

Supplementary Information for

## Engineered Contrast Agents in a Single Structure for $T_1$ - $T_2$ Dual Magnetic Resonance Imaging

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## 1. Materials characterization

**Table S1** Physical and chemical properties of Gd-Fe containing contrast agents prepared in present work.<sup>a</sup>

Sample	Molecular Formula	Composition <sup>b</sup>			Gd:Fe <sup>b</sup> (at)	<i>d</i> TEM <sup>c</sup> (nm)	<i>d</i> DLS (nm) <sup>d</sup>		Morphology <sup>c</sup> (crosses:rods)	Area BET (m <sup>2</sup> g <sup>-1</sup> )	Z-Potential (mV)
		Gd (%)	Fe (%)	Si (%)			Water	PBS			
<b>GF-1</b>	Gd(H <sub>2</sub> O) <sub>4</sub> [Fe(CN) <sub>6</sub> ]	47.2	52.8	0	0.89	94.3±35.5	<i>nd</i>	<i>nd</i>	1:99	<i>nd</i>	<i>nd</i>
<b>GF-2</b>	Gd(H <sub>2</sub> O) <sub>4</sub> [Fe(CN) <sub>6</sub> ]	50.9	49.1	0	1.04	262.1±43.5	<i>nd</i>	<i>nd</i>	36:64	<i>nd</i>	<i>nd</i>
<b>GFS-1</b>	Gd(H <sub>2</sub> O) <sub>4</sub> [Fe(CN) <sub>6</sub> ]@SiO <sub>2</sub>	32.2	35.3	32.4	0.91	106.7±33.5	124.7±81.9/0.431	492.7±71.4/0.786	3:97	61.4	-12.3
<b>GFS-2</b>	Gd(H <sub>2</sub> O) <sub>4</sub> [Fe(CN) <sub>6</sub> ]@SiO <sub>2</sub>	39.9	41.1	19.0	1.28	230.3±35.5	343.7±154.1/0.201	806.8±231.6/0.620	32:68	57.6	-15.5
<b>GFS-1@PEG</b>	Gd(H <sub>2</sub> O) <sub>4</sub> [Fe(CN) <sub>6</sub> ]@SiO <sub>2</sub> @PEG <sub>3</sub>	34.5	36.4	29.1	1.20	<i>nd</i>	111.4±62.7/0.317	299.3±58.9/0.328	3:97	4.8	-8.7
<b>GFS-2@PEG</b>	Gd(H <sub>2</sub> O) <sub>4</sub> [Fe(CN) <sub>6</sub> ]@SiO <sub>2</sub> @PEG <sub>3</sub>	40.8	41.4	17.9	1.26	<i>nd</i>	228.9±79.3/0.120	600.3±147.4/0.357	32:68	27.7	-10.7

