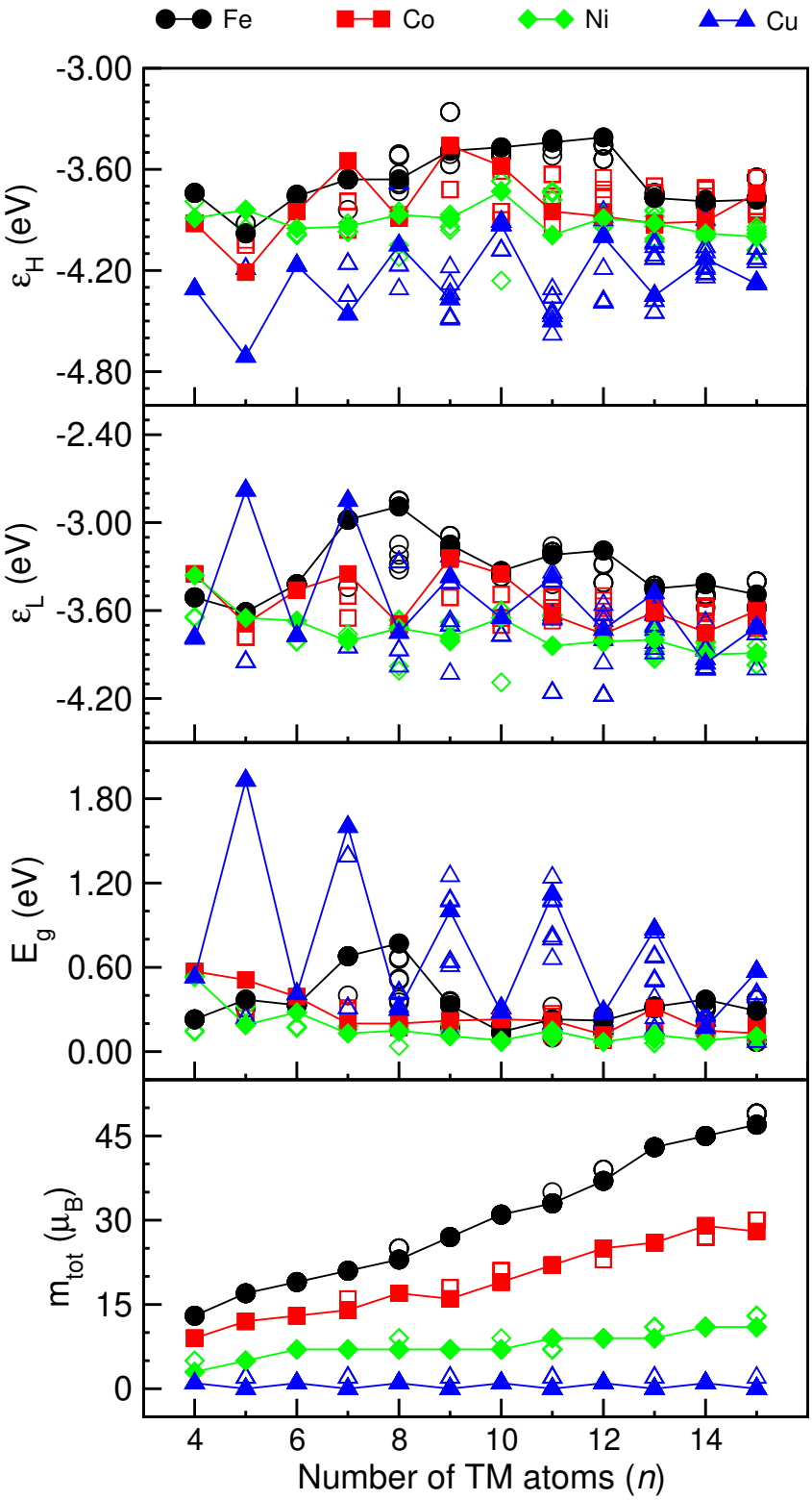

Figure S16: Energetic properties for H/TM$_n$ systems where $n = 4 - 15$ and TM=Fe, Co, Ni, Cu.

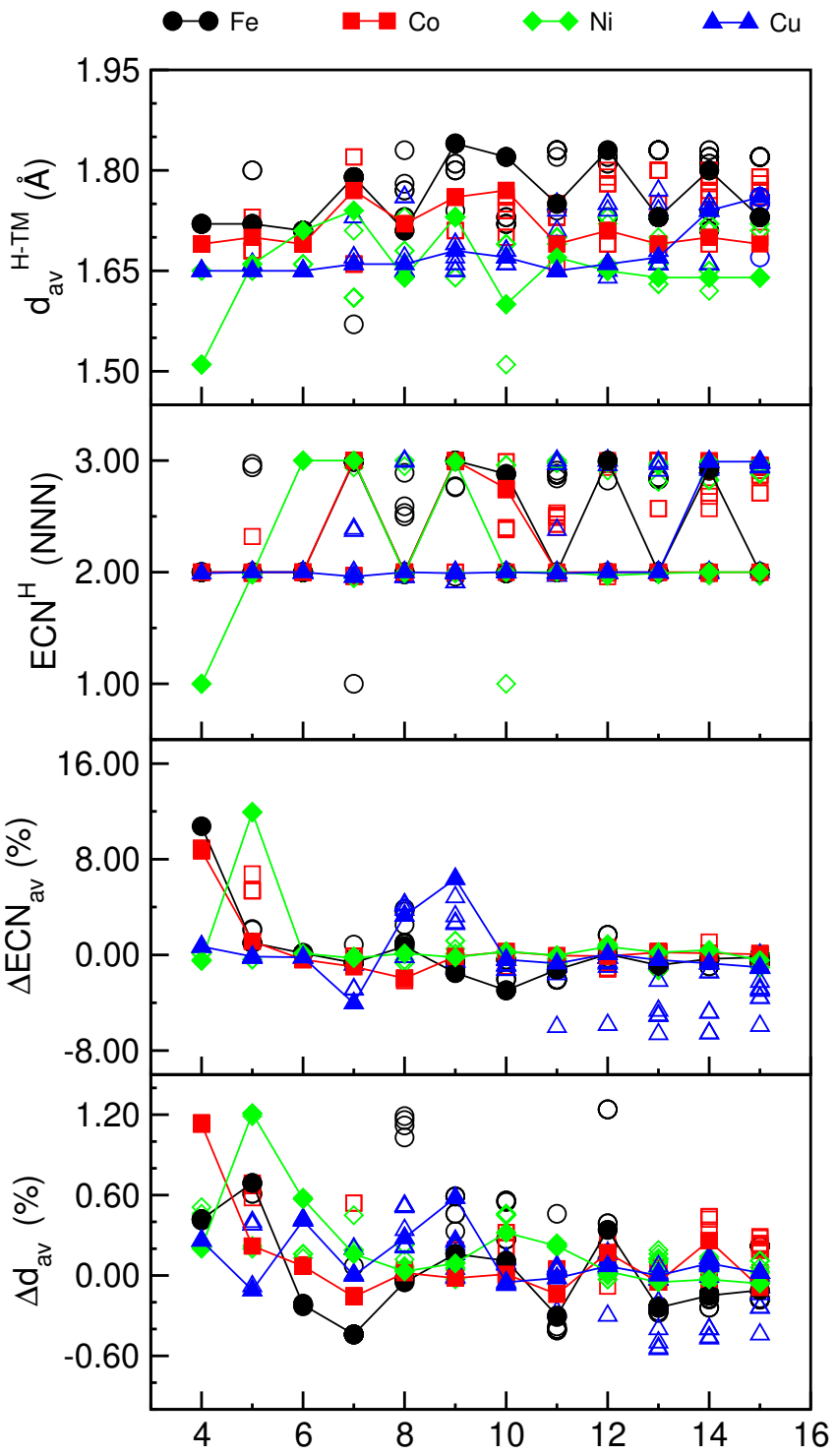

Figure S17: Structural properties for H/TM$_n$ systems where $n = 4 - 15$ and TM = Fe, Co, Ni, Cu.

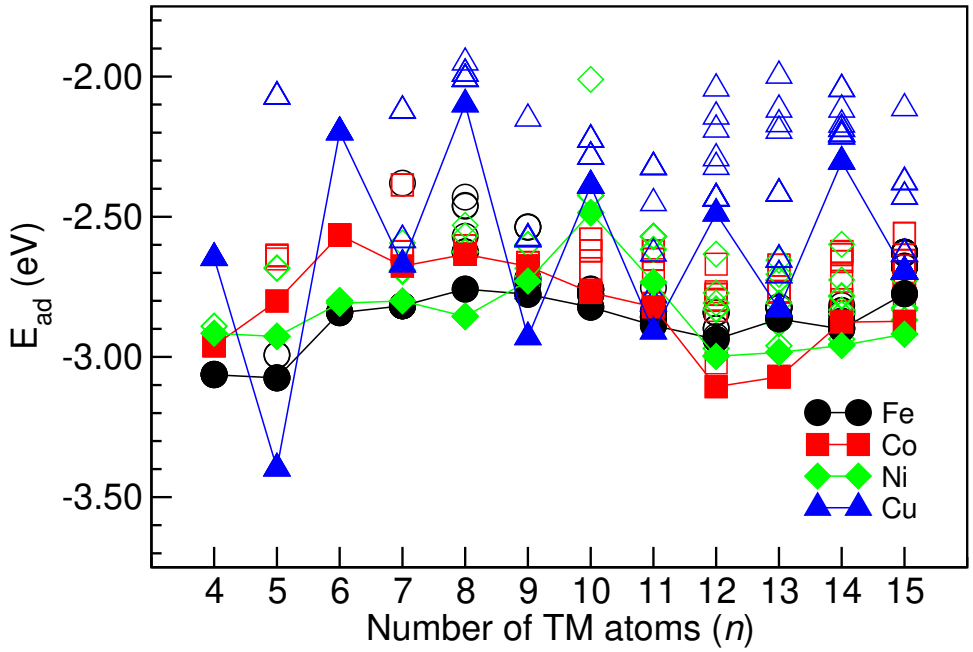

Figure S18: Adsorption energy for the H/TM$_n$ systems where $n = 4 - 15$ and TM = Fe, Co, Ni, and Cu.

- The adsorption of H preferable occurs on Cu_5 cluster, followed bay Co_{12} and Co_{13}, for the remaining $n < 12$, it presents similar magnitudes for Ni_n and Fe_n, except for $n = 9$.

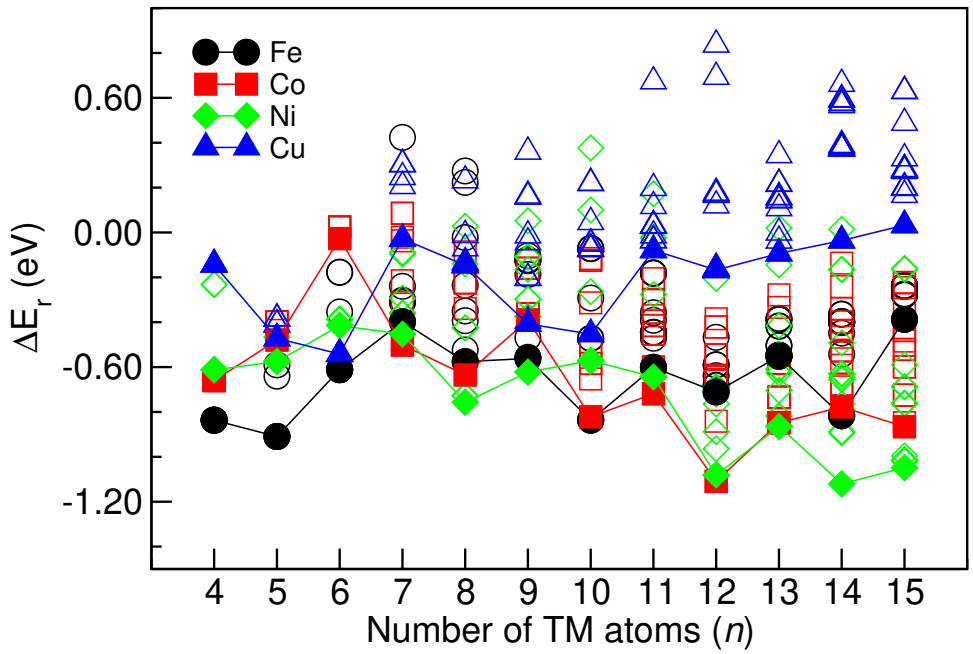

Figure S19: Reaction energy for the first CH_4 dehydrogenation step on the TM$_n$ clusters where $n = 4 - 15$ and TM = Fe, Co, Ni, Cu.

- The reaction energy (ΔE_r) indicates that the formation of the adsorbed CH_3+H product from the gas-phase molecules preferable occurs on Ni_n clusters for higher n values $n = 12-15$, where is observed a competition with Co_{11}, Co_{12}, Fe_{12}, and Co_{13}.

- For $6 < n < 12$ E_r has similar magnitudes between Fe_n, Co_n and Ni_n clusters.
- However, for $n < 7$, the reaction preferably occurs on Fe clusters.
- For almost all values of n, the reaction is not so favorable on Cu_n clusters, except for Cu_5 and Cu_6, and Cu_9 and Cu_{10} which has a larger and comparative values of $|E_r|$. However, for the other Cu_n clusters the magnitude of E_r is the smallest ones, nearing positive values.

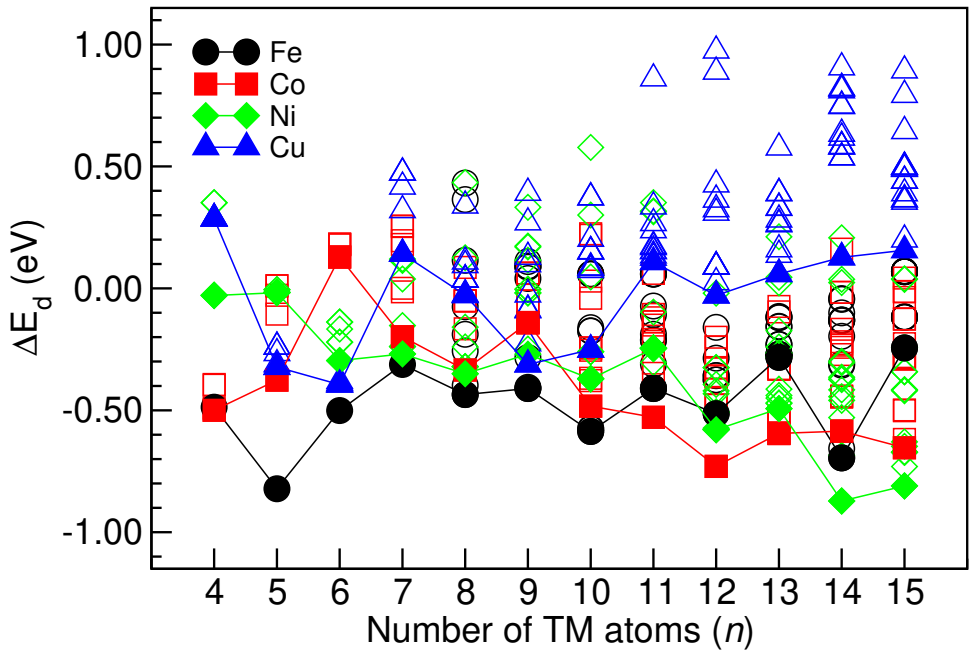

Figure S20: Dissociation energy for the first CH_4 dehydrogenation step on the TM_n clusters where $n = 4-15$ and TM = Fe, Co, Ni, Cu.

- The dissociation energy (ΔE_d) evaluates the exotermicity ($\Delta E_d < 0$) and endotermicity ($\Delta E_d > 0$) between two consecutive steps for an elementary reaction, in this case the dissociation of methane, i.e., $CH_4 \rightarrow CH_3$+H.
- For almost all values of n, $\Delta E_d < 0$, except for Cu_4, Co_6, and Cu_{11}-Cu_{15}, indicating that the CH_4 dissociation is an endothermic process on almost all Cu_n clusters.
- For $4 < n < 11$, has the largest magnitude of ΔE_d for Fe_n clusters, while for $11-13$ the higher exotermicty is observed for Co clusters.
- Ni_{14} and Ni_{15}, along with Fe_5 presents the largest values of ΔE_d.

11 Reactivity Analysis: Unity Bond Index-Quadratic Exponential Potential Framework

We calculated also the reaction and dissociation energies, i.e., ΔE_r and ΔE_d, where the first one evaluates the energetic tendency to form the adsorbed CH_3+H dissociated species from the gas-phase CH_4 and TM_n molecules, while the second measures the thermodynamic regime, i.e., exothermic or endothermic, between the CH_4/TM_n and $(CH_3+H)/TM_n$ consecutive elementary reactions, i.e., $CH_4 \rightarrow CH_3$+H ΔE_r and ΔE_d are defined by the following equations:

$$\Delta E_r = E_{tot}^{(CH_3+H)/TM_n} - (E_{tot}^{CH_4\ lowest} + E_{tot}^{TM_n\ lowest}) \,, \tag{13}$$

$$\Delta E_d = E_{tot,i}^{(CH_3+H)/TM_n} - E_{tot,i}^{CH_4/TM_n} \,. \tag{14}$$

The Unity Bond Index-Quadratic Exponential Potential (UBI-QEP) method, previously named BOC-MEP, is a hybrid semi-empirical/DFT method to access the thermodynamics and kinetics properties of adsorbed systems. This method is useful when the adsorbent does not present significant geometrical changes after the adsorption process, i.e., the bond indexes are conserved as unity after the adsorption process, in which directly implies that the adsorbates are relative small in comparison with the adsorbent species. In this work the UBI-QEP method was used to estimate the activation energies barriers (E_a) for $CH_4 \rightarrow CH_3$+H elementary reaction on the TM_n clusters, by employing the following formula:

$$E_a^{CH_4} = \phi \left(D_{ad}^{CH_4} + \frac{E_{ad}^{CH_3}.E_{ad}^{H}}{E_{ad}^{CH_3} + E_{ad}^{H}} \right) \,, \tag{15}$$

where ϕ is the bond index ($0 < \phi < 1$). $E_{ad}^{CH_3}$ and E_{ad}^{H} are the adsorption energies calculated for CH_3 and H species, respectively. Further, for all $E_a^{CH_4}$ calculations, we used a bond index of 0.5, which is the assumed value from the original method, implying an interpolation of the half-cross section between the potential energy surfaces of the gas-phase and adsorbed systems. $D_{ad}^{CH_4}$ is defined as the dissociation energy of the CH_4 adsorbed specie, and given by the following equation:

$$D_{ad}^{CH_4} = D_{gas-phase}^{CH_4\text{-}CH_3+H} + E_{ad}^{CH_4} - E_{ad}^{CH_3} - E_{ad}^{H} \,, \tag{16}$$

where $D_{gas-phase}^{CH_4\text{-}CH_3+H}$ is the dissociation energy of the gas-phase CH_4 specie towards the formation of CH_3+H. Thus, $D_{gas-phase}^{CH_4\text{-}CH_3+H}$ is defined as:

$$D_{gas-phase}^{CH_4\text{-}CH_3+H} = E_{tot\ lowest}^{CH_4} - E_{tot\ lowest}^{CH_3} - E_{tot\ lowest}^{H} \,. \tag{17}$$

where, $E_{tot\ lowest}^{CH_4}$, $E_{tot\ lowest}^{CH_3}$, $E_{tot\ lowest}^{H}$, are the DFT total energy for the gas-phase CH_4, CH_3 and H molecules, respectively. Thus, $D_{gas-phase}^{CH_4\text{-}CH_3+H}$ is the most sensitive parameter to the employed level of theory, therefore zero point energy (ZPE) corrections and higher order electronic correlation methods are required to obtain an accurate value, where experimental values can be also used if available in the literature.

