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Design of radionuclide separations based on MD simulations

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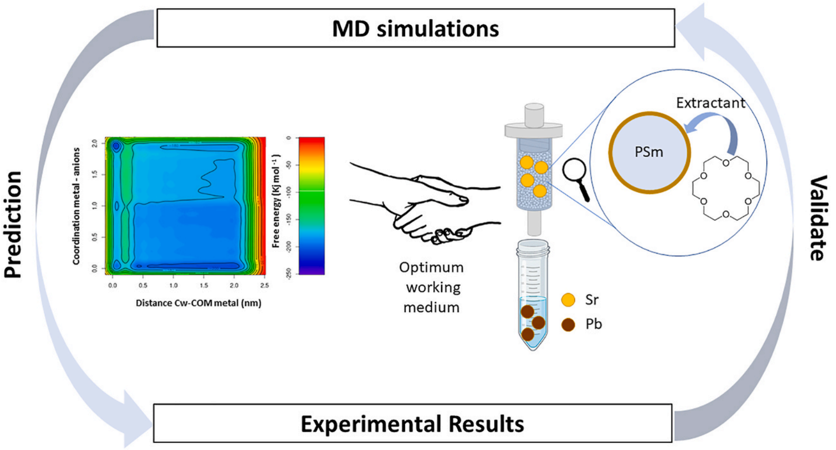
HIGHLIGHTS

- New methodology for radionuclide separation desing.
- MD simulations for ^{90}Sr and ^{210}Pb separation with crown-ether resins.
- Thermodynamic understanding of the anion affinity effect.

GRAPHICAL ABSTRACT



ABSTRACT

Background: The development of selective materials for radionuclide separation is often a long and costly process, requiring labor-intensive chemical separations and extensive optimization. To streamline the development of selective materials, this study explores MD simulations to accelerate the identification of optimal separation conditions, extractant-radionuclide affinity, reducing the need for extensive experimental trials.

Results: We assessed the effectiveness of MD simulations using the challenging system of Sr^{2+} and Pb^{2+} with 18-crown-6 crown ether, focusing on the influence of different working media, including nitric, hydrochloric, formic, acetic, and perchloric acids, as well as potassium thiocyanate. The simulation results were validated experimentally by measuring the distribution weight ratios (D_W) of Sr^{2+} and Pb^{2+} using crown ether immobilized on a polymeric surface. Our findings demonstrate a strong correlation between MD predictions and experimental data, particularly highlighting acetic acid as a medium where Sr^{2+} forms stable complexes, while Pb^{2+} does not.

Significance: This study confirms the suitability of MD simulations as a reliable tool for predicting the selectivity of extractants, enabling faster development of new scintillating and non-scintillating resins. By reducing the time and resources needed for experimental optimization, this approach offers a more efficient pathway for the development of advanced materials for radionuclide separation.

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1. Introduction

Liquid scintillation counting (LSC) is one of the primary techniques used for the determination of radionuclides, especially for beta and alpha emitters [1,2]. LSC is highly valued for its simplicity and high detection efficiencies. However, it lacks selectivity, requiring labour-intensive, costly, and time-consuming chemical separation processes to isolate the target radionuclide from interfering species present in the sample. These separation procedures frequently involve solid-phase extraction using chromatographic resins, which, under optimized conditions, can selectively isolate the radionuclide of interest [3–8]. After separation, the solution containing the isolated radionuclide is mixed with a scintillation cocktail for measurement in the detector.

Despite the existence of well-established procedures for the accurate isolation and quantification of many radionuclides, there remain challenges in determining certain radionuclides due to the absence of effective separation procedures and selective materials. Examples of these challenging systems include the separation of ^{55}Fe from ^{60}Co or ^{63}Ni, ^{90}Sr from ^{210}Pb, and ^{241}Am from ^{152}Eu or ^{244}Cm. Furthermore, conventional LSC methods generate mixed hazardous and radioactive wastes which are difficult and costly to manage, posing significant environmental concerns.

To address these limitations and develop more efficient analytical strategies, plastic scintillation resins (PSresins) have emerged over the past decade [9,10]. PSresins consist of a selective extractant immobilized on the surface of plastic scintillation microspheres (PSm) [11]. These resins integrate the chemical separation and measurement steps into a single process, eliminating the need for elution and thus reducing sample preparation complexity and eliminating the mixed waste generation. However, potential interferences have to be completely removed from the cartridge performing rinses to avoid activity overestimation, meaning that separation steps must be adapted to the PSresin working mode. Nevertheless, PSresins, as well as standard resins, are not available for all radionuclides of interest, meaning that challenges related to chemical separation persist despite the reduction in waste generation.

