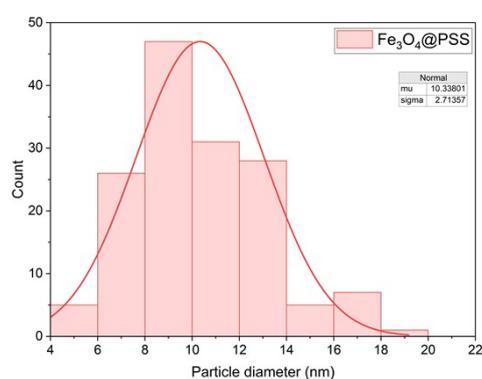


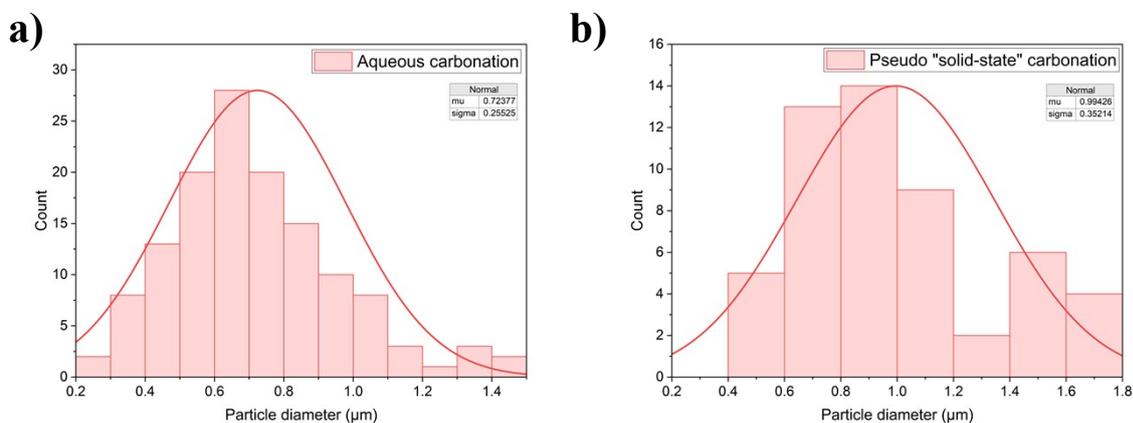
## Supporting Information

**Table S1.** Zeta potential of  $\text{Fe}_3\text{O}_4$  and  $\text{Fe}_3\text{O}_4@\text{PSS}$  data.

Sample	Mean Zeta Potential [mV]	Area [%]	Standard Deviation [mV]
$\text{Fe}_3\text{O}_4$	+6.87	100.0	14.6
$\text{Fe}_3\text{O}_4@\text{PSS}$	-14.6	100.0	11.6

