Theoretical study of CO_2 capture mechanisms of SrO and $Sr(OH)_2 \cdot nH_2O$ (n=0,1,8)

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CO$_2$ Capture Using SrO

➢ **Reactive capture uses metal oxides and solid carbon to capture/convert CO$_2$**

Capture: Thermally heating a mixture of CO$_2$ and metal oxide forms a stable metal carbonate:

CO$_2$ + MO → MCO$_3$

Conversion: Subsequently heating the MCO$_3$ and solid carbon produces carbon monoxide and regenerates the sorbent
1. MCO$_3$ + C → 2CO + MO (commercially useful)
2. MCO$_3$ → CO$_2$ + MO

➢ CaO has been used due to relatively low-cost and abundance

➢ **SrCO$_3$/SrO showed much better CO selectivity than CaCO$_3$/CaO**
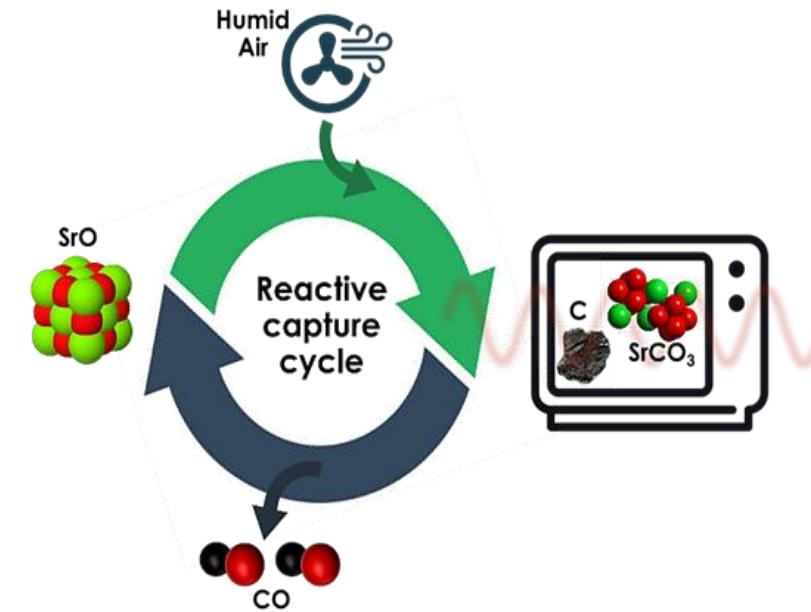
CO$_2$ Absorption on Surfaces

➢ **SrO can react with H$_2$O in flue gas to from Sr(OH)$_2$.nH$_2$O**

➢ **CO$_2$ adsorption first happens on the surface before CO$_2$ diffuses into the bulk**

$$SrO \xrightarrow{H_2O} Sr(OH)_2 \xrightarrow{H_2O} Sr(OH)_2 \cdot 1H_2O \xrightarrow{7H_2O} Sr(OH)_2 \cdot 8H_2O$$