The development of new selective materials and new chemical separation procedures involves multiple stages, each requiring significant time, labour, and resources. Initially, the extractant, must be synthesized with enough yield and purity, and immobilized on an inert or scintillating support, in the optimal proportion, to produce the selective material. Subsequently, the interaction between the radionuclide and the extractant is studied to determine its affinity. The efficacy of the target radionuclide's extraction is influenced by the composition of the working medium, requiring extensive batch studies to identify optimal conditions. Once an effective working medium is established, potential interferences are studied to evaluate the selectivity of the material and determine the achievable decontamination factors [9,12–16]. Following these analyses, the separation process is optimized by determining the appropriate loading medium and additional rinses needed to retain the target radionuclide while removing interferences. This process can take several months or even years due to the long duration of required experiments and their inability to be conducted in parallel, some of them.

To streamline this development process, computational simulations can be employed to explore extractant molecules, understand interaction mechanisms, and assess the influence of different working media before initiating experimental work, thereby saving time and reducing costs. Techniques such as Density Functional Theory (DFT) and Molecular Dynamics (MD) simulations are particularly valuable in this regard. DFT provides detailed insights into the electronic interactions between radionuclides and extractants, helping to predict binding affinities and selectivity. However, DFT is limited to static, idealized conditions without accounting for temperature, pressure, or solvent effects. In contrast, MD simulations can model the dynamic behaviour of these systems, including the impact of temperature and solvent interactions, which are crucial for realistic separation processes. Consequently, in this work, the suitability and accuracy of MD simulations for the development of selective resins are assessed by predicting the affinity of an extractant for a radionuclide, in various working media, and validating the results experimentally. The study focuses on the separation of Sr^{2+} and Pb^{2+} using crown ethers. This system is very well known, with the Sr-resin being widely used for the separation of ^{90}Sr from common interferences in strong HNO_3 [17]. While Pb^{2+} is more strongly retained than Sr^{2+}, this does not pose an issue, as ^{90}Sr can be selectively eluted in low HNO_3 conditions. ^{90}Sr analysis with crown ether PSresin is also feasible with the advantages of no mixed waste generation [10,18]. However, when ^{210}Pb is also present, it cannot be rinsed without removing ^{90}Sr. Consequently, the measurement of ^{90}Sr in the presence of ^{210}Pb is not possible unless a previous precipitation separation is applied [19]. The theoretical study of the interaction of Sr^{2+} and Pb^{2+} using crown ethers in different media will be not only useful to determine in which medium ^{90}Sr can be separated from ^{210}Pb in a crown ether PSresin, aiming for environmental waste samples such as contaminated water, soil, and effluents from nuclear power plants, but also to evaluate if this strategy could be valid for the design of new extractants and methods for radionuclide separation as well as to understand the mechanism of interaction between extractant and radionuclides.

Previous computational studies using DFT have explored the stability of crown ethers within various alkaline elements, focusing on the influence of cavity size and electron-withdrawing effects on their structures [20–23]. While these approaches provide precise electronic insights, they are typically conducted at 0 K, neglecting critical factors such as water solvation and entropic contributions. These factors are essential for a complete understanding, as they significantly impact stability and reactivity at non-zero temperatures. Thus, the final ion-crown ether conformation predicted by DFT may not accurately reflect the behaviour under more realistic conditions that include solvent and temperature effects.

Therefore, the present study uses MD simulations to evaluate the binding affinities of Sr^{2+} and Pb^{2+} with 18-crown-6 ether across different working media. This approach provides a more realistic representation of the interactions, accounting for solvent environments, conformational flexibility, and thermal effects. Six working media (nitric acid, hydrochloric acid, formic acid, acetic acid, perchloric acid, and potassium thiocyanate) have been simulated and subsequently validated experimentally using the 18-crown-6 ether immobilized on an inert polymeric surface. The simulation results are compared with experimental distribution weight ratios (D_w), a widely used metric for assessing the affinity of metals for resins in specific media.

