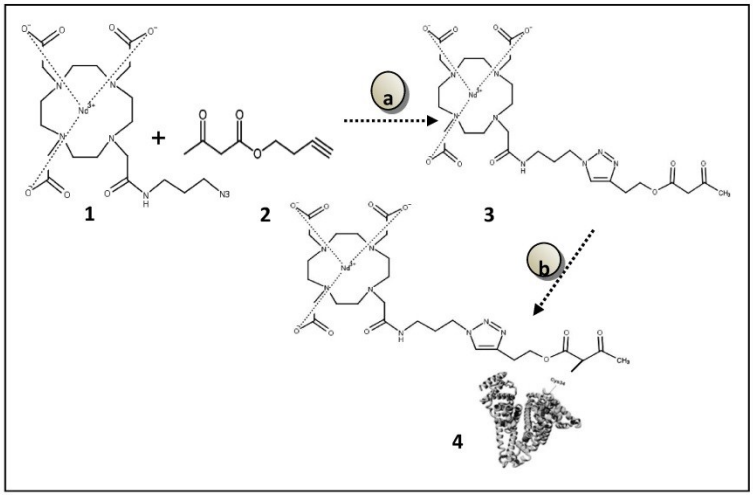
Legend of schemes

Scheme. S1 Reaction scheme for the preparation and labeling of sulfenic acid (SA) with Ln-DOTA-KE. Metallated Azide- DOTA (**1**) undergo a click reaction with alkyne ß-ketoester (KE) (**2**) to produce Ln-DOTA-KE (**3**).Click reaction conditions (**a**): azide: alkyne (2:1), THPTA: Cu(II)SO_4(5:1), sodium ascorbate (5 mM) and TEAA (100 mM, pH= 7.00), sonication in darkness for 1 h. Prepared Ln-DOTA-KE (**3**) is used after purification for SA labelling (**4**) where hydrogen peroxide is used as an oxidizing agent, SA labelling conditions (**b**): (H_2O_2) (5 mM) (8- fold excess to cysteine), Urea (8M), Ln-DOTA-KE (30- fold excess to cysteine) and THAM buffer 100mM, pH=8.4, 4 h, shaking at R.T.



Supplementary Information

Figure S1. Mass spectrum obtained by electrospray ionization quadrupole time-of- flight (ESI-q-TOF) mass spectrometry for Ln-DOTA-KE. The Ln metal used was standard Nd (expected *m/z* Nd-DOTA-KE 780.2095). Mass error for Nd-DOTA-KE (Δm)= -0.38 ppm, where Δm=(mass error/exact mass) $\times 10^6$.

