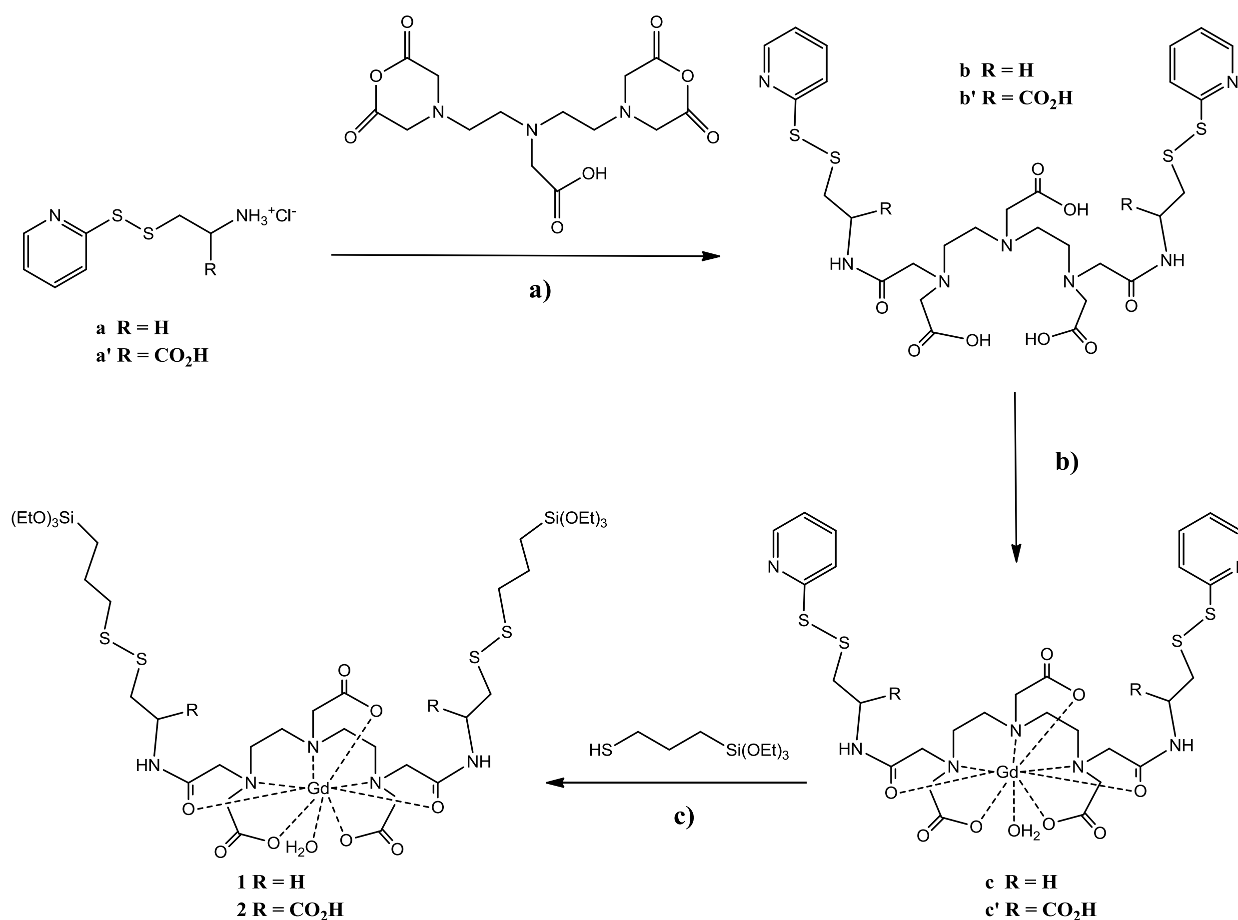
Materials and Methods. All reagents were purchased from Sigma Aldrich and used without further purification, except for 3-mercaptopropyltriethoxysilane (MP-TES), which were purchased from Gelest. Thermogravimetric analysis (TGA) was carried out with a Shimadzu TGA-50 equipped with a platinum pan using a heating rate of 5 ℃/min under air. A Hitachi 4700 Field Emission Scanning Electron Microscope (SEM) was used to determine particle size and morphology. A Cressington 108 Auto Sputter Coater equipped with an Au/Pd (80/20) target and MTM-10 thickness monitor was used to coat the sample with a thin conductive layer (~3 nm) before SEM imaging. Each SEM sample was prepared by suspending the nanoparticles in ethanol. A drop of the suspension was placed on a glass slide, and the solvent was allowed to evaporate. Gd^{3+} ion concentration was measured using a Varian 820-MS inductively-coupled plasma mass spectrometer. Dynamic light scattering (DLS) and zeta potential measurements were carried out using a Malvern Instruments Zetasizer Nano. MR images were acquired on a Siemens 3T Biograph mMR scanner (Siemens Medical Systems, Erlagen, Germany) with an mMR Head/Neck coil. A 3D FLASH sequence was utilized to compute T_1 maps with seven different flip angles (2, 5, 10, 20, 30, 40, and 60). Imaging parameters were: FOV=190 x 190 x 64 mm^3, matrix size= 128 x 128 x 32, TR/TE= 4.0/1.41 ms; total data acquisition time was less than 5 minutes. A 2-D multiple echo spin echo sequence was used to estimate T_2 maps. In total, 32 echoes with an echo spacing of 7.5 ms were obtained. The first echo time was 7.5 ms. TR was 3000 ms. FOV and matrix size were set to 190 x 190 mm^2 and 128 x 128. The slice thickness was 2 mm, and the total data acquisition time was about 4 min.

