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Dendrimer-coated magnetic particles for radionuclide separation

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Abstract

Magnetic particles were synthesised for radionuclide removal from nuclear wastes by magnetic separation. Dendrimers with terminal amino groups attached to the particle surface were used to bind chelating groups for lanthanides and actinides. This led to a 50–400-fold increase of the distribution coefficients for europium and americium in comparison to the reference particles without the dendrimers. Back-extraction studies have demonstrated the possibility of multiple particle recycling.

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1. Introduction

The recovery of lanthanides and actinides from high-level nuclear wastes, generated as a result of

the nuclear fuel cycle and the development and production of nuclear weapons, is an area of worldwide concern. Magnetic particles offer new perspectives for cost-effective and simple processes for nuclear waste minimisation and recycling efforts.

High activity wastes, which contain minor actinides with long half-lives, are currently vitrified for long-term storage in geological repositories. In

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order to decrease the potential toxicity of this waste, the selective removal of minor actinides in order to destroy them by transmutation, is an important task. The fundamental studies of Nuñez et al. [1] and Kaminski et al. [2] about the magnetically assisted chemical separation technique with extractant-coated magnetic particles [3–5] initiated our alternative strategy of covalent attachment of chelators on the surface of magnetic particles for lanthanide and actinide separation from high activity liquid wastes. These magnetic particle-based techniques avoid the use of large volumes of organic solvent, but combine the selectivity of solvent extraction with improved phase separation in the magnetic field, resulting in a system providing only a small volume of high-level waste. The magnetic particles can be directly vitrified or—even better—stripped, to enable their reuse in an automated process.

The highly efficient, neutral, organophosphorous ligand octyl phenyl *N,N*-diisobutyl carbamoylmethyl phosphine oxide is used in the TRUEx process for extraction of lanthanides and actinides with a selectivity for actinides (e.g. americium (Am)) over lighter lanthanides (e.g. europium (Eu)) [2]. With the covalent binding of calix [4] arenes bearing four CMPO (this abbreviation is used for carbamoylmethyl phosphine oxides in general) groups at the wide rim, it was clearly shown that a preorganisation of CMPO functions on a macrocyclic platform followed by attachment of this assembly to the particle surface, is advantageous [6].

Our new approach to achieve a further increase of the lanthanide and actinide extraction capacity of magnetic particles utilises the introduction of starburst dendrimers on the particle surface. Those dendrimer-coated particles have a high potential for immobilisation of a large variety of selective chelators in a very high density on the particle surface [7]. Here, we report on the synthesis and extraction studies of dendrimer-coated particles bearing CMPO, picolinamide [8], and *N,N*-dimethyldiglycolamide (DMDGA) groups, as tetraoctyldiglycolamide (TODGA) [9] analogous derivatives, on the surface. The extraction studies for Eu and Am were carried out under highly acidic conditions with Eu and Am solutions found

in real waste. The possibility of multiple use of the magnetic particles to save process costs is demonstrated by back-extraction studies. Potential cooperative effects on the efficiency and selectivity of the radionuclide complexation were checked by the introduction of different chelating groups on the surface of dendrimer-coated particles.

2. Methods and results

2.1. Dendrimer-coated magnetic silica particles

Magnetic silica particles were prepared by encapsulation of iron oxide into the pores of highly porous silica particles followed by introduction of amino groups on the particle surface (Fig. 1). These primary aliphatic amino groups were derivatised with cyanuric chloride to provide magnetic particles **M0DCT** having dichlorotriazinyl groups on the particle surface (these particles (product-code: 58-39-105) are available under <http://www.micromod.de>). For the attachment of starburst dendrimers (DAB-Am-16, 32 or 64) **1–3**, which contain 16, 32 or 64 primary amino groups in their outer sphere, the dichlorotriazinyl functionalised magnetic silica particles **M0DCT** were suspended in chloroform and shaken with the dendrimers (50 μmol of dendrimer **1**, **2** or **3** per g of particles) at room temperature in the presence of triethylamine (50 μmol per g of particles) to neutralise/capture the hydrogen chloride. After successive washing of the particles with chloroform, ethanol, and water assisted by magnetic separation, the density of amino groups on the particle surface was measured by streaming potential measurements and spectrophotometric determination using rhodamine B [10].

