



Salt-free separation of $^{241}\text{Am}(\text{III})$ from lanthanides by highly stable macroporous silica-polymer based $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ adsorbent

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Abstract

Macroporous $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ was used for separating $^{241}\text{Am}(\text{III})$ from lanthanides. Good selectivity towards $^{241}\text{Am}(\text{III})$ was achieved in 0.1–3 M HNO_3 . Adsorption equilibrium of $^{241}\text{Am}(\text{III})$ was obtained within 1 h in 0.1 M HNO_3 solution. $^{241}\text{Am}(\text{III})$ was efficiently separated from lanthanides through column experiment in both 0.1 M HNO_3 and 0.01 M $\text{HNO}_3\text{-}0.34$ M NaNO_3 solution. It still performed excellently at 207 kGy under γ irradiation in nitrate medium. In a word, $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ is a very promising candidate for separating MA from Ln that even a salt-free Ln&MA separation process may be established such as from 0.1 M HNO_3 based on it.

Keywords Minor actinides · BTP · HLLW · Irradiation · Extraction chromatography

Introduction

With the development of nuclear energy, the accumulated spent fuel increases year by year. For the purpose of sustainable development of nuclear energy and reducing the long-term radioactive waste to a minimum, it is urgent to dispose the spent fuel correctly and efficiently. The potential radiotoxicity of high level liquid waste (HLLW) from the current industrial plutonium uranium recovery by extraction (PUREX) process, which only separates 99.5% of U and Pu from spent fuel, still exceeds 10,000 years to reach the natural uranium ore radiotoxicity level. However, if further removal of minor actinides (MA) from HLLW followed by converting it into short-lived radionuclides, which is the so called Partition and Transmutation strategy (P&T), is

performed, the time will reduce to 300–500 years. Before transmutation, minor actinides must be separated from the neutron-absorbing elements from HLLW. Lanthanides (Ln), which account for about one-third of the fission products and are much higher than those of MA in a order of magnitude in contents, are strong neutron-absorbing elements and predominantly exist as trivalent cations in acidic aqueous solutions and have similar ionic radii with MA(III) making their chemistry properties very similar, resulting in the mutual separation of MA(III) and Ln(III) very difficult. The separation of trivalent Ln(III) and MA(III) has therefore become a challenging key issue in HLLW reprocessing.

Various materials have been prepared and used for MA(III) and Ln(III) separation, such as CMPO [1], TODGA [2], TPTZ [3], Cyanex 301 [4], TRPO [5], BTP [6, 7], BTBP [6, 8], BTPPhen [9], etc. But as HLLW generated by PUREX process is in high acidity (about 2–5 M, M = mol/L) with strong radioactivity and various elements co-existence, which proposes high requirements for materials used for directly separating MA from HLLW in the aspects of stability and selectivity, few of the extractants mentioned above can meet the demands. For example, CMPO and TODGA exhibits good stability in high acidity solution but poor selectivity between MA(III) and Ln(III) [1, 10–12]. Cyanex 301 has high selectivity towards MA(III) over Ln(III) only in very low acid solution such as $\text{pH} > 3$ with NaNO_3 added meantime [4]. TRPO exhibits good co-extraction ability towards MA and Ln in 0.8–1.2 M HNO_3 but it needs 5 M

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HNO₃ solution to strip MA and Ln from the loaded organic phase into liquid phase [13]. For the ligands of BTP, BTBP, BTPPhen [6], good selectivity between MA(III) and Ln(III) is obtained which has been widely recognized, most of which exhibits poor stability in high acid solution under irradiation, but there are some exceptions in the cases of ligands with cyclic side structures at the 5,6 positions of triazinyl rings avoiding α -H which is the weak parts under acid or irradiation [14], such as CyMe₄-BTBP [15, 16], CyMe₄-BTPPhen [15], Me₂-CA-BTP [17]. The extraction performance of both CyMe₄-BTBP and CyMe₄-BTPPhen towards Am and Eu show no decrease when the γ absorption dose is 300 kGy in 1 M HNO₃ solution [15] and are used as the reference extractants in the SANEX process [16].

Me₂-CA-BTP/SiO₂-P is a kind of adsorbent prepared in our previous work based on Me₂-CA-BTP (Fig. 1) [17–19]. It exhibits good adsorption selectivity towards ²⁴¹Am(III) over other typical fission products including Ln(III) in 3 M HNO₃ solution, and good stability in 1 M and 3 M HNO₃ solution even under γ irradiation [17]. Moreover, Me₂-CA-BTP/SiO₂-P also exhibits good adsorption selectivity towards ²⁴¹Am(III) over Ln(III) in low acid—NaNO₃ mediums [19]. In a word, Me₂-CA-BTP/SiO₂-P is a promising material for MA separation from the aspects of directly separating MA from HLLW and the mutual separation of MA&Ln, so it is worth studied throughly. Furthermore, except the direct separation process, the most widely accepted concept for MA separation nowadays is the two-step separation process, i.e. firstly MA and Ln are co-separated from other fission products from high acid HLLW, and secondly, MA and Ln is mutual separated from each other in low acid medium with salting-out agent added if necessary. So the possible application of Me₂-CA-BTP/SiO₂-P in the two-step MA separation process (low acid or low acid—NaNO₃ medium) is also worth study. The paper would firstly study the adsorption behavior of Me₂-CA-BTP/SiO₂-P towards other actinides, such as U, Pu, which has not been reported before. Then it would mainly focus on the stability and continuous column separation performance using radionuclides in low acid—NaNO₃ mediums. Column experiments using radionuclides is the most intuitive way to reflect the separation effect so it is very important.