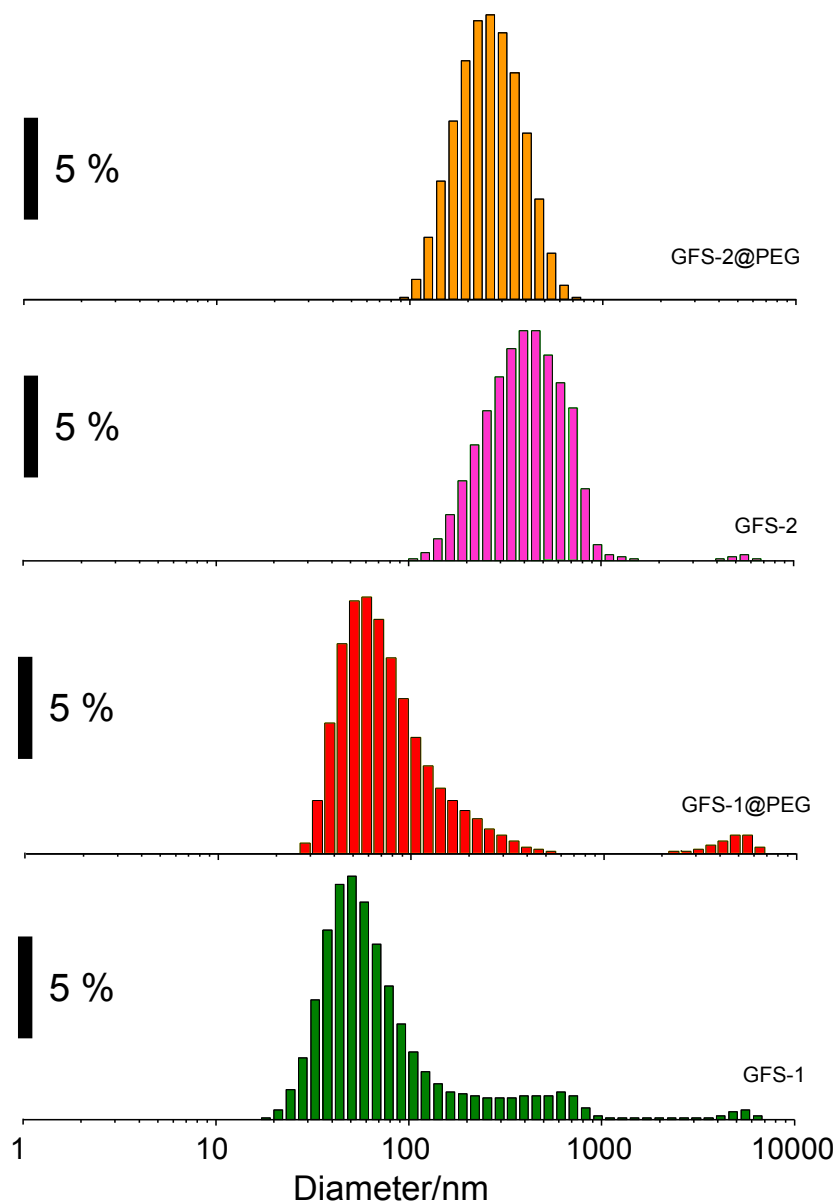
<sup>a</sup> *nd* = not determined. GF samples are fully soluble in aqueous medium (unable DLS and Z-Potential determinations), and show no external porosity (no N<sub>2</sub> adsorption). Also, PEGylated GFS materials present some blurry TEM images, making unfeasible to obtain accurate particle diameter measurements.

<sup>b</sup> As determined from the EDS study.

<sup>c</sup> TEM particle diameter (Avg ± SD). As determined by TEM measurement of at least 250 particles.