CO$_2$ CO$_2$ CO$_2$ CO$_2$

SrCO$_3$
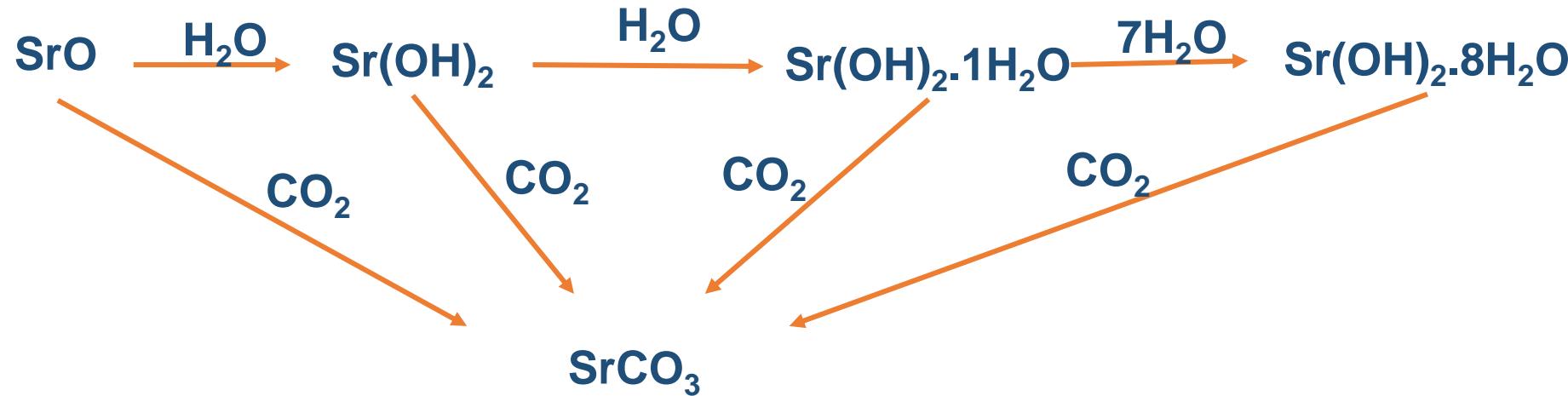
Bulk Crystal Parameters

Crystal	Calculated	Experiment
SrO	5.17x5.17x5.17 Å	5.16x5.16x5.16 Å
$Sr(OH)_2$	9.84x3.94x6.03 Å	9.89x3.92x6.12 Å
$Sr(OH)_2.1H_2O$	8.94x 8.95x11.37 Å	3.65x6.20x6.71 Å
$Sr(OH)_2.8H_2O$	8.94x 8.95x11.37 Å	9.02x9.02x11.61 Å
$SrCO_3$	5.19x 8.51x 6.05 Å	5.09x8.36x6.00 Å