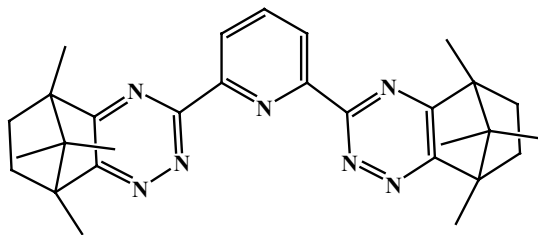


Fig. 1 Chemical structure of Me₂-CA-BTP (C₂₉H₃₅N₇)

Experimental

Reagents

Lanthanide nitrates were of analytical grade purchased from Sinopharm Chemical Reagent Co., Ltd., China. ²⁴¹Am(III), ¹⁵²Eu(III), ²³⁸U(VI) and ²³⁹Pu(IV) were from stock solutions of China institute of atomic energy and its concentration in the solution after dilution for adsorption were 1000 Bq/mL (3.27 × 10⁻⁸ M), 1000 Bq/mL (1.02 × 10⁻⁹ M), 1.00 × 10⁻³ M, 50 Bq/mL (9.11 × 10⁻⁸ M) respectively. The acetonitrile (CH₃CN) used was chromatographically pure. Other reagents such as nitric acid, sodium nitrate, etc. were of analytical grade and used without further treatment. The Me₂-CA-BTP/SiO₂-P adsorbent was synthesized in our previous study by impregnating Me₂-CA-BTP dissolved in CH₂Cl₂ into macroporous silica polymer under reduced pressure with the content of Me₂-CA-BTP about 33 wt% [18, 19]. The liquid solutions were prepared with ultra-pure water of 18 M Ω cm resistance.

Batch adsorption experiment

The effects of HNO₃ concentration and contact time on Me₂-CA-BTP/SiO₂-P adsorption towards actinides and lanthanides were studied by batch experiment. 0.1 g Me₂-CA-BTP/SiO₂-P was combined with 5 mL aqueous solution in a 10 mL plastic centrifuge tube with screw cap. The mixture was shaken mechanically in an air bath shaker at 300 rpm at 25 °C. The solid and liquid phases were separated by centrifugation. The radioactivities of ²⁴¹Am(III) and ¹⁵²Eu(III) were determined by high-purity germanium multichannel gamma spectrometer (CANBERRA) at 59.5 keV and 121.78 keV respectively. The radioactivity of ²³⁹Pu(IV) was measured by the Super Low Level Liquid Scintillation Analyzer (PE Tri-Carb 3170). The concentration of ²³⁸U(VI) was measured by UV spectrophotometer (Lab Tech UV1000/1100). The distribution coefficient K_d (mL g⁻¹), separation factor SF_{A/B}, and adsorption efficiency E (%) are calculated by Eqs. (1), (2), and (3), respectively:

$$K_d = \frac{A_0 - A_e}{A_e} \times \frac{V}{W} \quad (1)$$

$$SF_{A/B} = K_{dA}/K_{dB} \quad (2)$$

$$E = \frac{(A_0 - A)}{A} \times 100\% \quad (3)$$

where A_0 , A_e denote the radioactivities of metal ions in the aqueous phase before and after adsorption, respectively,

Bq mL⁻¹. V (mL) indicates the volume of aqueous phase and W (g) is the mass of dry Me₂-CA-BTP/SiO₂-P adsorbent.

Column separation experiment

Column separation experiment was carried out using a glass column of $\Phi 5$ mm * h500 mm. 5 g Me₂-CA-BTP/SiO₂-P was transferred to the column in the slurry state under atmosphere with the bed volume about 9.8 mL. The column behaviors of Me₂-CA-BTP/SiO₂-P towards ²⁴¹Am(III) and Ln(III) in both 0.1 M HNO₃ and pH2—0.34 M NaNO₃ solutions were studied respectively. Using the case of 0.1 M HNO₃ as the feed solution as an example, firstly, the packed column was pre-equilibrated with 20 mL 0.1 M HNO₃ solution. Then 10 mL 0.1 M HNO₃ containing 2 mM Ln (La, Ce, Pr, Nd, Sm, Eu, Gd, Dy) for each and 500 Bq/mL ²⁴¹Am(III) as feed solution flow through the column. Followed by about 40 mL 0.1 M HNO₃, about 10 mL 0.01 M HNO₃, and finally 0.001 M HNO₃–0.01 M DTPA solutions respectively flow through the column to elute the residual or poor adsorbed Ln(III) and strongly adsorbed ²⁴¹Am(III). The mobile phase was pumped at about 0.1 mL/min and the effluents were collected by a fractional collector. The concentrations of Ln in the effluents were determined by inductively coupled plasma-optical emission spectrometer (ICP-AES: Shimadzu ICPS-7510) while ²⁴¹Am(III) was determined by high-purity germanium multichannel gamma spectrometer.

