Electronic Supplementary Materials for

Perovskite Oxides as A New Family of Tunable CO_2 Sorbents

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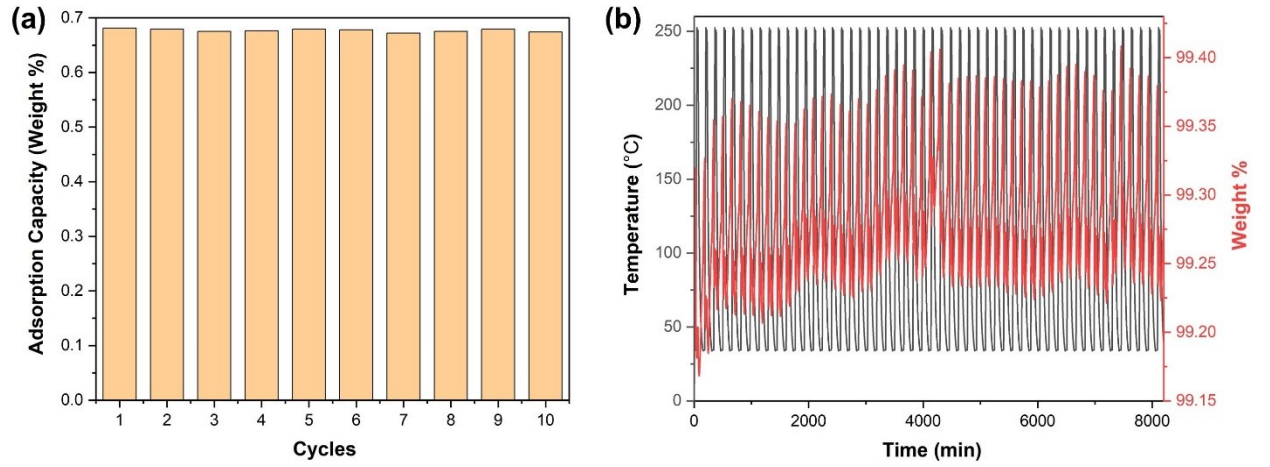

Figure S1- TGA results of $Sr_xLa_{1-x}FeO_3$ perovskite oxide. (a) Cyclic CO2 uptake of $Sr_{0.2}La_{0.8}FeO_3$ (Electrospun) of at 35 °C using CO2/Ar (10/90) via a gravimetric method. (b) Cyclic CO_2 uptake of $Sr_{0.2}La_{0.8}FeO_3$ (Electrospun) of at 35 °C using 0.4 % CO_2 via a gravimetric method.