Table S30: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Fe_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Fe_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
4	−0.35	−2.43	−3.06	−0.76	0.30
	−0.35	−2.44	−3.06	−0.77	0.30
	−0.35	−2.44	−3.06	−0.76	0.30
	−0.35	−2.44	−3.06	−0.76	0.30
5	−0.45	−2.44	−2.99	−0.59	0.38
	−0.44	−2.35	−2.99	−0.52	0.40
	−0.10	−2.59	−3.07	−1.18	0.11
	−0.09	−2.58	−3.07	−1.18	0.11
6	0.00	−2.30	−2.84	−0.74	0.26
	0.00	−2.30	−2.84	−0.75	0.26
	−0.11	−2.36	−2.84	−0.70	0.29
	−0.11	−2.11	−2.84	−0.45	0.38
	−0.11	−2.11	−2.84	−0.45	0.38
	−0.11	−2.15	−2.84	−0.49	0.37
7	−0.15	−1.99	−2.38	0.17	0.63
	0.07	−2.33	−2.82	−0.83	0.22
	−0.15	−2.07	−2.82	−0.35	0.42
	−0.15	−1.99	−2.82	−0.27	0.45
	−0.15	−1.99	−2.82	−0.27	0.45
	−0.15	−1.99	−2.82	2.55	0.45
	−0.12	−2.23	−2.82	−0.54	0.35
8	−0.16	−2.19	−2.43	−0.07	0.54
	−0.14	−2.02	−2.46	0.05	0.58
	−0.14	−2.24	−2.63	−0.34	0.44
	−0.16	−1.94	−2.62	−0.02	0.55
	−0.16	−2.06	−2.63	−0.14	0.51
	−0.14	−2.03	−2.46	0.03	0.57
	−0.13	−2.14	−2.76	−0.39	0.41
	−0.14	−2.21	−2.57	−0.25	0.47
9	−0.21	−2.37	−2.78	−0.54	0.37
	−0.21	−2.18	−2.54	−0.12	0.53
	−0.21	−2.18	−2.54	−0.12	0.53
	−0.21	−2.18	−2.54	−0.11	0.53
	−0.23	−2.37	−2.72	−0.47	0.40
	−0.18	−2.11	−2.72	−0.25	0.47
	−0.15	−2.05	−2.78	−0.29	0.45

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Table S30: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Fe_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Fe_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.15	−2.05	−2.74	−0.25	0.46
10	−0.13	−2.00	−2.78	−0.26	0.45
	−0.13	−2.00	−2.78	−0.26	0.45
	−0.13	−2.00	−2.78	−0.26	0.45
	−0.13	−2.27	−2.76	−0.51	0.37
	−0.30	−2.59	−2.82	−0.73	0.31
	−0.30	−2.59	−2.82	−0.72	0.31
	−0.26	−2.49	−2.76	−0.60	0.35
	−0.26	−2.49	−2.76	−0.60	0.35
	−0.25	−2.49	−2.76	−0.61	0.35
11	−0.24	−2.25	−2.86	−0.48	0.39
	−0.24	−2.25	−2.86	−0.48	0.39
	−0.24	−2.25	−2.86	−0.48	0.39
	−0.29	−2.48	−2.83	−0.64	0.34
	−0.28	−2.48	−2.75	−0.56	0.37
	−0.23	−2.37	−2.86	−0.61	0.34
	−0.23	−2.38	−2.86	−0.61	0.34
	−0.14	−2.38	−2.86	−0.71	0.29
	−0.28	−2.48	−2.86	−0.67	0.33
	−0.19	−2.48	−2.89	−0.79	0.27
	−0.19	−2.48	−2.89	−0.79	0.27
12	−0.31	−2.58	−2.84	−0.73	0.31
	−0.23	−2.39	−2.84	−0.61	0.34
	−0.23	−2.39	−2.84	−0.61	0.34
	−0.35	−2.56	−2.94	−0.75	0.31
	−0.31	−2.58	−2.90	−0.78	0.29
	−0.31	−2.58	−2.90	−0.78	0.29
	−0.20	−2.44	−2.93	−0.78	0.27
13	−0.26	−2.35	−2.82	−0.52	0.38
	−0.27	−2.42	−2.82	−0.59	0.36
	−0.27	−2.42	−2.83	−0.58	0.36
	−0.26	−2.37	−2.82	−0.54	0.37
	−0.26	−2.37	−2.82	−0.54	0.37
	−0.27	−2.42	−2.86	−0.62	0.35
	−0.27	−2.42	−2.86	−0.62	0.35
	−0.28	−2.42	−2.86	−0.62	0.35
	−0.26	−2.35	−2.86	−0.55	0.37

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Table S30: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Fe_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Fe_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.28	−2.42	−2.86	−0.62	0.35
	−0.27	−2.42	−2.86	−0.62	0.35
	−0.28	−2.42	−2.86	−0.62	0.35
	−0.27	−2.42	−2.86	−0.62	0.34
14	−0.32	−2.30	−2.85	−0.44	0.42
	−0.32	−2.30	−2.85	−0.43	0.42
	−0.27	−2.37	−2.90	−0.61	0.35
	−0.20	−2.37	−2.83	−0.62	0.34
	−0.26	−2.36	−2.87	−0.58	0.36
	−0.27	−2.37	−2.87	−0.58	0.36
	−0.27	−2.37	−2.87	−0.58	0.36
	−0.30	−2.37	−2.83	−0.51	0.39
	−0.13	−2.40	−2.82	−0.70	0.30
	−0.23	−2.39	−2.90	−0.67	0.32
	−0.16	−2.29	−2.90	−0.64	0.32
	−0.12	−2.34	−2.90	−0.72	0.29
15	−0.31	−2.22	−2.63	−0.15	0.53
	−0.31	−2.22	−2.63	−0.15	0.53
	−0.31	−2.22	−2.63	−0.15	0.53
	−0.31	−2.21	−2.63	−0.14	0.53
	−0.31	−2.21	−2.63	−0.15	0.53
	−0.31	−2.22	−2.63	−0.15	0.53
	−0.31	−2.22	−2.63	−0.15	0.53
	−0.31	−2.21	−2.63	−0.14	0.53
	−0.31	−2.22	−2.63	−0.15	0.53
	−0.31	−2.22	−2.63	−0.15	0.53
	−0.16	−2.37	−2.68	−0.50	0.38
	−0.16	−2.37	−2.68	−0.50	0.38
	−0.16	−2.37	−2.68	−0.50	0.38
	−0.16	−2.38	−2.68	−0.51	0.38
	−0.14	−2.36	−2.77	−0.60	0.34

Table S31: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Co_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Co_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
4	−0.16	−2.60	−2.96	−1.01	0.19
	−0.26	−2.60	−2.96	−0.90	0.24
	−0.16	−2.60	−2.96	−1.01	0.19
	−0.26	−2.60	−2.96	−0.90	0.24
5	−0.41	−2.49	−2.64	−0.33	0.47
	−0.41	−2.49	−2.64	−0.33	0.48
	−0.29	−2.52	−2.64	−0.48	0.41
	−0.41	−2.55	−2.80	−0.56	0.39
	−0.10	−2.31	−2.65	−0.47	0.38
6	−0.16	−2.34	−2.57	−0.36	0.43
	−0.16	−2.34	−2.57	−0.36	0.43
	−0.16	−2.34	−2.56	−0.36	0.43
	−0.16	−2.34	−2.56	−0.36	0.43
	−0.16	−2.34	−2.57	−0.36	0.43
	−0.16	−2.34	−2.57	−0.36	0.43
7	−0.17	−2.26	−2.62	−0.32	0.45
	−0.17	−2.26	−2.62	−0.32	0.44
	−0.22	−2.36	−2.39	−0.14	0.52
	−0.22	−2.36	−2.39	−0.14	0.52
	−0.22	−2.36	−2.68	−0.43	0.41
	−0.30	−2.26	−2.63	−0.19	0.51
8	−0.30	−2.31	−2.62	−0.24	0.49
	−0.17	−2.02	−2.60	−0.06	0.54
	−0.17	−2.02	−2.63	−0.10	0.52
	−0.17	−2.02	−2.63	−0.09	0.53
	−0.17	−2.02	−2.63	−0.09	0.53
	−0.17	−2.19	−2.60	−0.23	0.48
	−0.30	−2.38	−2.63	−0.32	0.46
9	−0.22	−2.25	−2.66	−0.30	0.46
	−0.22	−2.25	−2.66	−0.30	0.46
	−0.22	−2.25	−2.66	−0.30	0.46
	−0.22	−2.25	−2.68	−0.32	0.45
	−0.22	−2.25	−2.68	−0.31	0.45
	−0.22	−2.25	−2.68	−0.31	0.45
	−0.34	−2.41	−2.68	−0.36	0.45
	−0.34	−2.41	−2.68	−0.36	0.45

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Table S31: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Co_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Co_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
10	−0.34	−2.30	−2.58	−0.15	0.53
	−0.34	−2.30	−2.58	−0.15	0.53
	−0.15	−2.14	−2.68	−0.28	0.45
	−0.27	−2.51	−2.62	−0.47	0.40
	−0.15	−2.39	−2.77	−0.62	0.33
	−0.27	−2.50	−2.77	−0.61	0.35
	−0.34	−2.30	−2.62	−0.19	0.52
	−0.34	−2.30	−2.62	−0.19	0.52
	−0.27	−2.39	−2.68	−0.41	0.43
	−0.34	−2.30	−2.77	−0.34	0.46
11	−0.27	−2.16	−2.81	−0.32	0.45
	−0.19	−2.56	−2.81	−0.79	0.27
	−0.15	−2.56	−2.81	−0.83	0.25
	−0.25	−2.40	−2.62	−0.38	0.44
	−0.28	−2.56	−2.62	−0.52	0.39
	−0.28	−2.56	−2.62	−0.52	0.39
	−0.15	−2.59	−2.65	−0.71	0.30
	−0.24	−2.59	−2.65	−0.61	0.35
	−0.25	−2.40	−2.69	−0.45	0.41
	−0.19	−2.33	−2.82	−0.57	0.35
12	−0.18	−2.33	−2.80	−0.55	0.36
	−0.18	−2.33	−2.80	−0.56	0.36
	−0.18	−2.12	−2.80	−0.35	0.43
	−0.22	−2.53	−2.78	−0.69	0.31
	−0.22	−2.49	−2.78	−0.65	0.33
	−0.22	−2.49	−2.78	−0.65	0.33
	−0.19	−2.24	−2.79	−0.46	0.39
	−0.30	−2.35	−3.02	−0.69	0.32
	−0.30	−2.48	−2.67	−0.46	0.41
	−0.18	−2.24	−2.83	−0.50	0.37
	−0.36	−2.42	−2.77	−0.44	0.43
	−0.38	−2.55	−3.11	−0.89	0.25
13	−0.20	−2.25	−2.67	−0.34	0.44
	−0.24	−2.53	−2.67	−0.57	0.36
	−0.25	−2.45	−3.07	−0.87	0.24
	−0.25	−2.45	−3.07	−0.87	0.24
	−0.25	−2.45	−3.07	−0.88	0.24

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Table S31: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Co_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Co_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.25	−2.44	−3.07	−0.87	0.24
	−0.21	−2.15	−2.77	−0.32	0.44
	−0.20	−2.15	−2.77	−0.32	0.44
	−0.20	−2.15	−2.77	−0.32	0.44
	−0.21	−2.15	−2.77	−0.32	0.44
	−0.25	−2.45	−2.77	−0.57	0.36
	−0.25	−2.78	−2.77	−0.91	0.24
	−0.25	−2.78	−2.77	−0.91	0.24
14	−0.30	−2.38	−2.88	−0.57	0.36
	−0.23	−2.53	−2.84	−0.75	0.29
	−0.32	−2.48	−2.64	−0.40	0.44
	−0.23	−2.53	−2.66	−0.57	0.36
	−0.21	−2.41	−2.84	−0.66	0.32
	−0.25	−2.32	−2.73	−0.41	0.42
	−0.30	−2.39	−2.63	−0.33	0.46
	−0.29	−2.38	−2.66	−0.37	0.44
	−0.32	−2.47	−2.63	−0.39	0.44
	−0.23	−2.77	−2.62	−0.77	0.29
	−0.23	−2.76	−2.62	−0.77	0.29
	−0.24	−2.77	−2.63	−0.77	0.29
	−0.19	−2.30	−2.80	−0.51	0.37
	−0.19	−2.30	−2.80	−0.51	0.37
15	−0.21	−2.23	−2.56	−0.19	0.50
	−0.21	−2.23	−2.56	−0.19	0.50
	−0.21	−2.38	−2.68	−0.46	0.40
	−0.36	−2.55	−2.68	−0.48	0.41
	−0.31	−2.37	−2.75	−0.42	0.43
	−0.31	−2.37	−2.75	−0.42	0.43
	−0.26	−2.38	−2.76	−0.50	0.39
	−0.24	−2.43	−2.66	−0.46	0.41
	−0.24	−2.43	−2.66	−0.46	0.40
	−0.31	−2.37	−2.75	−0.42	0.43
	−0.31	−2.37	−2.75	−0.42	0.43
	−0.22	−2.54	−2.70	−0.62	0.34
	−0.23	−2.54	−2.70	−0.62	0.34
	−0.23	−2.54	−2.87	−0.79	0.28
	−0.21	−2.69	−2.80	−0.89	0.24

Table S32: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Ni_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Ni_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
4	−0.58	−2.80	−2.92	−0.75	0.34
	−0.58	−2.80	−2.92	−0.75	0.34
	−0.58	−2.81	−2.89	−0.72	0.35
	−0.58	−2.81	−2.89	−0.72	0.35
5	−0.46	−2.29	−2.68	−0.12	0.56
	−0.46	−2.29	−2.68	−0.12	0.56
	−0.56	−2.57	−2.93	−0.55	0.41
	−0.56	−2.53	−2.93	−0.51	0.42
	−0.56	−2.53	−2.93	−0.51	0.42
6	−0.25	−2.39	−2.80	−0.56	0.37
	−0.17	−2.39	−2.80	−0.64	0.33
	−0.17	−2.39	−2.80	−0.64	0.33
	−0.25	−2.39	−2.80	−0.55	0.37
	−0.12	−2.39	−2.81	−0.70	0.30
	−0.25	−2.39	−2.81	−0.57	0.36
7	−0.21	−2.31	−2.69	−0.40	0.42
	−0.21	−2.31	−2.69	−0.40	0.42
	−0.33	−2.20	−2.69	−0.18	0.52
	−0.21	−2.41	−2.69	−0.51	0.38
	−0.12	−2.41	−2.70	−0.59	0.34
	−0.21	−2.31	−2.59	−0.30	0.46
	−0.18	−2.41	−2.80	−0.64	0.33
8	−0.41	−2.23	−2.57	0.00	0.60
	−0.27	−2.27	−2.76	−0.37	0.44
	−0.18	−2.27	−2.62	−0.32	0.45
	−0.27	−2.27	−2.63	−0.24	0.49
	−0.41	−2.23	−2.85	−0.29	0.48
	−0.41	−2.23	−2.86	−0.29	0.48
	−0.41	−2.23	−2.86	−0.29	0.48
	−0.41	−2.23	−2.53	0.04	0.61
9	−0.28	−2.21	−2.73	−0.27	0.48
	−0.28	−2.21	−2.71	−0.25	0.48
	−0.28	−2.21	−2.71	−0.25	0.48
	−0.28	−2.21	−2.68	−0.22	0.49
	−0.28	−2.21	−2.60	−0.14	0.53
	−0.28	−2.21	−2.60	−0.14	0.53

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Table S32: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Ni_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Ni_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.37	−2.27	−2.73	−0.24	0.50
	−0.37	−2.27	−2.73	−0.24	0.50
	−0.37	−2.27	−2.73	−0.24	0.50
10	−0.20	−1.95	−2.01	0.63	0.81
	−0.20	−1.86	−2.42	0.31	0.68
	−0.31	−1.97	−2.48	0.25	0.67
	−0.31	−1.97	−2.42	0.31	0.70
	−0.20	−1.97	−2.42	0.20	0.64
	−0.31	−1.97	−2.42	0.31	0.70
	−0.20	−2.21	−2.49	−0.10	0.53
11	−0.18	−1.91	−2.62	0.05	0.57
	−0.34	−2.09	−2.73	−0.09	0.55
	−0.34	−2.09	−2.73	−0.10	0.54
	−0.18	−1.95	−2.62	0.00	0.56
	−0.40	−2.56	−2.57	−0.35	0.47
	−0.40	−2.57	−2.57	−0.35	0.47
	−0.40	−2.57	−2.57	−0.35	0.47
	−0.34	−2.09	−2.57	0.07	0.61
	−0.40	−2.57	−2.57	−0.35	0.47
	−0.40	−2.57	−2.57	−0.35	0.47
	−0.40	−2.57	−2.57	−0.35	0.47
12	−0.19	−2.36	−2.63	−0.42	0.41
	−0.18	−2.79	−2.77	−0.99	0.20
	−0.37	−2.31	−3.00	−0.55	0.38
	−0.37	−2.31	−3.00	−0.55	0.38
	−0.30	−2.31	−3.00	−0.62	0.34
	−0.30	−2.31	−3.00	−0.62	0.34
	−0.33	−2.83	−2.83	−0.94	0.24
	−0.38	−2.72	−2.81	−0.76	0.31
	−0.39	−2.29	−2.83	−0.34	0.46
	−0.50	−2.55	−2.97	−0.62	0.37
	−0.50	−2.54	−2.97	−0.62	0.37
	−0.50	−2.54	−2.97	−0.62	0.37
13	−0.19	−1.98	−2.81	−0.20	0.48
	−0.19	−1.98	−2.98	−0.38	0.40
	−0.44	−2.32	−2.66	−0.15	0.54
	−0.30	−2.69	−2.71	−0.70	0.32