Stability evaluation

The gamma-ray irradiation stability of Me₂-CA-BTP/SiO₂-P in both 0.1 M HNO₃ and pH2-nitrate mediums was evaluated by ⁶⁰Co γ source. 0.3 g Me₂-CA-BTP/SiO₂-P and 15 mL aqueous solution (e.g., 0.1 M HNO₃ solution) were put together in a 40 mL glass vial with a cap and irradiated at the dose rate of 1 kGy/h. The total organic carbon (TOC) in the aqueous solution was determined by TOC analyzer (Shimadzu, TOC-V), which, in some degree, can partly reflect the dissolution of both decomposed and undecomposed Me₂-CA-BTP from the adsorbent into the liquid phase. The irradiated Me₂-CA-BTP/SiO₂-P was analyzed by UV/vis spectra (Shimadzu, UV-3600 Plus) and UPLC/Q-TOF-MS (Waters) to evaluate the changes in structure and content of Me₂-CA-BTP in Me₂-CA-BTP/SiO₂-P after irradiation. UV/vis spectra were performed in a wavelength range from 400 nm to 200 nm. The UPLC/Q-TOF-MS test was performed in the positive mode using ACQUITY UPLC BEH C₁₈ column. Furthermore, the adsorption of Me₂-CA-BTP/SiO₂-P towards ²⁴¹Am(III) and ¹⁵²Eu(III) after irradiation was further evaluated.

Results and discussion

Effect of HNO₃ concentration

In our previous study, the adsorption behaviors of Me₂-CA-BTP/SiO₂-P towards stable Ln(III), other typical FPs and trace amount of ²⁴¹Am(III) were studied that it exhibited good adsorption selectivity towards ²⁴¹Am(III) over Ln(III) and most other FPs [17–19]. Herein, the effect of initial HNO₃ concentration (pH4—4 M) on the adsorption of Me₂-CA-BTP/SiO₂-P towards trace amounts of ²³⁸U(VI), ²³⁹Pu(IV) was further studied. The results are shown in Fig. 2 combining with the results of ¹⁵²Eu(III) and ²⁴¹Am(III) obtained in Ref. [17]. Considering ²³⁹Pu(IV) hydrolyzes easily in low acidity solution, the effect of acidity on adsorption was performed in 0.5–4 M HNO₃ solution, and its uptake rate was over 97% in 0.5–2 M HNO₃ while it decreased as acidity further increased in 3–4 M HNO₃, which is similar with that of ²⁴¹Am(III). The reason may be that the adsorption of Me₂-CA-BTP/SiO₂-P towards both ²³⁹Pu(IV) and ²⁴¹Am(III) needs the participation of NO₃⁻ to keep charge balance. As the radiotoxicity, it is difficult to conduct in-depth analysis with analytical instruments, so stable Ln(III) in the Ref. [7] was used as a simulated elements of MA(III), and XPS analysis was performed on Ln-loaded isoBu-BTP/SiO₂-P where NO₃⁻ was observed [7]. But as HNO₃ concentration further increases, the competition between HNO₃ and metal ions also increases which leads to the decrease of metal ion adsorption. In pH4—4 M HNO₃ solution, Me₂-CA-BTP/SiO₂-P exhibited almost no adsorption towards ²³⁸U(VI), which is consistent with the results in reference that BTP has no adsorption or extraction ability towards U [20]. The adsorption towards ²⁴¹Am(III)

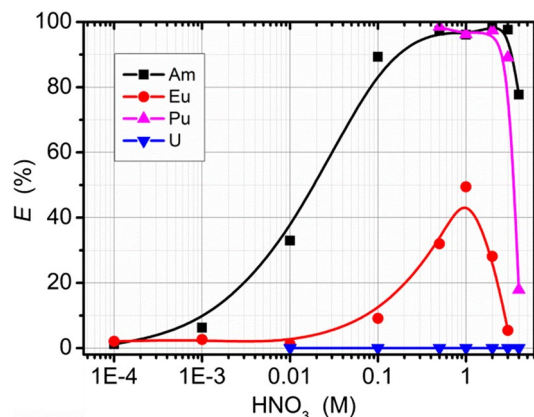


Fig. 2 Effect of HNO₃ concentration on Me₂-CA-BTP/SiO₂-P adsorption towards ²⁴¹Am(III), ²³⁹Pu(IV), ²³⁸U(VI), and ¹⁵²Eu(III) (V/M: 5 mL/0.1 g, ²⁴¹Am(III) and ¹⁵²Eu(III): 1000 Bq mL⁻¹, ²³⁹Pu(IV): 50 Bq/mL, ²³⁸U(VI): 1 mM, 298 K, 24 h, shaking speed: 300 rpm)

was much higher than that of $^{152}\text{Eu}(\text{III})$ with $SF_{\text{Am/Eu}}$ over 72 in 0.1–3 M HNO_3 . In a word, $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ has good adsorption selectivity towards $^{239}\text{Pu}(\text{IV})$ and $^{241}\text{Am}(\text{III})$ over $\text{Eu}(\text{III})$.