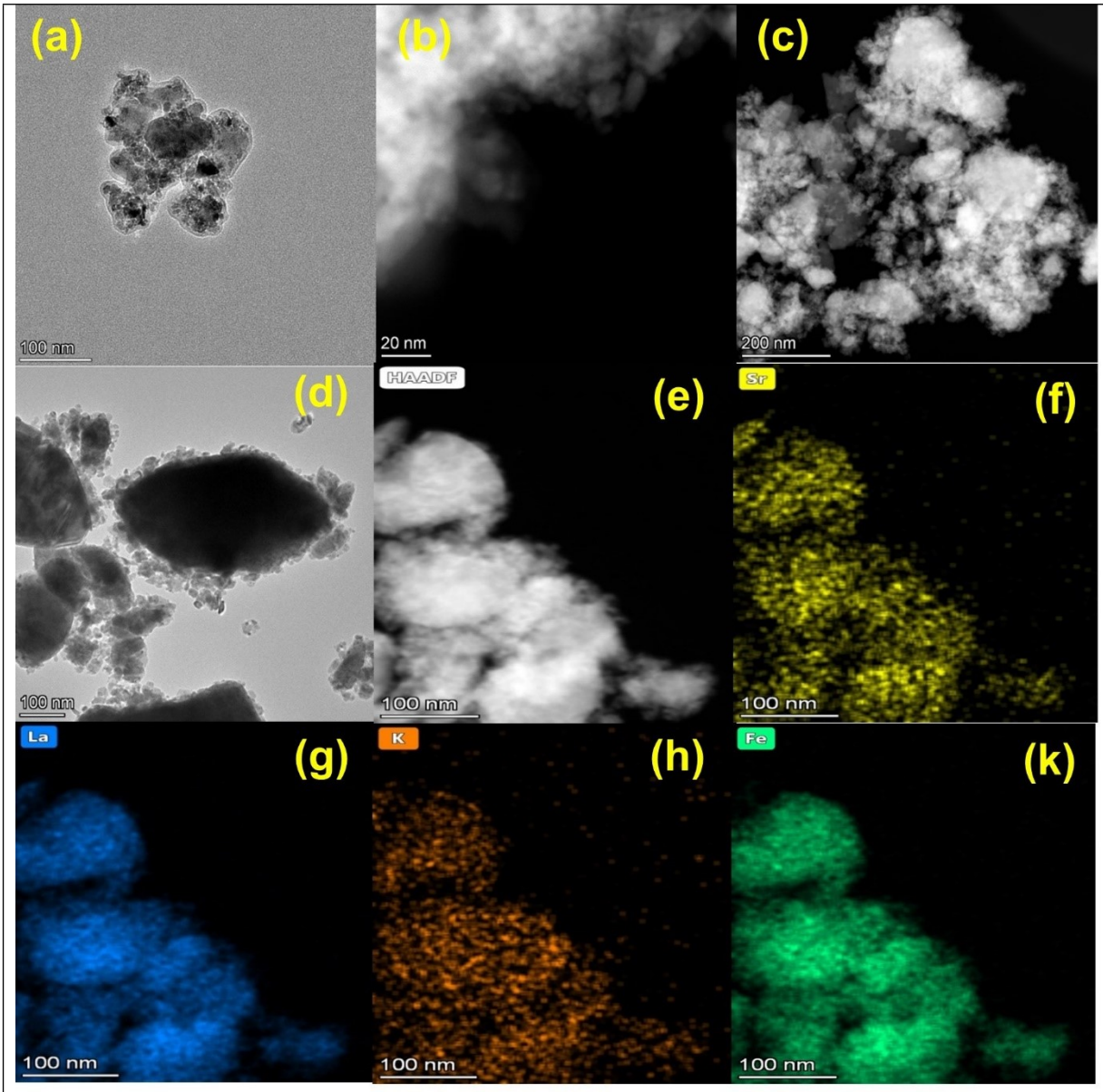

Figure S2- TEM results of Sr0.2 La0.8 Fe O3- Reactive grinding (a-d)TEM images (e-k) Elemental mapping by EDS from TEM.

