

## Supplementary Information

### Flame spheroidisation of dense and porous $\text{Ca}_2\text{Fe}_2\text{O}_5$ microspheres

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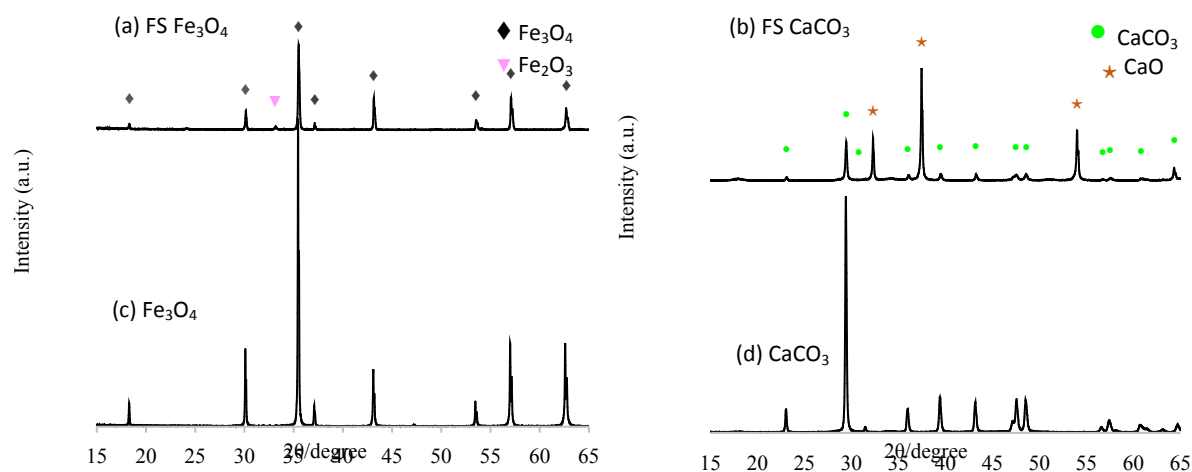
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## 1.0 Structural Analysis

Complementary XRD investigations were performed to establish the crystalline structures of the  $\text{Fe}_3\text{O}_4$  and  $\text{CaCO}_3$  powders, both pre and post FS-processing, to clarify the structural transformations of these materials when processed individually (Figure S1). The structures of the starting materials were confirmed as  $\text{Fe}_3\text{O}_4$  and  $\text{CaCO}_3$  (Figures S1c and S1d, respectively). FS-processed magnetite powders (in the form of solid microspheres from SEM observations) were found to remain predominantly as  $\text{Fe}_3\text{O}_4$ , with some evidence for the development of trace amounts of  $\text{Fe}_2\text{O}_3$  (Figure S1a). FS-processed porogen exhibited diffraction peaks attributable to both  $\text{CaCO}_3$  and  $\text{CaO}$  (Figure S1b).



**Fig. S1.** XRD data for: (a) FS-processed  $\text{Fe}_3\text{O}_4$ ; (b) FS-processed  $\text{CaCO}_3$ ; (c)  $\text{Fe}_3\text{O}_4$  starting powder; and (d)  $\text{CaCO}_3$  starting porogen

## 2.0 Magnetic expression

A simple experiment using a magnet confirmed the sieved FS-processed  $\text{Ca}_2\text{Fe}_2\text{O}_5$  products to indeed be magnetic (Figure S2).



**Fig. S2.** Magnet placed next to FS-processed  $\text{Fe}_3\text{O}_4$ : $\text{CaCO}_3$ , following sieving, demonstrating the  $\text{Ca}_2\text{Fe}_2\text{O}_5$  microsphere products to be magnetic.

