1 Materials and Methods

1.1 Peptide-Peptoid Synthesis

Rink amide MBHA resin, HBTU, Oxymapure, and Fmoc-amino acids were obtained from Anaspec Corporation (Fremont, CA). DMF, DCM, and NMP were obtained from VWR International. DMF was dried over activated alumina column prior to use. \(N,N'\)-
Diisopropylethlyamine (DIEA), acetic anhydride, bromoacetic acid, and propargyl amine were obtained from Sigma-Aldrich. Unless otherwise noted, all other reagents were obtained from Sigma and used without further purification.

Peptoid linker synthesis was conducted using the procedure of Zuckerman with the following modifications. The synthesis was conducted at a 0.5 mmol scale in a fritted shaker vessel. The resin was deprotected with piperidine (25 mL, 30% in DMF) for 10 minutes. A solution of bromoacetic acid (10 mL, 0.6 M) in dry DMF and 1.05 mL of diisopropylcarbodiimde, was allowed to react for 1 minute and was added to the resin. After 30 minutes, a second addition of bromoacetic acid was performed and the solution shaken for an additional 45 minutes. When ninhydrin confirmed successful addition, the resin was washed with DMF and DCM and a solution of propargylamine (5 mL, 1.2 M) was added to the resin and agitated overnight. Applying a few drops a solution of chloranil (1% in DMF) and solution of acetaldehyde (1% DMF) to a few of the resin beads produced a green color—indicating the presence of a secondary amine.

Coupling peptoid linker to amino acid was conducted using 7.9 equivalents of HBTU to 8 equivalents fmoc-amino acid (glycine or glutamic acid) and 12 equivalents of DIEA. All other amino acids were added using a CS Bio Co (Model CS136) peptide synthesizer. For amino acid coupling, 4 equivalents of amino acid, 4 equivalents of Oxymapure, and 4 equivalents of DIC were used. Between each coupling step, a solution of 10% acetic anhydride in DCM was added to cap unreacted amines.

Synthesis of the azide-functionalized macrocycle was conducted as previously described. To couple the macrocycle and peptide amphiphile, 10 mg of sodium ascorbate and 2
mg of Tris[(1-benzyl-1H-1,2,3-triazol-4-yl)methyl]amine TBTA was added to a round bottom flask. Two equivalents of azide to crude PA were added to the flask in minimum volumes of Millipore water and DMSO respectively. The reaction proceeded over 24 hours at 50 °C in a mixture of 6:5:1 DMSO/H₂O/DIEA. DIEA was removed by rotary evaporation and the DMSO/water mixture removed via lyophilization. The resulting orange-yellow powder was purified using reverse-phase HPLC in H₂O/MeCN with 0.1% NH₄OH in the mobile phase.

1.2 Quantification of Gd(III) Concentration Using Inductively Coupled Mass Spectrometry (ICP-MS)

All concentrations used in relaxivity determination were obtained using ICP-MS. Samples were digested using ACS reagent grade nitric acid (70%) for a least 2 hours at 70°C. Samples were diluted in filtered, deionized water. A standard containing the elements Bi, Ho, In, Li, Sc, Tb, and Y was added to a final concentration of 5 ng/mL (Inorganic Ventures, Christiansburg, VA, USA). Final nitric acid concentration was 3%. The instrument was calibrated using a serial dilution of Gd(III) standards (Inorganic Ventures, Christiansburg, VA, USA) with nitric acid and internal standard concentrations identical to the samples. Calibration was conducted with 1.000, 5.000, 10.00, 20.00, 50.00, 100.0, and 200.0 ng/mL Gd(III) standards.

For Gd(III) analysis, mouse leg muscles were excised and digested in 1mL of nitric acid per gram of tissue in Teflon tubes. A Milestone EthosEZ microwave digestion system (Shelton, CT, USA) was used to digest the samples, ramping to 120°C over 30 minutes, then holding at 120°C for an additional 30 minutes. Digest was diluted and measured using the same conditions as above.

