SUPPLEMENTARY MATERIAL

Computational study on the adsorption of arsenic pollutants on graphene-based single-atom iron adsorbents

Kai Ma[a], Di Zheng[a], Weijie Yang[a], Chongchong Wu[b]*, Shuai Dong[a], Zhengyang Gao[a], Xiaojun Zhao[c]

[a] Department of Power Engineering, School of Energy, Power and Mechanical Engineering, North China Electric Power University, Baoding 071003, China

[b] CNOOC Research Institute of Refining and Petrochemicals, Beijing, 102200, PR China

[c] Department of Electrical Engineering, North China Electric Power University, Baoding 071003, China

* Chongchong Wu (wuchch6@cnooc.com.cn)

Table S1 The bond length of C-Fe or N-Fe (d, *Å*), adsorption height of Fe atom (h, *Å*), charge of Fe atom (Q,*e*), magnetic moment (*M*, *μB*) and binding energy (E_b , eV) for $Fe_{SA}@V_x$-N_y.

$Fe_{SA}@V_x$-N_y	d (*Å*)	h (*Å*)	Q (*e*)	*M* (*μB*)	E_b (eV)
$Fe_{SA}@V_s$-N_0	1.76	1.35	+0.69	0.00	-7.14
$Fe_{SA}@V_s$-N_3	1.78	1.23	+0.90	3.13	-4.41
$Fe_{SA}@V_d$-N_0	1.86	0.66	+0.90	2.22	-6.12
$Fe_{SA}@V_d$-N_4	1.89	0.05	+1.08	2.00	-7.14

Table S2 The bond length, electron transfer (Δq), magnetic moment (*M*, μB) and adsorption energy (E_{ads}) of As_2.

Properties	$Fe_{SA}@V_s$-N_0	$Fe_{SA}@V_s$-N_3	$Fe_{SA}@V_d$-N_0	$Fe_{SA}@V_d$-N_4
As(1)- As(2) (Å)	2.22	2.24	2.25	2.19
As(1)- Fe (Å)	2.43	2.36	2.36	2.49
As(2)- Fe (Å)	2.37	2.38	2.36	2.50
Δq-As2 (e)	+0.03	+0.27	-0.07	-0.06
Δq-Fe (e)	-0.62	-0.88	-0.70	-0.87
Δq-Gra (e)	+0.59	+0.61	+0.77	+0.93
M (μB)	0.00	3.00	1.16	0.00
E_{ads} (eV)	-1.83	-2.49	-1.66	-0.66

Table S3 The bond length, electron transfer (Δq), magnetic moment (M, μB) and adsorption energy (E_{ads}) of As_4.

Properties	$Fe_{SA}@V_s$-N_0	$Fe_{SA}@V_s$-N_3	$Fe_{SA}@V_d$-N_0	$Fe_{SA}@V_d$-N_4
As(1)- As(2) (Å)	2.47	2.48	2.48	2.45
As(1)- As(3) (Å)	2.47	2.99	2.82	2.49
As(1)- As(4) (Å)	2.48	2.48	2.47	2.46
As(2)- As(3) (Å)	2.57	2.48	2.48	2.46
As(2)- As(4) (Å)	2.57	2.44	2.46	2.46
As(3)- As(4) (Å)	2.57	2.48	2.47	2.46
As(1)- Fe (Å)	-	2.40	2.49	-
As(2)- Fe (Å)	2.46	-	-	2.93
As(3)- Fe (Å)	2.46	2.46	2.50	-
As(4)- Fe (Å)	2.46	-	-	-
Δq-As4 (e)	-0.08	+0.20	-0.10	-0.08
Δq-Fe (e)	-0.59	-0.88	-0.72	-1.05
Δq-Gra (e)	+0.67	+0.68	+0.82	+1.13
M (μB)	0.00	3.07	1.45	2.00
E_{ads} (eV)	-1.34	-1.59	-0.69	-0.06

Table S4 The bond length, electron transfer (Δq), magnetic moment (*M*, μB) and adsorption energy (E_{ads}) of AsO.

Properties	$Fe_{SA}@V_s$-N_0	$Fe_{SA}@V_s$-N_3	$Fe_{SA}@V_d$-N_0	$Fe_{SA}@V_d$-N_4
As- O (Å)	1.78	1.78	1.76	1.67
As- Fe (Å)	2.35	2.31	2.24	2.30
O- Fe (Å)	1.84	1.85	1.92	3.25
Δq-AsO (e)	+0.27	+0.47	+0.15	-0.01
Δq-Fe (e)	-0.81	-1.08	-0.82	-0.81
Δq-Gra (e)	+0.54	+0.61	+0.67	+0.82
M (μB)	1	2.07	0.00	0.97
E_{ads} (eV)	-2.32	-3.09	-2.10	-1.71

Table S5 The bond length, electron transfer (Δq), magnetic moment (M, μB) and adsorption energy (E_{ads}) of AsH_3.