The streaming potential measurements use polyelectrolyte titrations of the magnetic particle suspensions against a 0.001 N sodium polyethylene sulphonate standard solution. This anionic polyelectrolyte was added to the positively charged magnetic particles until the electric zero point of charge is reached. The amount of consumed polyelectrolyte solution was used to calculate the density of the amino groups on the particle surface (Table 1).

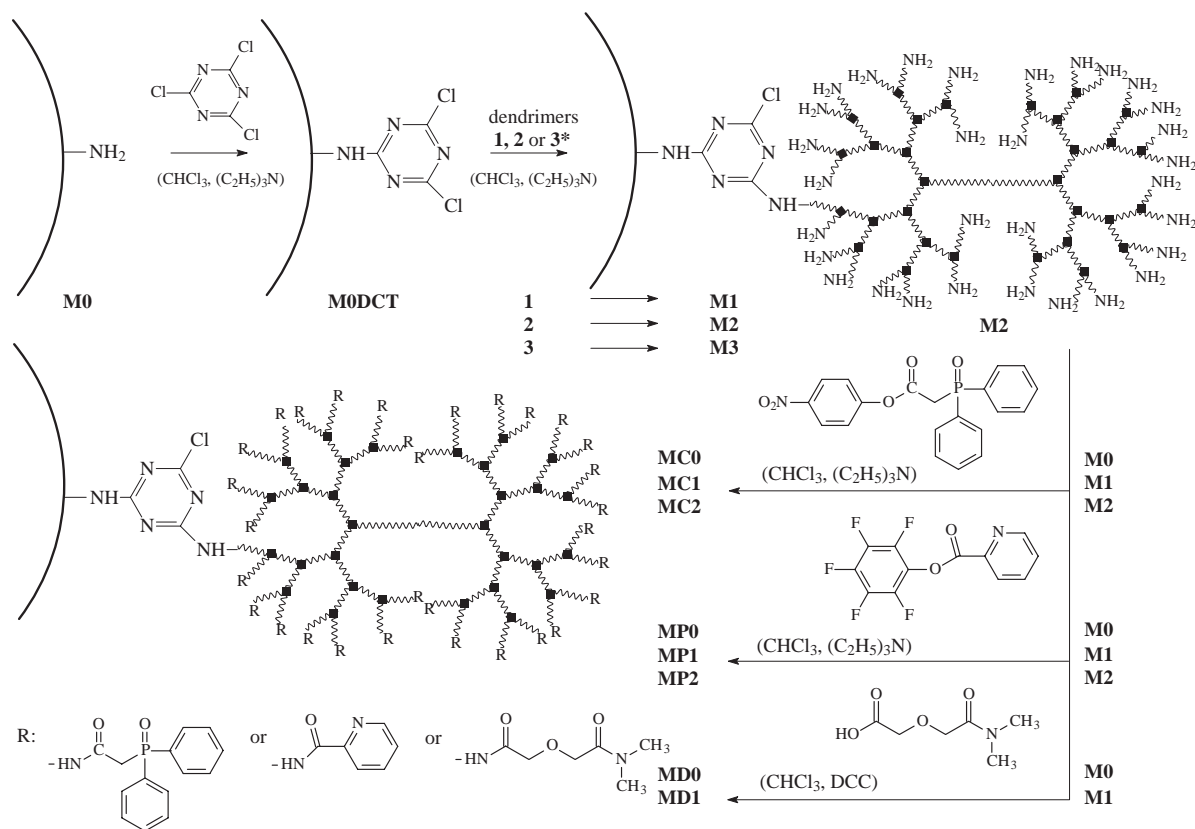


Fig. 1. Attachment of CMPO, picolinamide and DMDGA as TODGA analogous derivative on the surface of dendrimer-coated magnetic silica particles. Only the 4th generation dendrimer-coated magnetic particles (**M2**) were graphically represented for brevity. 1: DAB-Am-16 = polypropylene imine hexadecaamine dendrimer (3rd generation). 2: DAB-Am-32 = polypropylene imine dotriacontaamine dendrimer (4th generation). 3: DAB-Am-64 = polypropylene imine tetrahexacontaamine dendrimer (5th generation).

