Supporting Information

Oxygen Evolution Reaction on the NiFe-LDH/(Ni, Fe)OOH: Theoretical Insights into the effects of electronic structure and spin-state evolution

Qian Lin[1], Guangjun Nan[2], Dawei Fu[1], Liyan Xie[,1]*

1. Institute for Science and Applications of Molecular Ferroelectrics, Key Laboratory of the Ministry of Education for Advanced Catalysis Materials, Zhejiang Normal University, Jinhua, 321004, P. R. China.

2. Department of Physics, Zhejiang Normal University, Jinhua, Zhejiang 321004, P. R. China.

**Corresponding author.*

E-mail: liyanxie@zjnu.edu.cn

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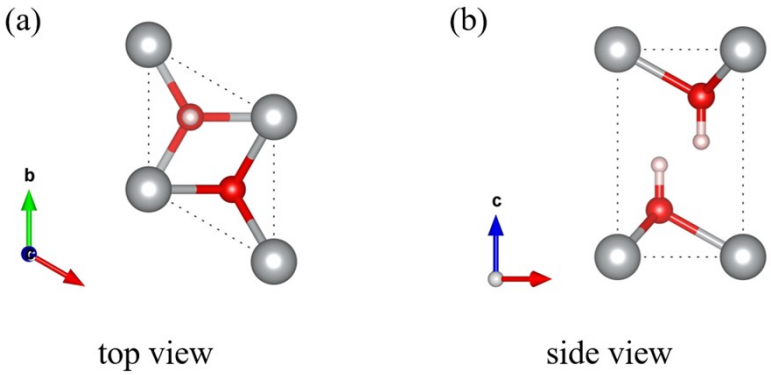
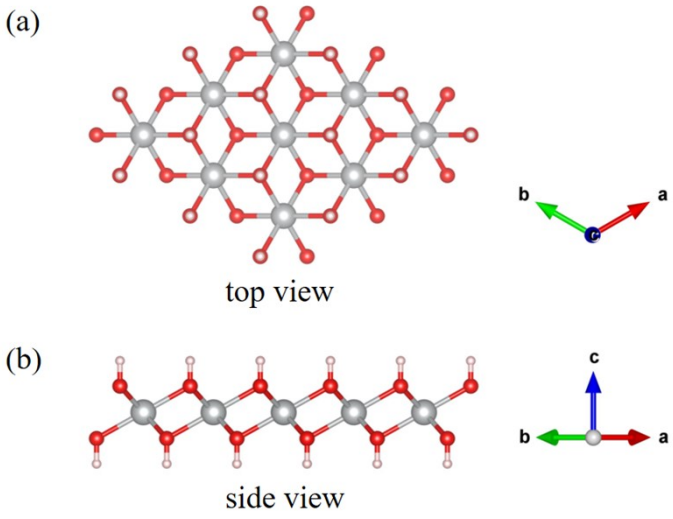


Figure S1. Atomic model structure of β-$Ni(OH)_2$ unit Cell, (a) top view, (b) side view. The pink, red and gray sphere present H, O and Ni, respectively.



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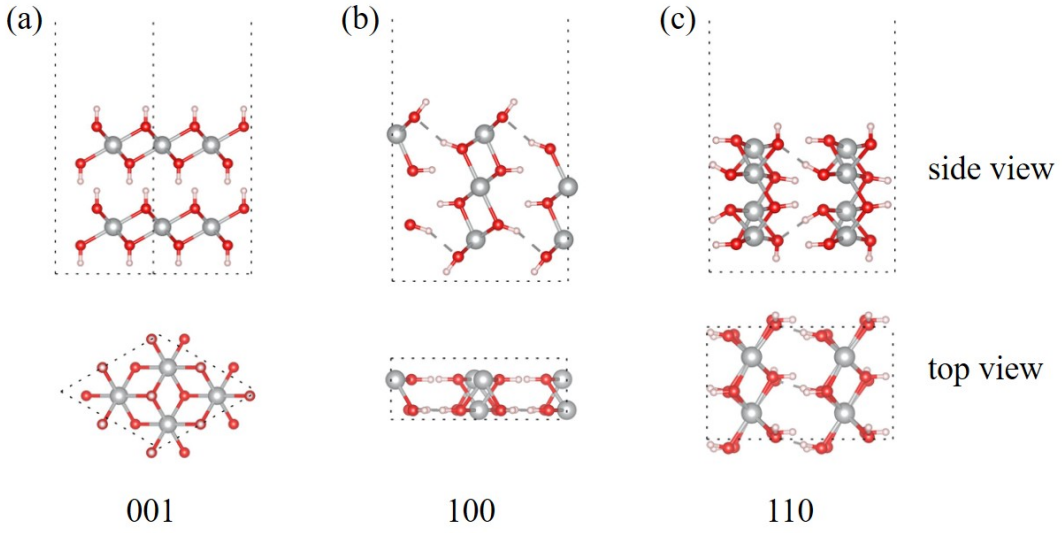