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Table S32: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Ni_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Ni_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.19	−2.28	−2.96	−0.66	0.32
	−0.44	−2.32	−2.98	−0.48	0.41
	−0.35	−2.69	−2.96	−0.91	0.25
	−0.44	−2.32	−2.98	−0.48	0.41
	−0.37	−2.70	−2.88	−0.82	0.29
	−0.38	−2.69	−2.88	−0.81	0.29
	−0.37	−2.69	−2.73	−0.66	0.35
	−0.39	−2.69	−2.73	−0.64	0.36
	−0.39	−2.70	−2.73	−0.64	0.36
14	−0.19	−1.93	−2.73	−0.07	0.53
	−0.20	−2.00	−2.96	−0.36	0.41
	−0.19	−1.98	−2.78	−0.18	0.49
	−0.19	−2.21	−2.96	−0.59	0.34
	−0.26	−2.59	−2.85	−0.79	0.28
	−0.26	−2.59	−2.85	−0.79	0.28
	−0.28	−2.27	−2.60	−0.20	0.51
	−0.35	−2.61	−2.83	−0.70	0.33
	−0.35	−2.68	−2.94	−0.88	0.26
	−0.34	−2.68	−2.90	−0.85	0.27
	−0.36	−2.58	−2.95	−0.78	0.30
	−0.20	−2.31	−2.94	−0.66	0.32
	−0.36	−2.19	−2.79	−0.22	0.50
	−0.25	−2.66	−2.96	−0.98	0.21
15	−0.20	−2.04	−2.92	−0.37	0.42
	−0.20	−2.20	−2.73	−0.33	0.44
	−0.35	−2.15	−2.73	−0.14	0.53
	−0.35	−2.15	−2.92	−0.34	0.45
	−0.35	−2.15	−2.92	−0.34	0.45
	−0.34	−2.65	−2.92	−0.83	0.28
	−0.35	−2.65	−2.92	−0.83	0.28
	−0.34	−2.20	−2.83	−0.30	0.47
	−0.37	−2.18	−2.87	−0.30	0.47
	−0.20	−2.61	−2.87	−0.88	0.24
	−0.37	−2.19	−2.87	−0.30	0.47
	−0.35	−2.16	−2.83	−0.25	0.49
	−0.35	−2.16	−2.83	−0.25	0.49
	−0.35	−2.16	−2.83	−0.25	0.49

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Table S32: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Ni_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Ni_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	-0.32	-2.54	-2.83	-0.66	0.34

Table S33: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Cu_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Cu_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
4	-0.43	-2.03	-2.65	0.14	0.65
	-0.43	-2.03	-2.65	0.14	0.65
	-0.43	-2.03	-2.65	0.14	0.65
5	-0.14	-2.40	-3.40	-1.26	0.07
	-0.14	-2.40	-3.40	-1.26	0.07
	-0.09	-2.97	-2.07	-0.56	0.33
	-0.14	-2.97	-2.07	-0.50	0.36
	-0.16	-2.97	-2.07	-0.49	0.37
6	-0.16	-1.61	-2.20	0.74	0.83
	-0.15	-1.61	-2.20	0.73	0.83
	-0.15	-1.61	-2.20	0.73	0.83
	-0.15	-1.61	-2.20	0.74	0.83
7	-0.17	-1.68	-2.58	0.29	0.66
	-0.17	-1.68	-2.58	0.29	0.66
	-0.17	-1.68	-2.67	0.20	0.62
	-0.11	-2.45	-2.12	-0.07	0.53
	-0.17	-1.68	-2.12	0.75	0.85
8	-0.11	-1.53	-1.99	0.97	0.92
	-0.11	-1.53	-2.10	0.86	0.87
	-0.18	-1.73	-2.01	0.83	0.88
	-0.11	-1.53	-1.99	0.97	0.92
	-0.11	-1.53	-1.95	1.01	0.93
	-0.18	-1.73	-2.01	0.83	0.88
	-0.18	-1.73	-2.01	0.83	0.88
	-0.18	-1.73	-2.01	0.83	0.88
9	-0.03	-2.40	-2.70	-0.68	0.30

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Table S33: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Cu_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Cu_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.11	−1.51	−2.58	0.40	0.68
	0.03	−2.40	−2.77	−0.81	0.24
	−0.11	−1.55	−2.15	0.80	0.85
	−0.02	−2.52	−2.58	−0.69	0.29
	−0.12	−2.48	−2.58	−0.55	0.36
	−0.11	−2.48	−2.58	−0.56	0.35
	−0.17	−1.67	−2.93	−0.04	0.51
	−0.10	−2.42	−2.75	−0.69	0.30
10	−0.15	−1.92	−2.39	0.23	0.65
	−0.15	−1.92	−2.39	0.23	0.65
	−0.16	−1.91	−2.39	0.25	0.65
	−0.20	−1.99	−2.29	0.32	0.69
	−0.20	−1.99	−2.29	0.32	0.69
	−0.20	−1.99	−2.29	0.32	0.69
	−0.16	−1.82	−2.23	0.50	0.75
	−0.16	−1.82	−2.23	0.50	0.75
	−0.15	−1.82	−2.22	0.49	0.75
	−0.20	−1.99	−2.39	0.21	0.65
11	−0.19	−2.08	−2.78	−0.28	0.45
	−0.14	−2.24	−2.63	−0.35	0.43
	−0.15	−1.89	−2.91	−0.26	0.44
	−0.09	−2.50	−2.32	−0.34	0.43
	−0.21	−2.50	−2.32	−0.23	0.49
	−0.14	−2.50	−2.32	−0.30	0.45
	−0.14	−2.38	−2.81	−0.65	0.32
	−0.15	−2.37	−2.81	−0.65	0.32
	−0.19	−2.02	−2.78	−0.22	0.48
	−0.19	−2.02	−2.78	−0.22	0.47
	−0.19	−2.08	−2.45	0.05	0.59
12	−0.14	−1.87	−2.32	0.34	0.69
	−0.19	−2.02	−2.14	0.42	0.73
	−0.25	−1.90	−2.49	0.25	0.66
	−0.19	−1.90	−2.19	0.49	0.75
	−0.16	−1.97	−2.29	0.28	0.67
	−0.19	−1.58	−2.04	0.96	0.92
	−0.25	−1.90	−2.44	0.30	0.69
	−0.25	−1.90	−2.43	0.30	0.69
	−0.25	−1.90	−2.43	0.31	0.69

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Table S33: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Cu_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Cu_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.25	−1.89	−2.44	0.32	0.69
	−0.14	−1.69	−2.43	0.41	0.70
	−0.19	−1.90	−2.43	0.25	0.66
13	−0.23	−2.19	−2.17	0.27	0.68
	−0.11	−2.23	−2.83	−0.56	0.35
	−0.11	−2.23	−2.83	−0.55	0.35
	−0.23	−2.19	−2.65	−0.22	0.49
	−0.23	−2.19	−2.65	−0.22	0.49
	−0.12	−1.65	−2.42	0.44	0.71
	−0.12	−1.65	−2.42	0.44	0.71
	−0.17	−2.27	−2.12	0.18	0.64
	−0.16	−2.15	−2.19	0.21	0.65
	−0.17	−2.26	−2.42	−0.12	0.52
	−0.15	−2.31	−2.00	0.23	0.65
14	−0.24	−1.69	−2.30	0.64	0.81
	−0.16	−1.80	−2.04	0.70	0.83
	−0.16	−1.80	−2.04	0.70	0.83
	−0.24	−1.73	−2.21	0.69	0.83
	−0.24	−1.73	−2.21	0.69	0.83
	−0.24	−1.73	−2.12	0.78	0.87
	−0.24	−1.69	−2.30	0.64	0.81
	−0.15	−1.78	−2.19	0.57	0.78
	−0.15	−1.78	−2.19	0.57	0.78
	−0.24	−1.69	−2.21	0.74	0.85
	−0.21	−1.68	−2.21	0.71	0.83
	−0.21	−1.68	−2.21	0.71	0.83
	−0.21	−1.68	−2.20	0.72	0.83
	−0.16	−1.70	−2.17	0.68	0.82
15	−0.16	−2.21	−2.43	−0.08	0.54
	−0.26	−1.94	−2.43	0.28	0.68
	−0.16	−2.21	−2.69	−0.36	0.43
	−0.16	−2.21	−2.69	−0.35	0.43
	−0.16	−2.21	−2.63	−0.29	0.45
	−0.16	−2.21	−2.63	−0.29	0.45
	−0.22	−1.94	−2.70	−0.02	0.55
	−0.22	−1.94	−2.70	−0.02	0.55
	−0.22	−1.94	−2.70	−0.02	0.55
	−0.22	−1.94	−2.38	0.29	0.68

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Table S33: Energetic parameters to calculate the activation energy for $CH_4 \rightarrow CH_3 + H$ reaction on the Cu_n clusters, considering all parent n CH_4 energy adsorption configurations and without the H co-adsorption effect. For all calculation $D^{CH_4}_{gas-phase}$ is a constant calculated value, i.e., -4.39 eV.

Cu_n	$E^{CH_4}_{ad}$ (eV)	$E^{CH_3}_{ad}$ (eV)	E^{H}_{ad} (eV)	$D^{CH_4}_{ad}$ (eV)	E_a (eV)
	−0.19	−2.30	−2.69	−0.42	0.41
	−0.16	−1.71	−2.69	0.15	0.60
	−0.17	−2.20	−2.11	0.24	0.66
	−0.19	−2.30	−2.63	−0.35	0.44
	−0.17	−1.97	−2.38	0.21	0.65
	−0.35	−2.16	−2.83	−0.25	0.49
	−0.32	−2.54	−2.83	−0.66	0.34

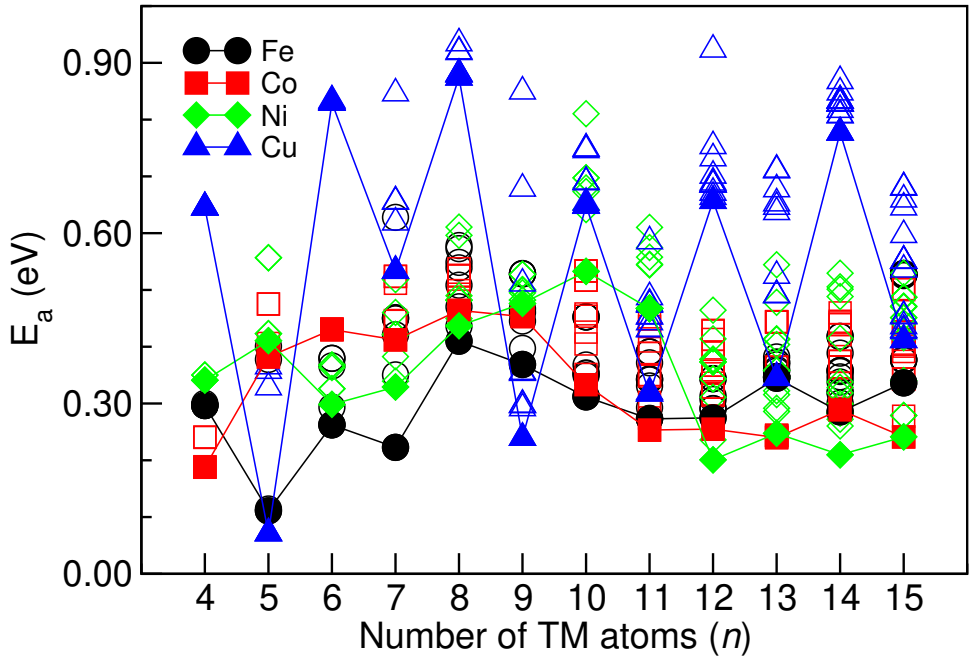

Figure S23: Activation energy barriers calculated with UBI-QEP method for the first CH_4 dehydrogenation step towards CH_3+H formation on TM_n clusters where $n = 4 - 15$ and TM = Fe, Co, Ni, Cu.

- The smallest E_a values are observed for Cu_5 and Fe_5 clusters, followed by Co_4 cluster.
- For $5 < n < 9$, Fe_n clusters present the lowest value of E_a, while for $n = 10$ and $n = 11$ Fe_n and Co_n clusters present similar magnitudes of E_a.
- Above $n = 11$, Ni_n clusters present the lowest values of E_a while the largest magnitude are observed for Cu_n clusters, where the only exceptions occur for Cu_5, Cu_9, and Cu_{11}.

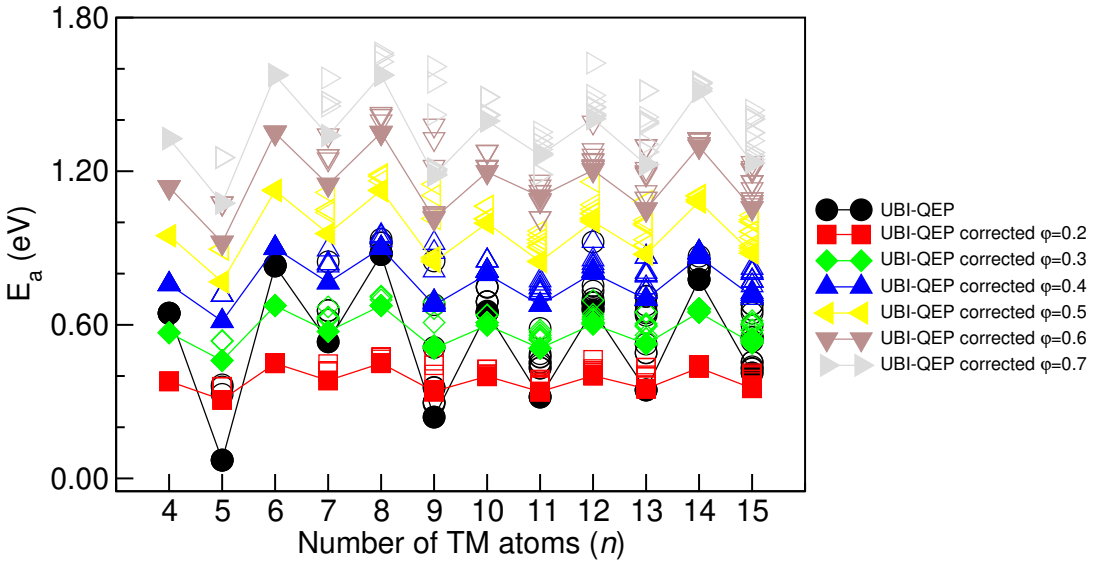

Figure S24: Activation energy barriers calculated with UBI-QEP method and Matteo–Reuster correction for the first CH_4 dehydrogenation step towards CH_3+H formation on Cu_n clusters where $n = 4$-15.

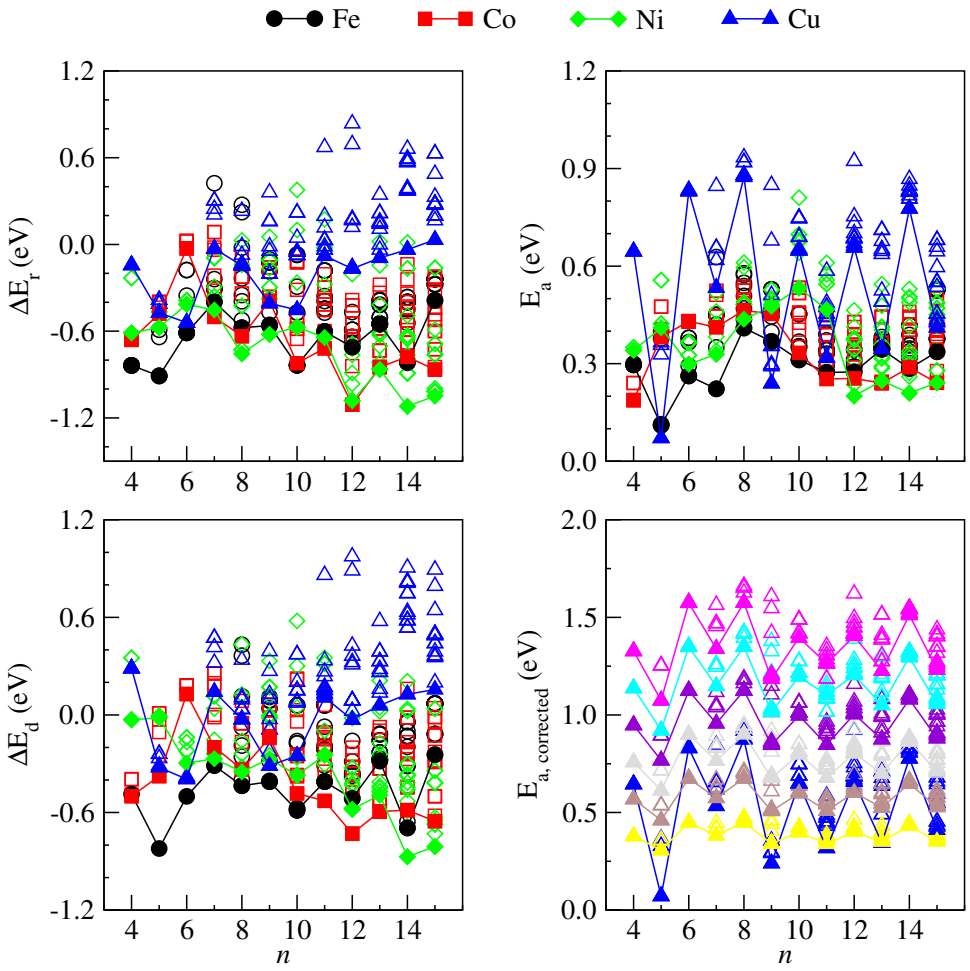

Figure S25: Energetic parameters, reaction energy (ΔE_r), dissociation energy (ΔE_d), and activation energy (E_a), for the CH_4 dissociation on the TM_n clusters where $n = 4 - 15$ and TM = Fe, Co, Ni and Cu. The Matteo–Reuster UBI-QEP correction for Cu_n clusters ($E_{a,corrected}$) are present in function of the ϕ parameter, where ϕ = 0.2 (yellow), 0.3 (brown), 0.4 (gray), 0.5 (violet), 0.6 (ciane), and 0.7 (magenta).