Effect of contact time on adsorption towards $^{241}\text{Am}(\text{III})$ and $^{152}\text{Eu}(\text{III})$

According to the above study, $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ exhibits good adsorption selectivity towards $^{241}\text{Am}(\text{III})$ over $^{152}\text{Eu}(\text{III})$ in 0.1 M HNO_3 solution without salting-out agent added, which indicates the possibility of establishing a salt-free Ln&MA separation process in 0.1 M HNO_3 solution and will decrease the second radioactive waste significantly. The effect of contact time on adsorption towards $^{241}\text{Am}(\text{III})$ and $^{152}\text{Eu}(\text{III})$ in 0.1 M HNO_3 solution was studied with the results shown in Fig. 3. The adsorption equilibrium towards $^{241}\text{Am}(\text{III})$ can almost be obtained within 1 h with the uptake rate $^{241}\text{Am}(\text{III})$ about 90% and $SF_{\text{Am/Eu}}$ over 75 as

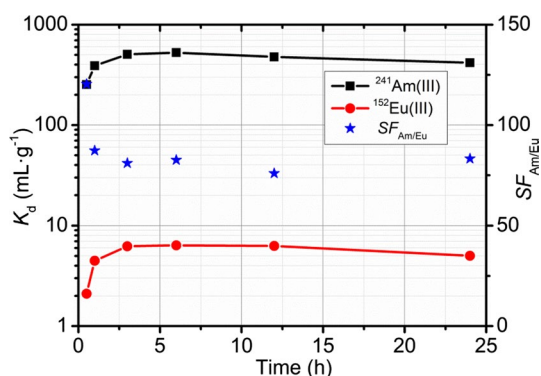


Fig. 3 Effect of time on $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ adsorption towards $^{241}\text{Am}(\text{III})$ and $^{152}\text{Eu}(\text{III})$ in 0.1 M HNO_3 solution (V/M: 5 mL/0.1 g, [M]: 1000 Bq/mL, Temp.: 25 °C, shaking speed: 300 rpm)

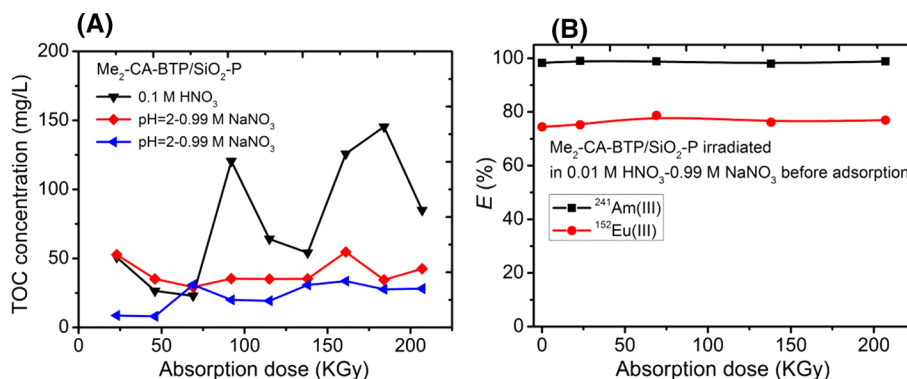


Fig. 4 Effect of γ absorption dose on **a** TOC in the liquid phase, **b** adsorption towards $^{241}\text{Am}(\text{III})$ and $^{152}\text{Eu}(\text{III})$ after irradiated in 0.01 M HNO_3 –0.99 M NaNO_3 solution (γ irradiation conditions: Co-60 source, dose rate: 1 kGy/h, V/M: 15 mL/0.3 g; TOC test con-

ditions: diluting the liquid solution 50 times before test; Adsorption conditions: V/M: 2.5 mL/0.05 g, [M]: 1000 Bq/mL, Temp.: 25 °C, time: 24 h, shaking speed: 300 rpm, medium: 0.01 M HNO_3 –0.99 M NaNO_3)