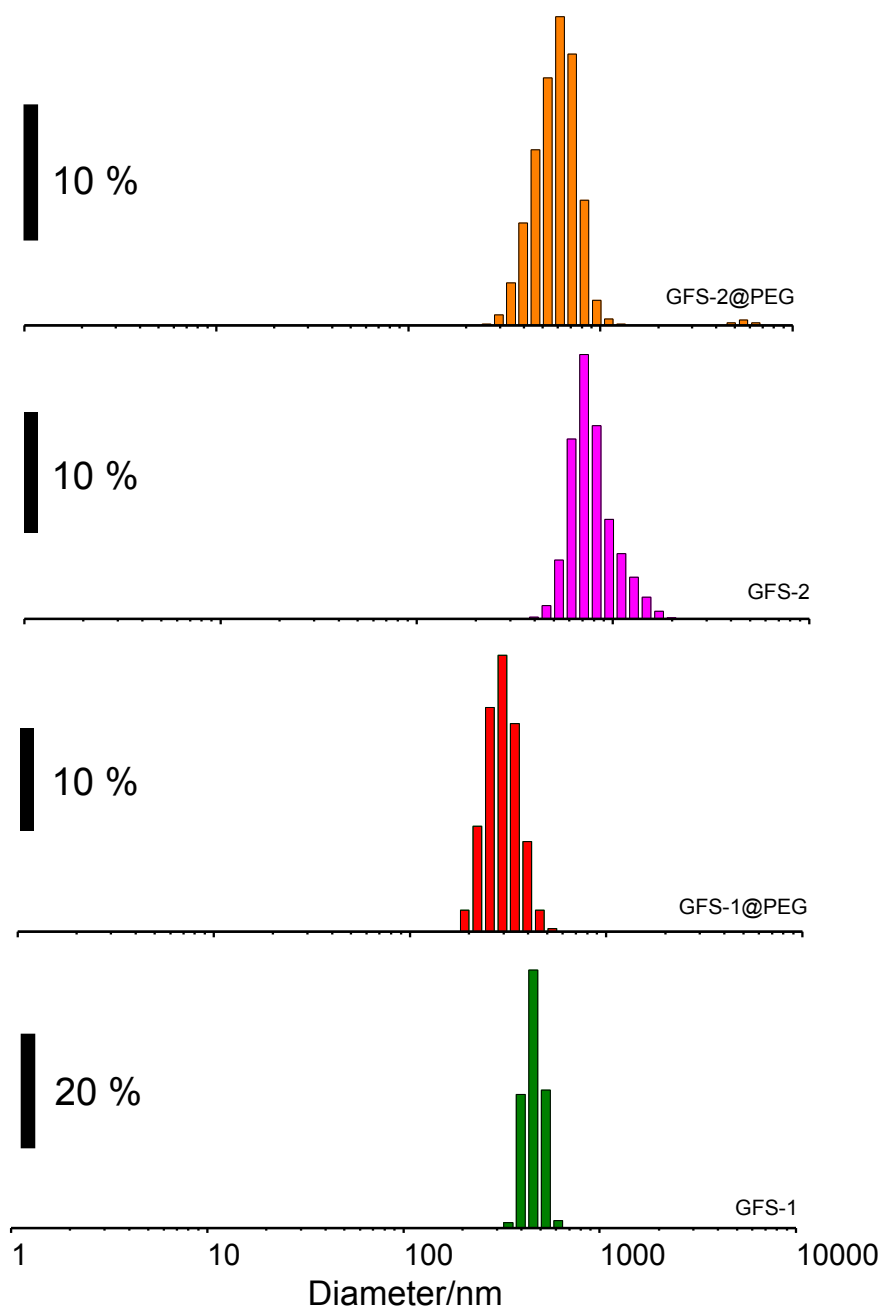
<sup>d</sup> DLS particle diameter (Avg ± SD)/Polydispersity Index. As determined by DLS.