- We observed that applying the Matteo–Reuster UBI-QEP correction, the only difference observed in comparison with the original UBI-QEP approach, is the displacement of E_a magnitude towards largest values.

- Thus, the same behaviour is maintained, leading to the almost the same behavior of E_a by ranging ϕ from 0.2 to 0.7, where the lowest value of E_a is observed for Cu_5 followed by Cu_9 cluster.

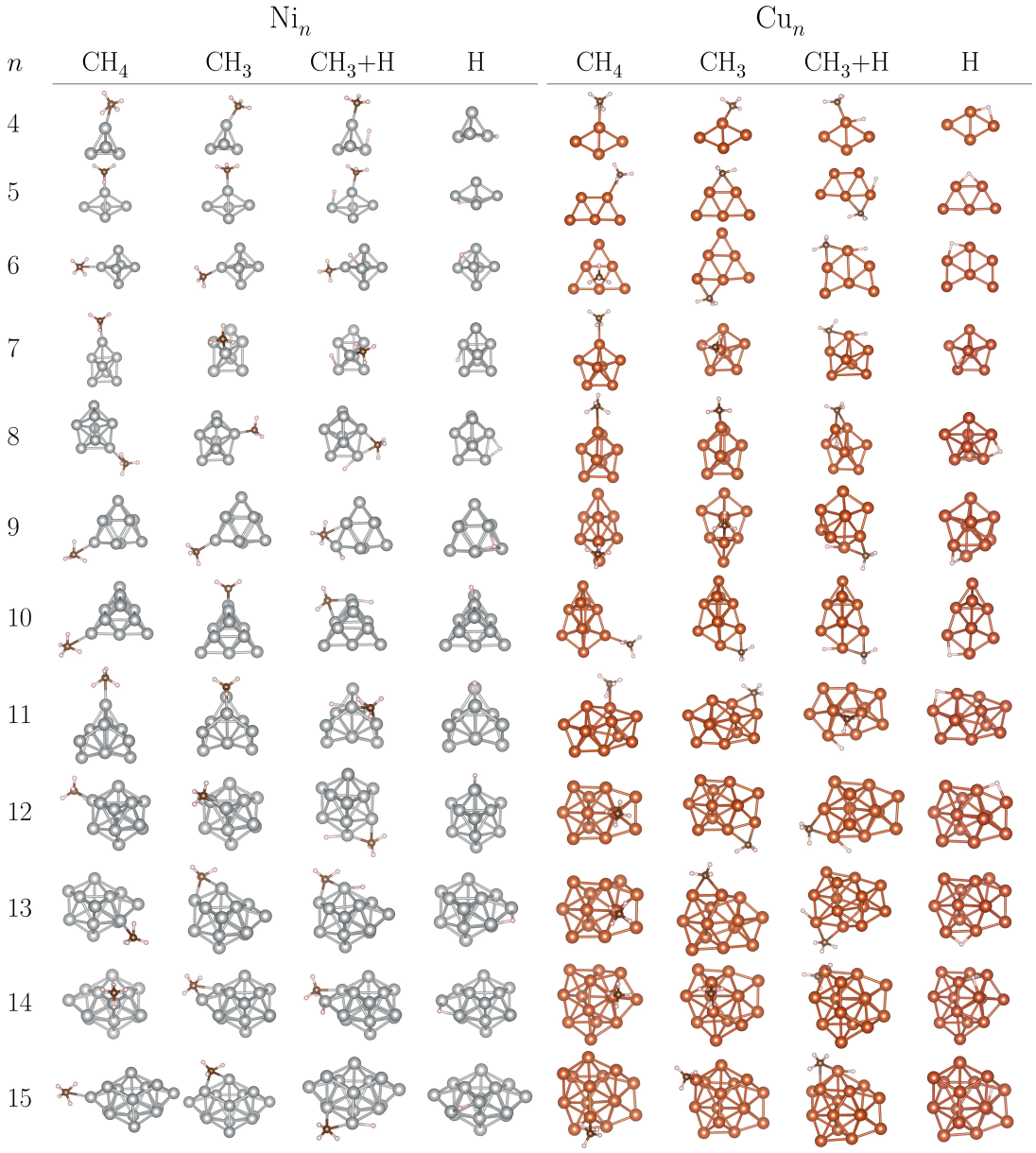

Figure S21: Lowest energy configurations for the adsorption of CH_4, CH_3, H and co-adsorbed CH_3+H on the TM_n clusters where $n = 4 - 15$ and TM = Ni and Cu.

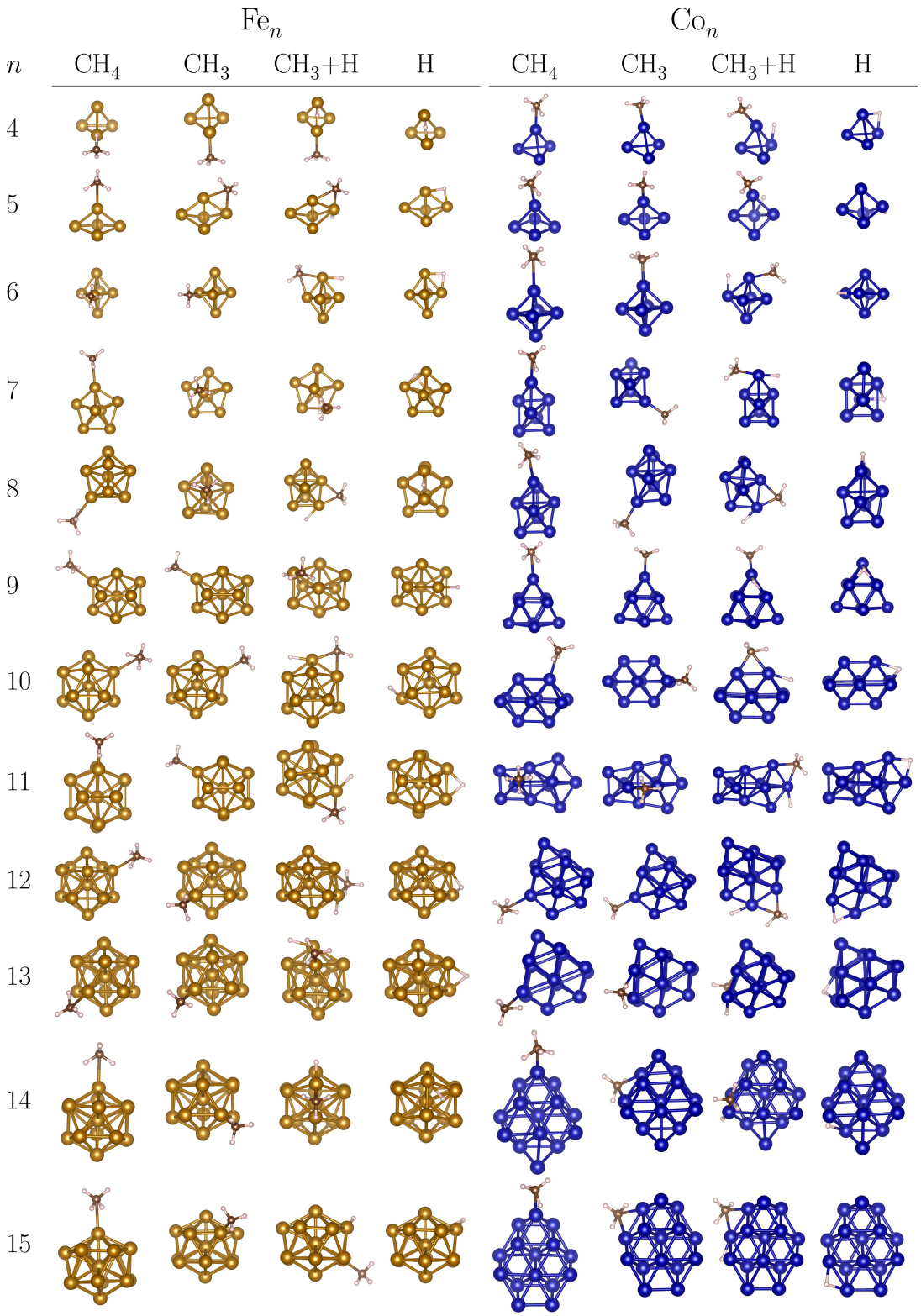

Figure S22: Lowest energy configurations for the adsorption of CH_4, CH_3, H and co-adsorbed CH_3+H on the TM_n clusters where $n = 4 - 15$ and TM = Fe and Co.

12 Spearman Rank Correlation Analysis

We employed the Spearman Rank Correlation Analysis to investigate the correlation between the structural, energetic and electronic properties with the adsorption and activation energies of all configurations. Therefore, the Spearman correlation coefficient is obtained as follows:

$$r_s = 1 - \frac{6\sum D_i^2}{n(n^2-1)}\,, \tag{18}$$

where n is the number of observations and D_i is the difference between each observation points. Thus, the value of r_s ranges from -1 to 1. (i) When $r_s \approx -1$, it means that the de/increase of one variable is associated with the in/decrease of the other variable, i.e., inverse correlation; (ii) $r_s \approx 0$ no-correlation is observed between the ranked data. $iii) r_s \approx 1$, a direct correlation is observed, which means that the two variables in/decrease together.

We investigated the following features for the Spearman rank correlation analysis of the adsorption and activation energy, i.e., E_{ad} and E_a, for the CH_4, CH_3, H, CH_3+H species on the TM_n for n = 4-15 and TM = Fe, Co, Ni, and Cu, as follows: relative total energy (ΔE_{tot}), total magnetic moment per atom (m_{tot}), eigenvalue of the HOMO state (ε_{homo}), eigenvalue of the LUMO state (ε_{lumo}), LUMO-HOMO energy gap (E_g), average effective coordination number of the cluster ($\mathrm{ECN}_{av}^{cluster}$), average bond length of the cluster ($d_{av}^{cluster}$), average HCH angle ($\theta_{av}^{\mathrm{HCH}}$), distance of the closest H to the closest TM to the molecule ($d^{\mathrm{H}_c-\mathrm{TM}_c}$), distance of the farthest H to the closest TM to the molecule ($d^{\mathrm{H}_f-\mathrm{TM}_c}$), distance of the C to the closest TM to the molecule ($d^{\mathrm{C}-\mathrm{TM}_c}$), dihedral angle ($\theta_{died}^{\mathrm{H}_f-\mathrm{C}-\mathrm{H}_c-\mathrm{TM}_c}$), coordination of the closest TM to the molecule (CN^{TM_c}), average bond length of the closest TM to the molecule ($\mathrm{d}_{av}^{TM_c}$), orientation angle ($\theta_{\mathrm{H}_c-\mathrm{C}-\mathrm{TM}_c}$), coordination of the C (CN^{C}), dissociation energy (D_{ab}), and adsorption coefficient (Q).

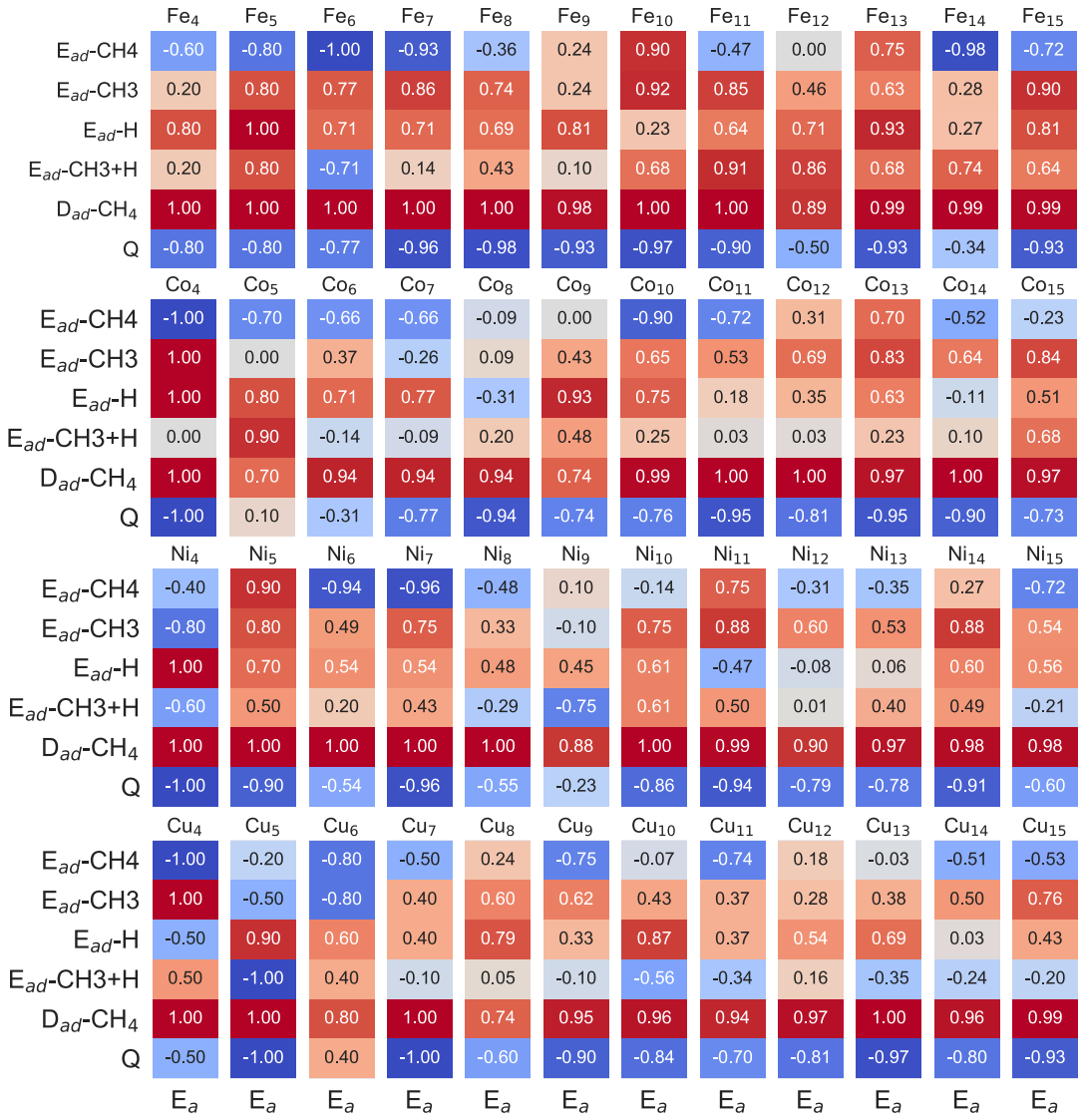

Figure S26: Spearman rank correlation analysis of the activation energy (E_a) for the $CH_4 \rightarrow CH_3$+H reaction.

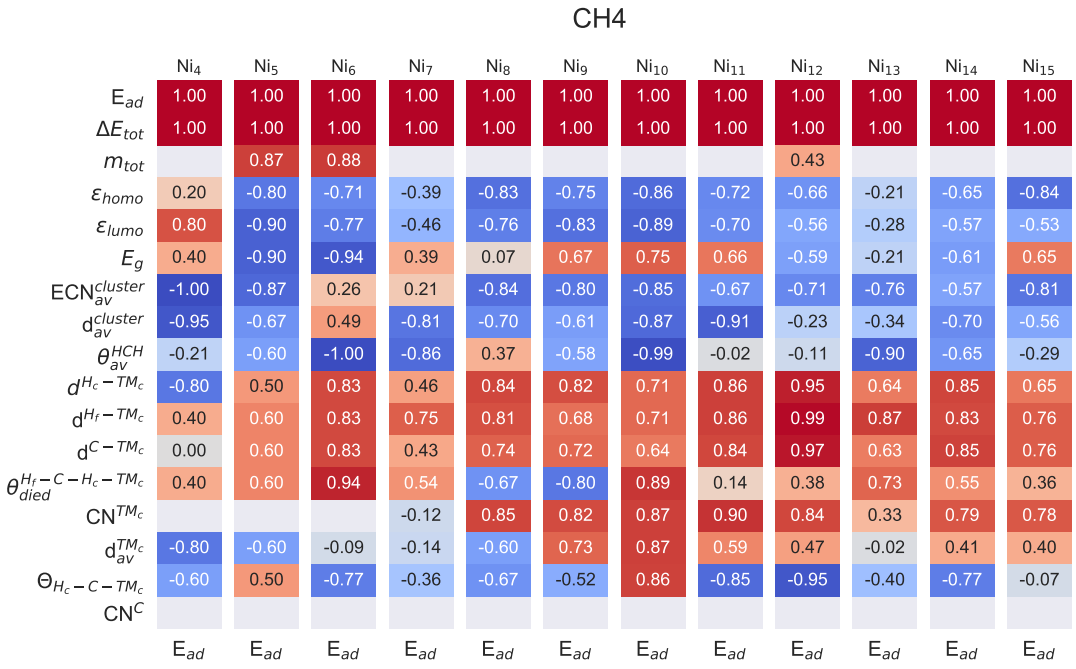

Figure S27: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_4 on the Ni_n clusters, where $n = 4 - 15$.

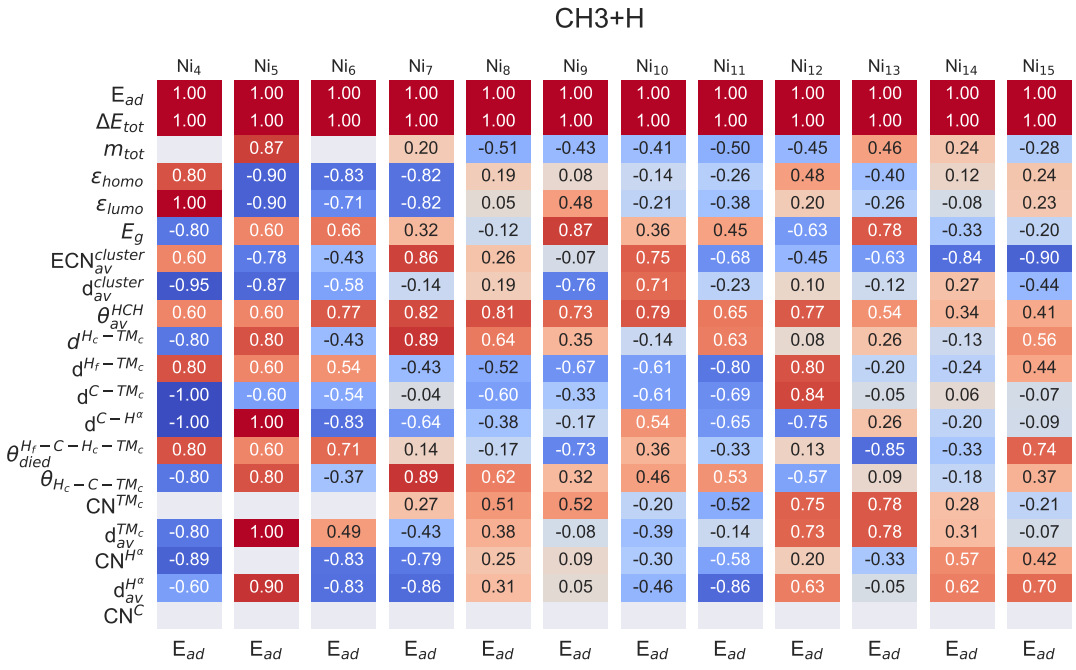

Figure S28: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3+H on the Ni_n clusters, where $n = 4 - 15$.