All measurements used a Thermo X series II ICP-MS (Thermo Fisher Scientific, Waltham, MA, USA) operating with an ESI SC-2 autosampler (Omaha, NE, USA).
acquisition consisted of one survey scan, utilizing 10 sweeps, and 3 peak-jumping measurements of 100 sweeps each. The two most common isotopes of Gd, $^{157}$Gd and $^{158}$Gd, were measured. $^{115}$In and $^{165}$Ho were measured as internal standards.

1.3 Purification

Reverse-phase, preparative HPLC was employed to purify the conjugates. A Varian Prostar 210 HPLC system supplied a 2% to 100% ACN to water gradient to a Phenomenex Gemini C18 column (5 μm particle size). To each solution was added 0.1% NH$_4$OH to maintain peptide solubility. Fractions were analyzed for product content on an Agilent 6510 Q-TOF MS, then recombined. ACN was removed with rotary evaporation, and the water then removed via lyophilization. The white, fluffy powders were stored dry at -20°C.
Table S1: HPLC methods for the purification of each contrast agent PA.

| Method | Elution Time (min) | % Acetonitrile | Method | Elution Time (min) | % Acetonitrile | Method | Elution Time (min) | % Acetonitrile | Method | Elution Time (min) | % Acetonitrile |
|--------|--------------------|----------------|--------|--------------------|----------------|--------|--------------------|----------------|--------|--------------------|----------------|
| PA1    | 15                 | 1.25           | PA2    | 22                 | 2.25           | PA3    | 22.5               | 1.25           | PA4    | 42                 | 1.25           |
| PA1    | 38                 | 3.0            | PA2    | 70                 | 4.0            | PA3    | 10                 | 4.0            | PA4    | 42                 | 4.0            |
| PA1    | 42                 | 1.25           | PA2    | 71                 | 1.5            | PA3    | 17                 | 9.7            | PA4    | 43                 | 9.7            |
| PA1    | 27                 | 6.6            | PA2    | 15                  | 0.6            | PA3    | 6                  | 0.6            | PA4    | 25                 | 0.6            |
| PA1    | 26                 | 4.7            | PA2    | 13                  | 1.7            | PA3    | 20                 | 4.4            | PA4    | 21                 | 4.4            |
| PA1    | 2                   | 2.2            | PA2    | 2                   | 2.2            | PA3    | 2                   | 2.2            | PA4    | 2                   | 2.2            |
| PA1    | 0                   | 0.2            | PA2    | 0                   | 0.2            | PA3    | 2                   | 0.2            | PA4    | 2                   | 0.2            |

TABLE S1: HPLC methods for the purification of each contrast agent PA.
Analytical HPLC was performed using Varian Prostar 500 HPLC equipped with a Varian 380LC ELSD system to generate the traces. An X-Bridge C18 column with dimensions 4.6x150mm and 5 μm pore size was used. NH₄OH solution at pH 10.3 was used as the polar mobile phase and acetonitrile as the nonpolar. A flow rate of 1 mL/min was used in all cases. Analytical traces are shown below, with peak elution times noted in the captions. Solutions were prepared at approximately 1 mM and injected using the following method:

Table S2: Analytical HPLC method used to verify agent purity. See Figure S17-S24 for Chromatograms and MALDI-MS results

| Time (min) | % ACN |
|-----------|-------|
| 0         | 5     |
| 8         | 5     |
| 55        | 100   |
| 65        | 100   |
| 75        | 5     |
| 80        | 5     |

1.5 MALDI-MS
A Bruker AutoFLex III high Resolution MALDI-TOF mass spectrometer was used to collect MALDI-MS spectra. Bruker Daltonics Prespotted AnchorChip 96 MALDI Targets were used as targets, prespotted with HCA.

1.6 TEM
For Cryogenic transmission electron microscopy (Cryo-TEM), two solutions of 1 mmol of each compound (PA1-4) were prepared at pH 7.4. Unannealed specimens were aged overnight prior to imaging. For imaging annealed specimens, the solution was annealed at 80°C for 30 minutes before being allowed to cool in the bath slowly overnight. Imaging was conducted on a JEOL 1230 microscope, operating at 100 kV. In a humidity and temperature controlled Vitrobot
VI, 5 μL of sample was deposited on a suspended grid, blotted, and plunged into liquid ethane to produce vitreous ice. Samples were transported in liquid nitrogen, and transferred to a Gatan 626 cryo holder, cooled with liquid nitrogen. The sample remained at liquid nitrogen temperatures during imaging for all samples.