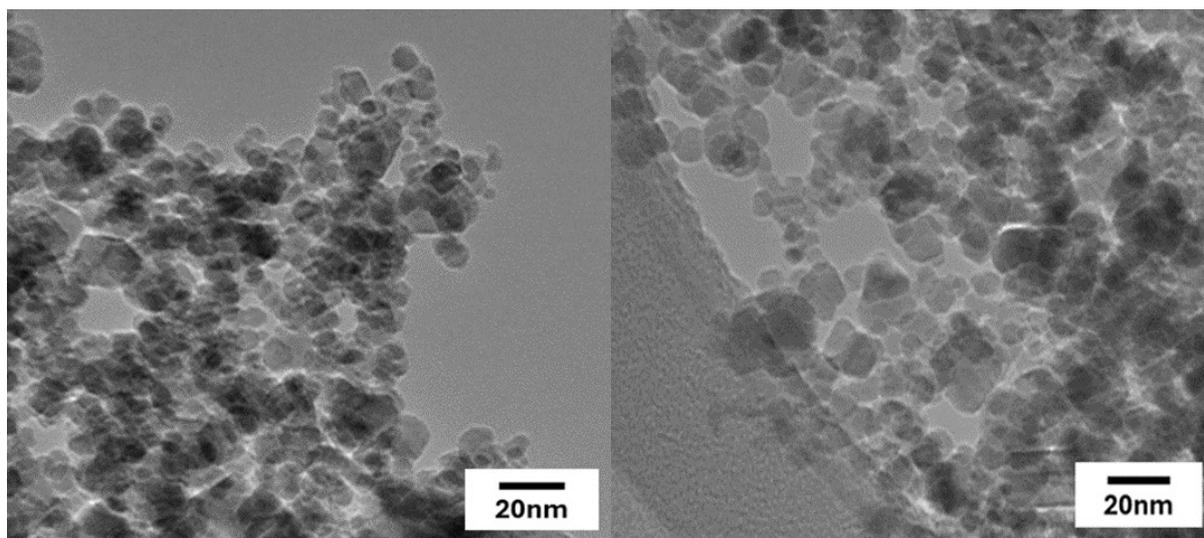


**Figure S1.** Graph of size distribution of  $\text{Fe}_3\text{O}_4@\text{PSS}$  MNPs from TEM images.

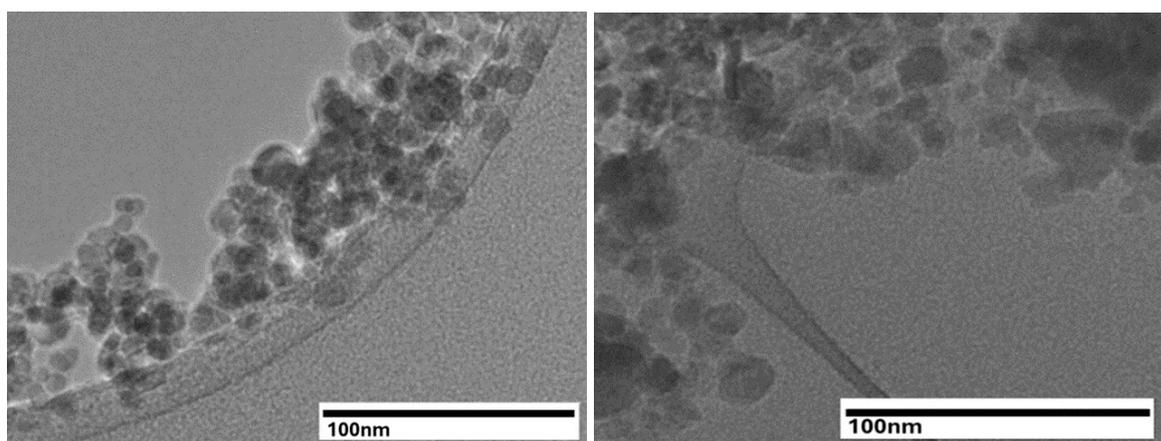


**Figure S2.** Graph of size distribution of  $\text{Fe}_3\text{O}_4@\text{PSS}@\text{CaCO}_3$  synthesised using a) aqueous and b) pseudo "solid-state" dry ice carbonation techniques from SEM images.

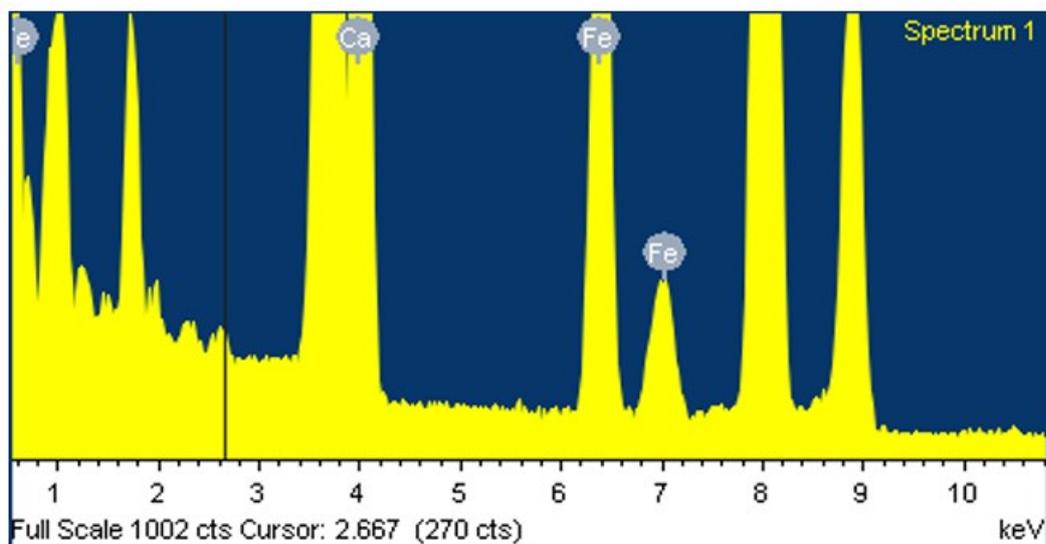
a)



b)