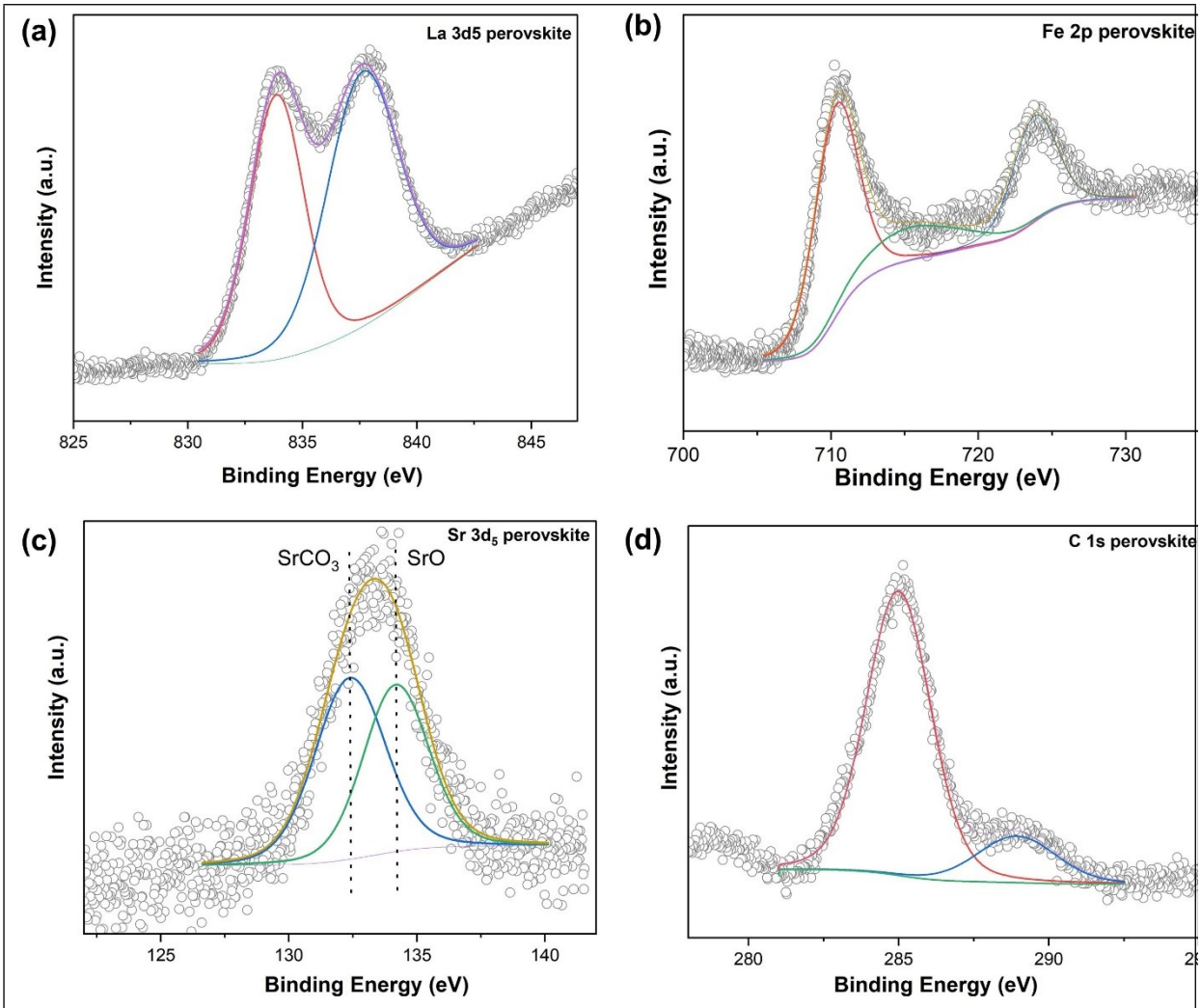

Figure S3- XPS results of $Sr_{0.2}La_{0.8}FeO_3$ perovskite oxide (Electrospinning method)