## 1.1. Dynamic Light Scattering (DLS)



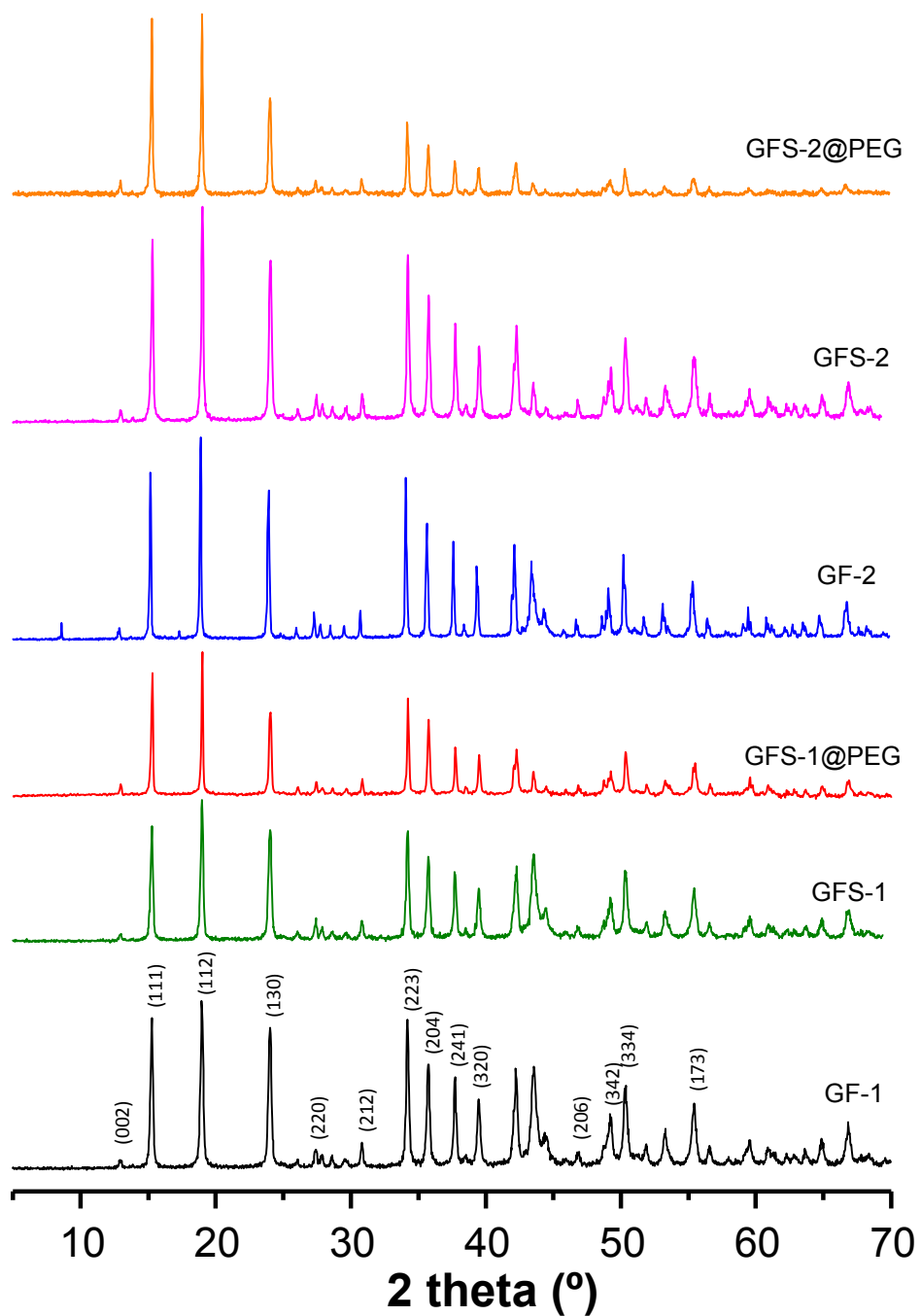
**Fig. S1a.** Particle hydrodynamic diameter of as-synthesized materials as determined in water by DLS (volume output).