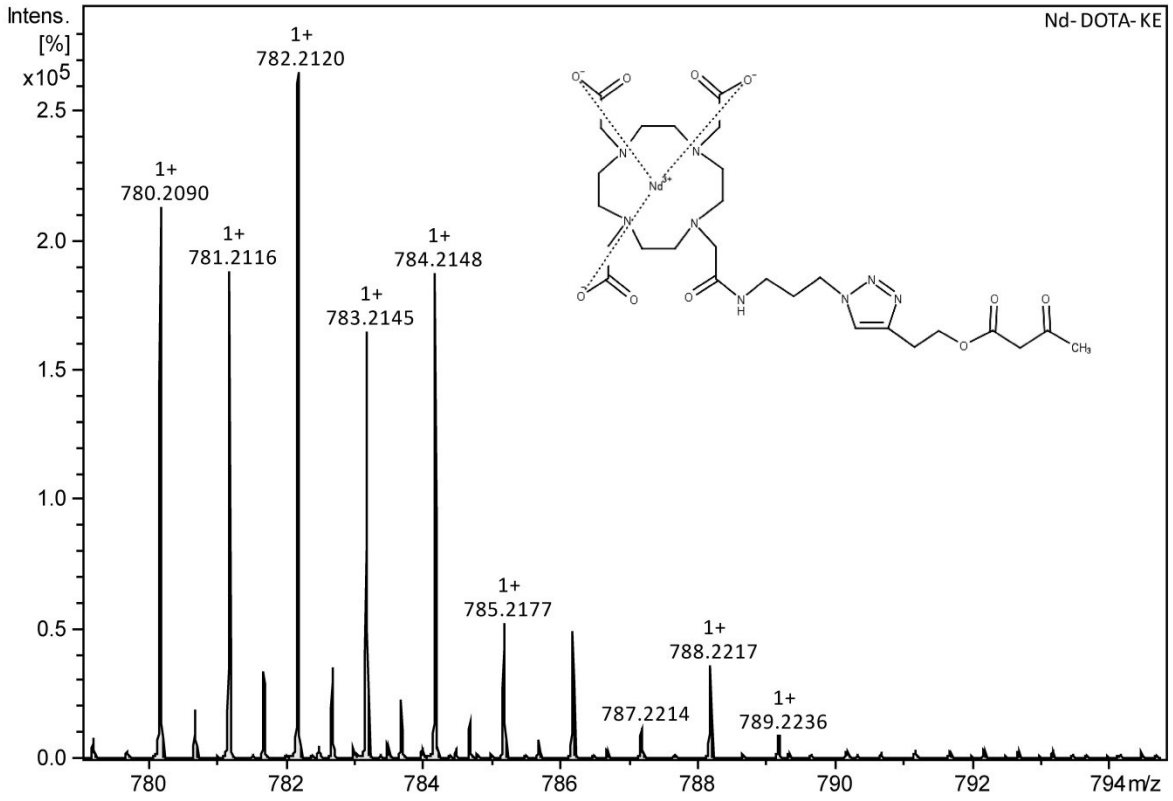


Figure S2. Chromatogram of Nd-DOTA-KE obtained by size exclusion chromatography-inductively coupled plasma mass spectrometry (SEC-ICP-MS). Monitored isotope was ^{142}Nd.

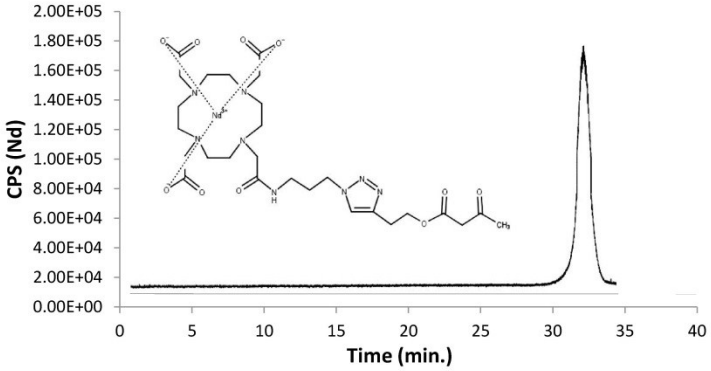


Figure S3. Mass spectrum obtained by electrospray ionization quadrupole time-of- flight (ESI-q-TOF) mass spectrometry for HSA obtained from the commercial standard.