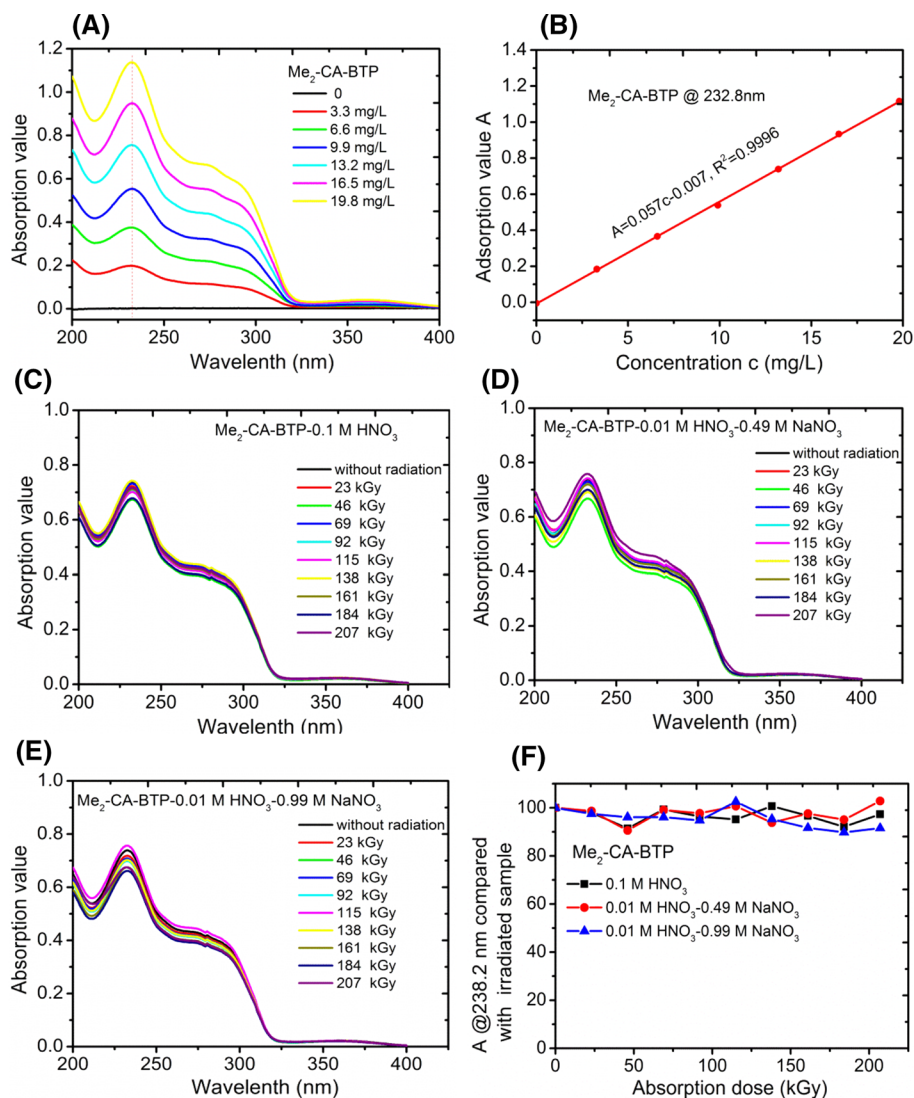
Stability evaluation

The γ irradiation stabilities of $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ in 0.1 M HNO_3 and low acid- NaNO_3 solutions were studied using ^{60}Co as γ source at the dose rate of 1 kGy/h. The total organic content (TOC) in the liquid phase after irradiation was analyzed with TOC less than 150 mg/L (Fig. 4a). Considering the support particle $\text{SiO}_2\text{-P}$ is very stable [21], so the TOC in the liquid phase is mainly from the dissolution of both decomposed and undecomposed of $\text{Me}_2\text{-CA-BTP}$ from the adsorbent. If all the $\text{Me}_2\text{-CA-BTP}$ was decomposed and dissolved in the liquid phase, the TOC should be 4820 mg/L. Compared with 4820 mg/L, the value of 150 mg/L is very small with the relative value less than 3%, which indicates good stability of $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$.

$\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ after irradiation was also used to adsorb $^{241}\text{Am}(\text{III})$ and $^{152}\text{Eu}(\text{III})$ from 0.01 M HNO_3 –0.99 M NaNO_3 solution (Fig. 4b). Compared with fresh $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ without irradiation corresponding to the absorption dose of 0 kGy, $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ after irradiation exhibited no obvious difference in $^{241}\text{Am}(\text{III})$ and $^{152}\text{Eu}(\text{III})$ adsorption with the uptake rates almost unchanged as the absorption dose increase.