Cell Lines: H460 human non-small lung cancer cells (ATC# HTB-177) were purchased from the Tissue Culture Facility of the Lineberger Comprehensive Cancer Center at the University of North Carolina at Chapel Hill. AsPC-1 human pancreatic adenocarcinoma cells (ATCC# CRL-1682) were received from the laboratory of Jen Jen Yeh at the University of North Carolina at Chapel Hill. Both cell lines were maintained at 37 °C with 5% of CO_2

and cultured in RPMI-1640 medium (Cellgro) with 10 % fetal bovine serum (Sigma) and 2 % penicillin/streptomycin (Sigma) according to ATCC recommendations. Murine monocyte cells (ATCC# TIB67) were purchased from the UNC Lineberger Comprehensive Cancer Center's Tissue Culture Facility. Cells were maintained in DMEM-H containing 10% FBS and 1% penicillin/streptomycin at 37 ℃ and 5% CO2.

Competitive binding assay. Confluent H-460 or AsPC-1 cells were trypsinized, and an aliquot of cell suspension was added to 7 mL RPMI-1640 media (Cellgro) in each 25 mL culture flask to obtain a cell density of 1.0 x 10^6 cells/flask. The flasks (five total) were incubated (37 °C, 5% CO_2) for 36 hours. To test the specific binding ability of the AA-PEG-PSQ-2 material, four of the cell culture flasks were then incubated in 5 mL fresh media containing the anisamide ligand (1 (two flasks), 5 and 10 mM) for 45 min. After that, the media was then removed and replaced with 5 mL fresh media containing either AA-PEG-PSQ-2 or PEG-PSQ-2 in a concentration of 100 μg/mL. The same procedure was followed for the cell culture flask (AA-PEG-PSQ-2) that were not incubated in the presence of anisamide ligand. The flasks were incubated (37 °C, 5 % CO_2) for 12 hours; cells were trypsinized, and centrifuged at 3,000 rpm for 15 minutes to obtain cell pellets. The amount of Gd^{3+} uptaken by AsPC-1 cells was determined by ICP-MS as described in the Experimental Section.



Scheme S1. Complexes **1** and **2** were synthesized by a multistep procedure. First, compound **a**/**a’** was obtained by a thiol displacement reaction with 2,2’-dipyridyl disulfide. **a)** The nucleophilic acyl addition of compound **a**/**a’** toward DTPA bis-anhydride affords compound **b**/**b’**. **b)** The metallation of compound **b**/**b’** with $GdCl_3$ under basic conditions produces complex **c**/**c’**. **c)** A thiol displacement reaction of complex **c**/**c’** with mercaptopropyl triethoxysilane affords complex **1**/**2**.