To determine the total density of amino groups, which are available on the particle surface, activated rhodamine B sulphonyl chloride (Lissamine[®], Molecular Probes) was covalently coupled with amino groups of particles **M0–M3** through a sulphonamide bond. For this, the particles (10 mg) were incubated for 1 h in a solution containing the rhodamine B derivative ($1 \mu\text{mol ml}^{-1}$) and pyridine (1% v/v) in dichloromethane (10 ml) under water-free conditions [10]. The disappearance of rhodamine B from the solution was monitored by absorption measurements at 570 nm. The spectrophotometric method led to higher densities of amino groups of dendrimer-coated particles in comparison to the polyelectrolyte titration (Table 1), because the small rhodamine B derivative can reach better the amino groups of the

Table 1

Comparison of the density of amino groups attached to the magnetic particles **M0–M3** (Fig. 1) measured by polyelectrolyte titration against 0.001 N sodium polyethylene sulphate and by spectrophotometric measurement of the rhodamine B binding [11]

Particle type	Density of amino groups by polyelectrolyte titration ($\mu\text{mol g}^{-1}$)	Density of amino groups by spectrophotometric measurement ($\mu\text{mol g}^{-1}$)
M0	52	55
M1	450–550	830–840
M2	500–590	840–850
M3	490–560	840–850

dendrimers which are not directly on the surface than the large polyelectrolyte molecules (MW: $19,100 \text{ g mol}^{-1}$). The increasing numbers of

terminal amino groups from dendrimer **1** (3rd generation) to dendrimer **3** (5th generation) did not lead to a substantial increase of the available amino groups on the surface of the dendrimer-coated particles **M1–M3** for further chelator binding due to steric problems. Therefore, the binding of chelators for lanthanides and actinides was focussed only on the magnetic particles **M1** and **M2**. Both types of measurements showed a good agreement for the amino group density of the reference particles **M0** without a dendrimer on the surface (Table 1), which supports the above mentioned explanation for the differences between the two determination methods observed with the dendrimer-coated particles **M1–M3**.

2.2. Modification of dendrimer-coated magnetic particles by CMPO

CMPO groups were attached to the surface of dendrimer-coated magnetic silica particles **M1** and **M2** by reaction of *p*-nitrophenyl(diphenylphosphoryl)acetate [11] with the terminal amino groups of the dendrimer-coated particles. To ensure a maximum CMPO density on the particle surface the dendrimer-coated particles (1.2 g) were shaken in chloroform (20 ml) with a large excess of *p*-nitrophenyl(diphenylphosphoryl)acetate (1.5 mmol) in the presence of stoichiometric amounts of triethylamine (Fig. 1). In a similar experiment reference particles **M0** without dendrimers on the surface were coated with CMPO groups. The reactions were monitored by spectrophotometric measurements of the increasing *p*-nitrophenol concentration in the chloroform phase at a wavelength of 310 nm. After 7 days the *p*-nitrophenol concentration remained constant, and the reactions were stopped by successive washing of the particles with chloroform, ethanol, and water, followed by drying in vacuum. UV-spectrophotometric measurements showed that all available amino groups of dendrimer-coated particles **M1** and **M2** which were found by the rhodamine B coupling (Table 1) reacted with *p*-nitrophenyl(diphenylphosphoryl)acetate to give a CMPO density of $820 \mu\text{mol g}^{-1}$ for **M1** particles and $860 \mu\text{mol g}^{-1}$ for **M2** particles.

The distribution coefficient K_D for solid/liquid extractions is defined as

$$K_D = \frac{(c_{L,0} - c_L) V_L}{c_L m_S}, \quad (1)$$

K_D is the distribution coefficient, m_S the mass of magnetic particles, V_L the volume of radionuclide solution, $c_{L,0}$ the initial activity of radionuclide solution, c_L the final activity of radionuclide solution.