Both UV and UPLC/Q-TOF-MS were further used to analyze $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ after irradiation. Fresh $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ was used as a standard sample. Acetonitrile (CH_3CN) was used to dissolve $\text{Me}_2\text{-CA-BTP}$ in the adsorbent while $\text{SiO}_2\text{-P}$ couldn't be dissolved. The overall absorption value and intensity increased with initial $\text{Me}_2\text{-CA-BTP}$ concentration shown in Figs. 5a and 6a. For UV analysis, the absorption peak at 232.8 nm is contributed to $\pi \rightarrow \pi^*$ transition of electrons from aromatic cycles, which is similar with that of $\text{CyMe}_4\text{-BTP}$, BTP-4Me , and

Fig. 5 UV spectra of Me₂-CA-BTP **a** Effect of Me₂-CA-BTP concentration, **b** absorption value at 232.8 nm versus Me₂-CA-BTP concentration, **c** irradiated in 0.1 M HNO₃ solution, **d** irradiated in 0.01 M HNO₃–0.49 M NaNO₃ solution, **e** irradiated in 0.01 M HNO₃–0.99 M NaNO₃ solution, **f** effect of γ absorption dose on Me₂-CA-BTP/SiO₂-P retention (γ irradiation conditions: Co-60 source, dose rate: 1 kGy/h, V/M: 15 mL/0.3 g; UV tests: 13.2 mg/L Me₂-CA-BTP in CNCH₃)



*i*Pr-BTP [22, 23] and is used as a characteristic peak for quantitative analysis. The absorption value at 232.8 nm versus Me₂-CA-BTP concentration is shown in Fig. 5b where a linear line is obtained and used for the retention calculation of Me₂-CA-BTP after irradiation. The UV spectra of Me₂-CA-BTP/SiO₂-P after irradiation in 0.1 M HNO₃, 0.01 M HNO₃–0.49 M NaNO₃, 0.01 M HNO₃–0.99 M NaNO₃ are shown in Fig. 5c–e respectively that the UV spectra of Me₂-CA-BTP/SiO₂-P are almost consistent with that of fresh Me₂-CA-BTP/SiO₂-P. The retention contents of Me₂-CA-BTP/SiO₂-P after irradiation were obtained according to the standard curve in Fig. 5b with the values over 90% shown in Fig. 5f.

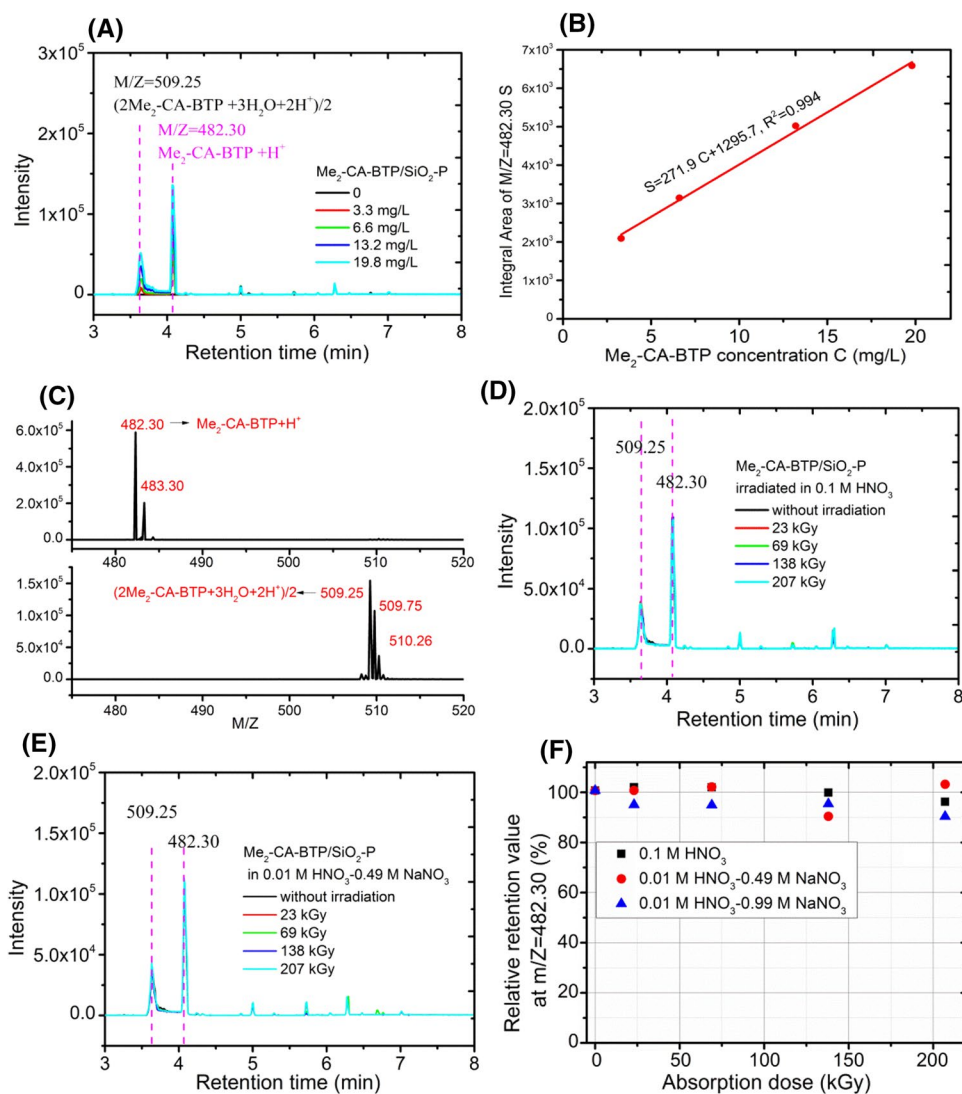
For UPLC/Q-TOF-MS analysis (Fig. 6), the mass-to-charge ratios of 509.25 and 482.30 shown in Fig. 6c are corresponding to $(2\text{Me}_2\text{-CA-BTP} + 3\text{H}_2\text{O} + 2\text{H}^+)/2$ and $\text{Me}_2\text{-CA-BTP} + \text{H}^+$ respectively, and herein $M/Z = 482.30$ is used for the retention calculation of Me₂-CA-BTP