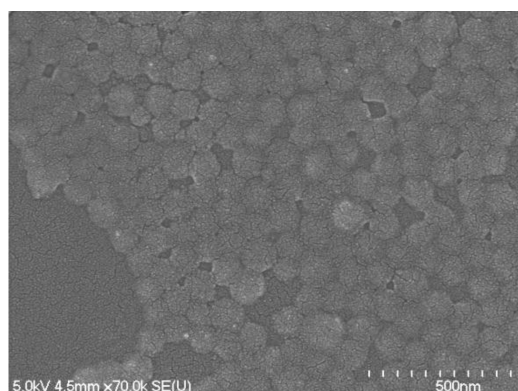


Figure S1. SEM image of PEG-PSQ-**2** nanoparticles.

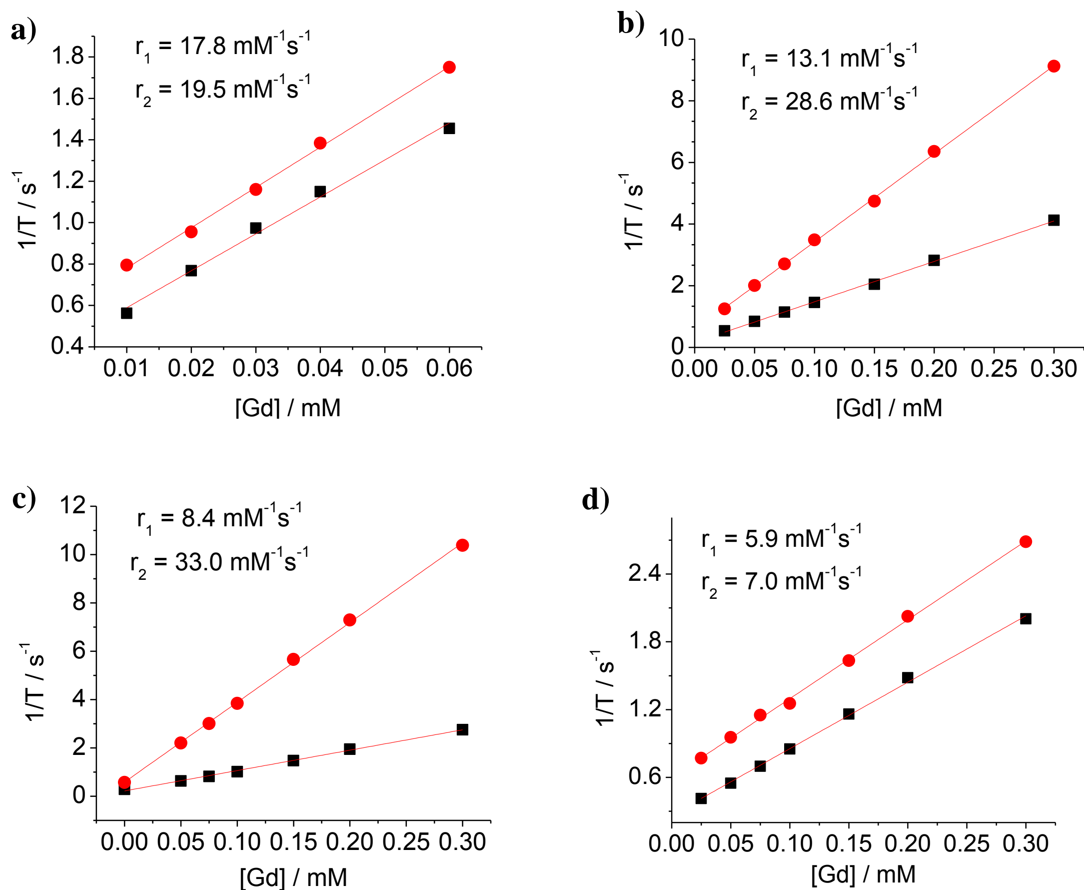


Figure S2. Longitudinal (r_1, squares) and transverse (r_2, circles) MR relaxivity plots for **a)** PSQ-**1**, **b)** PSQ-**2**, **c)** PEG-PSQ-**2**, and **d)** AA-PEG-PSQ-**2**.