Due to saturation phenomena, these K_D values are usually not constant and thus only values obtained under identical conditions (concentration in the liquid phase, amount of solid phase) can be compared. Therefore, all K_D for Eu and Am extraction with magnetic particles were determined under constant conditions: shaking 300 mg of magnetic particles (m_S) with 10 ml (V_L) of Eu or Am comprising a test solution of an activity of about 1400 kBq/L ($c_{L,0}$) for 1 h at room temperature. After magnetic separation of the particles the final radionuclide activity (c_L) was measured in the supernatant by γ -spectrometry. The results of the extraction experiments with CMPO modified particles **MC0–MC2** are summarised in Table 2. These extraction data show that the introduction of dendrimer spacers led to a significant increase of the distribution coefficients for Am and Eu by a factor of 20–50 at pH = 1.0 in comparison to the reference particle **M0** without a dendrimer spacer.

Back-extraction studies were carried out to demonstrate the possibility to recycle the particles. Therefore, 10 successive extraction/stripping steps of Eu from 3 M HNO₃ were studied with the magnetic particles **MC1**. The extraction steps were

Table 2
Distribution coefficients for Am and Eu extraction with microparticles **MC0–MC2** bearing CMPO groups on the surface

Particle type	K_D (Eu) (ml/g)	K_D (Am) (ml/g)	K_D (Am)/ K_D (Eu)
MC0	0.95	1.72	1.81
MC1	23.0	48.5	2.11
MC2	40.4	84.2	2.09

Extraction conditions: 3 M HNO₃ + ¹⁵²Eu + ²⁴¹Am, volume of aqueous phase: 10 ml, mass of particles: 300 mg, stirring time: 1 h, pH = 1.0 and temp.: 25 °C.

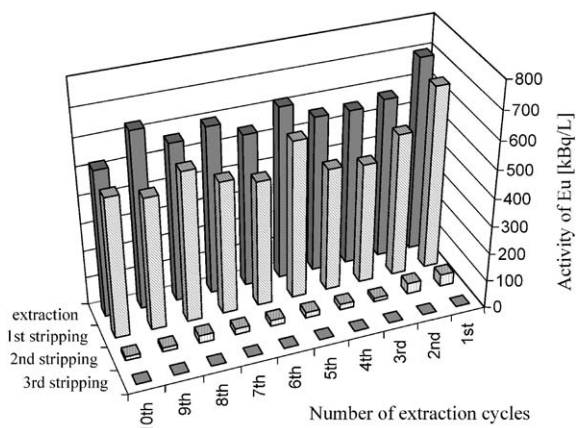


Fig. 2. Activity of extracted and back-extracted ^{152}Eu during 10 extraction/stripping cycles with particles **MC1** ($m_S = 300\text{ mg}$, $V_L = 10\text{ ml}$). After the 1st stripping the pH of the aqueous phase ranged between 1.1 and 1.4, after the 2nd one between 2.3 and 2.8, and after the 3rd one between 2.8 and 3.2.

performed according to the already described extraction experiment with 300 mg of particles **MC1** and 10 ml Eu solution with an activity of 1400 kBq/L. After magnetic separation of the particles Eu was back-extracted from the particle surface by shaking the particles three times with each 10 ml water for 10 min. After magnetic separation of the particles the radionuclide activity in the supernatants was measured by γ -spectrometry.

Generally, more than 90% of Eu activity was recovered by stripping the particles twice with demineralised water without any significant decrease of the Eu extraction capacity during 10 extraction/stripping cycles. The activity of extracted and back-extracted Eu at each step is represented in Fig. 2.

2.3. Modification of dendrimer-coated magnetic particles by picolinamide

Picolinamide groups were introduced on the surface of dendrimer-coated magnetic silica particles **M1–M2** by reaction of picolinic acid pentafluorophenyl ester [12] with the terminal amino groups of the dendrimer-coated particles under the same conditions as described for the **CMPO**

modification (Fig. 1). In an analogous experiment reference particles **M0** without dendrimers on the surface were coated with picolinamide groups, resulting in particles **MP0**.