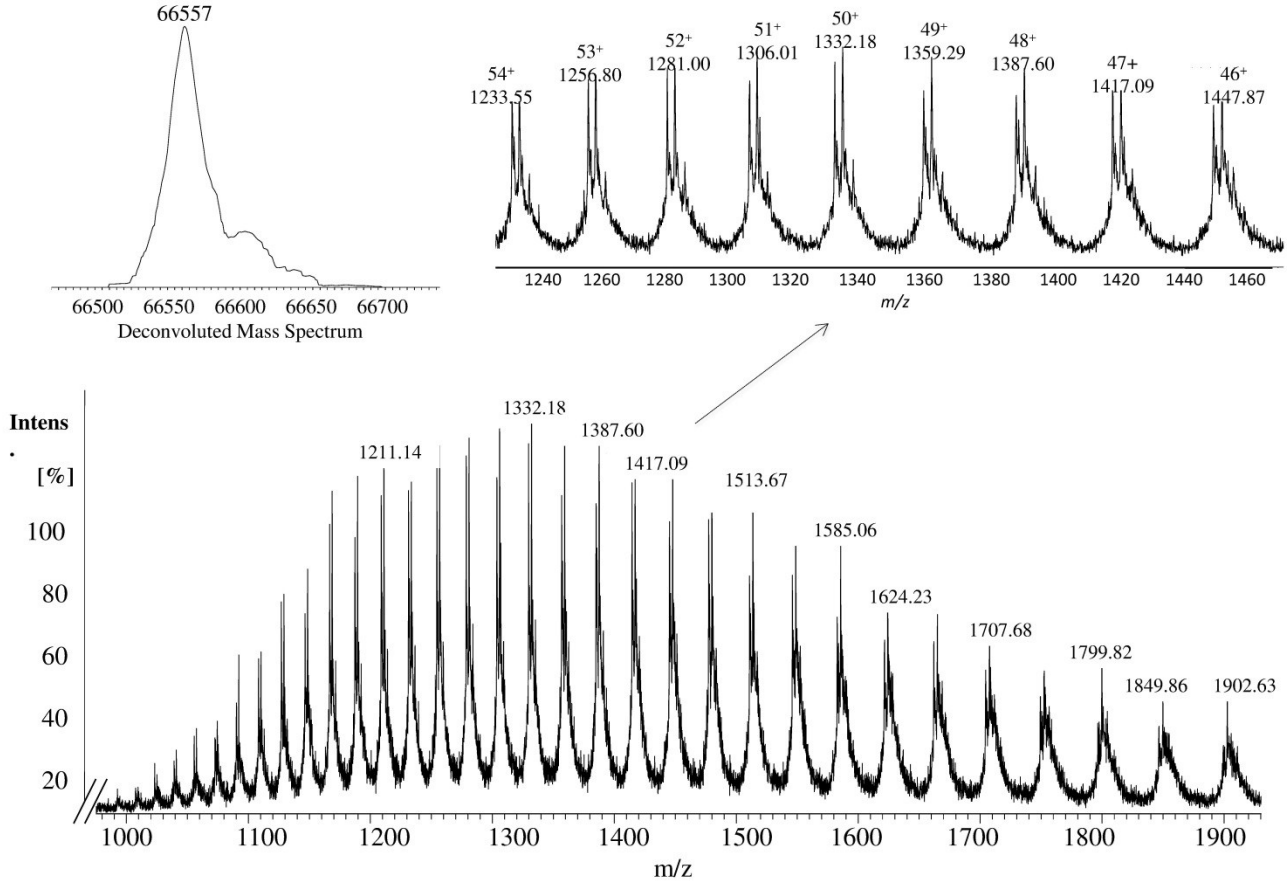


Figure S4. HPLC chromatogram for albumin purification from human serum with HiTrap™ Blue HP column, used binding buffer was 50 mM KH_2PO_4 (pH 7.00) and elution buffer was 50 mM KH_2PO_4 + 1.5 M KCl (pH 7.00). Signal was monitored at 280 nm, 10 µg of collected fractions A (plasma proteins) and B (albumin) were monitored by 10 % SDS- PAGE, band of purified albumin (fraction B) appeared at around 70 KDa in the inset.

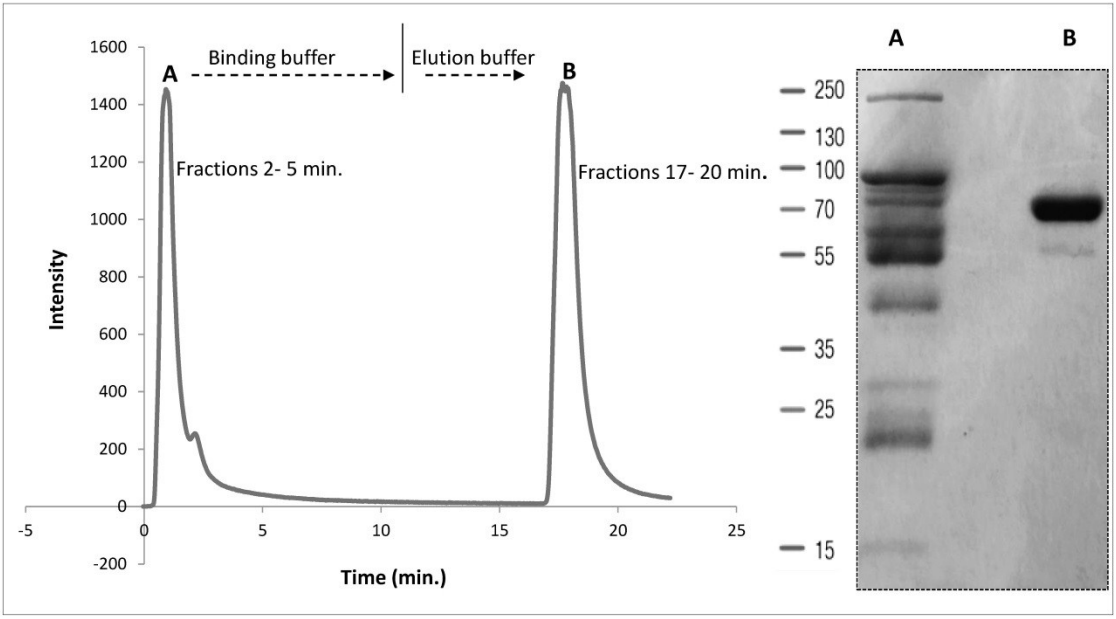


Figure S5.10% SDS- PAGE for 3µg of albumin band is shown for: (A) human serum and (B) purified albumin from human serum (C) Nd-DOTA-KE-HSA with 8 excess H_2O_2 and (D) Nd-DOTA-KE-HSA with 5 excess H_2O_2 .

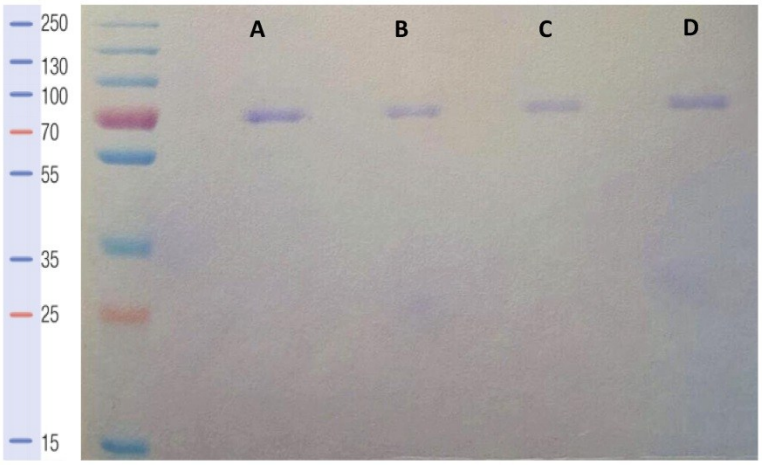


Figure S6. Error propagation plot for the standard ^{142}Nd and the spiking solution of ^{145}Nd.The error propagation plot represents the theoretical optimum ratio (R_m) that should present between the ^{145}Nd spiking solution and the samples with the standard ^{142}Nd in order to achieve the best precision for the measurement, where it was shown to be within 0.01 - 1.