**Supplementary Data**

**Table S1:** Compositional results (ICP-MS) of Al(OH)3 coated NPs without treatment.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Samples**  (***label***) | **Precursors ratio**  MFe2O4 to AlCl3  (M = Mn or Fe) | **Elements to detect (mmo/L)** | | | **Core-shell ratio**  MFe2O4 to Al(OH)3  (M = Mn or Fe) |
| Fe | Mn | Al |
| MnFe2O4@Al(OH)3  (***1***) | 1: 3 | 10.29 | 0.98 | 26.79 | 1: 7.3 |
| Fe3O4@Al(OH)3 (1:1)  (***2***) | 1: 1 | 7.79 | n/a | 3.05 | 1: 1.2 |
| Fe3O4@Al(OH)3 (1:2)  (***3***) | 1: 2 | 7.92 | n/a | 8.22 | 1 : 3.1 |
| Fe3O4@Al(OH)3 (1:3)  (***4***) | 1: 3 | 7.98 | n/a | 11.41 | 1 : 4.3 |

**Table S2.** DLS results for alumina coated samples before and after filtration

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Sample** | **Dh / nm** | | **Zeta potential / Mv** | | ***r1*** **/ mM-1s-1** | | ***r2*** **/ mM-1s-1** | |
| **Before** | **After** | **Before** | **After** | **before** | **after** | **before** | **After** |
| ***1*** | 21.0 | 50.7 | +72.3 | +55.9 | 1.47 | 0.65 | 21.4 | 18.0 |
| ***2*** | 18.2 | 50.8 | +70 | +52.5 | 1.65 | 5.36 | 60.5 | 116.6 |
| ***3*** | 21.0 | 49.8 | +49.8 | +38.9 | 3.54 | 3.7 | 81.6 | 121.9 |
| ***4*** | 396.1 | 458.0 | +27.0 | +18.4 | n/a | n/a | n/a | n/a |

**Table S3.** ICP-MS analysis of Al and Fe in pre-wash NPs colloids and the supernatant after wash.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
|  | | **Pre-wash**  **NPs colloid** | **1st wash**  **supernatant** | **2nd wash**  **supernatant** | **3rd wash**  **Supernatant** |
| ***2*** | [Fe] mM | 7.79 | Not detected | Not detected | Not detected |
| [Al] mM | 3.05 | Not detected | Not detected | Not detected |
| ***3*** | [Fe] mM | 7.92 | 0.13 | Not detected | Not detected |
| [Al] mM | 8.22 | 2.96 | 0.07 | Not detected |
| ***4*** | [Fe] mM | 7.98 | 0.44 | 0.01 | Not detected |
| [Al] mM | 11.04 | 5.55 | 0.15 | Not detected |

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**Figure S1.** IR spectrum evolution of Fe3O4@Al(OH)3 samples: **a)** as-synthesised Fe3O4 sample, **b)** NP ***2***, **c)** NP ***3***, and **d)** NP ***4***

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**Figure S2.** TEM images of Fe3O4 NPs and Fe3O4@Al(OH)3 NPs. **a)** Fe3O4 NPs prepared from hexane solution, **b)** NPs ***4*** prepared from aqueous solution, **c)** NPs ***3*** prepared from aqueous solution, and **d)** NPs ***2*** prepared from aqueous solution.



**Figure S3.** XRD pattern of samples: **a)** NP Fe3O4 and **b)** NP ***2***

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**Figure S4.** XPS spectrum of sample ***4***. **a)** Al2p level data, **b)** O1s level data, **c)** Fe2p level data, and **d)** full scan of XPS spectra.

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**Figure S5.** XPS spectra comparison of Fe3O4@Al(OH)3 samples (***2***- ***4***) with different core-shell ratio: **a)** NP ***2*,** 1:1; **b)** NP ***3***, 1:2; and **c)** NP ***4***, 1:3.

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**Figure S6.** Adsorption of non-radioactive 19F- by 5 mg ***1*** NPs in 5 ml NaF solution of different conditions, monitored by fluoride ion selective electrode.

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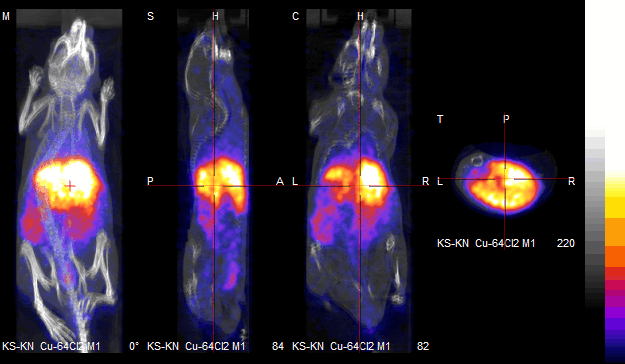
**Figure S7.** 18F-fluoride radio labelling of Fe3O4@Al(OH)3 NPs (***2***-***4***) varying the core-shell ratio. Pre-wash results showed that all three samples exhibited a low labelling efficiency, especially ***3*** and ***4***. A much higher radiolabelling efficiency were achieved after removal of unstable Al(OH)3 layer by washing with water 1 or 2 times.

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**Figure S8.** *In vivo*PET/MRI images of a normal young C57BL/6 mouse using 18F radiolabelled ***3***: **(a)** whole body PET image showing distribution of 18F 30 minutes post injection (maximum intensity projection, mice in prone position); **(b)** PET/MRI fused image (coronal section, 0-15 minutes); **(c)** PET/MRI fused image (coronal section, 105-120 minutes); **(d)** MR image prior to the injection of NPs, and **(e)** MR image post the injection of NPs, showing a darkening contrast at lung and live area. Due to the unstable Al(OH)3 shell, 18F-fluoride radioactivity was released from magnetic NPs ***3*** within 15 minutes and localised in bone.

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**Figure S9.** *In vivo*PET/CT images of a normal young C57BL/6 mouse using 18F radiolabelled ***1***: **a)** 0-15 minutes; **b)** 15-30 minutes; **c)** 30-45 minutes; **d)** 45-60 minutes; **e)** 60-75 minutes; **f)** 75-90 minutes; **g)** 90-105 minutes;and **h)** 105-120 minutes. NPs ***1*** obtained by a slow hydrolysis process has a better in vivo stability than NP ***3*** synthesised by a quick hydrolysis (Fig. S8), which is in consistent with the in vitro studies.



**Figure S10.** *In vivo*PET/CT images of a normal young C57BL/6 mouse using 64CuCl2 solution (0-30 minutes).

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**Figure S11.** DLS size distribution of NPs: **a)** Fe3O4 NPs in hexane; **b)** pre-washed NPs ***3*** in water; **c)** post-washed NPs ***3*** in water; and **d)** conjugates of post washed NPs ***3*** and BP-PEG (10K Da). [***3***] = [Fe3O4] ≈ 1 mg/L. Zeta potential was measured in neutral aqueous solution with a pH value ≈ 7.