

Supporting information for: Large scale computational screening and experimental discovery of novel materials for high temperature CO₂ capture

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Energy penalty calculations

If the temperatures for the carbonation and calcination reactors are set by the theoretical carbonation equilibrium of the material, the inlet p_{CO_2} (chosen to be 0.15 for a typical flue gas) and the outlet p_{CO_2} (chosen to be 1.00 for production of pure CO_2), then the total diverted energy from the power process per mole of CO_2 absorbed (m_{CO_2}) is given by:

$$Q_{\text{loss}} = \frac{\Delta H_{\text{calcination}} + C_p(T_{\text{calcination}} - T_{\text{carbonation}})}{m_{\text{CO}_2}} \quad (1)$$

where C_p is the molar heat capacity of the sorbent material, $T_{\text{calcination}}$ is the temperature of calcination, $T_{\text{carbonation}}$ is the temperature of carbonation and $\Delta H_{\text{calcination}} = -\Delta H_{\text{carbonation}}$. Assuming a Carnot efficiency, the work lost due to this diverted heat is

$$W_{\text{loss}} = Q_{\text{loss}} \left(1 - \frac{T_{\text{environment}}}{\min(T_{\text{calcination}}, T_h)} \right) \quad (2)$$

where $T_{\text{environment}}$ is chosen to be room temperature, 298 K, and T_h is the operating temperature of the power cycle, which was assumed to be 623 K.

However, there is a need to consider the amount of work that can be recovered by reintegrating the heat from the carbonator, which will reduce the total W_{loss} . The heat rejected by the CCS process is given by:

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$$Q_{recovered} = Q_{loss} - \Delta H_{CO_2,flow} \quad (3)$$

where $\Delta H_{CO_2,flow}$ is the net molar enthalpy flow between the CO₂ inlet and outlet. Therefore the recovered work from this heat can be calculated:

$$W_{recovered} = Q_{recovered} \left(1 - \frac{T_{environment}}{T_{carbonation}} \right) \quad (4)$$

and the total energy penalty, E_p is given by:

$$E_p = W_{loss} - W_{recovered} \quad (5)$$

Surface area analysis

The results from the BET SSA analysis are shown in Table S1.

Table S1: Results of the SSA and total pore volume measurements for the candidate materials studied here, along with other similar previously studied materials. The total pore volume was determined from N₂ absorption.

Compound	BET SSA (m ² /g)	Total pore volume (cm ³ /g)
Mg ₆ MnO ₈	1.6018	0.169809
Ca ₄ Nb ₂ O ₉	0.3813	0.033179
Na ₃ SbO ₄	0.3787	0.001239
Li ₅ FeO ₄	< 0.1	< 0.001
Li ₆ CoO ₄	1.00813	0.018272
Li ₄ SiO ₄	0.6778	0.004228
Li ₅ AlO ₄	2.7585	0.015195