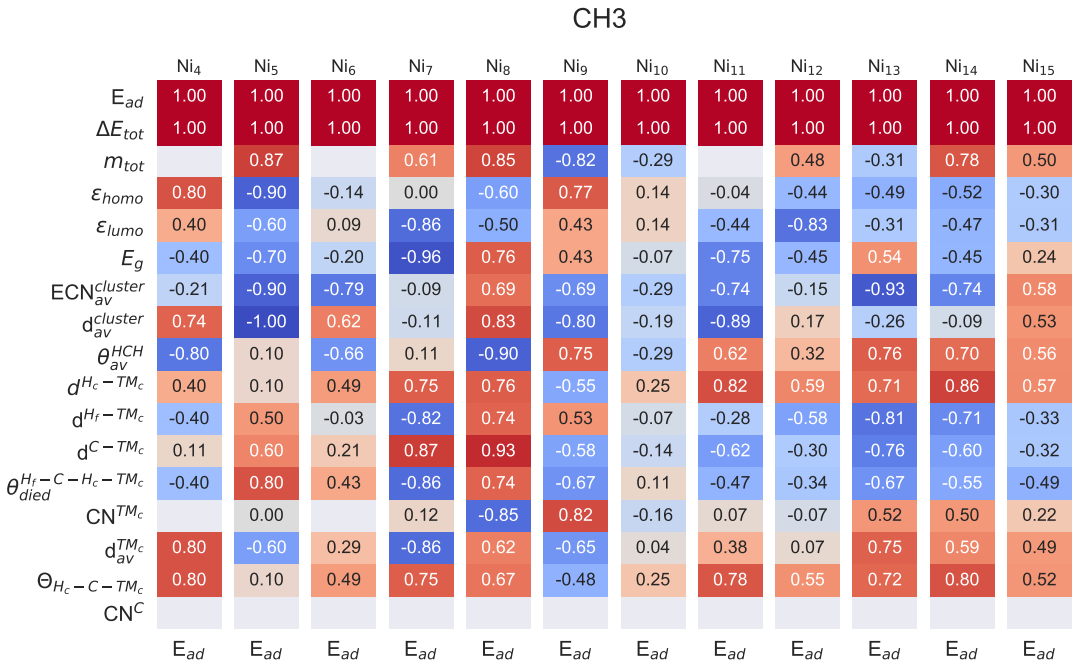

Figure S29: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3 on the Ni_n clusters, where $n = 4 - 15$.

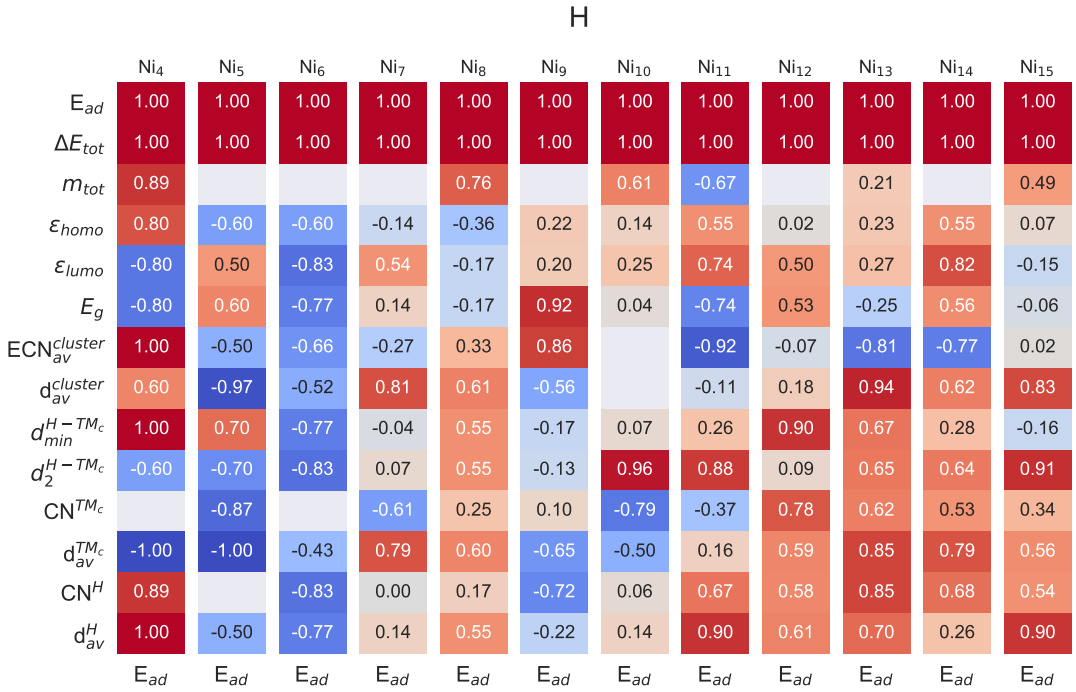

Figure S30: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for H on the Ni_n clusters, where $n = 4 - 15$.

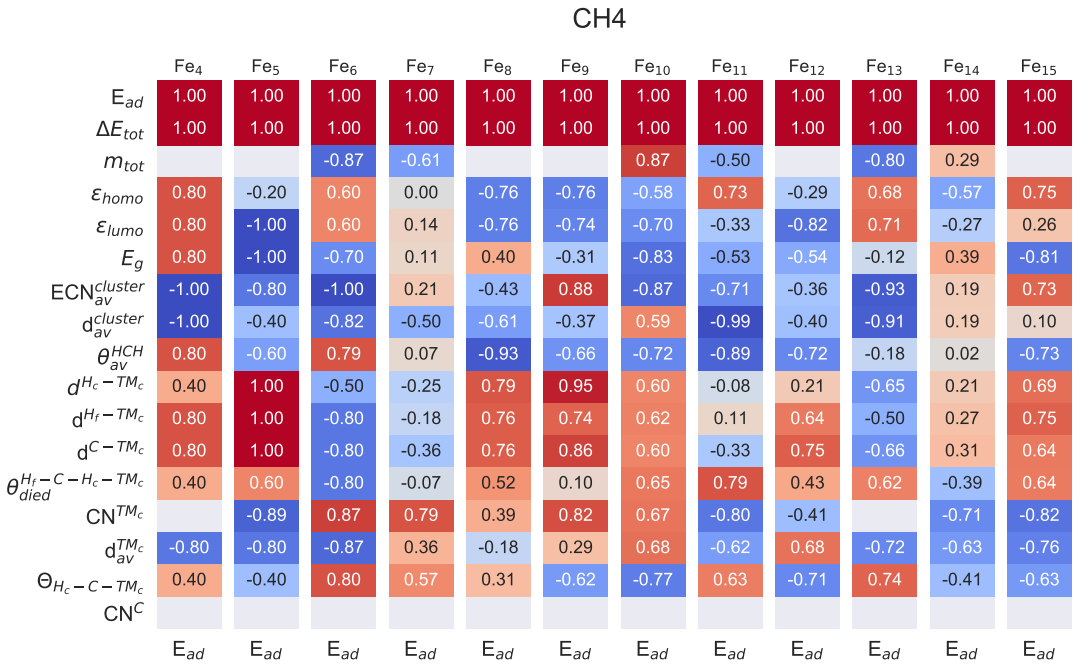

Figure S31: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_4 on the Fe_n clusters, where $n = 4 - 15$.

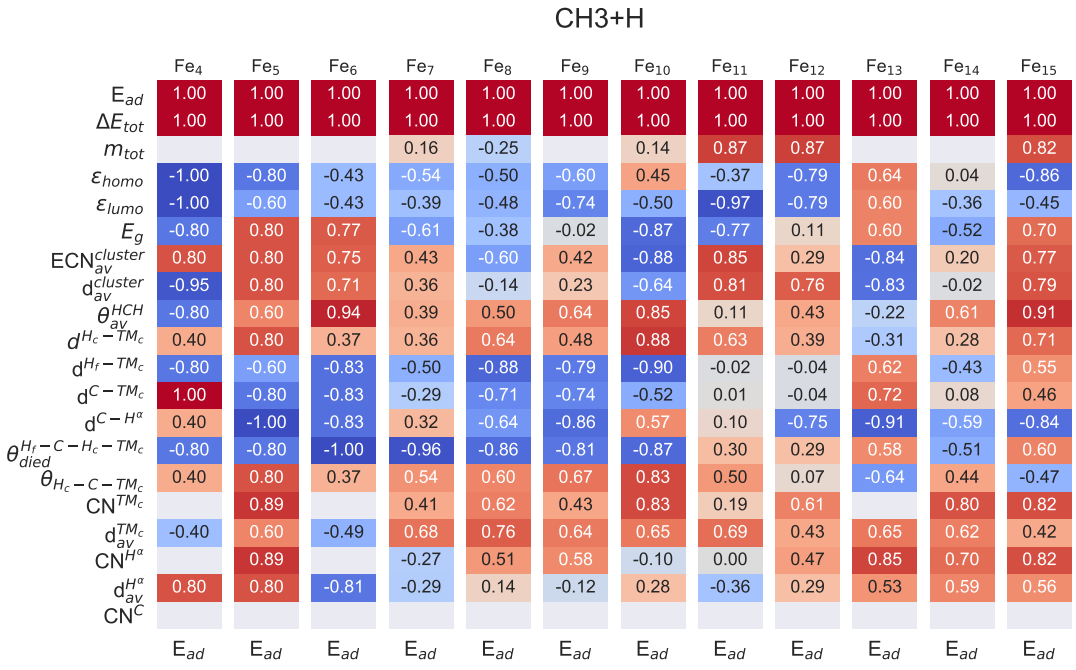

Figure S32: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3+H on the Fe_n clusters, where $n = 4 - 15$.

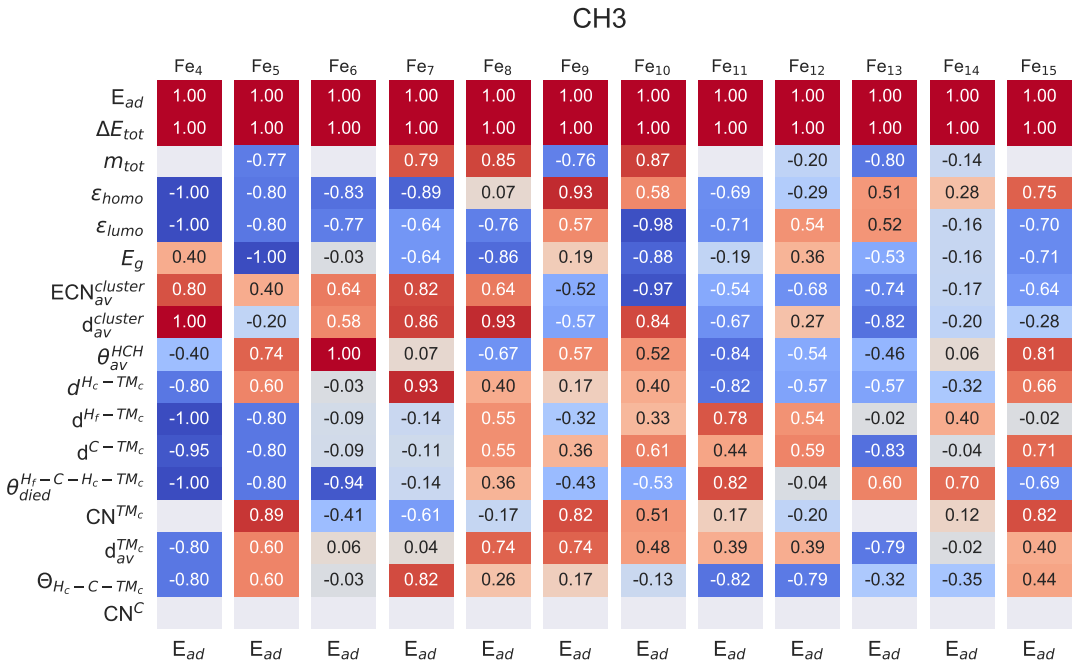

Figure S33: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3 on the Fe_n clusters, where $n = 4 - 15$.

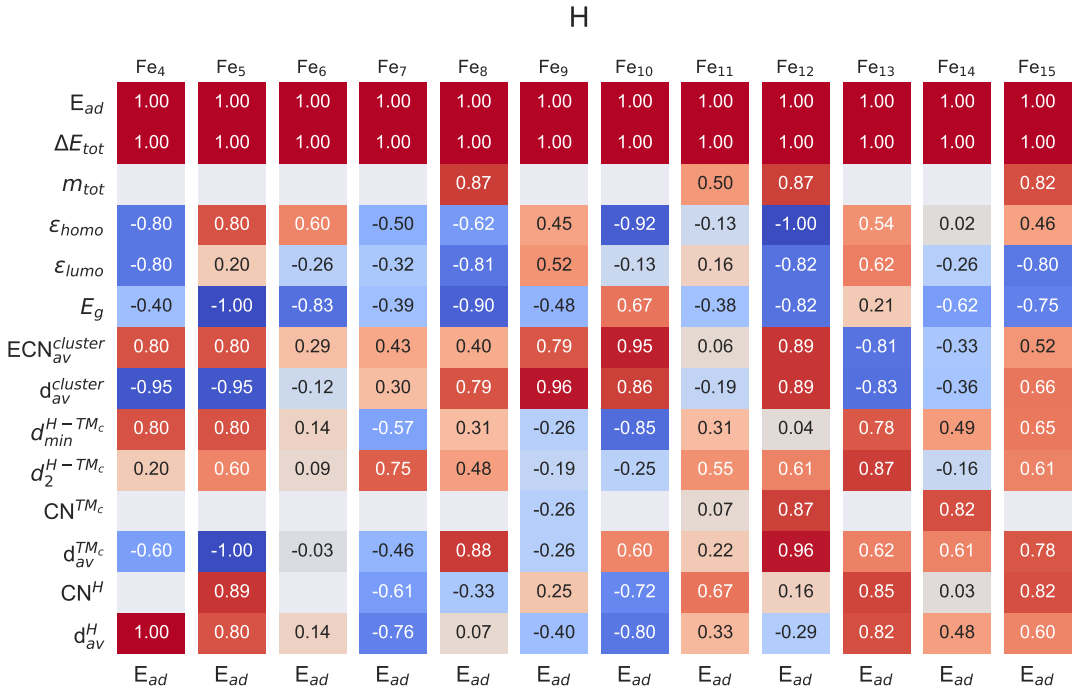

Figure S34: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for H on the Fe_n clusters, where $n = 4 - 15$.

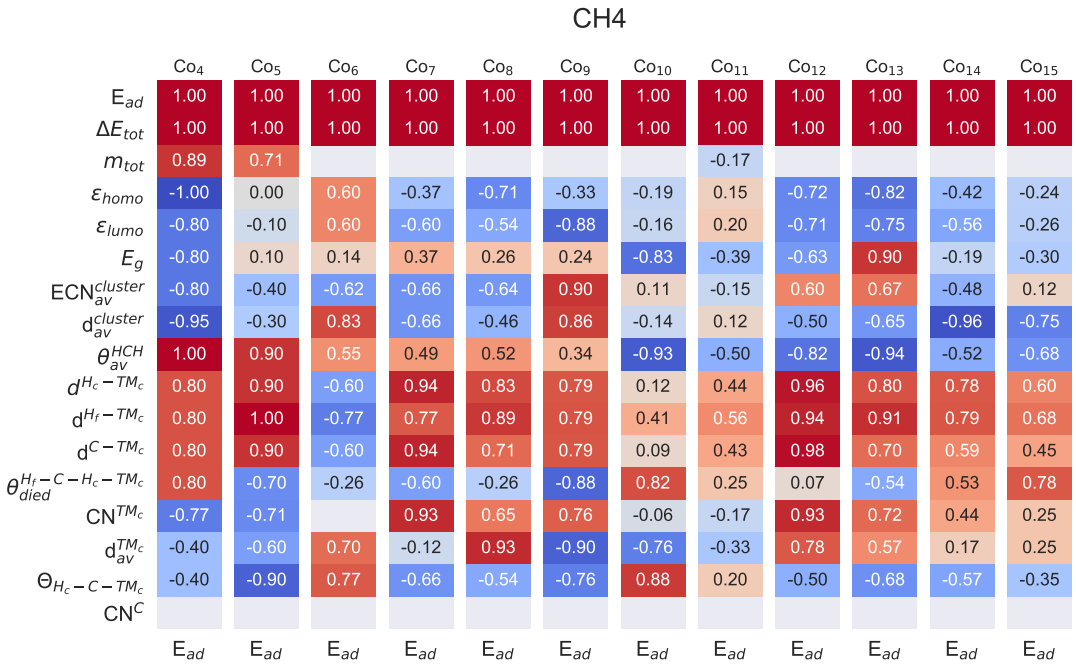

Figure S35: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_4 on the Co_n clusters, where $n = 4 - 15$.