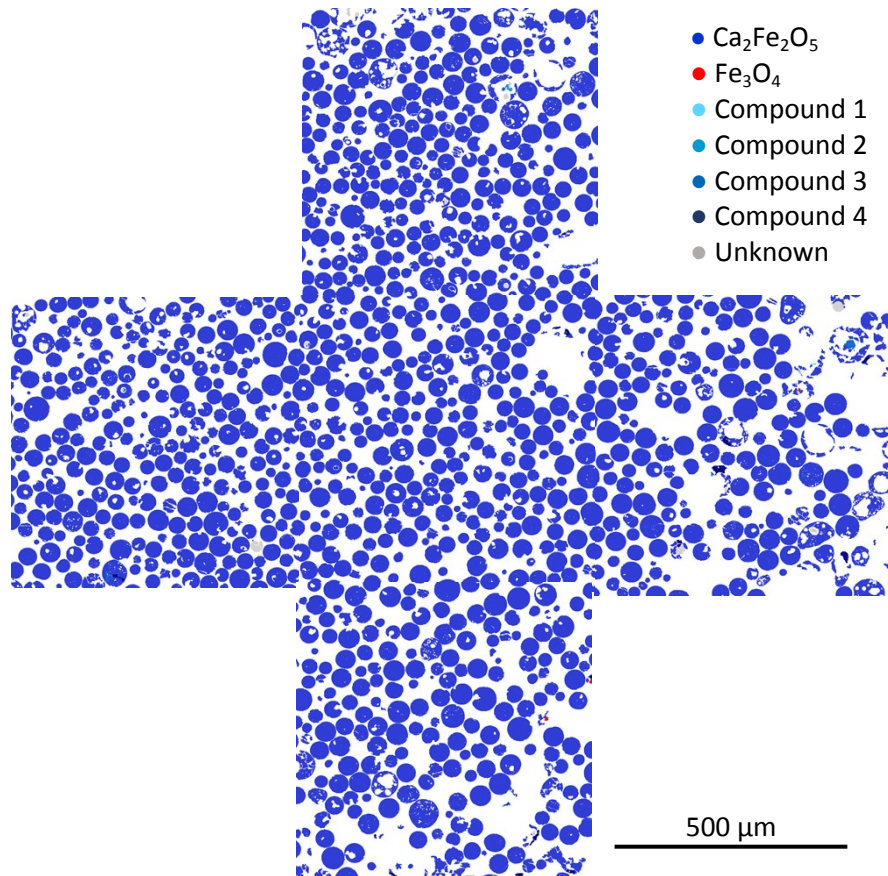
### 3.0 Mineral Mapping

**Table S1. Mineral reference**

Mineral	Ca%	Fe%	O%
Ca <sub>2</sub> Fe <sub>2</sub> O <sub>5</sub> (srebrodolskite)	19.1	57	23.9
Fe <sub>3</sub> O <sub>4</sub>	-	72.4	27.64
Compound 1	3.6	73.8	22.6
Compound 2	9.6	67.3	23.1
Compound 3	15.1	61.3	23.6
Compound 4	48.4	28.9	22.6

**Table S2. Modal mineralogy**

Mineral	Particles	Weight %
Ca <sub>2</sub> Fe <sub>2</sub> O <sub>5</sub> (srebrodolskite)	1501	<b>99.75</b>
Fe <sub>3</sub> O <sub>4</sub>	5	0.01
Compound 1	2	0.01
Compound 2	3	0.01
Compound 3	24	0.08
Compound 4	25	0.14
Unknown	46	0.00



**Fig. S3.** Full MLA compositional analysis of FS-processed Fe<sub>3</sub>O<sub>4</sub>:CaCO<sub>3</sub>, following sieving and sectioning, demonstrating very high levels of Ca<sub>2</sub>Fe<sub>2</sub>O<sub>5</sub>, along with very low levels of related CaFeO phases and Fe<sub>3</sub>O<sub>4</sub>.