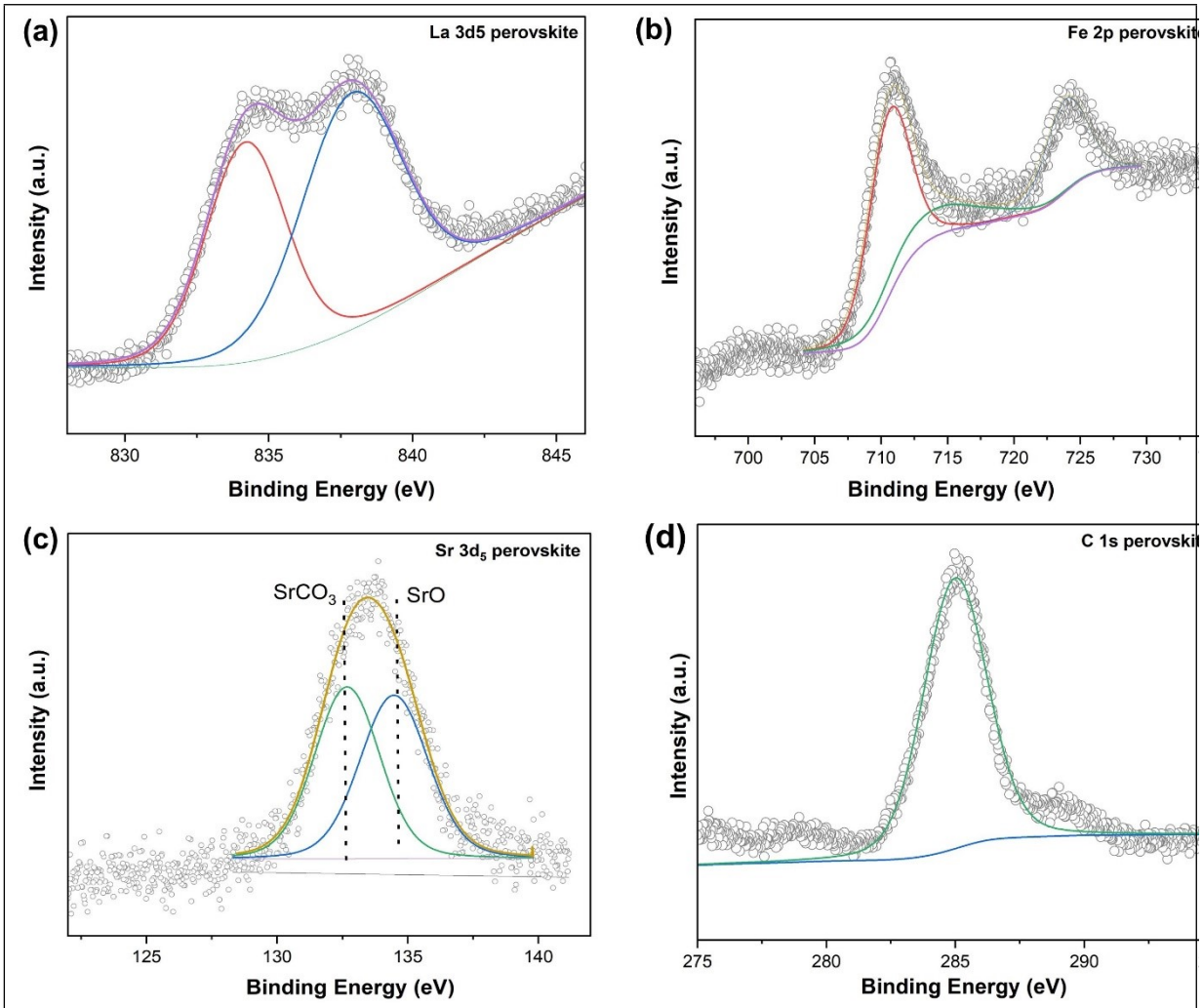

Figure S4- XPS results of $Sr_{0.2}La_{0.8}FeO_3$ perovskite oxide (salt-assisted grinding)