The extraction properties of picolinamide derivatives towards Eu^{3+} and Am^{3+} strongly depend on the pH value of the radionuclide solution. Therefore, the extraction properties of the picolinamide modified particles **MP0–MP2** were studied under the described standard conditions at different pH values of 1, 2 and 3 (Table 3). The introduction of dendrimer spacers between particle surface and picolinamide chelator led up to a 30-fold increase of the Am and Eu distribution coefficients. The K_D values for Am and Eu are extremely high at pH = 3 (up to 18,000 ml/g), but decrease significantly at lower pH values. This limits their use for the treatment of highly acidic nuclear wastes. Moreover, in some cases, interesting separation factors Am/Eu can be reached.

The K_D values for Eu and Am decrease at the same pH values from the **MP2** to the **MP3** particles with an increasing number of amino groups in the corresponding dendrimer spacers (Table 3). This may result from sterical disturbances of the radionuclide complexation, if the chelator density on the particle surface becomes too high.

Table 3

Distribution coefficients for Am and Eu extraction with microparticles **MP0–MP2** bearing picolinamide groups on the surface

Particle type	pH	K_D (Eu) (ml/g)	K_D (Am) (ml/g)	K_D (Am)/ K_D (Eu)
MP0	3	1780	2580	1.45
	2	31.1	105	3.39
	1	0	0	—
MP1	3	13,930	18,290	1.31
	2	1029	2235	2.17
	1	0	0	—
MP2	3	3300	11,220	3.51
	2	268	724	2.70
	1	0.26	0	—

Extraction conditions: HNO_3 (pH = 3) + ^{152}Eu + ^{241}Am , HNO_3 (pH = 2) + ^{152}Eu + ^{241}Am , HNO_3 (pH = 1) + ^{152}Eu + ^{241}Am , volume of aqueous phase: 10 ml, mass of particles: 300 mg, stirring time: 1 h, temp.: 25 °C.

2.4. Modification of dendrimer-coated magnetic particles by DMDGA

DMDGA groups were introduced as TODGA analogs on the surface of dendrimer-coated magnetic silica particles **M1** by reaction of *N,N*-dimethyl diglycolacid monoamide with the terminal amino groups of the dendrimer-coated particles. Therefore, diglycolic anhydride (5 g, 0.043 mol) was stirred overnight in dimethylamine (7.74 g, 0.172 mol) at room temperature under argon. The reaction mixture was evaporated to dryness giving the *N,N*-dimethyl monoamide of diglycolic acid as a white solid in quantitative yield. This DMDGA acid (1.5 mmol) was dissolved in 20 ml of chloroform and shaken with *N,N'*-dicyclohexylcarbodiimide for 1 h at room temperature. Dendrimer-coated particles **M1** (1.2 g) or reference particles **M0** (1.2 g) were added, and the suspension was shaken for 48 h at room temperature (Fig. 1). The resulting DMDGA modified particles **MD0** and **MD1** were successively washed with chloroform, ethanol, and water, and finally dried in vacuum.

The extraction properties of the DMDGA modified particles **MD0** and **MD1** towards Am^{3+} and Eu^{3+} at $\text{pH} = 1.0$ are given in Table 4. The increase of the Eu and Am distribution coefficients by factors of 350–470 was most pronounced for the introduction of the dendrimer spacer between particle surface and DMDGA derivative in comparison to the CMPO (Table 2) or picolinamide (Table 3) modification at the relevant pH value of 1. In contrast to the

CMPO and picolinamide modified particles with an Am over Eu selectivity of 1.3–3.5 (Tables 2–3) the DMDGA-bearing particles show a higher Eu selectivity with $K_D(\text{Eu})/K_D(\text{Am})$ ratios of 2.8.