SrO $Sr(OH)_2$ $Sr(OH)_2.1H_2O$ $Sr(OH)_2.8H_2O$ $SrCO_3$

Surface Stability

Most stable surfaces

	Surface
SrO	100
Sr(OH)$_2$	101
Sr(OH)$_2$.1H2O	010
Sr(OH)$_2$.8H$_2$O	001
SrCO$_3$	011
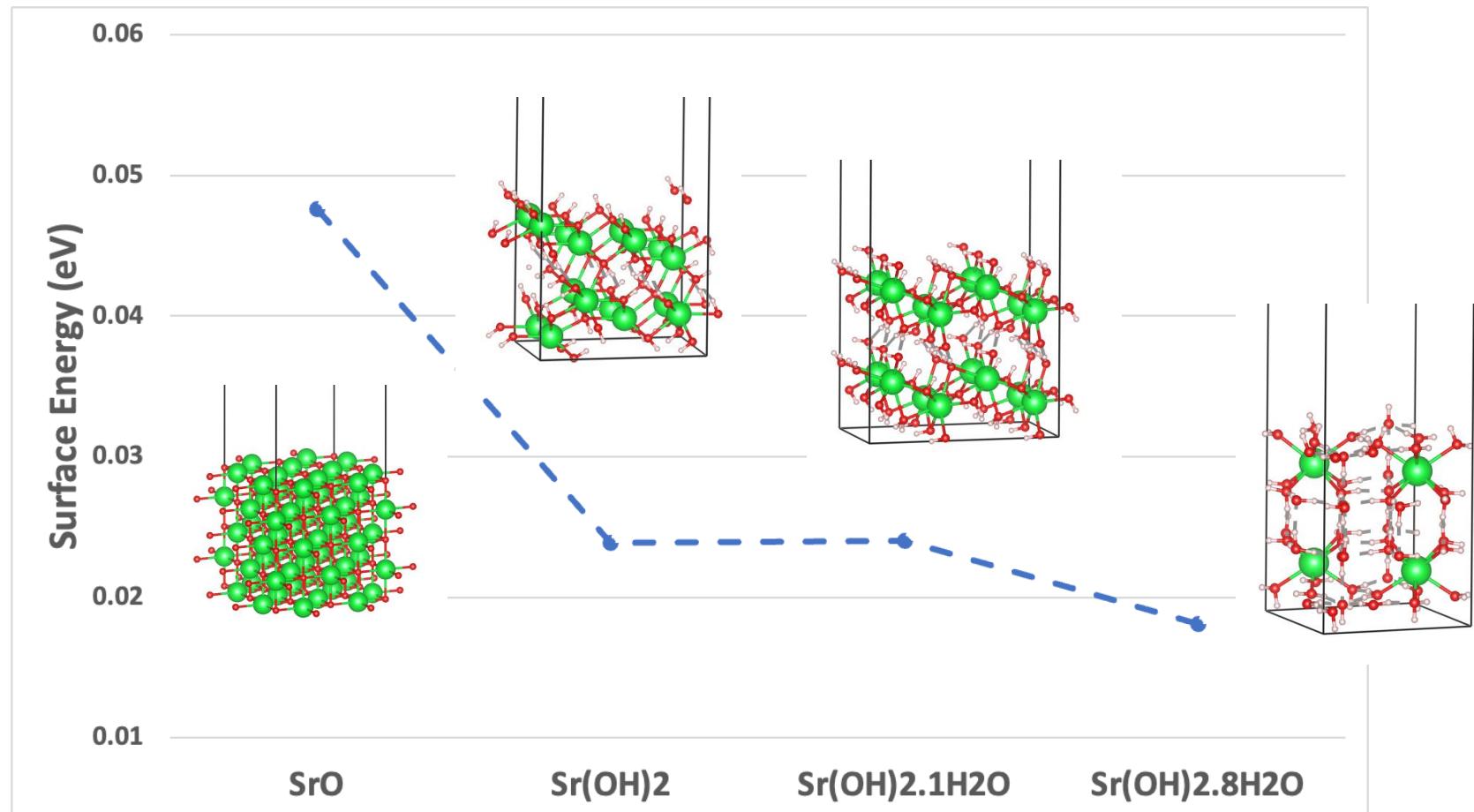
➤ **There is a drop in surface energy from SrO to Sr(OH)$_2$ surfaces and from Sr(OH)$_2$.1H$_2$O to Sr(OH)$_2$.8H$_2$O**