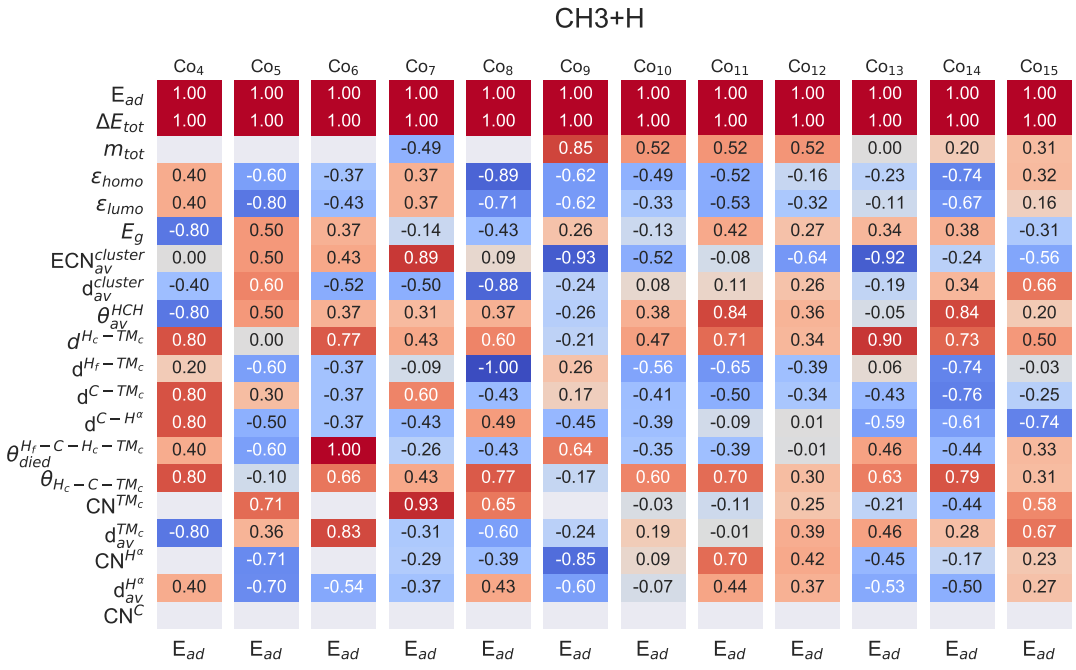

Figure S36: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3+H on the Co_n clusters, where $n = 4 - 15$.

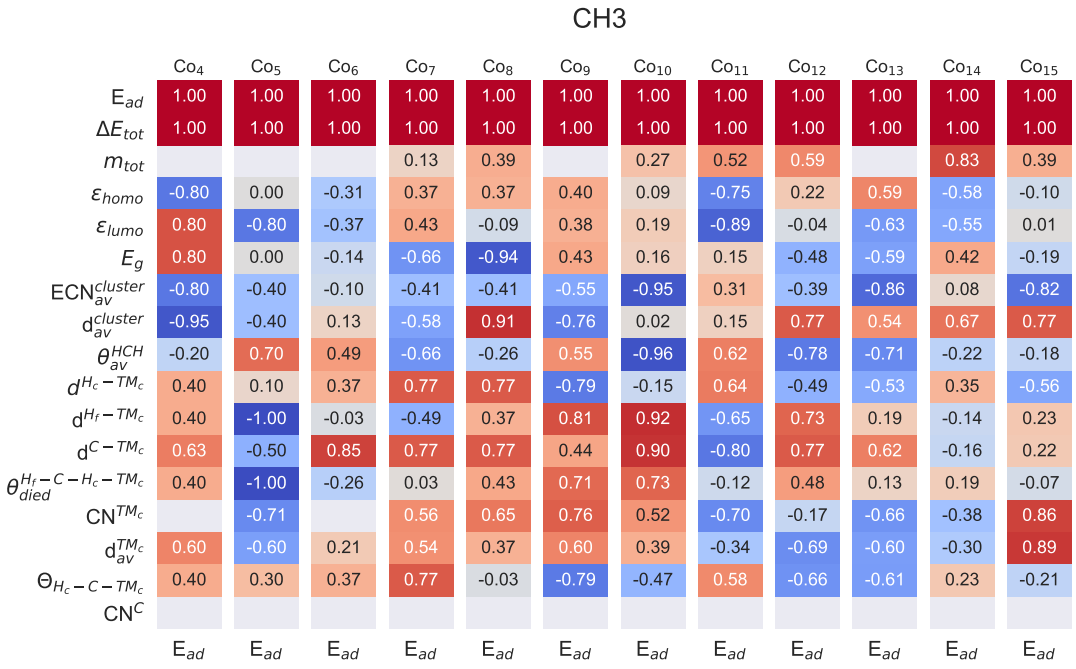

Figure S37: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3 on the Co_n clusters, where $n = 4 - 15$.

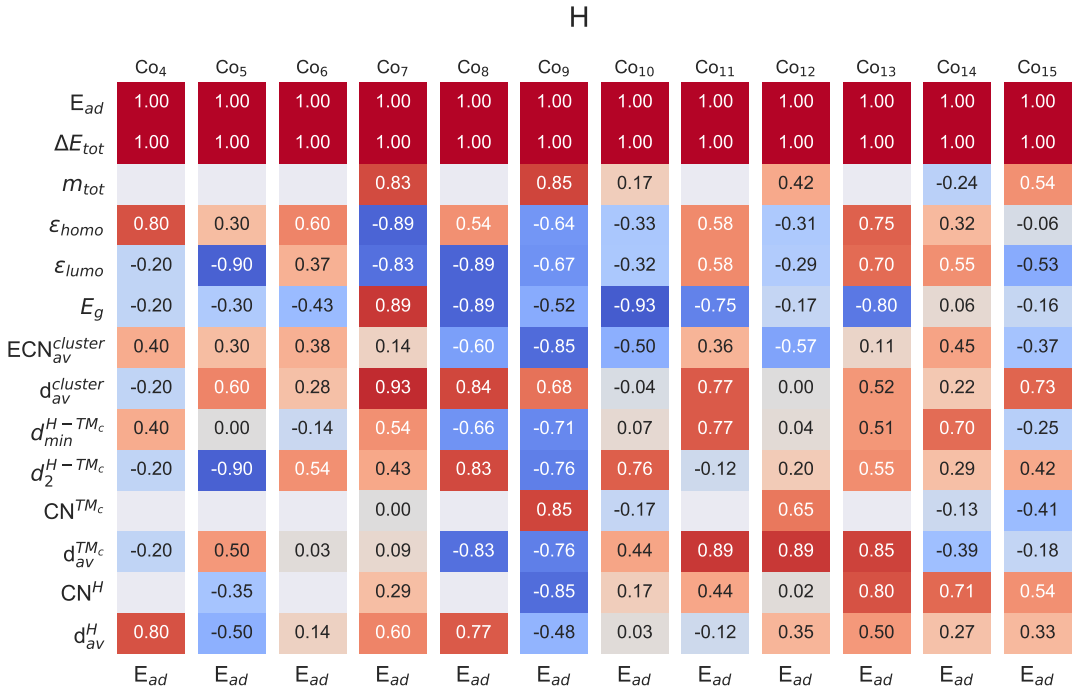

Figure S38: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for H on the Co_n clusters, where $n = 4 - 15$.

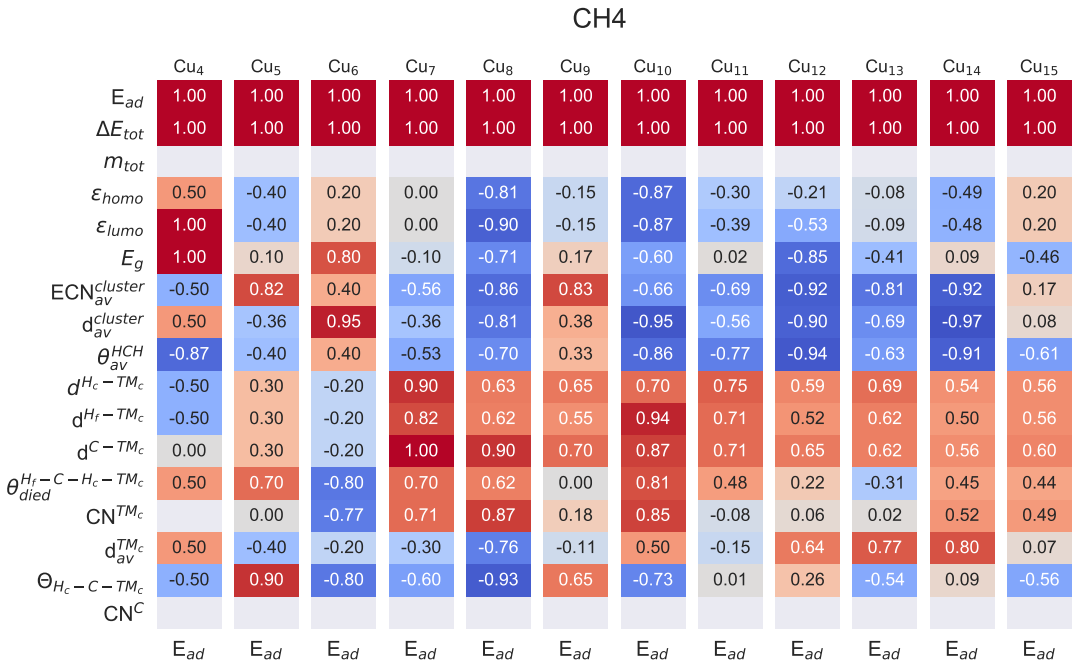

Figure S39: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_4 on the Cu_n clusters, where $n = 4 - 15$.

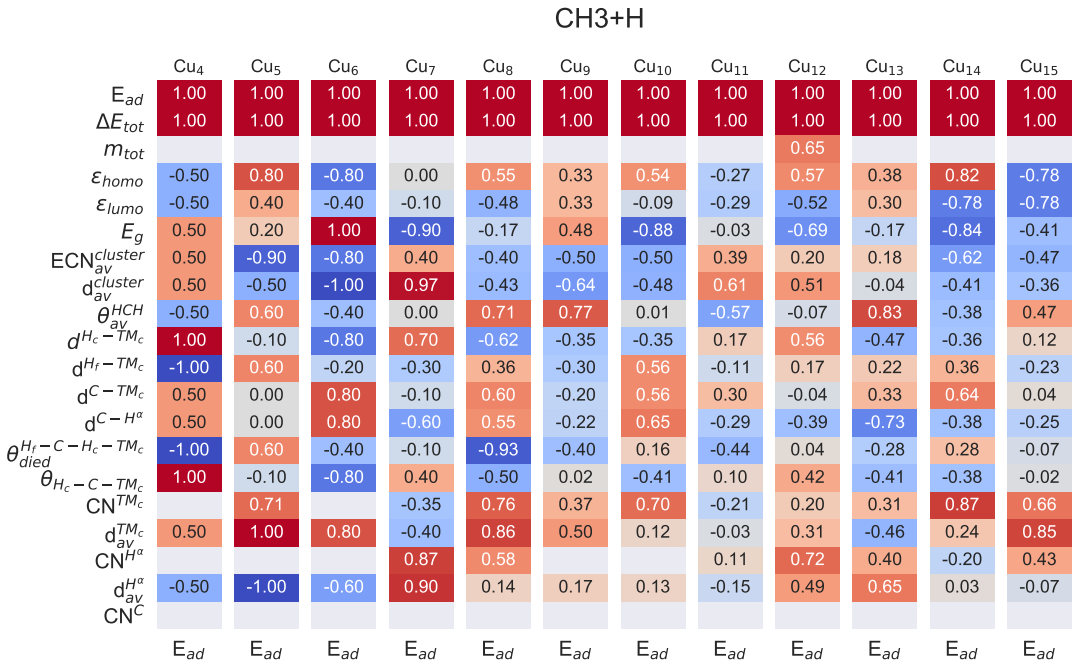

Figure S40: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3+H on the Cu_n clusters, where $n = 4 - 15$.

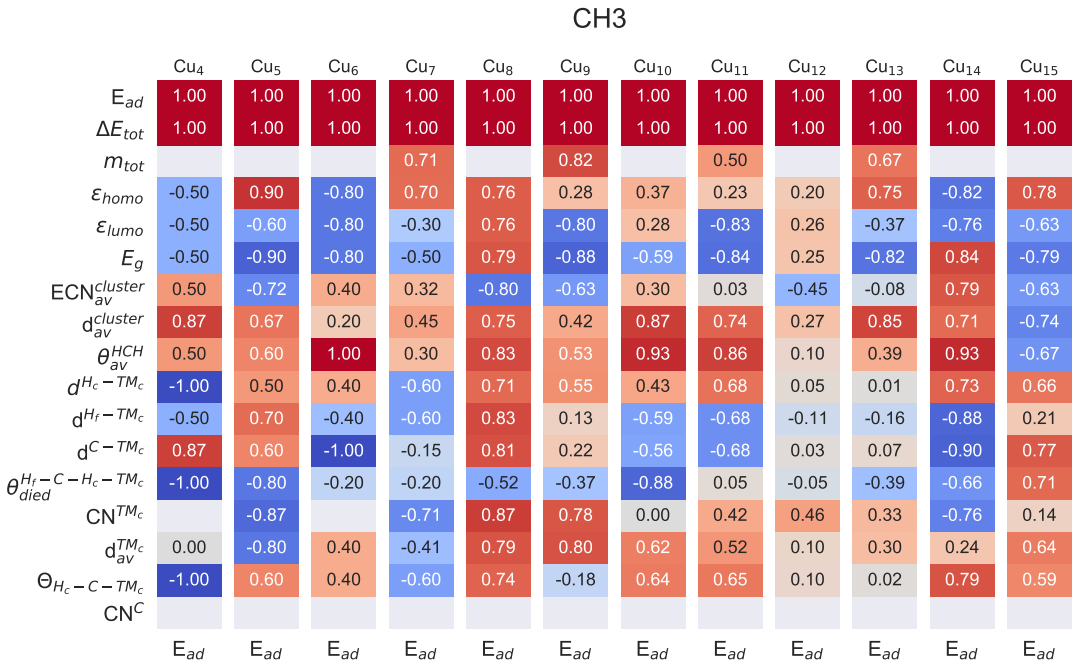

Figure S41: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for CH_3 on the Cu_n clusters, where $n = 4 - 15$.

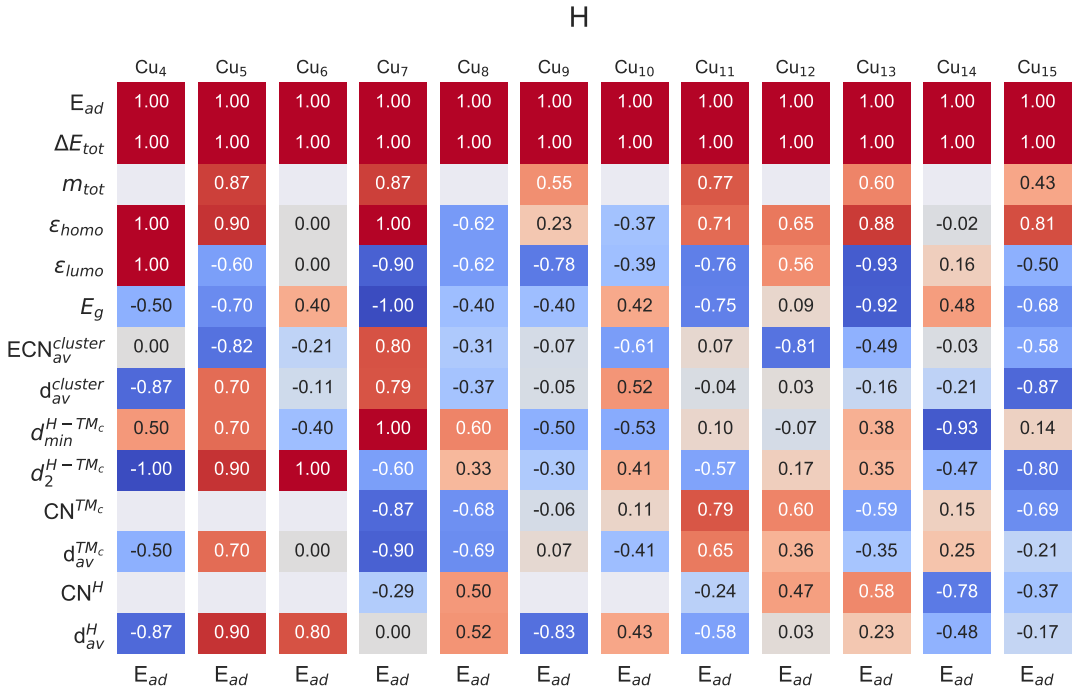

Figure S42: Spearman rank correlation analysis of the adsorption energy (E_{ad}) for H on the Cu_n clusters, where $n = 4 - 15$.

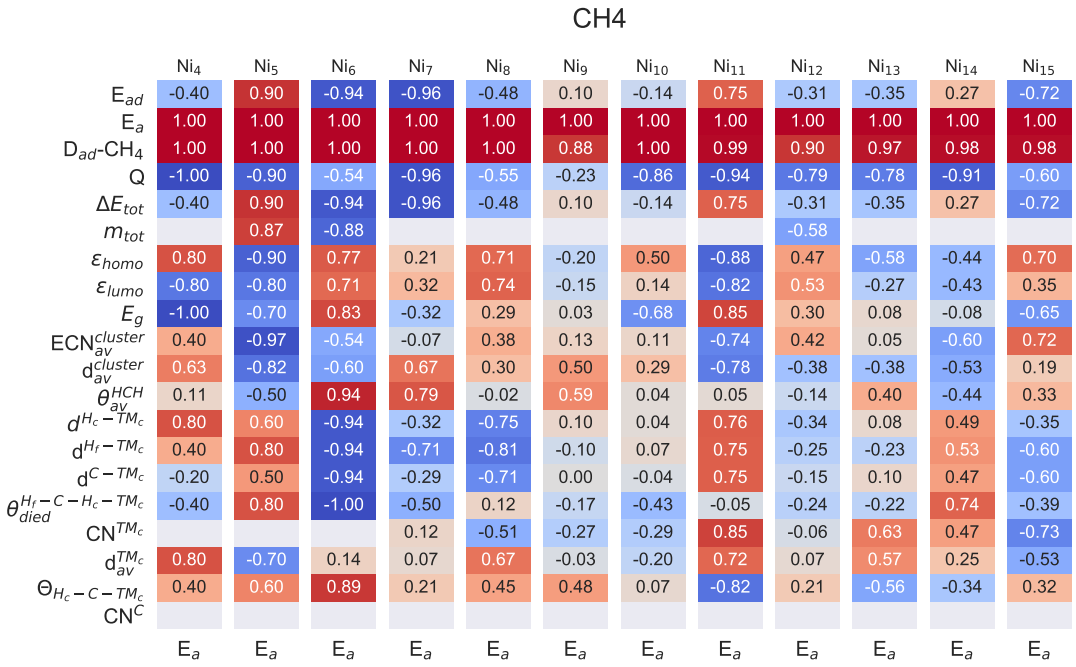

Figure S43: Spearman rank correlation analysis of the activation energy (E_a) for CH_4 on the Ni_n clusters, where $n = 4 - 15$.