with the standard curve of integral area versus concentration shown in Fig. 6b. The UPLC/Q-TOF-MS analysis of Me₂-CA-BTP/SiO₂-P after irradiation in 0.1 M HNO₃, 0.01 M HNO₃–0.49 M NaNO₃ are shown in Fig. 6d, e respectively that the elution curves of Me₂-CA-BTP/SiO₂-P are almost consistent with that of fresh Me₂-CA-BTP/SiO₂-P. The retention contents of Me₂-CA-BTP are over 90% in different mediums shown in Fig. 6f. In summary, the results above indicate good γ -irradiation stability of Me₂-CA-BTP/SiO₂-P in 0.1 M HNO₃ or low acid-NaNO₃ solution.

Column experiments

The column separation behaviors of Me₂-CA-BTP/SiO₂-P towards ²⁴¹Am(III) and Ln(III) in the feed solutions of both 0.01 M HNO₃–0.34 M NaNO₃ and 0.1 M HNO₃ mediums were studied with the results shown in Fig. 7, Tables 1 and 2. The effluent was collected since the feed liquid began to

Fig. 6 UPLC/Q-TOF-MS test results of Me₂-CA-BTP. **a** Effect of Me₂-CA-BTP concentration, **b** integral area of $M/Z=482.30$ versus Me₂-CA-BTP concentration, **c** mass spectra of Me₂-CA-BTP, **d** elution curve of Me₂-CA-BTP/SiO₂-P irradiated in 0.1 M HNO₃ solution, **e** elution curve of Me₂-CA-BTP/SiO₂-P irradiated in 0.01 M HNO₃–0.49 M NaNO₃ solution, **f** effect of γ absorption dose on Me₂-CA-BTP/SiO₂-P retention (γ irradiation conditions: Co-60 source, dose rate: 1 kGy/h, V/M: 15 mL/0.3 g; UPLC/MS-Q-TOF test: 13.2 mg/L Me₂-CA-BTP in CH₃CN)



be pumped into the column, so the effluent in step A was corresponding to the residual solution in the column used for pre-equilibrium. As seen, Me₂-CA-BTP/SiO₂-P exhibited almost no adsorption towards Y, La, Ce, Nd, Sm, Eu, Gd that they almost all flew out with the feed solution in step B and scrubbing solution of 0.1 M HNO₃ in step C. While it exhibited part adsorption towards Dy, although most of Dy flew out in step B, C, and D, but there were still few parts left in step E. But considering there are almost no Ln(III) heavier than and including Dy in HLLW, so the problem caused by Dy during MA(III) separation is almost negligible [24]. Finally, Me₂-CA-BTP/SiO₂-P exhibited good adsorption towards ²⁴¹Am that almost no ²⁴¹Am flew out in steps B, C, D. ²⁴¹Am was completely desorbed and separated from the other fission products Ln in step E using 0.001 M HNO₃–0.01 M DTPA (aminopolycarboxylate complexant diethylenetriamine-*N,N,N',N'',N'''*-pentaacetic acid). DTPA is a kind of actinide holding hydrophilic

reagents and can strong complex with An(III) which has been widely used in the TALSPEAK (Trivalent Actinide Lanthanide Separation by Phosphorus Extractants and Aqueous Complexes) process [14]. A high recovery yield of ²⁴¹Am was achieved with the value nearly 100% shown in Tables 1 and 2. The results above indicate Me₂-CA-BTP/SiO₂-P has the possibility to separate MA(III) from Ln(III), especially in the case of 0.1 M HNO₃ which makes it possible to establish an almost salt-free Ln&MA separation process.