CO$_2$ Absorption and Desorption Energy

Surface reaction	DFT Energy
SrO + CO$_2$	-1.7 eV / -164.069 KJ/mol
Sr(OH)$_2$ + CO$_2$	-0.48 eV / -46.3 KJ/mol
Sr(OH)$_2$.1H$_2$O + CO$_2$	-0.58 eV/ -40.01 KJ/mol
Sr(OH)$_2$.8H$_2$O + CO$_2$	-0.06 eV/ -5.8 KJ/mol
SrCO$_3$ => SrO + CO$_2$	3.16 eV/ 304.5 KJ/mol
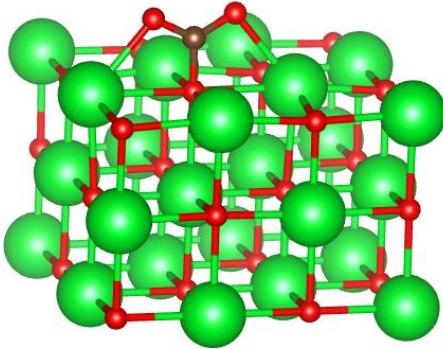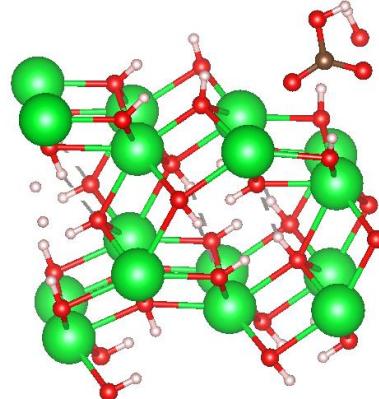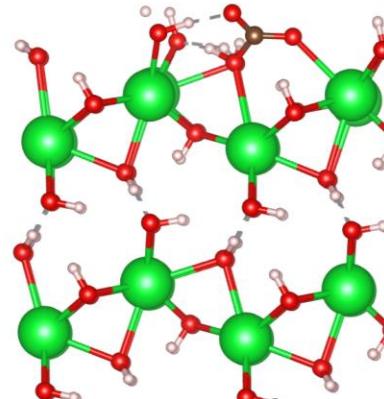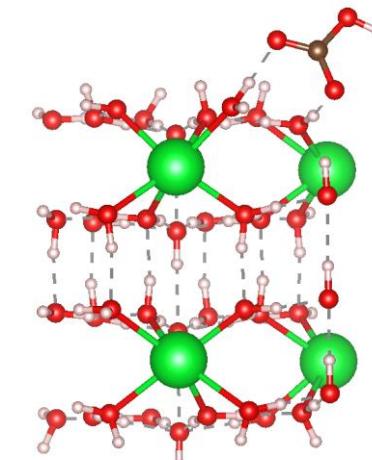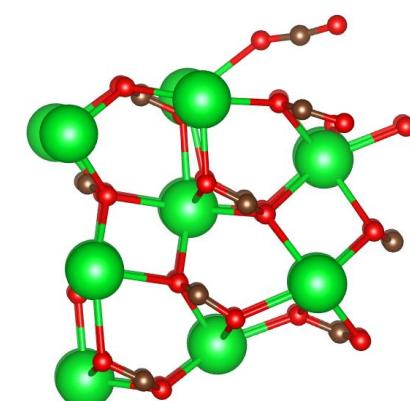
SrO   Sr(OH)$_2$ Sr(OH)$_2$.1H$_2$O   Sr(OH)$_2$.8H$_2$O   SrCO$_3$