1.7 Circular Dichroism
CD spectroscopy was performed with the same solutions used in cryo-EM, except they had been diluted 10x in Tris buffer to a final concentration of 0.1 mM. A J-715 Jasco Circular Dichroism spectrometer was used for these measurements. All measurements were taken at 37°C. Red shifts were calculated from the minima of the negative sigmoid peak characteristic of β-sheet structures. Each spectrum is the average of three scans.

1.8 Relaxation Measurements
T₁ Measurements were performed at 1.41T with a Bruker Minispect mq60 magnet, using an inversion recovery sequence. T₂ measurements were acquired using a Carr–Purcell–Meiboom–Gill (CPMG) pulse sequence. PAs were weighed out and dissolved in Millipore water to form 1 mg/mL solutions, and then these solutions were serial diluted to 0.5, 0.25, 0.125, 0.05, and 0.01 mg/mL concentrations. For each concentration, T₁ and T₂ were measured. These solution concentrations were then measured using ICP-MS for relaxivity determination. Samples were lyophilized and resuspended in 50 mM Tris and 150 mM NaCl buffer. These were then adjusted using small volumes of 0.1 M NaOH to reach pH 7.4. T₁ and ICP-MS measurements were then taken of these solutions. The relaxometer tubes were heated at 80°C for 30 min in a water bath. The bath was allowed to come to room temperature over several hours. T₁ times were measured. Finally, 100 μL of 20 mM CaCl₂ solution was added to each tube. The tubes were vortex-mixed and allowed to stand for 20 minutes. ICP-MS and T₁ measurements were conducted once more.
1.9 NMRD Profiles

The $^1$H NMRD profiles were analyzed assuming one water molecule regularly coordinated to the Gd(III) ion in the presence of freely diffusing outer-sphere water. The contribution of the coordinated water protons ($R_{1M}$) to the relaxation of bulk water protons ($R_1$) largely depends on the $\tau_M$ of coordinated water if $\tau_M > R_{1M}$, as expected for Gd(HP-DO3A), according to the following relationship:

$$R_1 = f_M(1/R_{1M} + \tau_M)^{-1} + R_{os}$$

where $f_M$ is the mole fraction of ligand nuclei in bound positions and $R_{os}$ is the outer-sphere contribution. $R_{1M}$ is calculated by considering the simultaneous presence of both static ($D$) and transient ($\Delta_t$) ZFS, as needed for Gd(III) complexes, through the “modified Florence” NMRD program,\textsuperscript{4-6} assuming slow rotation, \textit{i.e.} the molecular tumbling time $\tau_R$ is much longer than the electron relaxation time $T_{1e}$. The PA fiber’s micron-scale size assures that $\tau_R$ could be fixed to a large, constant value ($\geq 100$ ns) in the calculation of the relaxation profile.

The $^1$H NMRD profiles of PA 1 were repeated at both temperatures using the same $\Delta_t$ value of 0.0142 cm\textsuperscript{-1}, $D=0.046$ cm\textsuperscript{-1}, $\theta=42^\circ$ (angle between the z-axis of the ZFS tensor and the metal-water direction) and a correlation time for electron relaxation $\tau_{V}=24$ ps. A distance of closest approach of diffusing water molecules (d) of 7-10 Angstroms should be set, \textit{i.e.} somewhat larger than usually found in isolated Gd(III) chelates (3.5-4 Å). We expect the proximity of the peptide chains in the nanofibers precludes water approach to the Gd(III) chelates from many directions, thus decreasing the outer-sphere contribution $R_{os}$. The best-fit values of the other parameters are reported in Table 2.
A good fit of the high field region of the profiles could be obtained only by assuming the presence of local mobility, with a correlation time $\tau_{\text{local}}$, considered together with the global reorientation time $\tau_R$ using the Lipari-Szabo model free formalism$^{7,8}$. In this model, the order parameter $S^2$ tunes the degree of spatial restriction of the local motions, from totally free ($S^2=0$) to quenched ($S^2=1$).