Conclusions

To reduce the long-term radiotoxicity of HLLW, macroporous silica-polymer based adsorbent Me₂-CA-BTP/SiO₂-P was used for separating ²⁴¹Am(III) from lanthanides in nitrate medium. It exhibited good adsorption selectivity

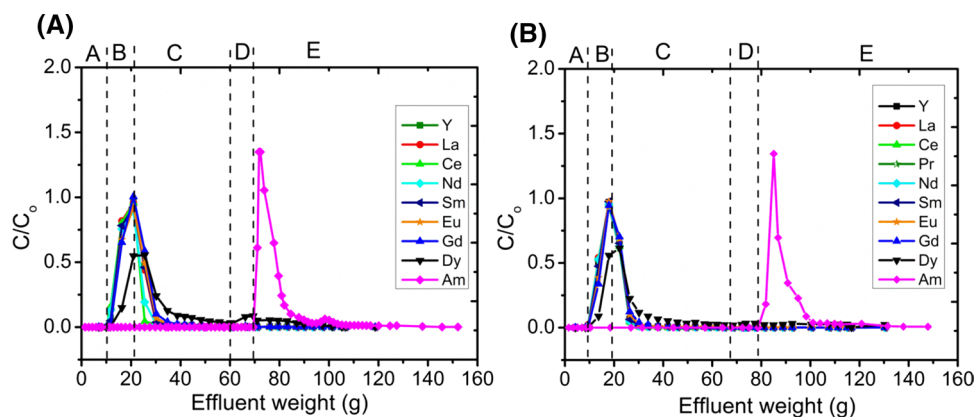


Fig. 7 Column experiment results of separating $^{241}\text{Am(III)}$ from **a** 0.01 M HNO_3 -0.34 M NaNO_3 , **b** 0.1 M HNO_3 simulated HLLW by $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ at 308 K (column: $\Phi 5\text{ mm} \times 50\text{ cm}$, 5 g $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$, Temp.: 308 K, liquid flow rate: about 0.2 mL/

min, A: 0.01 M HNO_3 -0.34 M NaNO_3 , B: 0.01 M HNO_3 -0.34 M NaNO_3 -2 mM Ln(III) -500 Bq/mL $^{241}\text{Am(III)}$ for **(a)** and 0.1 M HNO_3 -2 mM Ln(III) -500 Bq/mL $^{241}\text{Am(III)}$ for **(b)**, C: 0.1 M HNO_3 , D: 0.01 M HNO_3 , E: 0.001 M HNO_3 -0.01 M DTPA)

Table 1 Recovery yields of $^{241}\text{Am(III)}$ and Ln(III) in column experiments in 0.01 M HNO_3 -0.34 M NaNO_3 feed medium

	Y (%)	La (%)	Ce (%)	Nd (%)	Sm (%)	Eu (%)	Gd (%)	Dy (%)	Am (%)
B	75.2	78.3	79.2	72.7	71.4	65.5	68.0	28.7	0.00
C	23.4	20.1	4.2	10.3	22.4	26.1	32.6	50.4	0.00
D	0.2	0.1	0.2	0.1	0.2	0.3	0.5	6.9	0.02
E	0.4	0.1	0.1	0.2	0.3	0.6	0.8	8.2	99.49
Total recovery yield	99.3	98.7	83.7	83.3	94.2	92.6	102.0	94.3	99.51

Table 2 Recovery yields of $^{241}\text{Am(III)}$ and Ln(III) in column experiments in 0.1 M HNO_3 feed medium

	Y (%)	La (%)	Ce (%)	Pr (%)	Nd (%)	Sm (%)	Eu (%)	Gd (%)	Dy (%)	Am (%)
B	94.0	94.7	94.0	92.3	93.7	93.0	90.7	87.9	55.9	0.00
C	5.8	4.1	3.7	3.1	4.0	5.2	8.1	10.7	31.0	0.00
D	0.2	0.2	0.1	0.0	0.1	0.2	0.3	0.4	4.5	0.00
E	0.2	0.1	0.0	0.0	0.1	0.1	0.3	0.4	8.3	98.55
Total recovery yield	100.2	99.1	97.8	95.4	97.9	98.5	99.4	99.4	99.7	98.55

towards $^{241}\text{Am(III)}$ and $^{239}\text{Pu(IV)}$ over $^{152}\text{Eu(III)}$ with the uptake rates of $^{241}\text{Am(III)}$ and $^{239}\text{Pu(IV)}$ over 97% meantime in a wide nitric acid solution, such as 0.5–2 M, while almost no adsorption towards $^{238}\text{U(VI)}$. In the case of $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ adsorbing $^{241}\text{Am(III)}$ in 0.1 M HNO_3 solution, it took about 1 h to reach equilibrium and also kept high $\text{SF}_{\text{Am/Eu}}$ with the value over 75. In both 0.1 M HNO_3 and 0.01 M HNO_3 -0.34 M NaNO_3 feed solutions, $^{241}\text{Am(III)}$ was successfully separated from lanthanides by column experiment and high recovery yield over 98.55% was obtained meantime. Furthermore, $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ kept high effective content with the value over 90% and exhibited no decrease

in adsorption selectivity towards $^{241}\text{Am(III)}$ and $^{152}\text{Eu(III)}$ in 0.1 M HNO_3 and 0.01 M HNO_3 - NaNO_3 mediums when the Co-60 γ absorption dose was as high as 207 kGy at the dose rate of 1 kGy/h. In a word, $\text{Me}_2\text{-CA-BTP/SiO}_2\text{-P}$ is a very promising candidate for separating MA from Ln that even a soft free separation Ln&MA process can be established such as from 0.1 M HNO_3 based on it.

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