Supporting Information

First-principles Modeling of Ni$_4$M (M= Co, Fe and Mn) Alloys as Solid Oxide Fuel Cell Anode Catalyst for Methane Reforming

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Table S1. Ni$_4$Fe surface segregation energies (eV).

<table>
<thead>
<tr>
<th>Number of Fe atoms in each layer$^a$</th>
<th>Position of Fe atoms (see Figure 1)</th>
<th>Relative energy$^b$</th>
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$^a$The numbers in column 1 indicate a tale of Fe atoms from the bottom layer (1st layer) to the top layer (5th layer).

$^b$All energy values are relative to the 5-6-12-13 surface structure energy.
Table S2. Ni$_4$Co surface segregation energies (eV).

<table>
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$^a$The numbers in column 1 indicate a tale of Co atoms from the bottom layer (1st layer) to the top layer (5th layer)

$^b$All energy values are relative to the 5-6-12-13 surface structure energy.
Table S3. Ni$_4$Mn surface segregation energies (eV).

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$^a$The numbers in column 1 indicate a tale of Mn atoms from the bottom layer (1st layer) to the top layer (5th layer)

$^b$All energy values are relative to the 5-6-12-13 surface structure energy.
Table S4. CH$_3$ binding energies on Ni and Ni$_4$Fe surfaces.

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Table S5. CH$_2$ binding energies on Ni and Ni$_4$Fe surfaces.

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<th>Site</th>
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<th>Opt. spin</th>
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Table S6. CH binding energies on Ni and Ni$_4$Fe surfaces.

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<tr>
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<th>Opt. spin</th>
<th>Ni ave. spin</th>
<th>Fe ave. spin</th>
<th>Ni</th>
<th>$E_{\text{bond}}$ (kcal/mol)</th>
<th>Calc. spin</th>
<th>Opt. spin</th>
<th>Ni ave. spin</th>
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<td>F</td>
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<td>10.06</td>
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Table S7. C binding energies on Ni and Ni$_4$Fe surfaces.

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Table S8. H binding energies on Ni and Ni\textsubscript{4}Fe surfaces.

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<th>E\textsubscript{bond} (kcal/mol)</th>
<th>Opt. spin</th>
<th>Ni ave. spin</th>
<th>Fe ave. spin</th>
<th>Sites</th>
<th>E\textsubscript{bond} (kcal/mol)</th>
<th>Calc. spin</th>
<th>Opt. spin</th>
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Table S9. Binding energies (kcal/mol) of the reaction intermediates at f and h sites of Ni₄M alloys.

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