Electronic Supplementary Material (ESI) for Analyst

Supporting Information

A Smart T_1-weighted MRI Contrast Agent for Uranyl Cation based on a DNAzyme-Gadolinium Conjugate

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Experimental Details

Materials, equipment and DNA sequences

All DNA molecules with biotin or amine modifications were purchased from Integrated DNA Technologies, Inc. (Coralville, IA). The DNA was standard desalted by the vendor and used without further purification. NHS-activated DOTA was purchased from Macrocyclics, Inc. (Dallas, TX). Streptavidin and other chemicals were purchased from Sigma-Aldrich, Inc. or Fisher Scientific, Inc.

T_1 was measured on a Bruker Minispec mq 60 MRI contrast agent analyzer (1.5 T, Bruker), Varian NMR spectrometer (300 MHz), and 60 MHz - Varian EM360L NMR Spectrometer with Anasazi FT Upgrade. MRI measurements were acquired on a 1.5T GE Signa Horizon Echo Speed (1.5 T, GE) using the version 9.0 software package.

The sequences of the DNA strands used in this work are listed in Table S1.

Table S1. DNA sequences used

39E	5'-CCATCTCTTCAGTCGGGTAGTTAAACCGACCTTCAGACATAGTGAGT-biotin-3'
39S	5'-amine-ACTCACTATrAGGAAGAGATGG-3’

T1 measurement

39E-based contrast agent was prepared by dissolving the DOTA-Gd-coupled 39S and biotinylated 39E in 50 mM MES-Na (pH 5.5) buffer at a concentration of 30 μM. The solution was heated to 90 °C and cooled to ambient temperature over one hour. Streptavidin was then added into the solution at a concentration of 30 μM. Uranyl acetate (UO_2OAc_2) was added into the solution to reach different concentrations. EDTA (100 μM) was used to stop the reaction after 30 minutes. The sample was then subjected to T_1 measurement.

A Bruker Minispec mq 60 MRI contrast agent analyzer (1.5 T, Bruker) was used to measure the longitude relaxation time of water protons. The temperature was kept constant at 37 ˚C during all T_1 measurements. The parameters of the measurement are summarized in Table S2.

Table S2. Parameters for T1 measurements

First Duration	Last Duration	Data points	Duration Factor	Total Analysis	Del. Sam. Win.	Sam. Win.	Expon. Order
150 ms	15 s	10	1.668	12.58 min	0.03 ms	0.03 s	1