Properties	Fe/SV-GN	Fe/SV-N3	Fe/DV-GN	Fe/DV-N4
As- H(1) (Å)	1.53	1.54	1.53	1.53
As- H(2) (Å)	1.53	1.54	1.53	1.53
As- H(3) (Å)	1.53	1.54	1.53	1.53
As- Fe (Å)	2.45	2.38	2.45	2.61
Δq-AsH3 (e)	-0.12	-0.05	-0.18	-0.10
Δq-Fe (e)	-0.65	-0.84	-0.91	-1.05
Δq-Gra (e)	+0.77	+0.89	+1.09	+1.1
M (μB)	0.00	3.08	3.64	2.00
E_{ads} (eV)	-0.79	-0.98	-0.56	-0.15

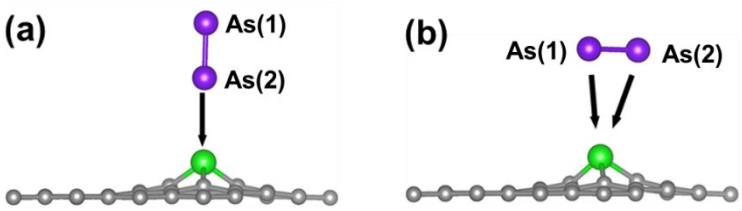

Fig. S1. (a) End-on, (b) side-on adsorption configurations of As_2 on $Fe_{SA}@V_x$-N_y.

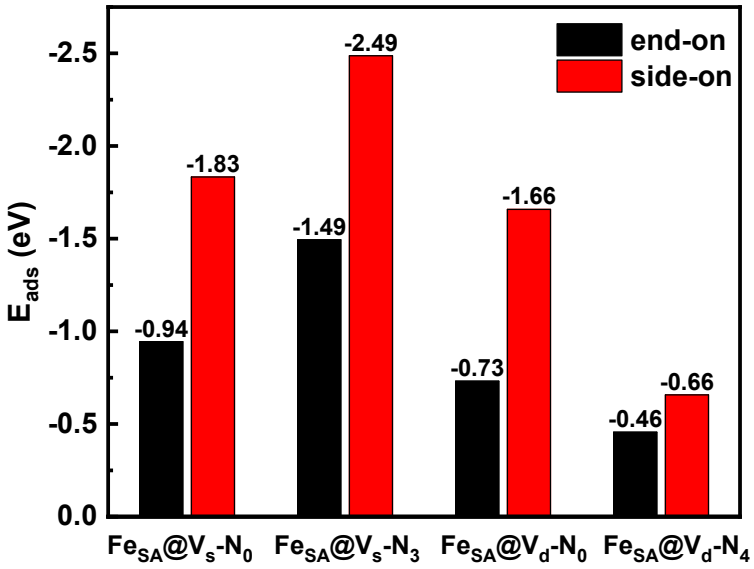

Fig. S2. Adsorption configurations of As_2 and corresponding adsorption energy.

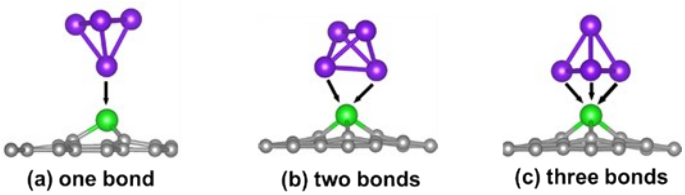

Fig. S3. Three possible adsorption configurations of As_4 on $Fe_{SA}@V_x$-N_y.

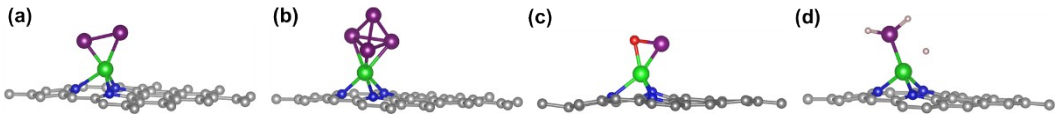

Fig. S4. (a) ~ (d) Final adsorption structures of As_2, As_4, AsO and AsH_3 after AIMD simulation at 1100 K.