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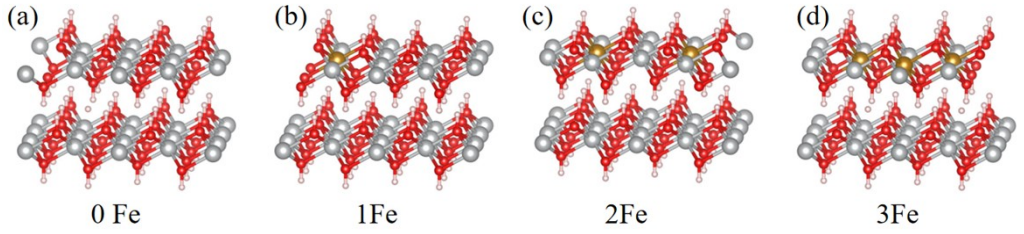

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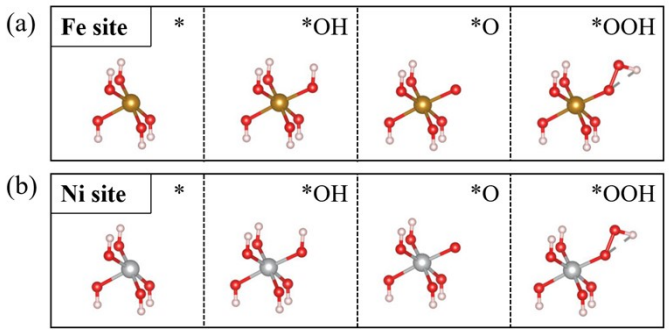

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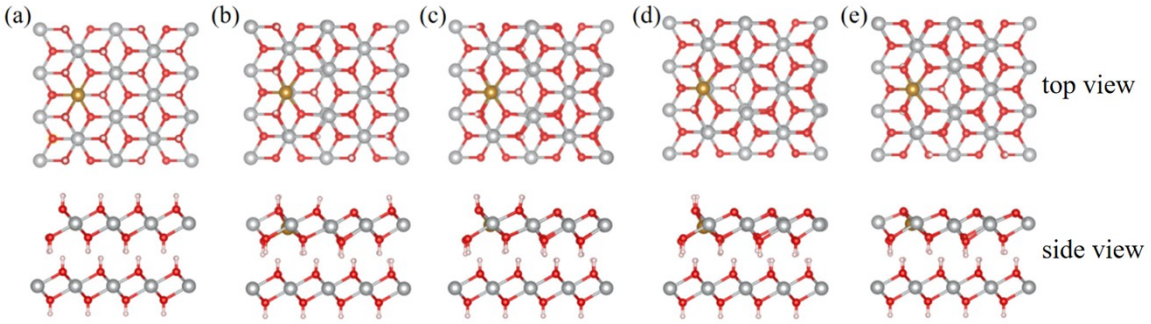