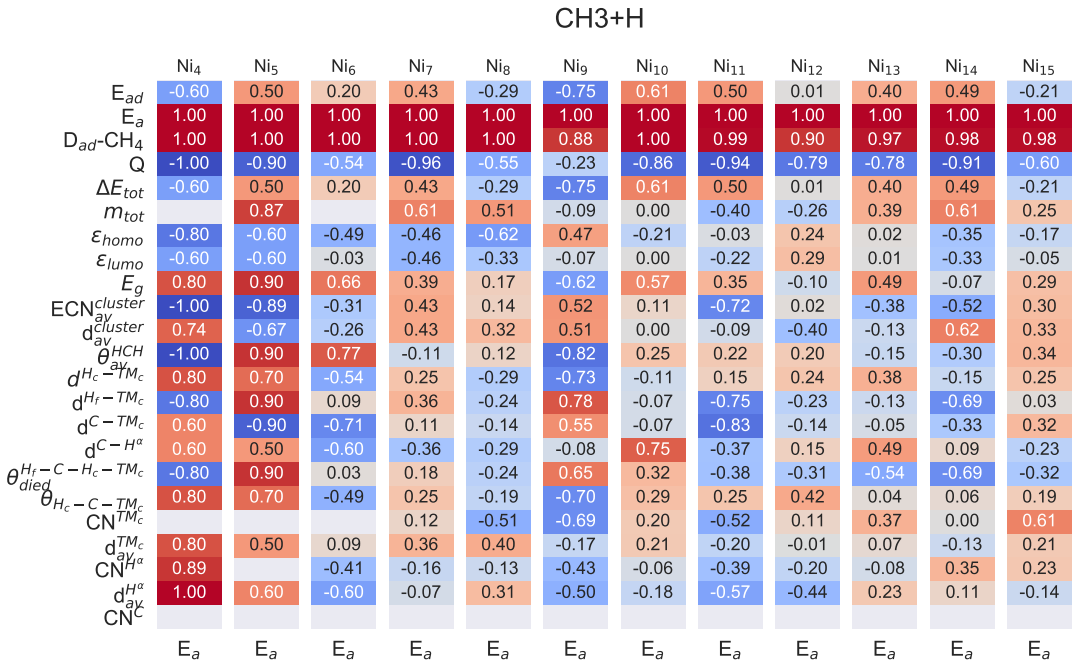

Figure S44: Spearman rank correlation analysis of the activation energy (E_a) for CH_3+H on the Ni_n clusters, where $n = 4 - 15$.

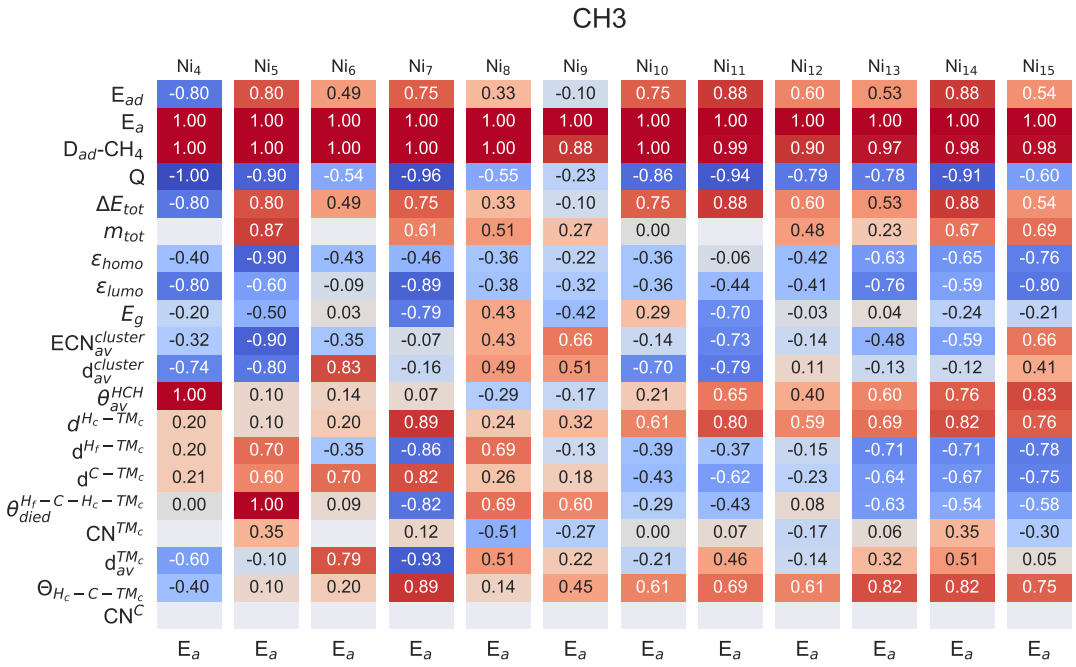

Figure S45: Spearman rank correlation analysis of the activation energy (E_a) for CH_3 on the Ni_n clusters, where $n = 4 - 15$.

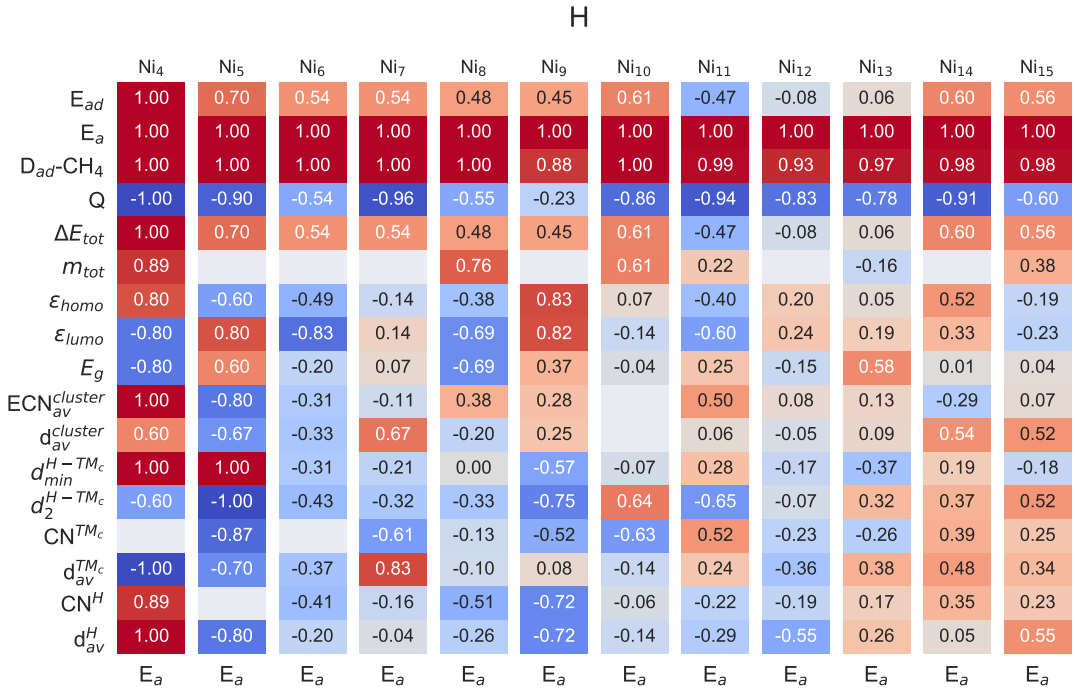

Figure S46: Spearman rank correlation analysis of the activation energy (E_a) for H on the Ni_n clusters, where $n = 4 - 15$.

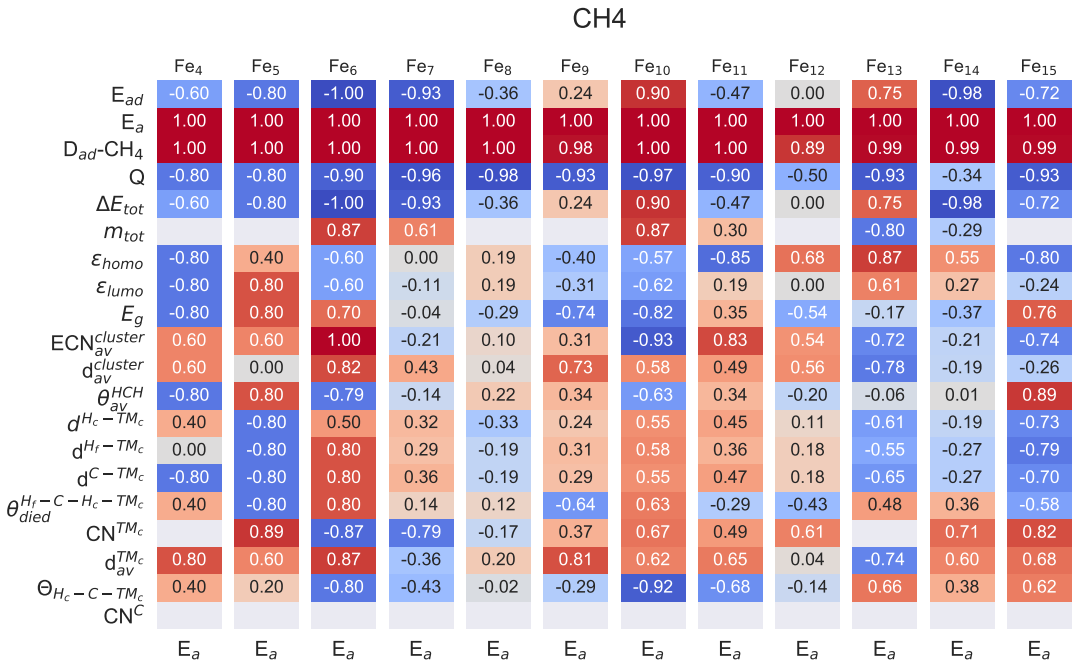

Figure S47: Spearman rank correlation analysis of the activation energy (E_a) for CH_4 on the Fe_n clusters, where $n = 4 - 15$.

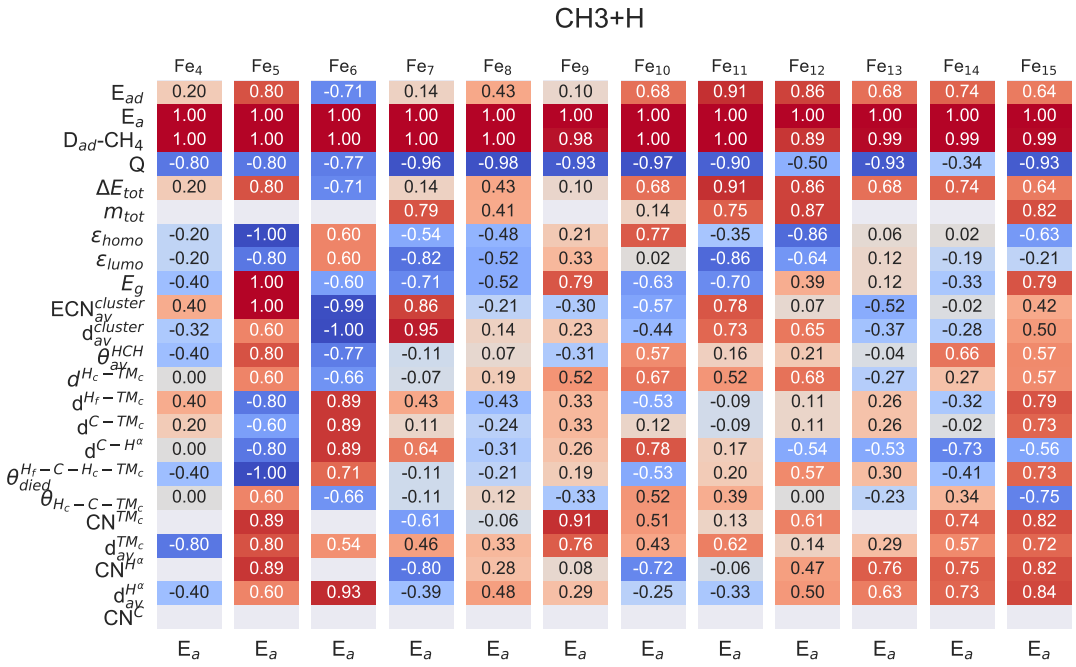

Figure S48: Spearman rank correlation analysis of the activation energy (E_a) for CH_3+H on the Fe_n clusters, where $n = 4 - 15$.

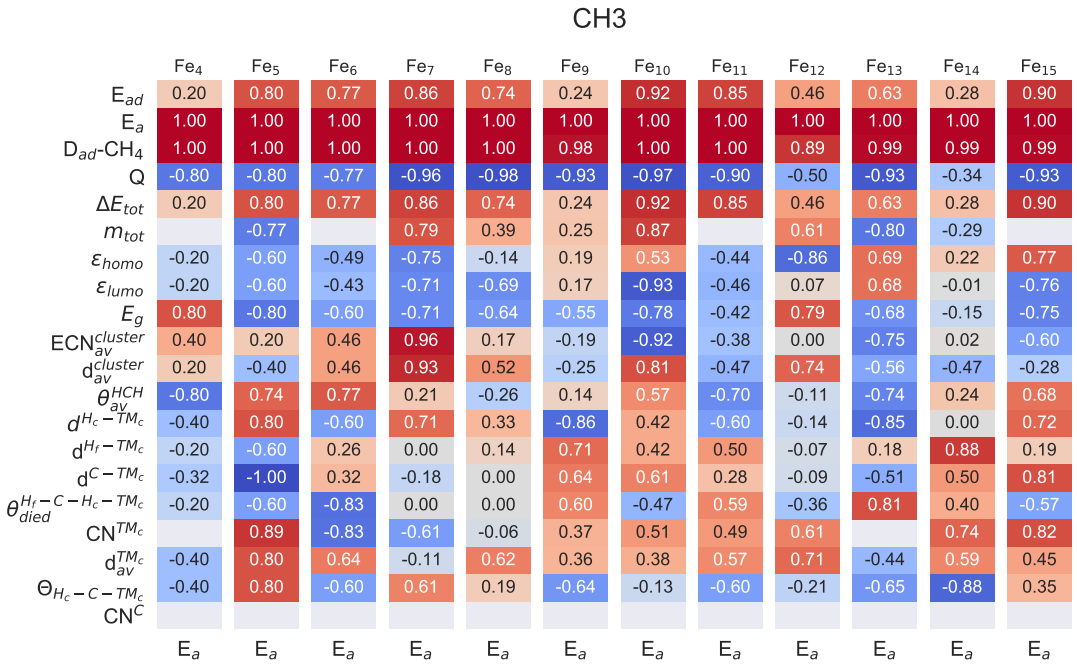

Figure S49: Spearman rank correlation analysis of the activation energy (E_a) for CH_3 on the Fe_n clusters, where $n = 4 - 15$.

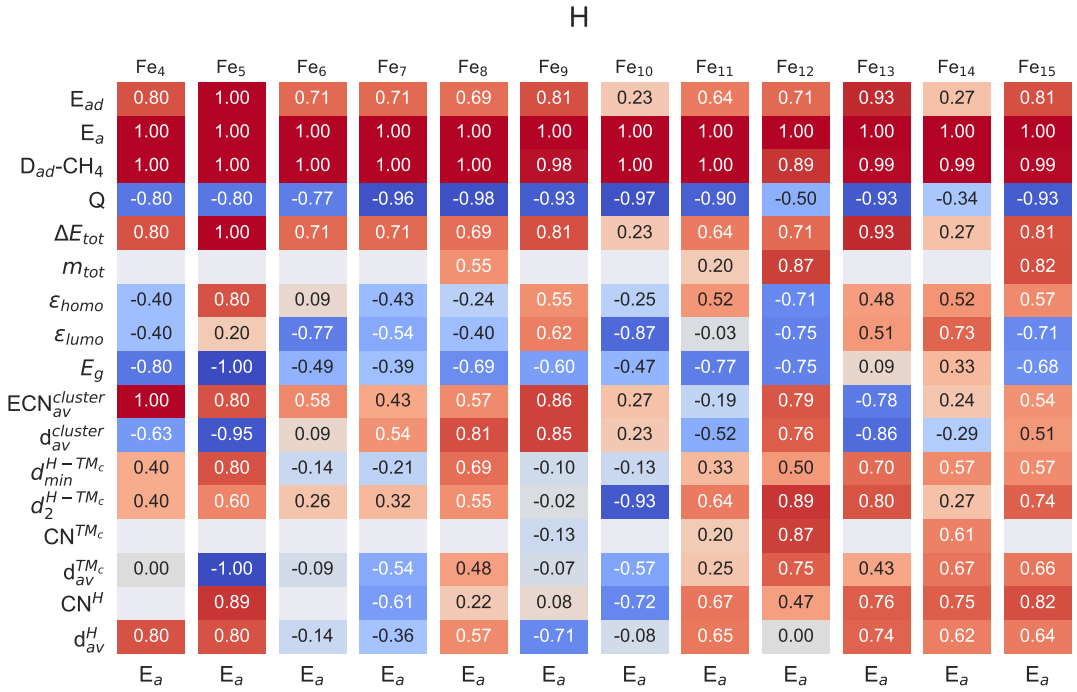

Figure S50: Spearman rank correlation analysis of the activation energy (E_a) for H on the Fe_n clusters, where $n = 4 - 15$.

