Solvothermal synthesis of Co$_x$Fe$_{3-x}$O$_4$ spheres and their microwave absorption properties

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Figure S1. The EDS spectra of different Co$_x$Fe$_{3-x}$O$_4$ spheres fabricated at 200°C, (a)-(d) and 300°C, (e). The atomic ratio of Co to Fe in original EG solution is 1:1 (a), 1:2 (b) and (e), 1:4(c) and 0 (d). The atomic ratio in spheres (molecular formula) is obtained from table 1. The remaining peaks in spectra are originated from sputtered Au or Pt film, which can improve the conductivity and emissivity of the specimen to get most informative SEM images.
Figure S2. XRD data of spheres C and D (spheres fabricated at 300°C, and post-treated with calcination at 700°C for two hours in air, similarly with sphere C) were acquired by X’ Pert Pro MPD multi-purpose X-ray diffraction system operated at a step size equal to 0.01° and time per step of 0.3 s. To compare the diffractograms, the same mass of powder is used in measurements and the phase analysis is conducted through software JADE 5. The inset shows the whole patterns in 2θ range of 0° - 80° and the main curve highlights two special peaks. The peak centered at 33.3° is the strongest one of hematite, whose area (counts) is 1551 and 1884 for sphere C and D separately. The peak centered at 35.655° actually can be divided into two parts which are the second strongest one of hematite and the strongest of cobalt ferrite respectively. The area (counts) is 6125 for sphere C and 4788 for sphere D. This denotes more hematite ingredient in sphere D than C. The hematite comes from the oxidization of ferrous ion. So there are more ferrous ions in sphere B than A. The results coincide with EDS analysis in table 1 for Figure S1 (b) and (c).

Figure S3. XRD patterns of Co\textsubscript{x}Fe\textsubscript{3-x}O\textsubscript{4} spheres synthesized at 200°C (CoFe\textsubscript{2}O\textsubscript{4}, Co\textsubscript{0.9}Fe\textsubscript{2.1}O\textsubscript{4} (sphere A), Co\textsubscript{0.73}Fe\textsubscript{2.27}O\textsubscript{4} and Fe\textsubscript{3}O\textsubscript{4}). The crystallinity is enhanced with decreased cobalt ratio.
Figure S4. FESEM images of \( \text{Co}_{x}\text{Fe}_{3-x}\text{O}_4 \) spheres synthesized by solvothermal method at 200°C temperatures: (a) and (b), \( \text{CoFe}_2\text{O}_4 \); (c) and (d), \( \text{Co}_{0.73}\text{Fe}_{2.27}\text{O}_4 \); (e) and (f), \( \text{Fe}_3\text{O}_4 \).

Figure S5. Magnetization hysteresis loops of different \( \text{Co}_{x}\text{Fe}_{3-x}\text{O}_4 \) spheres synthesized by solvothermal method at 200°C (\( \text{CoFe}_2\text{O}_4 \), \( \text{Co}_{0.9}\text{Fe}_{2.1}\text{O}_4 \) (sphere A), \( \text{Co}_{0.73}\text{Fe}_{2.27}\text{O}_4 \) and \( \text{Fe}_3\text{O}_4 \)).
Figure S6. Permittivity (a) and permeability (b) of composites containing different Co$_x$Fe$_{3-x}$O$_4$ spheres (CoFe$_2$O$_4$, Co$_{0.73}$Fe$_{2.27}$O$_4$ and Fe$_3$O$_4$, 75% weight ratio) and paraffin wax.