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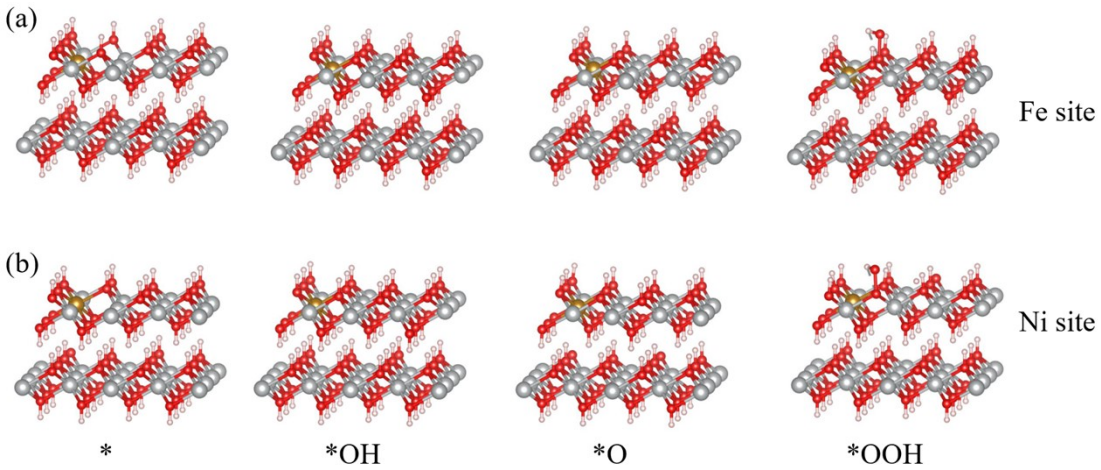

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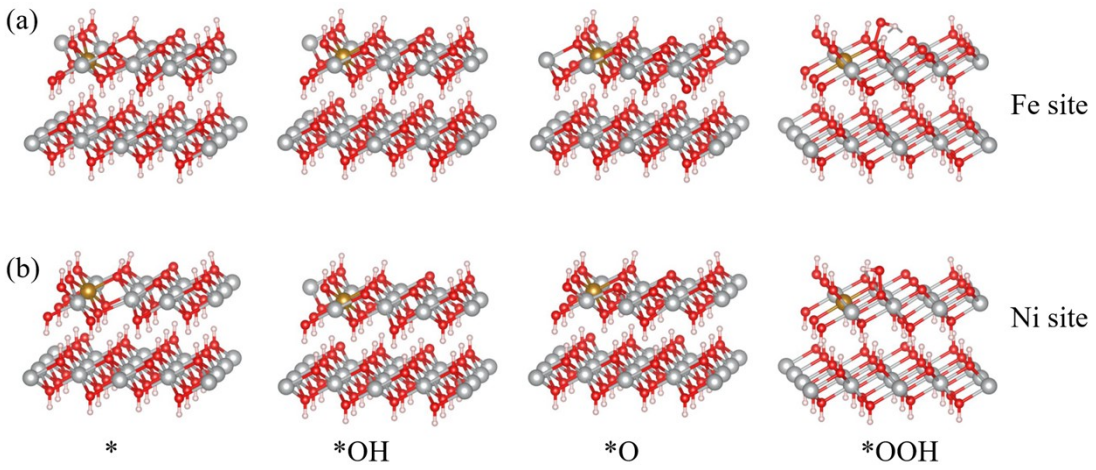

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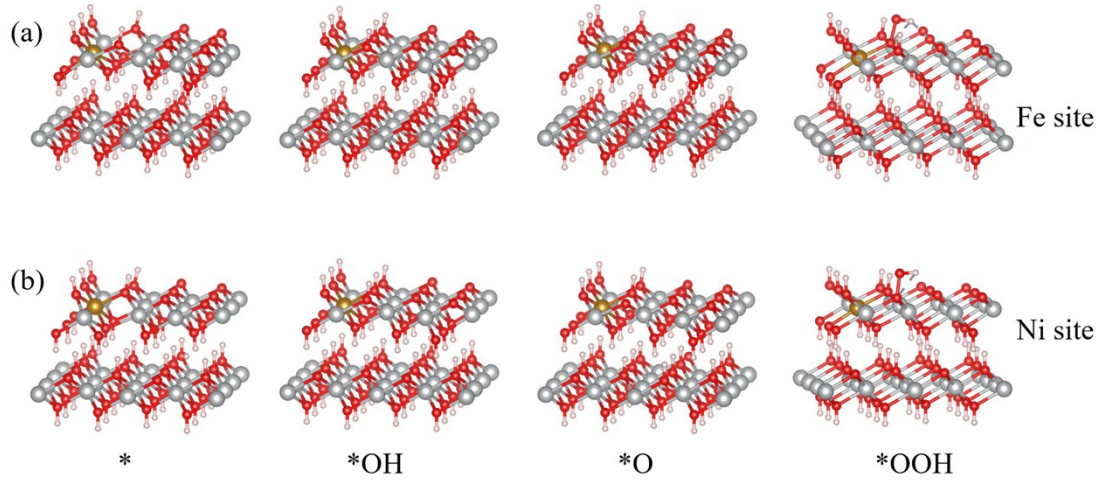