The $^1\text{H} \text{NMRD}$ profiles of the other compounds could then be reproduced by fitting only the parameters $\tau_M$, $S^2$, and $\tau_{\text{local}}$, with the constraint that $S^2$ should be the same at the two measurement temperatures (25 and 37°C). Finally, the $^1\text{H} \text{NMRD}$ profiles of the compounds after thermal annealing and in the presence of Ca(II) could be reproduced using the same set of parameters producing the profiles of the corresponding compound acquired without Ca(II), at the same temperature, and adjusting only the value of $\tau_M$.

1.10 Small Angle X-ray Scattering

SAXS measurement was performed at beam line 5ID-D, in the DuPont-Northwestern-Dow Collaborative Access team (DND-CAT) Synchrotron Research Center at the Advanced Photon Source, Argonne National Laboratory. The wavelength, selected with double-crystal monochromator, was 0.83 Å. A MAR CCD detector was positioned 245 cm behind the sample. Samples were loaded into 1.5 mm quartz capillaries and irradiated. All compounds (PAs 1-4) were prepared at 0.1 mM in solutions of Tris buffered saline (50mM Tris 150 mM NaCl).
2 Data and Results

2.1 Supplementary Structural Data
Table S3: Full SAXS fit parameters in buffered solution for PA 1-4.

|                     | PA-1    | PA-2    | PA-3     | PA-4     |
|---------------------|---------|---------|----------|----------|
| Scale               | 0.000845| 0.0015  | 4.80E-07 | 0.00038  |
| mean CORE radius (A)| 20.5    | 37      | 35       | 22       |
| radial polydispersity (sigma) | 0.01    | 0.15    | 0.0001   | 0.05     |
| CORE length (A)     | 20000   | 20000   | 100      | 10000    |
| radial shell thickness (A) | 40.3    | 19      | 15       | 27       |
| face shell thickness (A) | 0.5     | 10      | 1        | 0.1      |
| SLD core (A^-2)     | 9.25E-06| 8.90E-06| 6.33E-05 | 9.00E-06 |
| SLD shell (A^-2)    | 7.73E-06| 8.20E-06| 0.00016  | 6.90E-06 |
| SLD solvent (A^-2)  | 9.40E-06| 9.40E-06| 9.40E-06 | 9.40E-06 |
| incoh. bkg (cm^-1)  | 0.0015  | 0.0009  | 9.00E-07 | 0.00028  |
Figure S1: Cryo-EM images of each contrast agent PA after dissolution in 8 mM Tris buffer at pH 7.4. Structures indicated by black arrows.

A) PA1  B) PA2  C) PA3  D) PA4
Figure S2: Circular Dichroism of PA compounds. Bisignate peaks around 200nm and 216 are indicative of a betasheet morphology. All measurements performed at 0.1 mM agent in 10 mM Tris buffered 50 mM NaCl at pH 7.4.

Table S4: Red shifts of CD beta sheets. B-sheet red shifts are an indication of betasheet helicity. Red shift values for these compounds imply sheets of similar helicity.

| Compound                  | Red Shift(nm) |
|---------------------------|---------------|
| PA1: \( C_{16} V_3 A_3 E_3 G(Gd) \) | 3             |
| PA2: \( C_{16} V_3 A_3 E_3 (Gd) \)   | 3.5           |
| PA3: \( C_{16} V_3 A_3 E_3 G(Gd)_3 \) | N/A           |
| PA4: \( C_{16} V_4 A_3 E(Gd)E_2 \)   | 2             |

2.2 Relaxivity Measurements
Figure S3: PA1 T₂ Relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.

Table S5: PA2 T₂ Relaxivity Results.

|                          | Slope (mM⁻¹ s⁻¹) | Fit R² |
|--------------------------|-------------------|--------|
| Milipure H₂O             | 34±1              | .981   |
| Buffered at pH 7.4       | 30±4              | .866   |
| After Thermal Annealing  | 37±1              | .981   |
| After Addition of CaCl₂ | 40.0±0.1          | .9998  |

Figure S4: PA2 T₂ relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.