#### 4.0 Products before and after FS

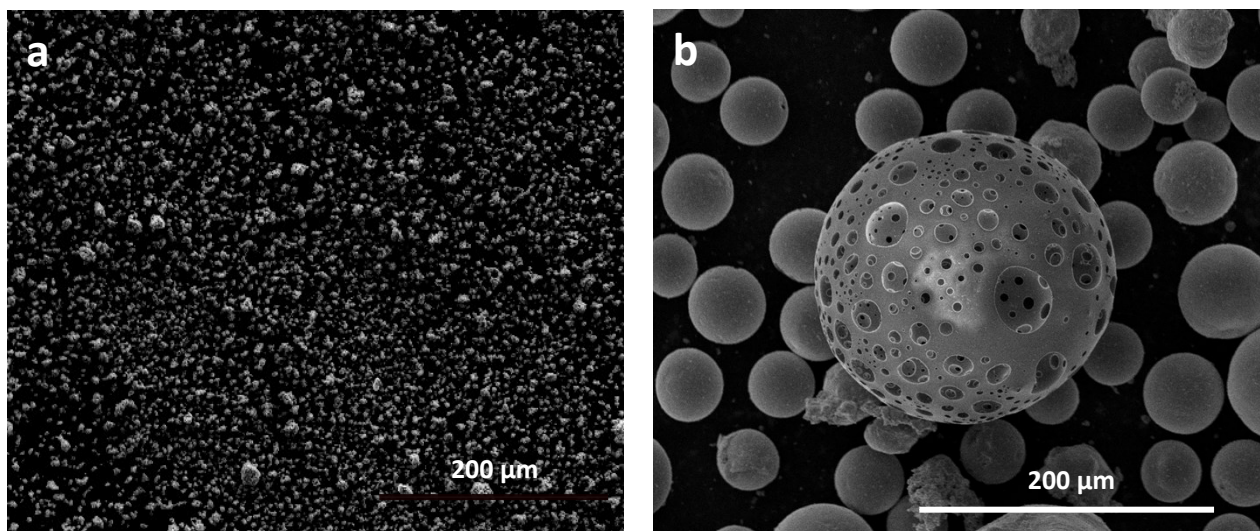


Fig. S4. a) Prepared  $\text{Fe}_3\text{O}_4 / \text{CaCO}_3$  mixture with PVA. Powder used for both FS and HT-XRD analysis; b)  $\text{Ca}_2\text{Fe}_2\text{O}_5$  porous microsphere (centre) and dense microspheres (background) after FS processing.

#### 5.0 Surface analysis

Complementary X-ray photoelectron spectroscopy (VG ESCALab Mark II X-ray Photoelectron and Scanning Auger Spectrometer; Al  $K\alpha$  source; 20 mA; 20 kV; step 1; No. of scans 2; dwell 0.2; pass energy 50 eV, Constant Analyser Energy mode) study was performed to analyse the surface chemistry of the FS-processed  $\text{Ca}_2\text{Fe}_2\text{O}_5$  porous and dense microspheres. Figure S5 presents a survey spectra of the srebrodolskite microspheres.

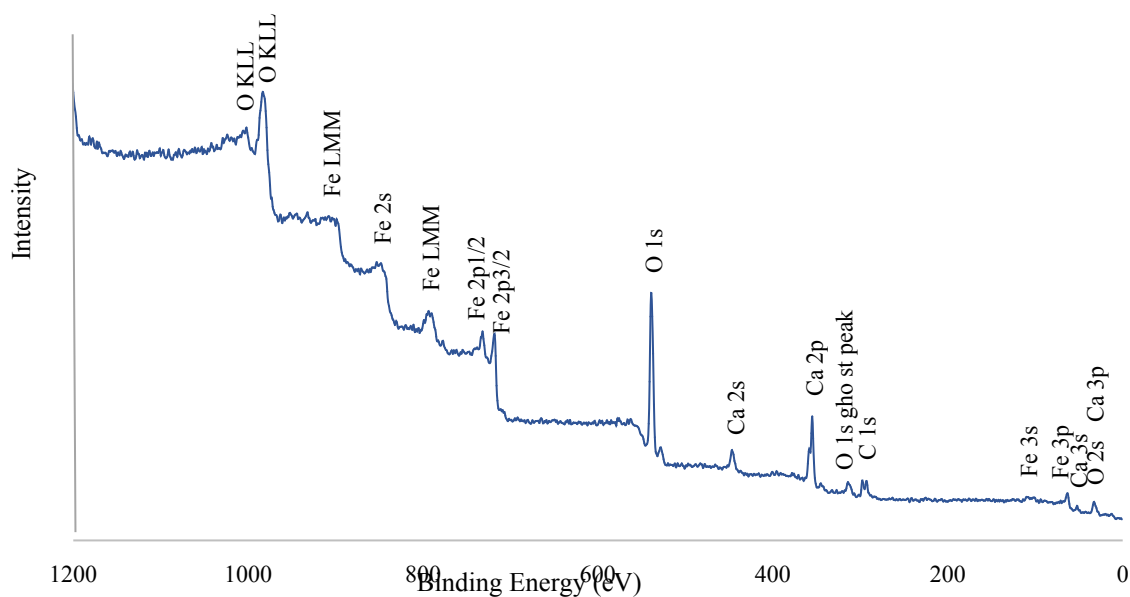


Fig. S5. XPS of FS-processed  $\text{Ca}_2\text{Fe}_2\text{O}_5$  porous and dense microspheres.