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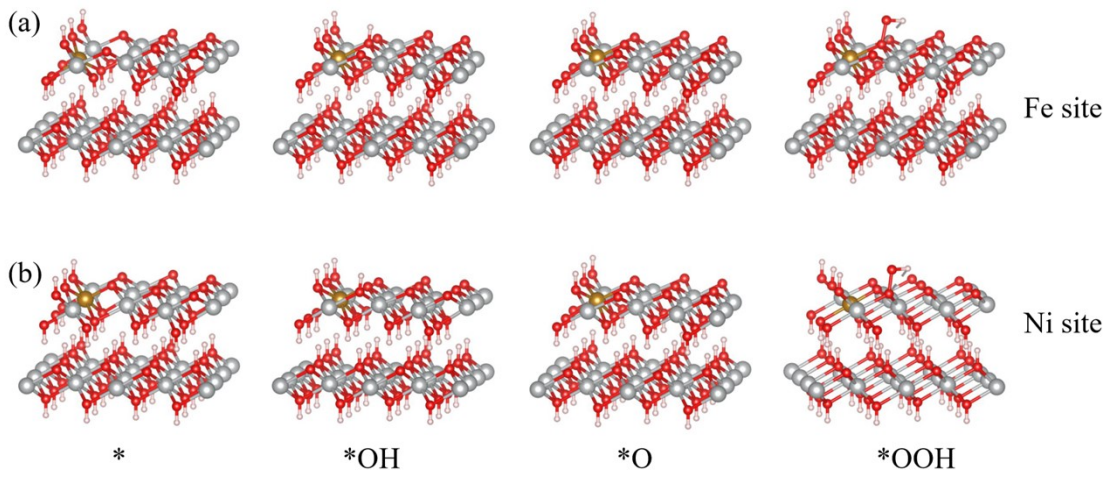