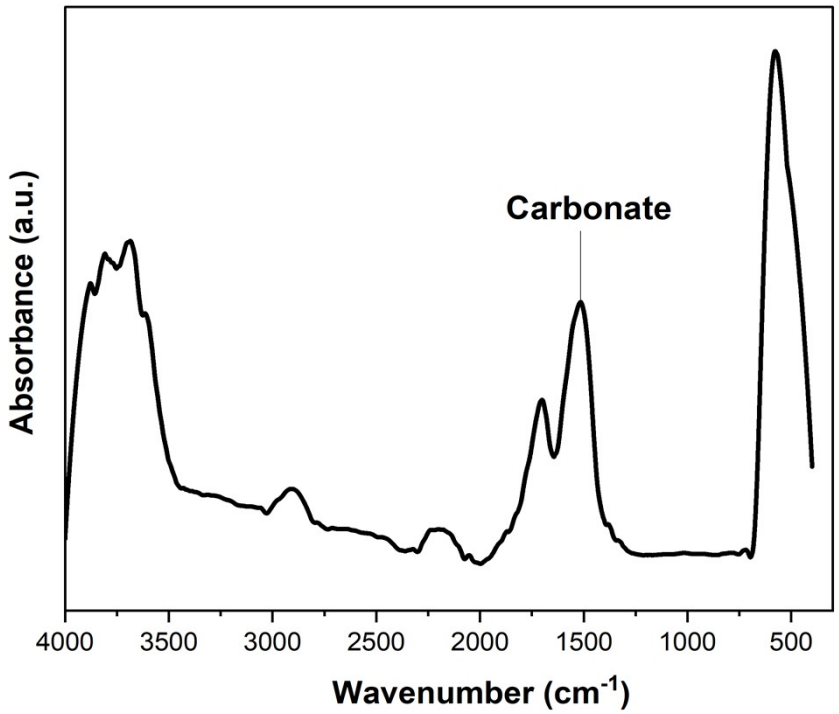

Figure S5- Ex-situ FTIR of $Sr_{0.2}La_{0.8}FeO_3$ at 25 C after CO_2 adsorption

To comprehend the bonding between carbon dioxide and adsorption sites in $Sr_{0.2}La_{0.8}FeO_3$ synthesized through electrospinning, ex-situ FTIR analysis was conducted. Ex-situ FTIR analysis was performed to assess the surface properties of the electrospun perovskite. To conduct *ex-situ* FTIR analysis, the initial CO_2 adsorption cycle utilized a gas mixture of CO_2/Ar (10% CO_2, 90% Ar) flowing at a rate of 200 mL/min within the TGA system. This process operated for 30 minutes at 35 °C. Subsequently, the treated sample was transferred to the IR for spectrum collection. **Figure S5** reveal distinct peaks corresponding to bicarbonate and carbonate compounds, which are observed in the spectral range of approximately 1300 to 1500 cm^{-1}.[39] The presence of carbonate at the surface of the sample is indicated by these prominent peaks. This observation is suggests that carbonates have formed on the perovskite material's surface.