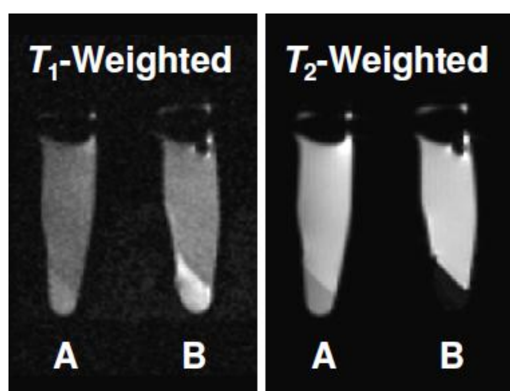
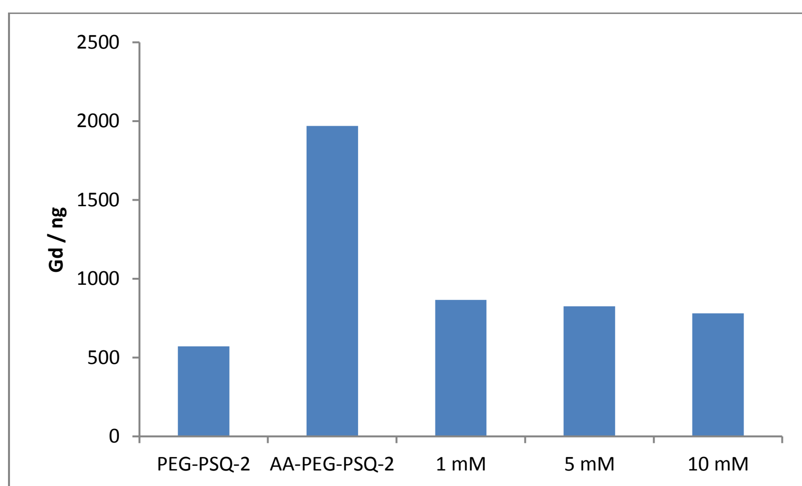


Figure S3. T_1- and T_2-weighted images of monocytes incubated with no nanoparticles (A) and PSQ-**1** nanoparticles (B).



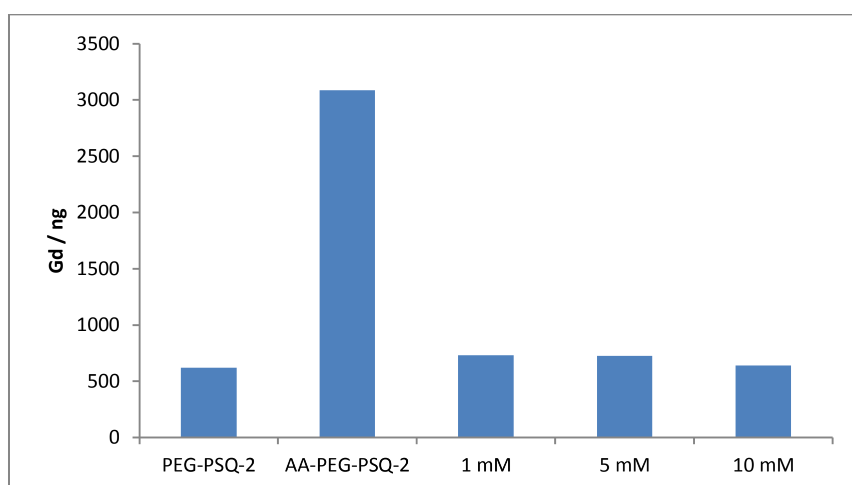


Figure S4. Anisamide competitive binding assays. The amounts of Gd^{3+} uptaken by H-460 (top) and AsPC-1 (bottom) cells were determined by ICP-MS. The graph shows the effects of the anisamide ligand on the uptake of AA-PEG-PSQ-2 as an indication that the internalization of this material is partially due to a receptor-specific endocytosis pathway. The first bar shows the internalization of PEG-PSQ-2 material in presence of 1 mM anisamide ligand. The second bar shows the internalization of AA-PEG-PSQ-2 material in the absence of anisamide ligand. The third, fourth, and fifth bars show the internalization of AA-PEG-PSQ-2 material by the cells that had been pre-incubated with the anisamide ligand at different concentrations (1, 5, and 10 mM, respectively).