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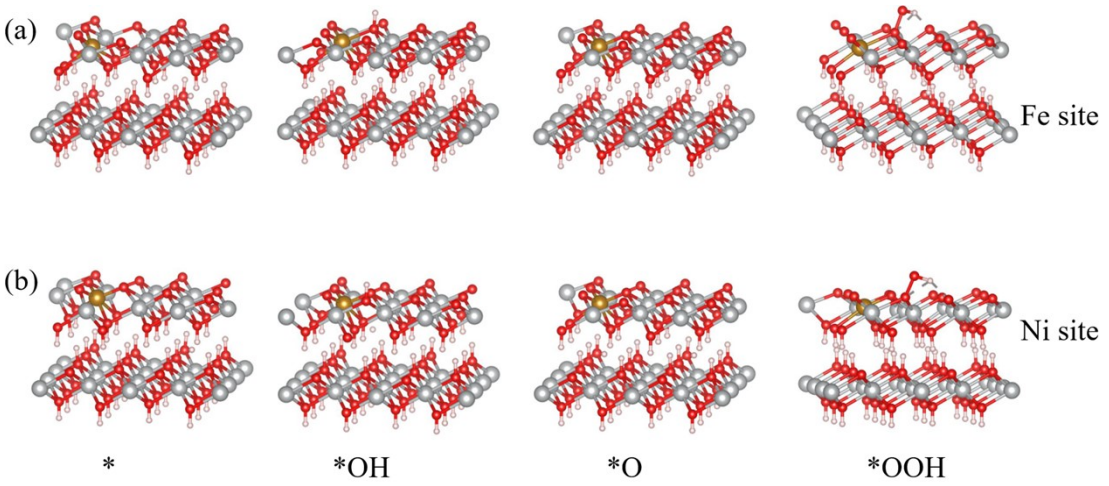
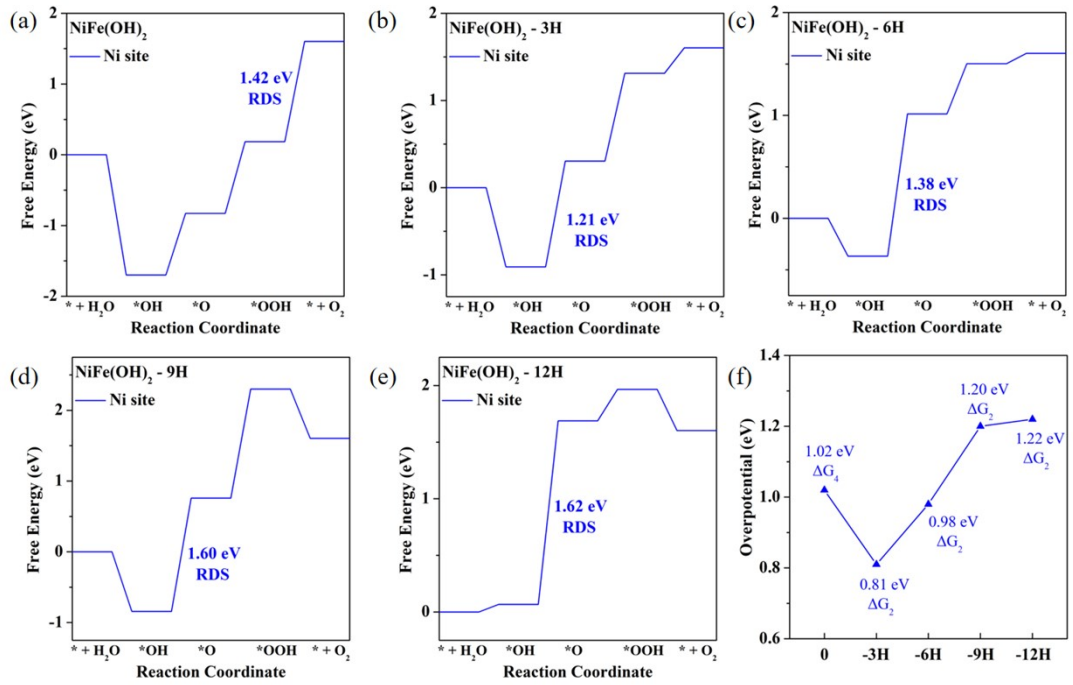


Figure S11. Atomic model structure of catalyst * and intermediates *OH, *O, and *OOH adsorption on the (a) Fe and (b) Ni site of $Ni(OH)_2$ - 12H surface. The pink, red, gray and golden sphere present H, O, Ni and Fe, respectively.



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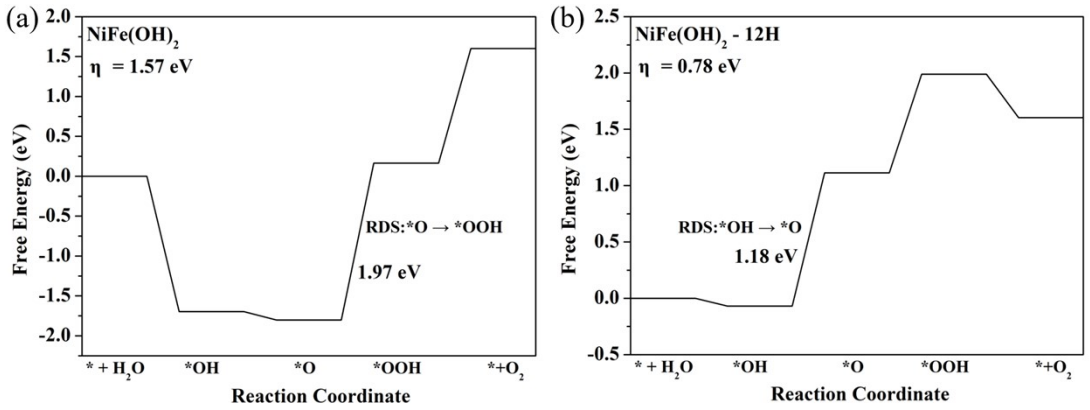