### 1.1. Dynamic Light Scattering (DLS) (cont.).



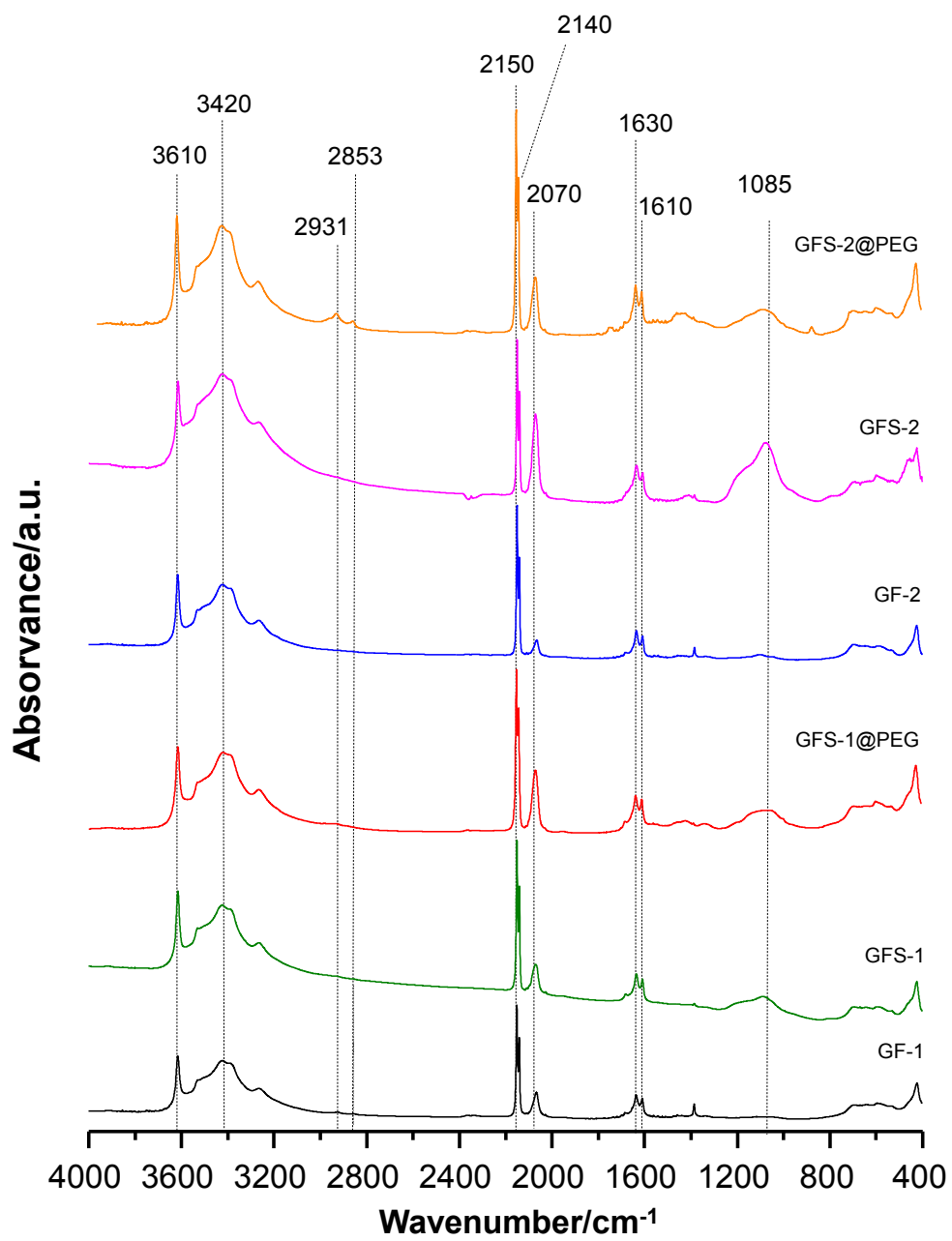
**Fig. S1b.** Particle hydrodynamic diameter of as-synthesized materials as determined in PBS by DLS (volume output).

## 1.2. Powder X-Ray diffraction



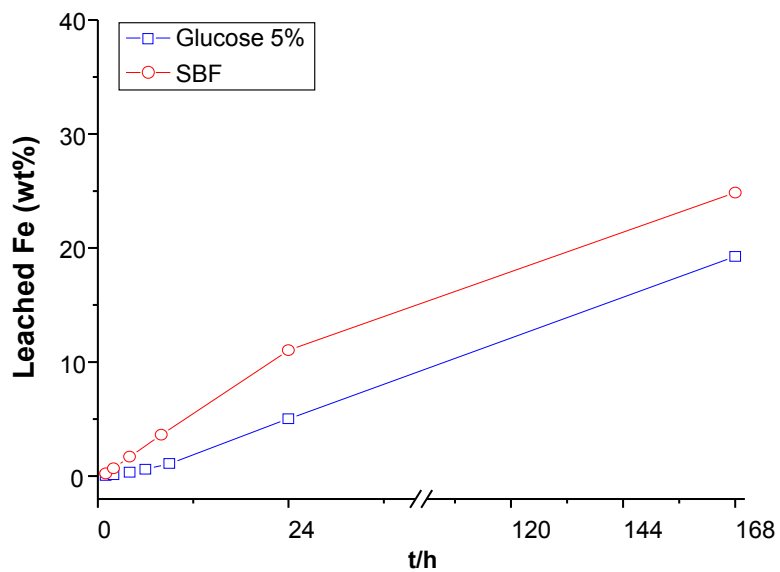
**Fig. S2.** Powder XRD patterns of as-made materials.