➢ **CO$_2$ adsorption energy on surfaces increases with H$_2$O content except for monohydrate**

CO$_2$ Surface Absorption Reaction Pathways
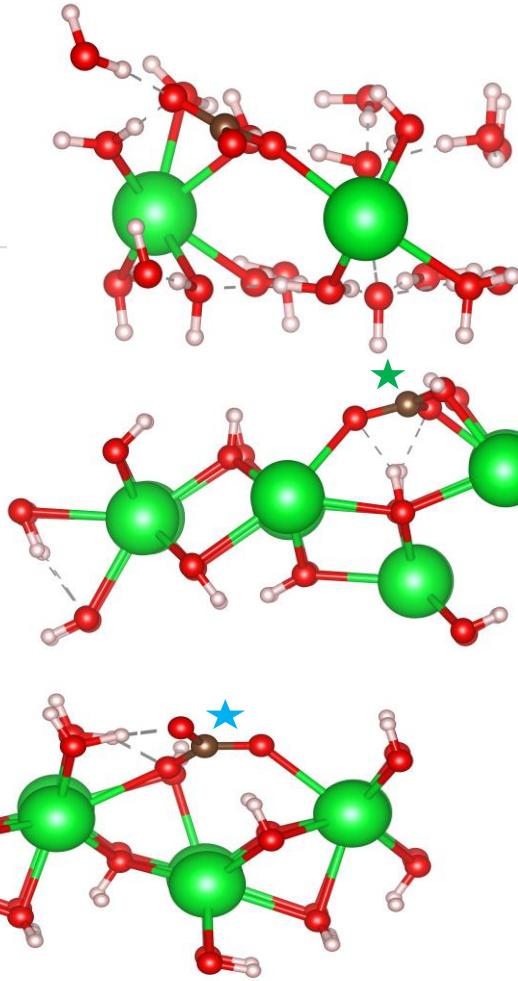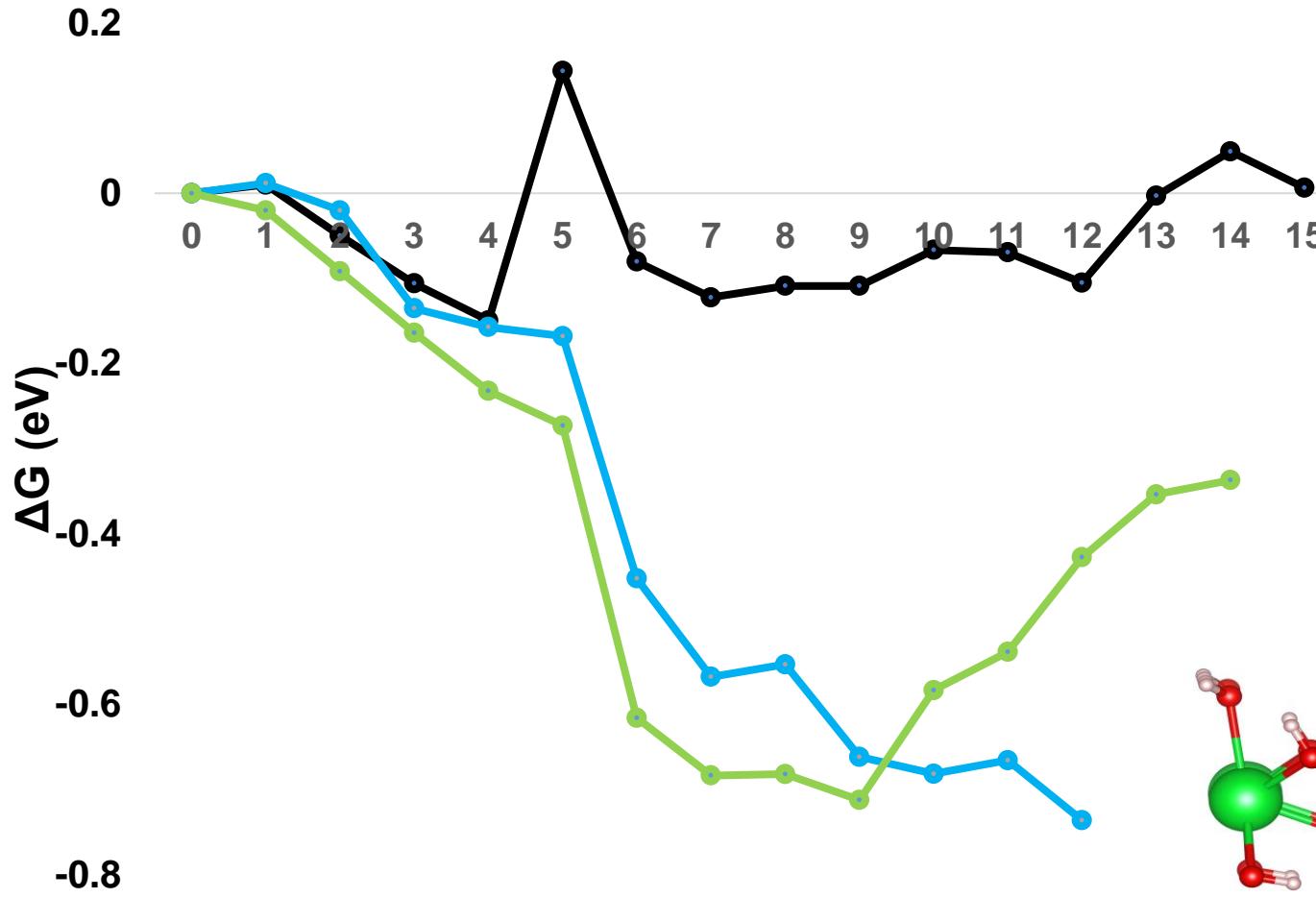
> Initial surface carbonation was most favorable on Sr(OH)$_2$*1H$_2$O because CO$_2$ formed a carbonate-like species without an increase in free energy