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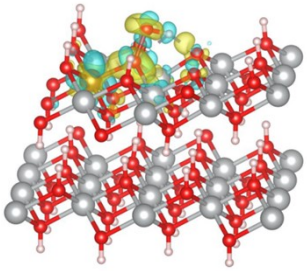

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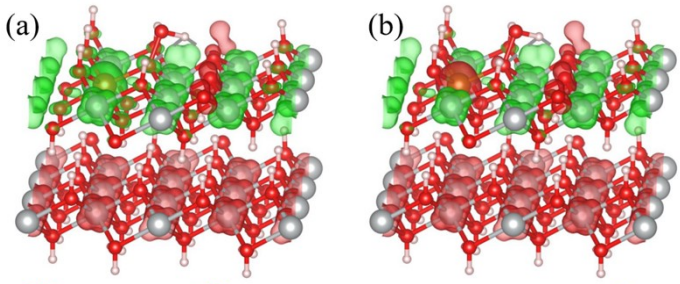

Figure S15. Spin density: (a) before and (b) after the Fe spin flip in the $NiFe(OH)_2$ - 3H adsorbing OOH, with the latter being energetically more favorable. The iso-surface is 0.02 electron/Å^3. (red: spin up; green: spin down). The pink, red, gray and golden sphere present H, O, Ni and Fe, respectively.

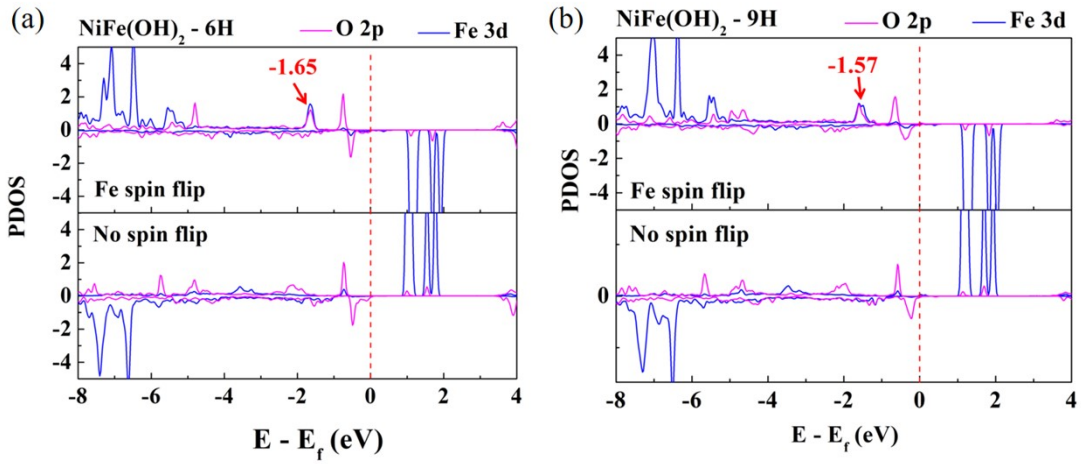

Figure S16. Computed PDOS of Fe-3d and O-2p (end O of OOH group) of OOH adsorption on (a) $NiFe(OH)_2$ - 6H, (b) $NiFe(OH)_2$ - 9H.

2. Supplementary Tables:

Table S1. The properties of $Ni(OH)_2$ (001), (100), and (110) surfaces.

Surface	Surface energy (γ, eV)	Coordination number (surface Ni)
(001)	0.023	6
(100)	0.042	5
(110)	0.055	4

Table S2: The calculated energies for different amounts of substitutional Fe ions are presented. E_{fer} and E_{anti} represent the total energies of the ferromagnetic and antiferromagnetic states, respectively.

Amount of Fe ions	E_{fer} /eV	E_{anti} /eV	E_{anti} - E_{fer} /eV
0	-605.34	-605.35	-0.01
1	-609.07	-609.09	-0.02
2	-613.67	-614.44	-0.77
3	-619.06	-619.08	-0.02