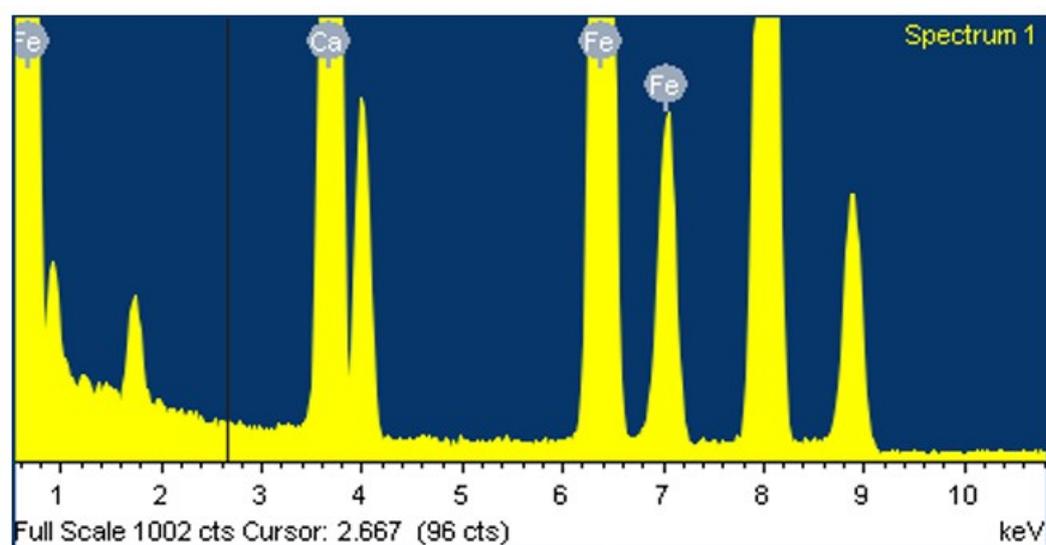
**Figure S3.** TEM images of Fe<sub>3</sub>O<sub>4</sub>@PSS@CaCO<sub>3</sub> synthesised using a) aqueous and b) pseudo solid-state dry ice-carbonation. (top two calcite rhombohedra using aqueous synthesis, bottom two star-like structures using solid state synthesis).



**Figure S4:** EDX analysis for particles using pseudo “solid-state” synthesis.

**Table S2:** EDX analysis for particles using pseudo “solid-state” synthesis.

Element	Weight %	Atomic %
C K	7.64	16.70
O K	24.26	39.81
Ca K	62.14	40.69
Fe K	5.96	2.80



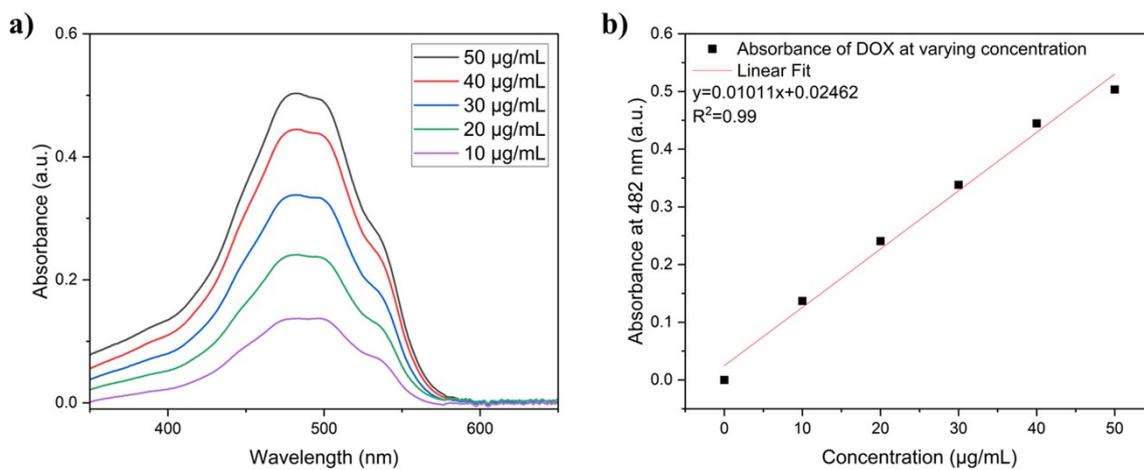
**Figure S5:** EDX analysis for particles using aqueous carbonation synthesis.