Table S6: PA1 T₂ Relaxivity Results.

|                          | Slope (mM⁻¹ s⁻¹) | Fit R² |
|--------------------------|-------------------|--------|
|                          |                   |        |
Figure S5: PA3 T<sub>2</sub> relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.

Table S7: PA1 T<sub>2</sub> Relaxivity Results.

|                        | Slope (mM<sup>-1</sup> s<sup>-1</sup>) | Fit R<sup>2</sup> |
|------------------------|----------------------------------------|------------------|
| Milipure H<sub>2</sub>O | 30±2                                    | 0.932            |
| Buffered at pH 7.4     | 25.3±0.2                                | 0.999            |
| After Thermal Annealing| 25.2±0.2                                | 0.999            |
| After Addition of CaCl<sub>2</sub> | 25±1                                    | 0.957            |
Figure S6: PA4 T₂ relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.

Table S8: PA4 T₂ Relaxivity Results.

|                  | Slope (mM⁻¹ s⁻¹) | Fit R² |
|------------------|-------------------|--------|
| Milipure H₂O     | 31±2              | .932   |
| Buffered at pH 7.4 | 25.3±0.2         | .999   |
| After Thermal Annealing | 25.2±0.2     | .999   |
| After Addition of CaCl₂ | 25±1           | .957   |

Figure S7: PA1 T₁ relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.
Table S9: PA1 T₁ Relaxivity Results.

|                           | Slope (mM⁻¹ s⁻¹) | Fit R² |
|---------------------------|-------------------|--------|
| Milipure H₂O              | 16.5±0.5          | 0.985  |
| Buffered at pH 7.4        | 16.2±2            | 0.876  |
| After Thermal Annealing   | 19.0±0.6          | 0.986  |
| After Addition of CaCl₂   | 18.4±0.2          | 0.998  |

Figure S8: PA2 T₁ relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37°C and 1.41T.

Table S10: PA2 T₁ Relaxivity Results.

|                           | Slope (mM⁻¹ s⁻¹) | Fit R² |
|---------------------------|-------------------|--------|
| Milipure H₂O              | 17.3±0.8          | 0.968  |
| Buffered at pH 7.4        | 16.7±0.2          | 0.997  |
| After Thermal Annealing   | 16.9±0.2          | 0.998  |
| After Addition of CaCl₂   | 21.7±0.1          | 0.9995 |
Figure S9: PA3 $T_1$ relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.

Table S11: PA3 $T_1$ Relaxivity Results.

|                          | Slope (mM$^{-1}$ s$^{-1}$) | Fit $R^2$ |
|--------------------------|----------------------------|-----------|
| Milipure H$_2$O          | 16.4±0.6                   | 0.985     |
| Buffered at pH 7.4       | 15.8±0.5                   | 0.993     |
| After Thermal Annealing  | 16.4±0.6                   | 0.984     |
| After Addition of CaCl$_2$ | 16.5±0.8                   | 0.975     |
Figure S10: PA4 T₁ relaxivity determination. The slope of the linear fit corresponds to the compound relaxivity. All measurements conducted at 37° C and 1.41T.

Table S12: PA4 T₁ Relaxivity Results.

|                          | Slope (mM⁻¹ s⁻¹) | Fit R² |
|--------------------------|------------------|--------|
| Milipure H₂O             | 16.8±1           | 0.931  |
| Buffered at pH 7.4       | 15.6±0.1         | 0.999  |
| After Thermal Annealing  | 16.0±0.1         | 0.999  |
| After Addition of CaCl₂  | 18.2±1           | 0.933  |

2.3 NMRD Profiles

Figure S11: All NMRD profiles measured for PA1
Figure S12: All NMRD profiles and fits obtained for PA2.