### 1.3. FTIR Spectroscopy



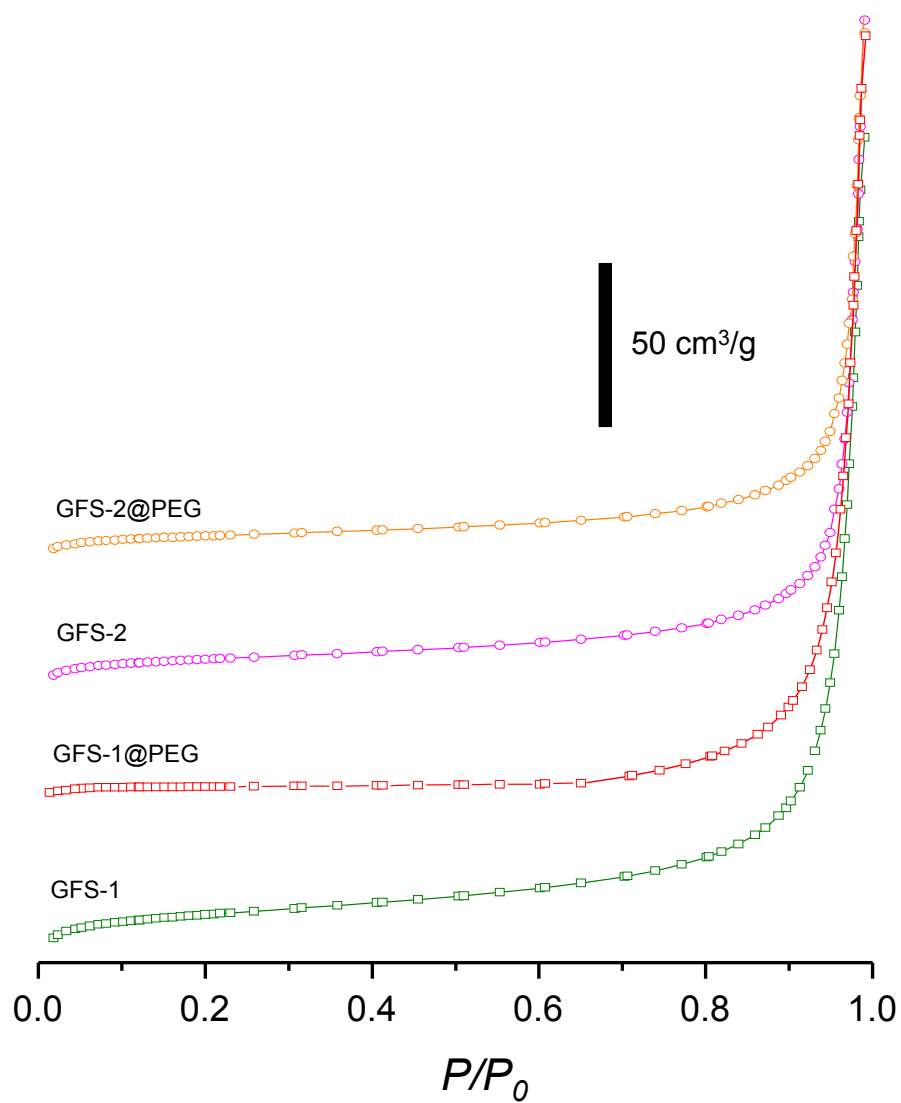
**Fig. S3.** FTIR spectra of as-synthesized materials.

