Supporting Information

Selective catalytic reduction of NO with NH$_3$ over Mo-Fe/Beta catalysts: The effect of Mo loading amounts

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1. H-Beta synthesis

H-Beta zeolite with a Si/Al ratio of 30 was prepared by dynamic hydrothermal synthesis using tetraethyl ammonium hydroxide (TEAOH) as a template. The solution for zeolite synthesis was prepared by mixing sodium hydroxide (AR), tetraethyl ammonium hydroxide (20 wt%), aluminum isopropoxide (98 wt%), silica sol (30 wt%) and deionized water with molar composition: 0.35Na$_2$O: 4.5(TEA)$_2$O: 0.25Al$_2$O$_3$: 25SiO$_2$: 295H$_2$O. The mixture was stirred for 24 h and then transferred to a Teflon autoclave and maintained at 100 °C for 3 days. The product was filtered, washed and dried at 110 °C for 12 h and calcined at 550 °C for 6 h to get Na-Beta. H-Beta was obtained by treating Na-Beta with 0.1 mol/L NH$_4$NO$_3$ at 80 °C for 8 h under vigorous stirring, then the solid product was dried at 110 °C for 2 h, and calcined at 450 °C for 3 h in air atmosphere.

2. Results

![Top view of Fe terminated surface](a)

![Side view of Fe terminated surface](b)

**Fig. S1.** (a) Top view of Fe terminated surface. (b) Side view of Fe terminated surface. Red and blue spheres represent O, and Fe atoms, respectively.
Fig. S2. The magnetic charge density plot of Fe$_2$O$_3$. Red and blue spheres represent O, and Fe atoms, respectively. The yellow and grey balls around the Fe atoms represent the positive and negative charge, respectively.

As shown in Fig. S2, the range of the magnetic charge of the model of Fe terminated Fe$_2$O$_3$ (001) surface is + − − +, which is the most favored magnetic configuration for α-Fe$_2$O$_3$ [1, 2].

Fig. S3. Stable structures for MoO$_x$/Fe$_2$O$_3$ (001) (top and side views) with the top five layers shown. Green and blue spheres represent Mo, and Fe atoms, respectively. The
big and small red spheres represent the surface and bulk O atoms, respectively. The pink spheres represent the O atoms of MoO\textsubscript{x} species.

To obtain a sufficiently stable structure of supported MoO\textsubscript{x} (x = 1-4) using density functional theory, we define the binding energy (BE) for the oxidation of Mo species referenced against gas-phase O\textsubscript{2} via Eq. (3)

\[
BE = E(\text{MoO}_x/\text{Fe}_2\text{O}_3) - E(\text{MoO}_{x-1}/\text{Fe}_2\text{O}_3) - 1/2 E(\text{O}_2)
\]  

(3)

The optimized constructions of MoO, MoO\textsubscript{2}, MoO\textsubscript{3} and MoO\textsubscript{4} on the Fe\textsubscript{2}O\textsubscript{3} surface are shown in Figure S3, which displayed the most stable binding configuration for the Mo species. The systematic study results are summarized in Table S2. The energy of MoO\textsubscript{2} obtaining an O atom is numeric equivalent of the energy of MoO\textsubscript{3} losing an O atom. Thus, MoO\textsubscript{3} is identified as the most stable structure supported on the surface of Fe\textsubscript{2}O\textsubscript{3}.

**Figure S4.** Stable structures for Mo\textsubscript{2}O\textsubscript{x} on the surface of Fe\textsubscript{2}O\textsubscript{3} (001). Green and blue spheres represent Mo, and Fe atoms, respectively. The big and small red spheres represent the surface and bulk O atoms, respectively. The pink spheres represent the O atoms of MoO\textsubscript{x} species.
In order to establish the stable model of two Mo atoms, we explored the stable construction of Mo$_2$O$_x$. Based on Eq. (3), Mo$_2$O$_4$ binds with an O atom to form Mo$_2$O$_5$ species, releasing energy of 2.76 eV. As well, Mo$_2$O$_5$ binds with an O atom to form Mo$_2$O$_6$ species, releasing energy of 2.34 eV. Thus, the formation of Mo$_2$O$_6$ is favorable in thermodynamics with the increasing of Mo loadings.

Fig. S5. Top view and side view of stable structure (MoO$_3$)$_2$ and (MoO$_3$)$_3$ on the surface of Fe$_2$O$_3$. Green and blue spheres represent Mo, and Fe atoms, respectively. The big and small red spheres represent the surface and bulk O atoms, respectively. The pink spheres represent the O atoms of MoO$_x$ species.
Table S1. Structural parameters for the structure MoO₅/Fe₂O₃ (001).

<table>
<thead>
<tr>
<th>MoOₓ</th>
<th>EₓE (eV)</th>
<th>dₓMo-O (Å)</th>
<th>dₓFe-O (Å)</th>
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<tr>
<td>MoO</td>
<td>—</td>
<td>1.724</td>
<td>—</td>
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<tr>
<td>MoO₂</td>
<td>-1.96</td>
<td>1.815, 1.828</td>
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<td>MoO₃</td>
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<td>MoO₄</td>
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<td>1.715, 1.841, 1.842, 1.842</td>
<td>1.941, 1.942, 1.943</td>
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References