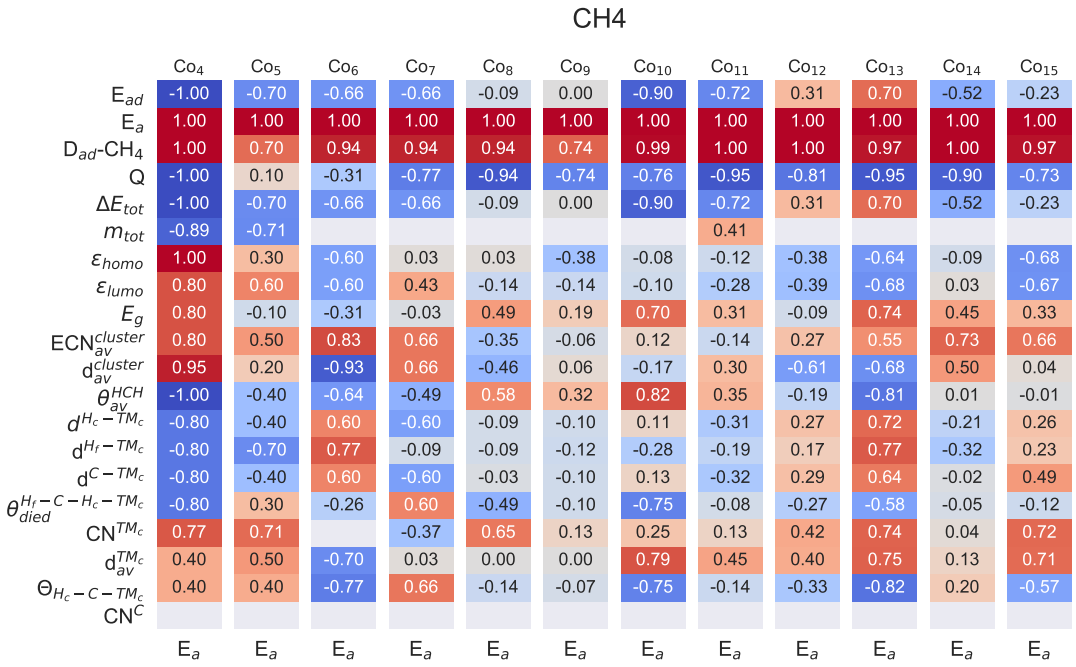

Figure S51: Spearman rank correlation analysis of the activation energy (E_a) for CH_4 on the Co_n clusters, where $n = 4 - 15$.

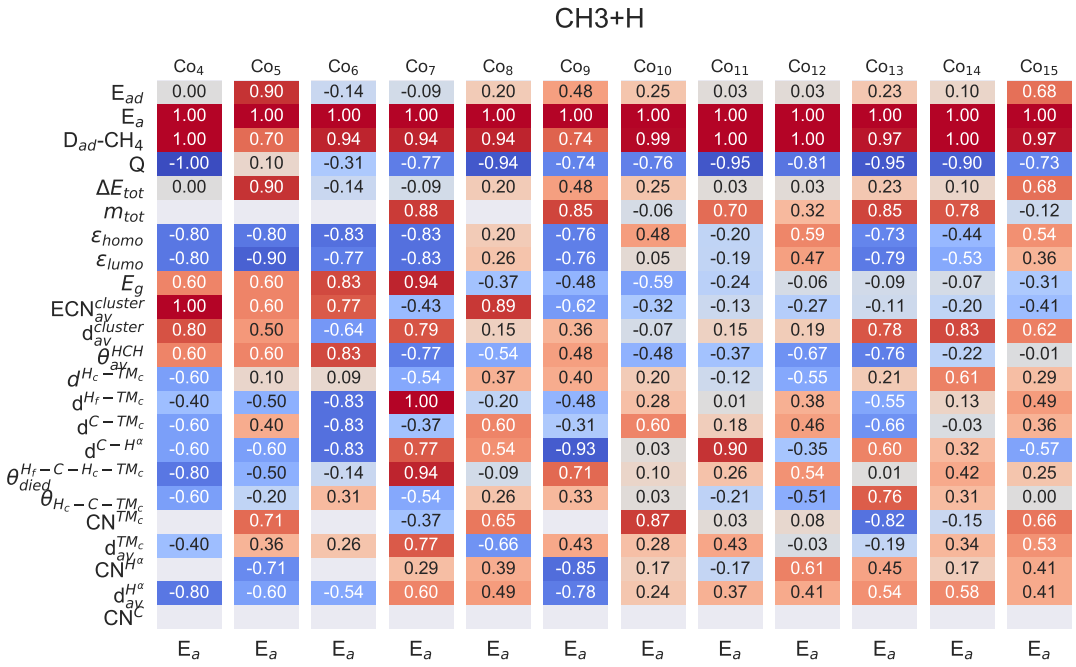

Figure S52: Spearman rank correlation analysis of the activation energy (E_a) for CH_3+H on the Co_n clusters, where $n = 4 - 15$.

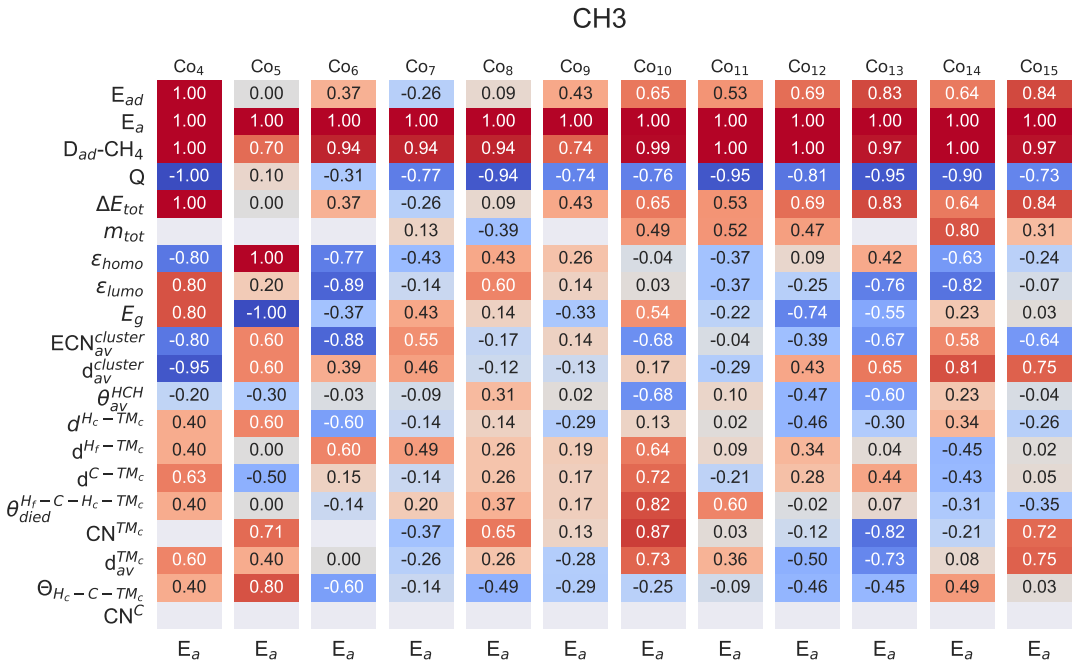

Figure S53: Spearman rank correlation analysis of the activation energy (E_a) for CH_3 on the Co_n clusters, where $n = 4 - 15$.

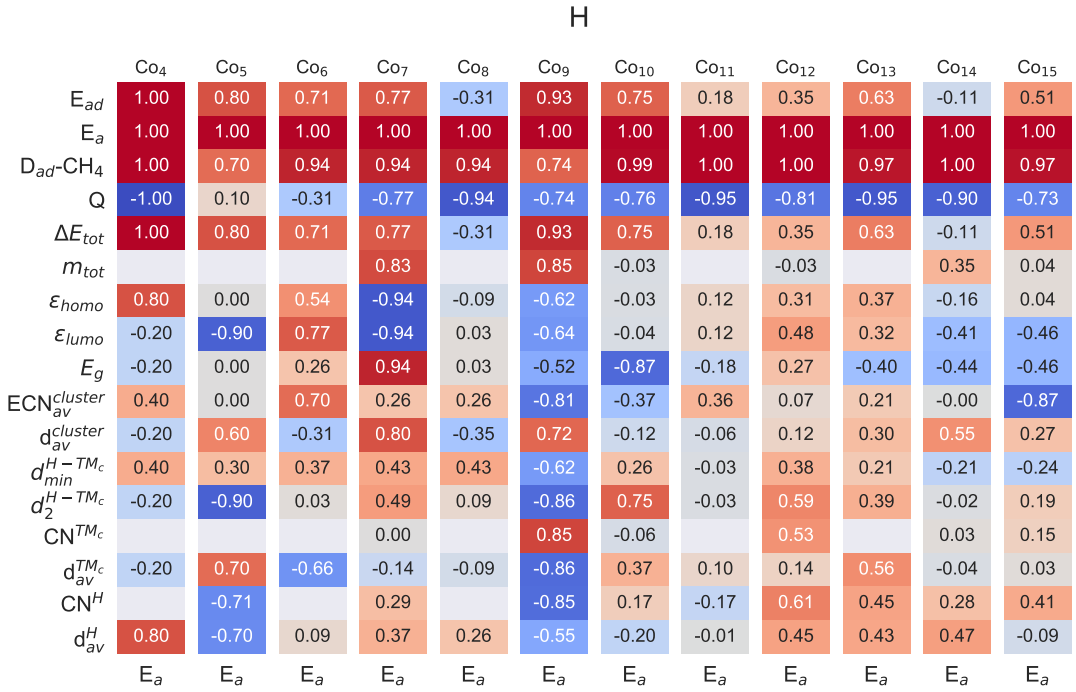

Figure S54: Spearman rank correlation analysis of the activation energy (E_a) for H on the Co_n clusters, where $n = 4 - 15$.

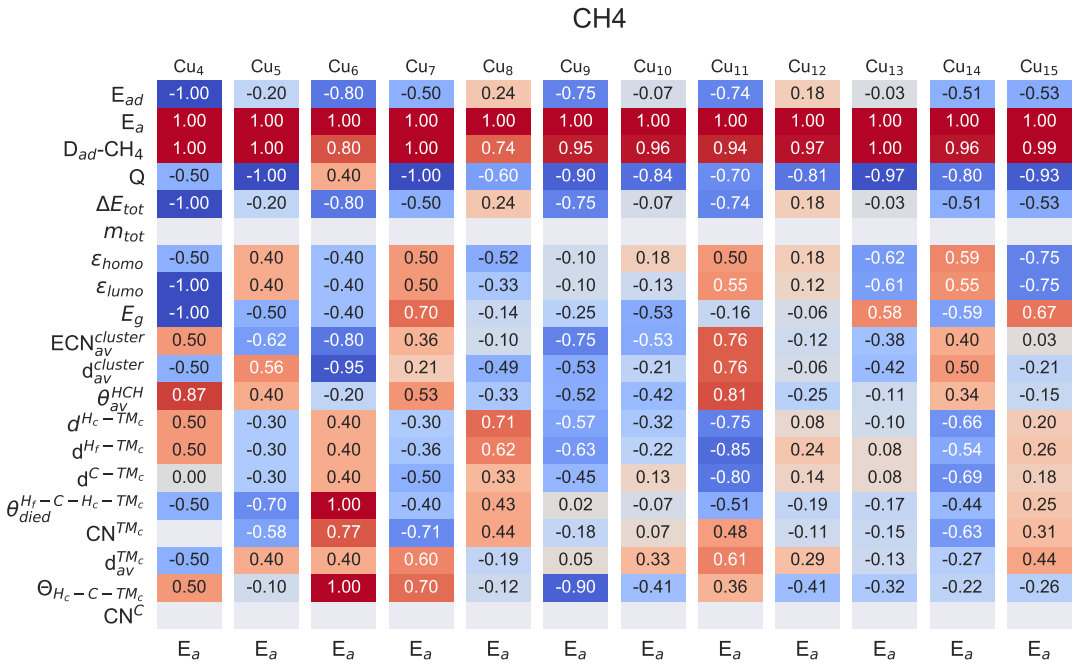

Figure S55: Spearman rank correlation analysis of the activation energy (E_a) for CH_4 on the Cu_n clusters, where $n = 4 - 15$.

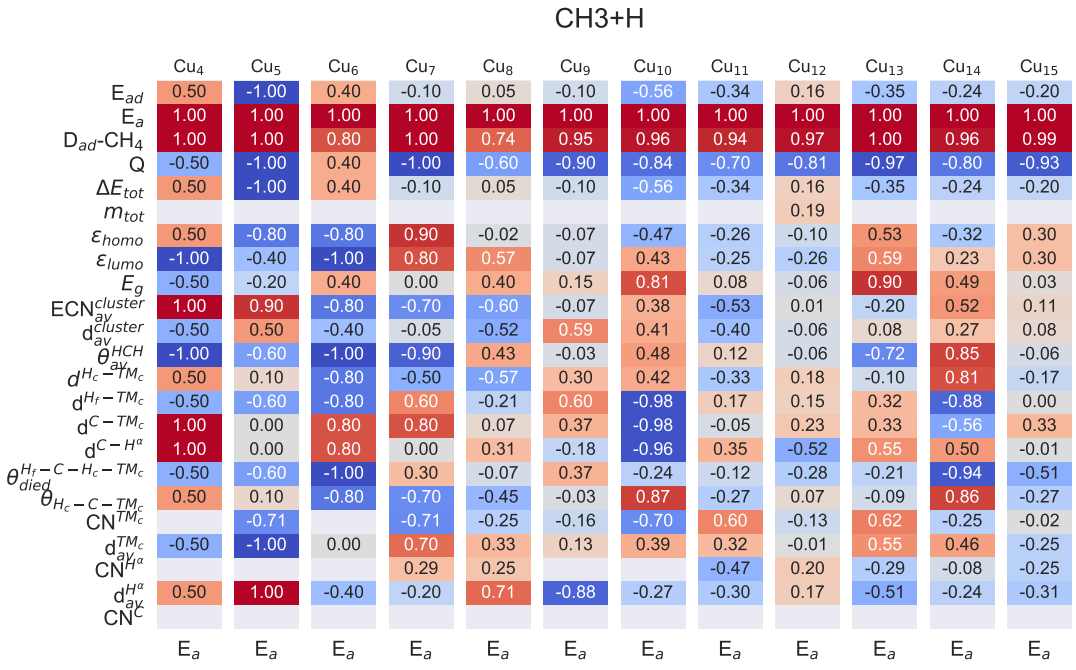

Figure S56: Spearman rank correlation analysis of the activation energy (E_a) for CH_3+H on the Cu_n clusters, where $n = 4 - 15$.

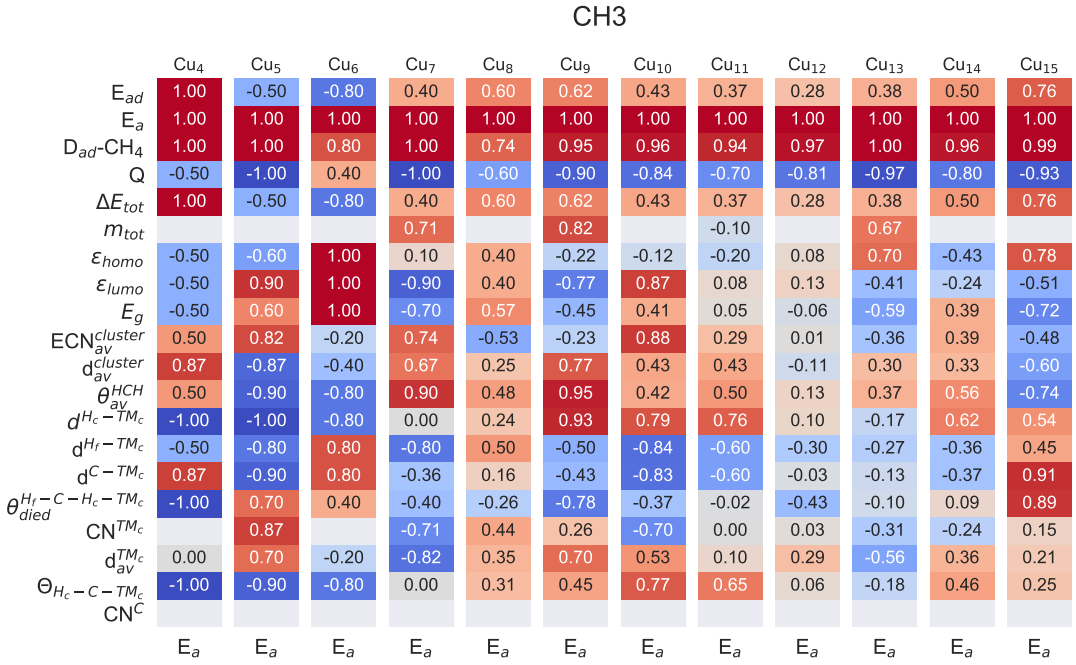

Figure S57: Spearman rank correlation analysis of the activation energy (E_a) for CH_3 on the Cu_n clusters, where $n = 4 - 15$.

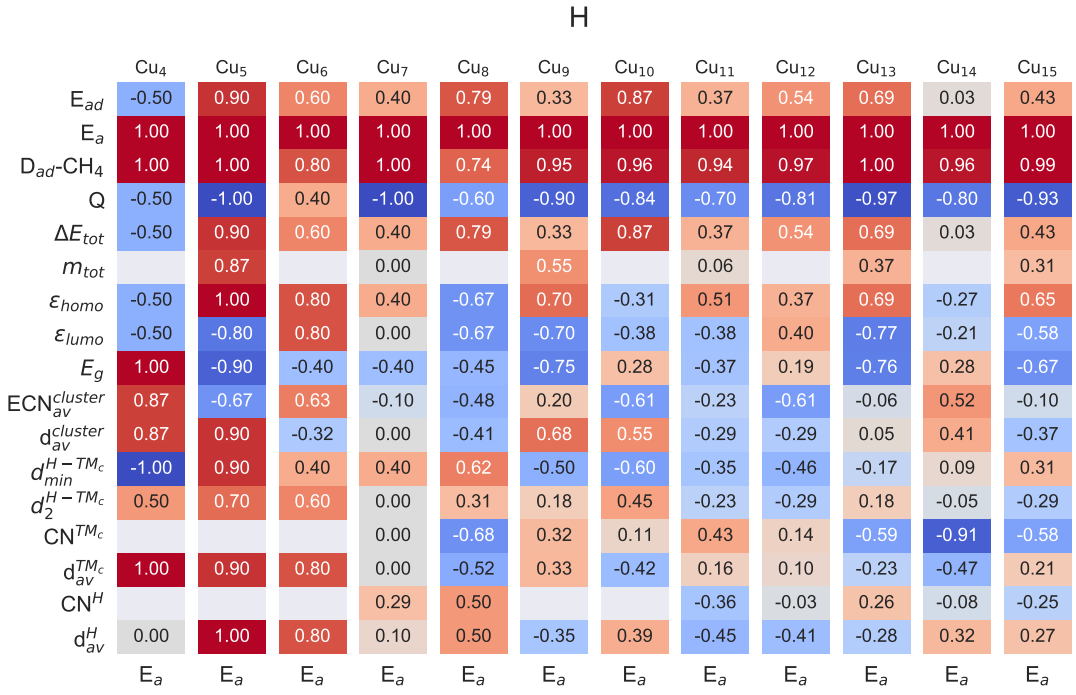

Figure S58: Spearman rank correlation analysis of the activation energy (E_a) for H on the Cu_n clusters, where $n = 4 - 15$.

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