#### 1.4. Stability in isotonic solution.



**Fig. S4.** Representation of leached metal cation ( $\text{Fe}^{3+}$ ) by GFS-1@PEG sample after incubation at 37 °C in glucose 5% solution (as determined by ICP) or SBF (as determined by ICP/MS).

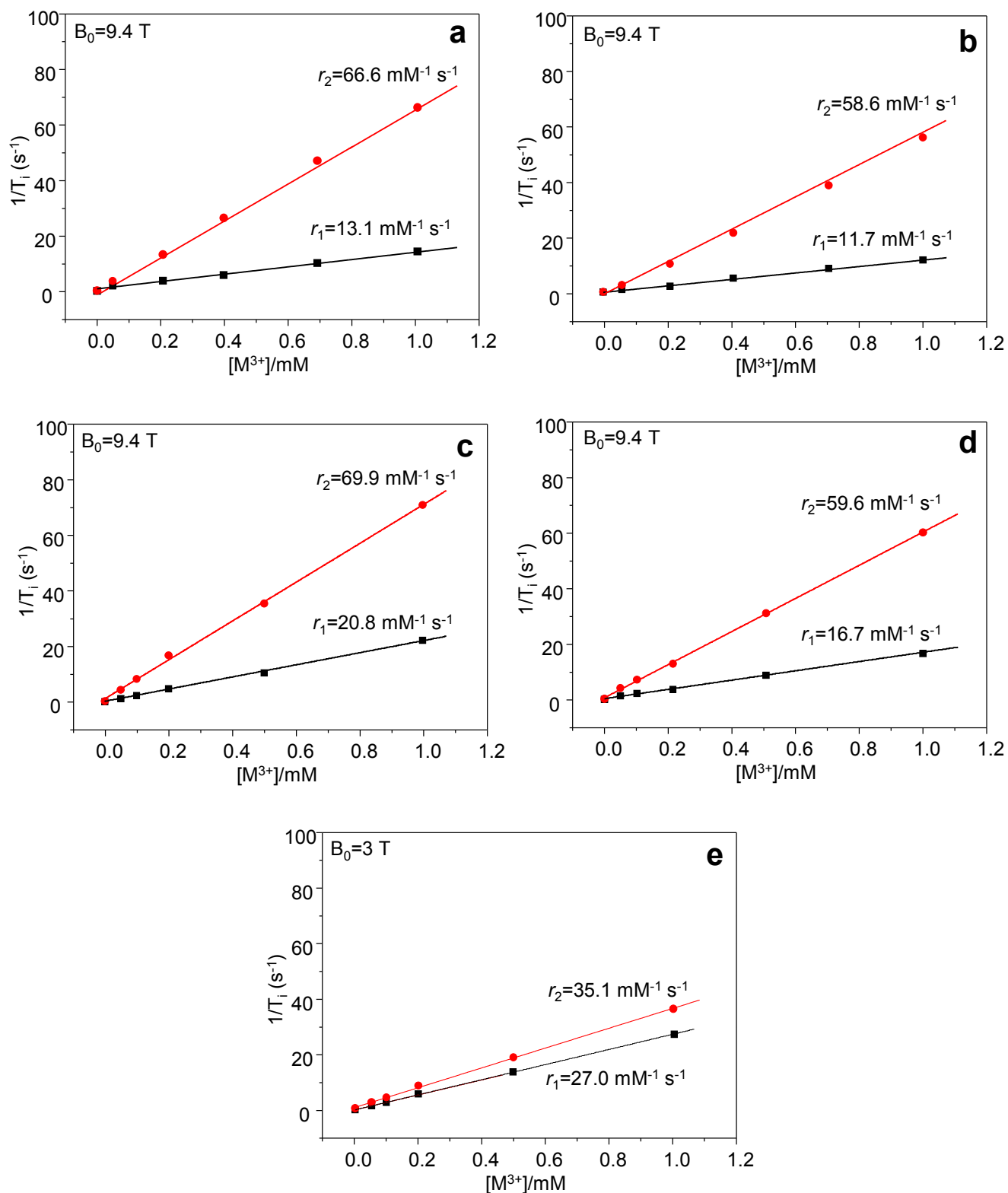
## 1.5. Nitrogen Adsorption Isotherms



**Fig. S5.** BET nitrogen adsorption isotherms of as-prepared materials.

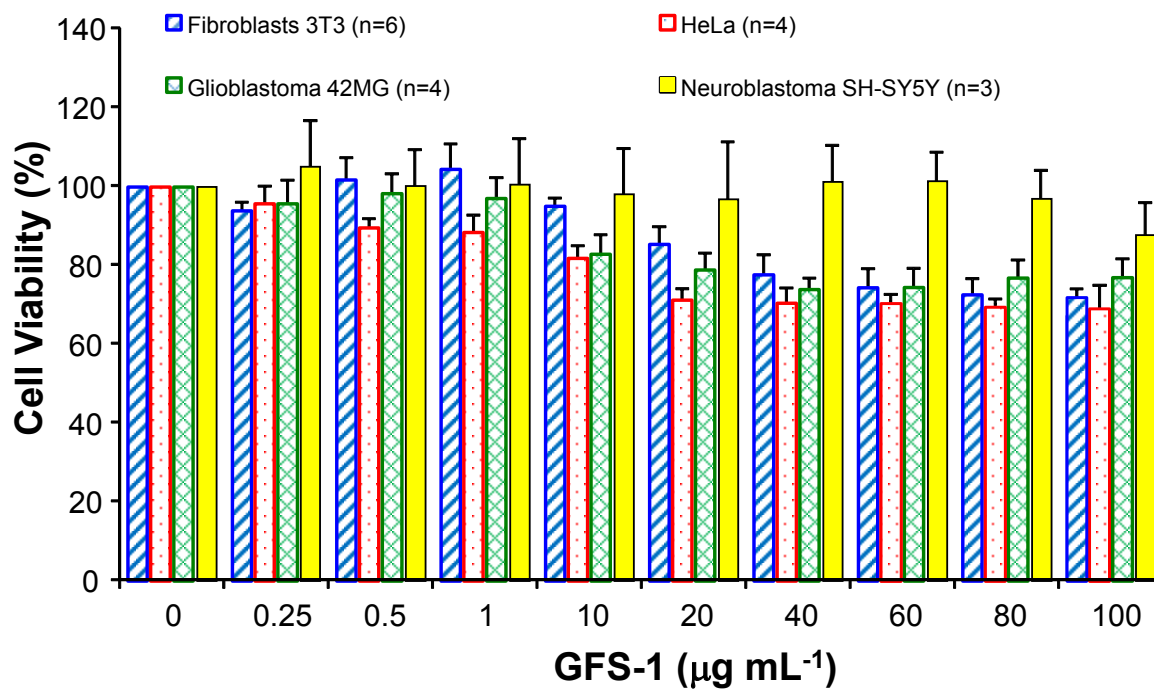


## 2. Relaxivity Measurements and *In Vitro* MRI



**Fig. S6.**  $T_1$  (black dots) and  $T_2$  (red dots) relaxation rate measurements vs concentration ( $M=Gd$  for  $T_1$ ,  $Fe$  for  $T_2$ ) of GFS aqueous solutions (0.1 % xanthan gum) at high (a-d, 9.4 T) and medium (e, 3 T) magnetic field. (a) GFS-1. (b) GFS-2. (c) GFS-1@PEG. (d) GFS-2@PEG. (e) GFS-1@PEG.

### 3. Cytotoxicity Study



**Fig. S7.** *In vitro* MTT cell viability assays in different cell lines incubated with variable concentrations of GFS-1 magnetic nanorods (mean±SEM). *n*=number of experiments.