**Table S3:** EDX analysis for particles using aqueous carbonation synthesis.

Element	Weight %	Atomic %
C K	8.90	17.99
O K	35.25	53.49
Ca K	24.84	15.05
Fe K	31.01	13.48

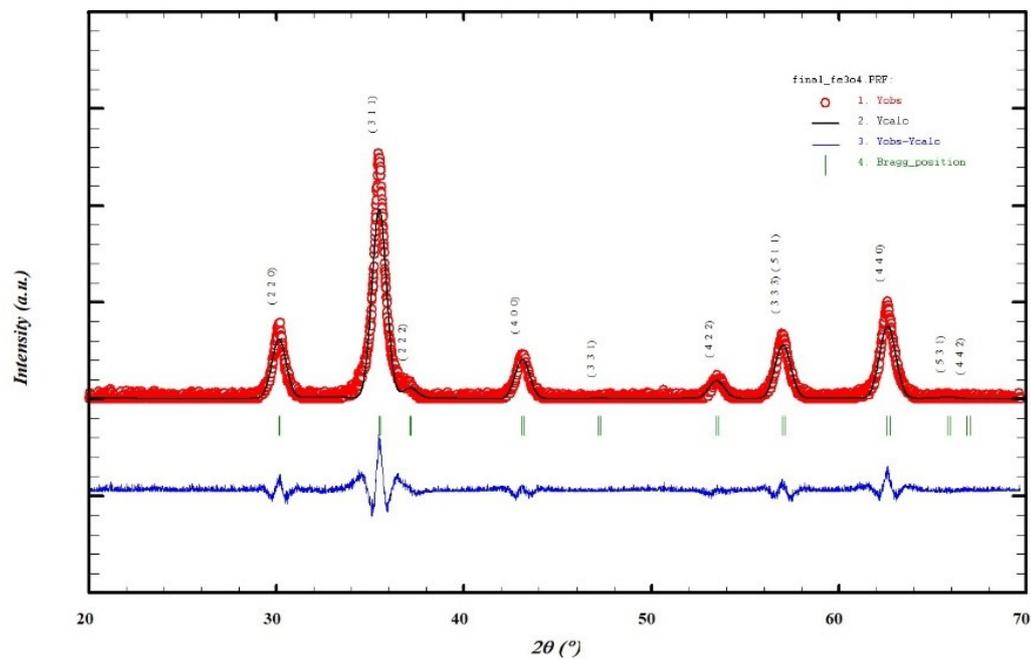
**Table S4:** Intruded volume and BET surface area for both synthetic methods

Synthetic route	Porosimetry intruded volume (cc/g)	BET surface area (m <sup>2</sup> /g)
Pseudo "solid state" carbonation	3.1	26
Aqueous carbonation	1.0	7