CO$_2$ Bulk Reaction Thermodynamics
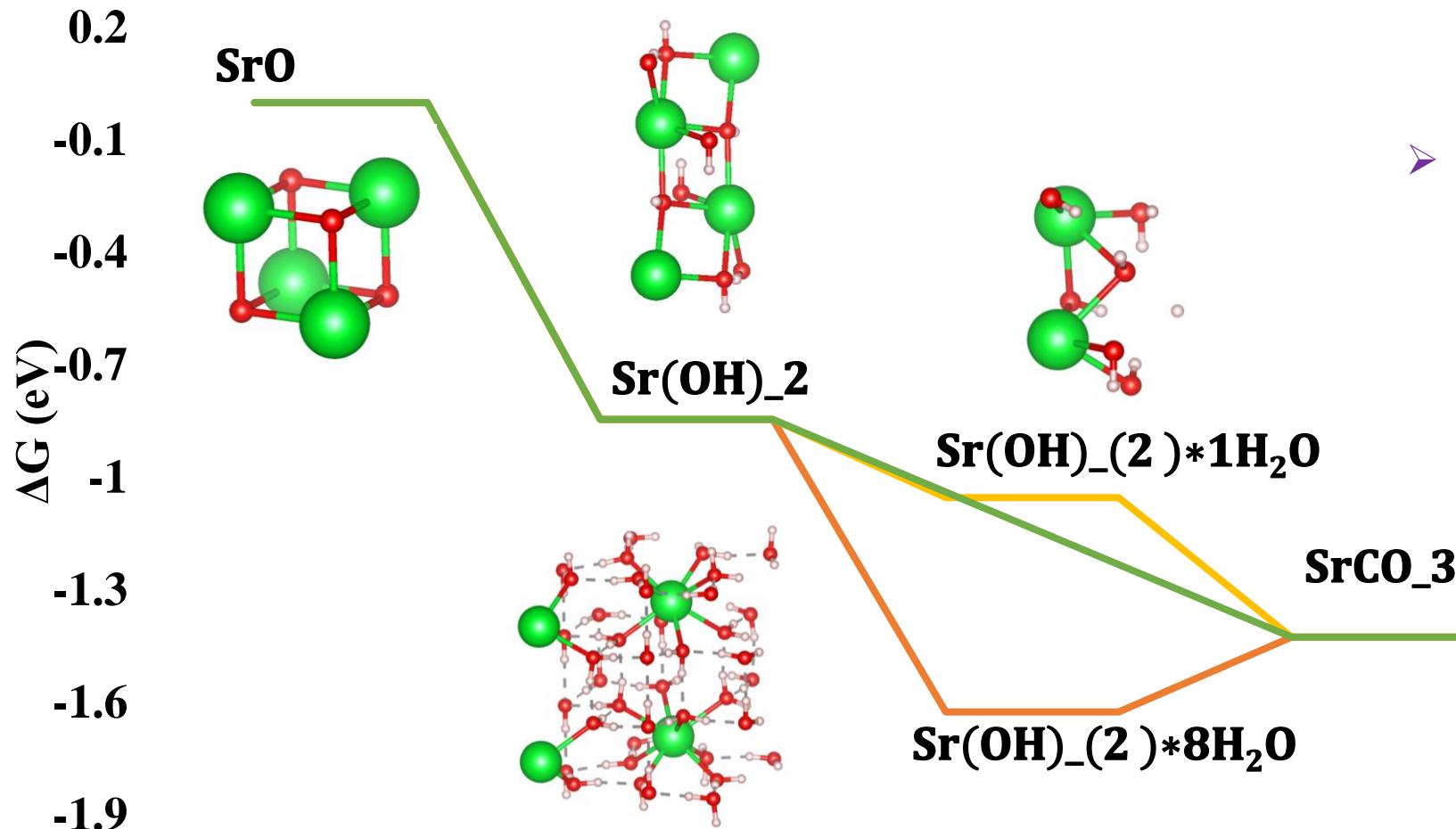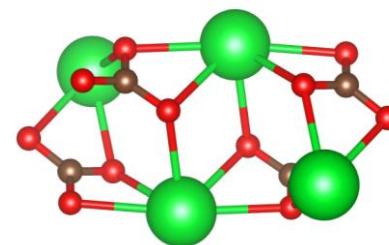
Conclusion

- We examined the adsorption reactions of CO_2 on SrO, and $Sr(OH)_2 \cdot nH_2O$ surfaces

- The predicted energies of CO_2 adsorption on surfaces follows the order of increasing H_2O content except for monohydrate

- The carbonation of monohydrate surface is barrierless, whereas the CO_2 reaction on the $Sr(OH)_2$ and $Sr(OH)_2 \cdot 8H_2O$ surfaces follows pathways with activation barriers of 0.4 eV and 0.3 eV respectively.

- The dissociation of CO_2 from $SrCO_3$ (011) surface needs energy of 304.5 kJ mol^{-1}.

- The thermodynamics of bulk reaction is also addressed

Acknowledgement

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THANK YOU!