Back-extraction studies were carried out with the DMDGA-dendrimer particles **MD1**. Two extraction/stripping cycles were studied using standard conditions for the extraction and three washings with each 10 ml of mineral water for back-extraction. The Am and Eu distribution coefficients do not show any significant decrease of the extraction capacity of the particles.

2.5. Magnetic particles bearing dendrimers with different chelating sites on the surface

First attempts were made to immobilize different types of chelators on the surface of dendrimer-coated magnetic particles to investigate possible co-operative effects on the radionuclide complexation. Thus CMPO groups and the TODGA analogous derivative DMDGA were attached on dendrimer-coated particles with varying CMPO/DMDGA ratios.

A constant mass of particles **M1** (1 g) was suspended in 20 ml of chloroform. After addition of increasing amounts of *p*-nitrophenyl(diphenylphosphoryl)acetate (0–400 μmol , Table 5) and corresponding stoichiometric amounts of triethylamine the particle suspension was shaken for 7 days at room temperature. The density of covalently attached CMPO on the dendrimer-particle surface was measured by monitoring the *p*-nitrophenol release into the liquid phase by UV-spectrophotometry (Table 5). After three times of washing the particles with chloroform and subsequent drying in vacuum, the remaining free amino groups on the particle surface were reacted with carbodiimide activated DMDGA acid. Therefore different amounts of DMDGA acid (400–550 μmol , Table 5) were dissolved in 20 ml of chloroform. After addition of corresponding stoichiometric amounts of *N,N'*-dicyclohexylcarbodiimide the solutions were shaken for 1 h at room temperature and added to the dendrimer particles with different densities of CMPO groups according to Table 5. The suspensions were shaken for 48 h at room temperature, and successively

Table 4

Distribution coefficients for Am and Eu extraction with microparticles **MD0** and **MD1** bearing DMDGA as TODGA analogous groups on the surface

Particle type	$K_D(\text{Eu})$ (ml/g)	$K_D(\text{Am})$ (ml/g)	$K_D(\text{Am})/$ $K_D(\text{Eu})$
MD0	1.48	0.40	0.27
MD1	523	189	0.36

Extraction conditions are given in Table 2. After the 1st extraction with particles **MD1** 95% of the initial ^{152}Eu activity and 90% of the initial ^{241}Am activity were back-extracted from the particles by successive washings with 3×10 ml mineral water. The washed particles were re-used for the 2nd extraction.

Table 5

Comparison of the supplied amounts of *p*-nitrophenyl(diphenylphosphoryl)acetate (CMPO derivative) and carbodiimide activated *N,N*-dimethyl diglycolic acid monoamide with the spectrophotometrically (analytical) measured amounts of covalently bound ligands at the attachment of the two different ligands in varying ratios on the dendrimer-coated magnetic particles **M1**

Particle type	c (CMPO) ($\mu\text{mol/g}$) (supplied)	c (CMPO) ($\mu\text{mol/g}$) (analytical)	c (DMDGA) ($\mu\text{mol/g}$) (supplied)	c (DMDGA) ($\mu\text{mol/g}$) (analytical)	c (CMPO + DMDGA) ($\mu\text{mol/g}$) (analytical)
MCD1-1	400	395	0	0	395
MCD1-2	200	199	400	144	343
MCD1-3	100	99	550	307	406
MCD1-4	50	50	550	358	408
MCD1-5	25	25	550	490	515
MCD1-6	12.5	12.5	550	490	502
MCD1-7	6.25	6.25	550	511	517
MCD1-8	0	0	550	440	440

Table 6

Distribution coefficients for Am and Eu extraction with microparticles **MCD1-1–MCD1-8** bearing CMPO and TODGA analogous groups (DMDGA) with varying ratios on the particle surface (extraction conditions are given in Table 2)

Particle type	Immobilized chelators % CMPO/% DMDGA (analytical)	K_D (Eu) (ml/g)	K_D (Am) (ml/g)	K_D (Am)/ K_D (Eu)
MCD1-1	100/0	129	163	1.27
MCD1-2	58/42	87	88	1.01
MCD1-3	24/76	125	92	0.74
MCD1-4	12/88	130	92	0.71
MCD1-5	4.8/95.2	162	98	0.60
MCD1-6	2.5/97.5	128	70	0.55
MCD1-7	1.2/98.8	127	58	0.45
MCD1-8	0/100	170	54	0.31

washed with chloroform, ethanol, and water, followed by drying of the particles in vacuum. The amount of covalently attached DMDGA on the dendrimer-particle surface was measured by monitoring its disappearance from the liquid phase by UV-spectrophotometry at 243 nm (Table 5).