Figure S13: All NMRD profiles and fits obtained for PA3.
Figure S14: All NMRD profiles and fits obtained for PA4.
Figure S15: NMRD profiles acquired for C_{16}V_{3}A_{3}E_{3}-NH_{2} as a control for peptide relaxation contribution.
Figure S16: NMRD Profiles acquired for the parent Gd(III) chelate in solution to establish a no-assembly control. Profiles agree with published results for the ‘ProN3’ (GdHPDO3A) chelate.\textsuperscript{3}

2.4 Chemical Characterization
Figure S17: MALDI-MS for PA1.

Figure S18: Analytical Trace PA1. Peak maximum at 27.8 minutes.

Figure S19: MALDI-MS of PA2
Figure S20: PA2 Analytical HPLC Trace. Peak maximum at 35.6 minutes.

Figure S21: MALDI-MS of PA3
Figure S22: Analytical Trace PA3. Peak maximum at 4.9 minutes.

Figure S23: PA4 MALDI
Figure S24: PA4 analytical HPLC trace. Peak maximum at 36.26 minutes.

3 Animal Experiments

Figure S25: Photograph of modified syringe pump used for gel tract injections.
| Volume(microL) | Mouse # | ng Gd in Tissue | Weight of tissue (g) | ng Gd/g tissue | Amount of Gd injected (ng) | Percent Gd remaining after 4 days | Agent |
|---------------|---------|-----------------|----------------------|----------------|---------------------------|----------------------------------|-------|
| 4             | LL      | 214             | 0.30                 | 248            | 709                       | 365                              | PA1   |
| 4             | RL      | 238             | 0.28                 | 187            | 845                       | 365                              | E3    |
| 4             | LL      | 179             | 0.31                 | 122            | 672                       | 365                              | PA1   |
| 4             | RL      | 249             | 0.37                 | 168            | 672                       | 365                              | E3    |
| 4             | LL      | 244             | 0.29                 | 181            | 672                       | 365                              | PA1   |
| 4             | RL      | 249             | 0.30                 | 168            | 817                       | 365                              | PA1   |
| 4             | LL      | 244             | 0.37                 | 168            | 2367                      | 365                              | PA3   |
| 4             | RL      | 249             | 0.29                 | 168            | 2367                      | 365                              | PA3   |
| 4             | LL      | 238             | 0.37                 | 181            | 2048                      | 365                              | PA3   |
| 4             | RL      | 238             | 0.29                 | 181            | 2048                      | 365                              | PA3   |
| 4             | LL      | 244             | 0.37                 | 168            | 1401                      | 365                              | E3    |
| 4             | RL      | 249             | 0.29                 | 168            | 1401                      | 365                              | E3    |
| 4             | LL      | 244             | 0.37                 | 168            | 1245                      | 365                              | E3    |
| 4             | RL      | 249             | 0.29                 | 168            | 1245                      | 365                              | E3    |
| 4             | LL      | 238             | 0.37                 | 181            | 8                        | 0                                 | NA    |
| 4             | RL      | 238             | 0.37                 | 181            | 8                        | 0                                 | NA    |

Table S13: Table of ICP-MS results from mouse intramuscular injections. RL=Right leg, LL=Left Leg. E3 is unfunctionalized (“filler”) PA.
Figure S27: Representative images of mouse legs receiving injections of 1 wt% $\text{C}_{16}\text{V}_{3}\text{A}_{3}\text{E}_{3}\cdot\text{NH}_2$. Day 0 and day 4 after injections shown. Each figure row represents a unique leg. Anatomical images and T1 maps have identical geometry to facilitate comparison.
Figure S28: Representative images of mouse legs receiving injections of 10% PA 1. Day 0 and day 4 after injections shown. Each figure row represents a unique leg. Anatomical images and T1 maps have identical geometry to facilitate comparison.
Figure S29: Representative images of mouse legs receiving injections of 10% PA 3 in filler PA. Day 0 and day 4 after injections shown. Each figure row represents a unique leg. Anatomical images and T1 maps have identical geometry to facilitate comparison.
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