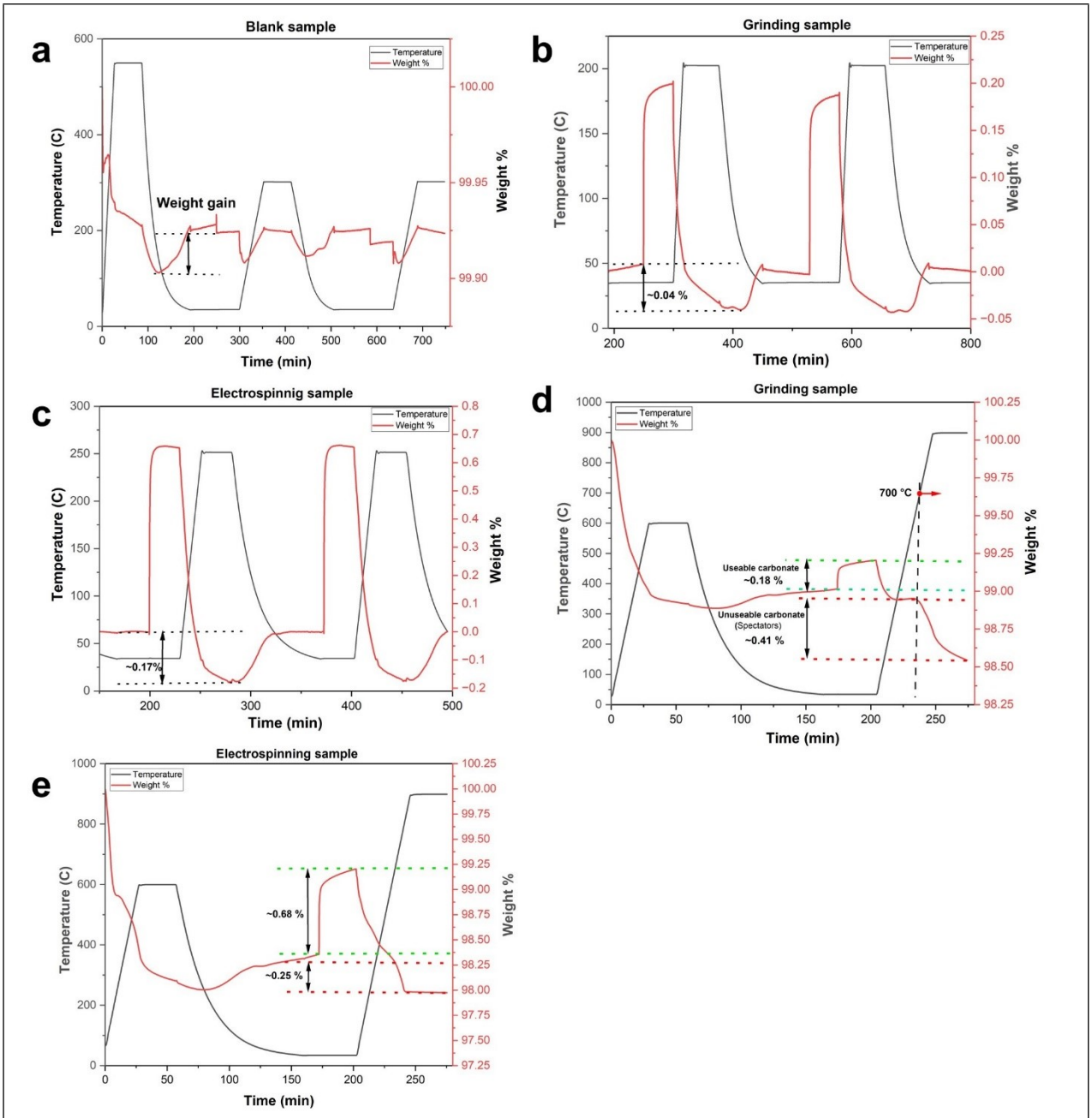

Figure S6- (a) Blank sample used to test weight gain in the TGA. (b-c) First two cycles of CO_2 adsorption on $Sr_{0.2}La_{0.8}FeO_3$ in TGA for both the ground and electrospun samples. (d-e) Temperature-programmed desorption of $Sr_{0.2}La_{0.8}FeO_3$ for ground and electrospun samples to identify different types of carbonates.

Figure S6 shows with increasing temperature after CO_2 adsorption (up to 900°C), different types of carbonates were identified in the electrospun and ground samples. In the electrospun sample, the weight difference between the point before CO_2 injection and after CO_2 desorption at the desorption temperature (~0.17%, **Figure S6(c)**) is quite close to the weight loss observed at 900°C (~0.25%, **Figure S6(e)**). In contrast, the ground sample shows a much larger difference: the weight loss at the desorption temperature is ~0.04% (**Figure S6(b)**), while it

increases to ~0.41% at 900°C (**Figure S6(d)**). This suggests that a significant portion of the carbonates in the ground sample behaves as spectator species, only releasing after 700°C. This again highlights the possibility that some of the $SrCO_3$ formed may act as a spectator phase without participating in reversible CO_2 sorption.

Figure S6a shows the weight gain for the blank sample (YSZ) run in the TGA. The instrument exhibits some weight gain, particularly at lower temperatures (near room temperature) between two temperature points. This confirms that the small weight gain observed during the pretreatment step is due to the instrument itself, not the samples.

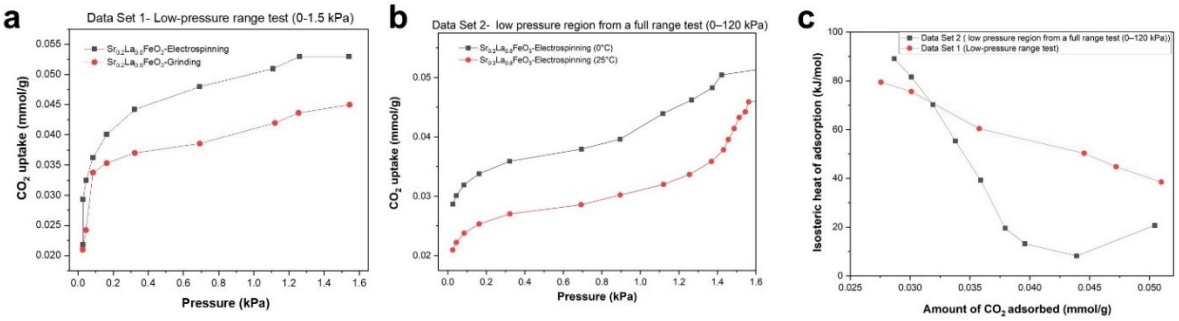

Figure R2- Comparison of the two data sets for the low-pressure range of the CO_2 isotherm. (a) CO_2 adsorption isotherms of $Sr_{0.2}La_{0.8}FeO_3$ at 0 and 25 C (Data set 1) (b) CO_2 adsorption isotherms of $Sr_{0.2}La_{0.8}FeO_3$ at 0 and 25 C (Data set 2) (c) Comparison of CO_2 isosteric heats of adsorption between Data Set 1 and Data Set 2.

We conducted two tests for CO_2 uptake on the electrospun sample: a low-pressure range test (0 – 1.5 kPa) and a full-pressure range test. Both tests covered the low-pressure region between 0 and 1.5 kPa, as shown in **Figures S7a** and **S7b**. The results reveal minor inconsistencies in instrument measurements within this range. However, both data sets consistently indicate that the adsorption process in this region is dominated by chemisorption. This conclusion is supported by **Figure S7c**, where the average enthalpy in both data sets exceeds 60 kJ/mol at adsorption capacity below 0.035 mmol/g. Both tests were performed on the same batch of the sample under identical experimental conditions.