The Am/Eu extraction results show that increasing amounts of DMDGA groups lead to an increasing selectivity of the particles for Eu over Am, which is typical for particles containing only the DMDGA functions on the surface (Table 6). In comparison to the synthesis and extraction studies of the particles **MC1** with only CMPO groups (Table 2) or **MD1** with only DMDGA groups (Table 4) on the surface of the dendrimer particles, the introduction of a mixture of CMPO and DMDGA groups did not result in a complete saturation of the binding sites of the dendrimer

particles **M1**. It is interesting that a lower concentration of *p*-nitrophenyl(diphenylphosphoryl)acetate of 400 μmol per g particles **MCD1-1** leads to higher Am and Eu distribution coefficients (Table 6) than a large excess of 1.25 mmol per g particles **MC1** (Table 2). Apparently, there is an optimal density of chelating CMPO sites on the particle surface to achieve a maximum radio-nuclide binding. If the density of CMPO sites becomes too high the co-ordination sites of the neighbouring CMPO groups may disturb each other. For the corresponding particle modification with DMDGA, the Am and Eu distribution coefficients increase with higher densities of DMDGA groups on the particle surface (Tables 4 and 6). No disturbance of the extraction process could be detected by complete saturation of the **M1** particle surface with DMDGA derivatives. At

a ligand ratio of 58% CMPO/42% TODGA the lowest distribution coefficient for Eu was measured, and the selectivity between Am and Eu was lost (Table 6). Finally, the distribution coefficients for Am and Eu could be improved by combination of CMPO and DMDGA derivatives on the particle surface, if the percentage of CMPO on the particle surface is higher than 4%.

Analogous experiments were carried out by combining the TODGA analogous derivative with picolinamide structures on the M1 particle surface, or with CMPO and picolinamide structures. In all cases a decrease of the extraction capacity of the dendrimer-coated magnetic particles was observed in comparison with the dendrimer-coated particles bearing only one type of chelator.

3. Conclusions

The covalent attachment of dendrimers with a high number of terminal amino groups on the surface of magnetic silica particles allowed the introduction of selective chelators for radionuclides in a high density. The dendrimer-coated particles were used as universal platform for the covalent binding of chelator molecules like CMPO, picolinamide or TODGA analogous amides. The introduction of the dendrimer spacer led to a 50–400-fold increase of the distribution coefficients for the Eu and Am extraction in comparison to the reference particles without the dendrimer. Picolinamide derivatised dendrimer particles have a low pH stability of the Am and Eu complexes at $\text{pH} < 3$ but a slightly higher Am/Eu selectivity. This limits their usefulness in nuclear waste remediation for the treatment of highly acidic nuclear wastes.

Dendrimer-coated particles were functionalised also with two different types of chelators to study possible co-operative effects of the radionuclide complexation on the particle surface. The variation of the CMPO/DMDGA ratio on the surface of the dendrimer particles did not influence the size range of the Am and Eu distribution coefficients significantly in comparison to the dendrimer

particles bearing only CMPO or only DMDGA on the surface. However, the selectivity for Am over Eu, which is typical for CMPO dendrimer particles, was reduced by increasing DMDGA/CMPO ratios until an inversion of the selectivity to Eu over Am was reached.

Back-extraction experiments with CMPO- or DMDGA-bearing dendrimer-coated magnetic particles demonstrated the possibility of multiple use of the magnetic particles, which makes their application for radionuclide extraction from nuclear wastes an interesting alternative to conventional liquid–liquid extractions.

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