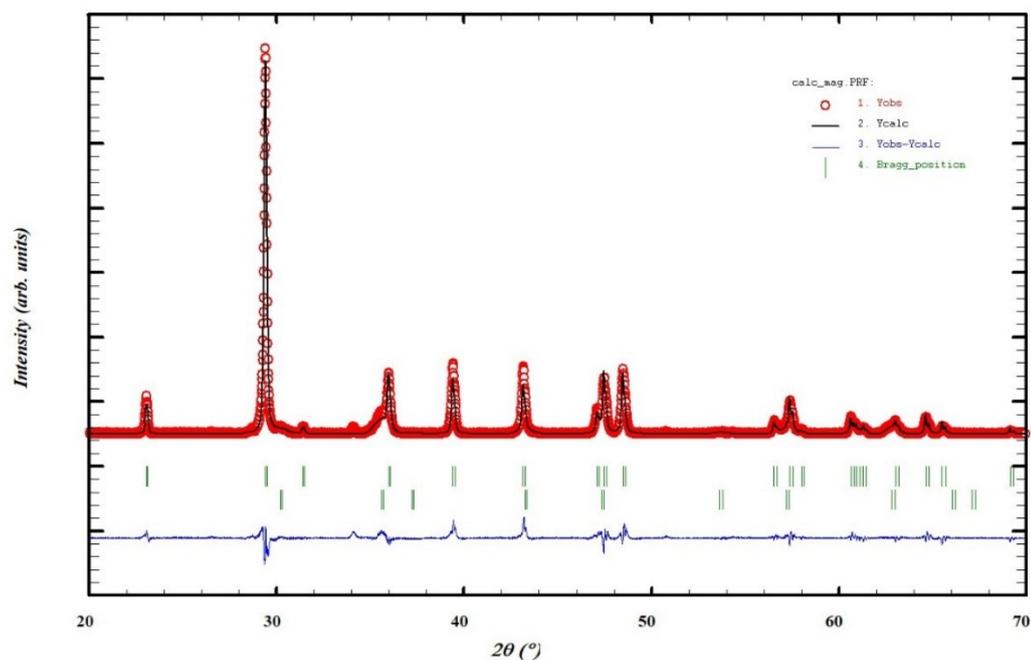
**Figure S6:** (a) UV-Vis spectra of DOX at varying concentration and (b) corresponding standard curve of DOX solution, calibrated at 482 nm.

### Rietveld Refinement of Fe<sub>3</sub>O<sub>4</sub>@PSS



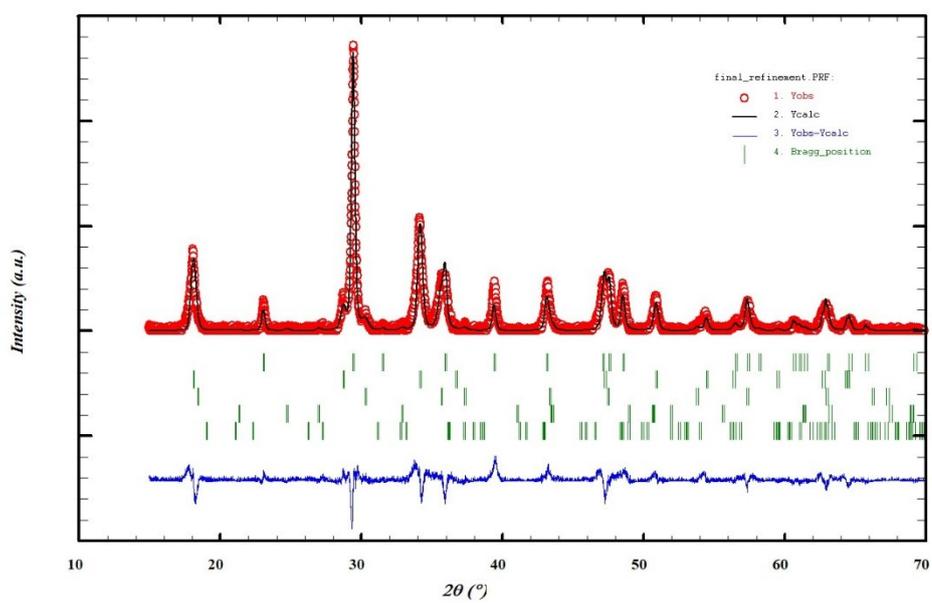
**Figure S7:** Rietveld refinement of Fe<sub>3</sub>O<sub>4</sub>@PSS MNPs.

### Rietveld Refinement for Aqueous Carbonation Approach



**Figure S8:** Rietveld refinement of Fe<sub>3</sub>O<sub>4</sub>@PSS@CaCO<sub>3</sub> sample synthesised using the aqueous carbonation approach.

### Rietveld Refinement of Pseudo "Solid-State" Approach



**Figure S9:** Rietveld refinement of  $\text{Fe}_3\text{O}_4@PSS@CaCO_3$  sample synthesised using the solvent-free carbonation approach.