2. Results & discussion

2.1. Studied system

We investigate the stability of the complexes formed between the 18-crown-6 crown ether and Sr^{2+} or Pb^{2+} in six different working media that are of common use in laboratories: nitric acid (HNO_3) the common working media for ^{90}Sr separations with crown ethers, hydrochloric acid (HCl), formic acid (HCOOH), acetic acid (CH_3COOH), perchloric acid ($HClO_4$), and potassium thiocyanate (KSCN).

18-crown-6 is the core section of the crown ether commonly used in strontium selective resins, (4,4′(5')-di-*t*-butylcyclohexano-18-crown-6). This section is selected since it constitutes the essential binding component with the ion (refer to the top panel of Fig. 1). The presence of substituents on the crown ether can influence the binding affinity, depending on the functional groups of the substituents, but the general trend will remain the same, especially if the substituents are based on alkenes chains [24,25]. In a similar manner, to simplify the simulation, no diluents were considered in spite that it may have also some influence. By this way, focusing only on the essential part of the crown ether, the computational cost of the assessment of the theoretical procedure may be reduced, providing a simple but efficient tool to study extractant-metal interaction in aqueous medium in presence of anions.

18-crown-6 consists of a cyclic arrangement of six ethylene oxide (–CH_2CH_2O–) units, forming a flexible macrocyclic ligand. The six oxygen atoms in the ring, each with lone pairs of electrons, are strategically positioned to coordinate with a central cation via ion-dipole interactions (see the middle panel of Fig. 1). The cavity size of 18-crown-6, defined by the effective internal diameter of the macrocyclic ring, is approximately 2.6–3.2 Å. The geometrical compatibility between the cation and the cavity, coupled with the optimal alignment of the oxygen atoms' electron lone pairs, results in the formation of highly stable complexes.

In general, crown ethers are known for their inherent flexibility, which allows them to adapt their cavity size to accommodate ions of varying sizes. This flexibility has been studied extensively, particularly through DFT simulations, which have shown that crown ethers can modify their structure to host ions ranging in size from small lithium ions to larger cesium ions. In the case of 18-crown-6, its flexible backbone enables it to fold and adjust in response to the ionic radius of the guest cation, effectively trapping it within the cavity. Studies in the gas phase have demonstrated that this unrestricted flexibility allows crown ethers to achieve optimal conformations for ion binding, resulting in a strong complexation affinity for ions across a wide range of radii [26,27].

However, the flexibility of 18-crown-6 is influenced by environmental conditions, such as solvent and temperature. In solution, the presence of solvent molecules and thermal motion introduces some restrictions on the crown ether's ability to fold, slightly limiting the extent to which it can adapt its structure [26]. Even with these restrictions, 18-crown-6 retains enough conformational adaptability to form stable complexes with ions of similar ionic radii, such as Sr^{2+} and Pb^{2+}, as demonstrated in various studies [21,23,28–32].

2.2. Free energies landscapes of crown ether-metal complexes in solvent

Free energy calculations are used to estimate the Gibbs free energy change associated with a specific process. In this case, the binding of metal ions (like Pb^{2+} or Sr^{2+}) with crown ethers in the presence of anions in the solvent, provides a measure of the binding strength between molecules. A negative value for the Gibbs free energy change indicates that the process is thermodynamically favorable (the metal ion and the crown ether form a stable complex). The larger the absolute value obtained the stronger and more stable the interaction is, suggesting strong binding affinities. In practical terms, a higher binding energy makes the metal-crown ether complex more stable, which can influence its behavior in processes such as separation or purification. The presence of counter anions due to the working media, can enhance or not this binding by stabilizing the complex, further increasing or decreasing the affinity between the crown ether and the metal ion [29]. Therefore, by understanding these free energy values, we can predict how different the crown ethers will behave in practical applications.

2.2.1. Simulation process definition: nitrates as counterion

We initially conducted a 400 ns molecular dynamics (MD) simulation of Sr^{2+} or Pb^{2+} together with 18-crown-6 and two nitrate molecules, solvated with approximately 2500 SPC/E water molecules. The 12-6 Lennard-Jones is a widely used potential model for nonbonded interactions, however, when dealing with highly charged metal ions, it faces significant limitations, as it does not account for ion-induced dipole interactions, underestimating the strength of ion-water interactions and, as a consequence, lacking in accuracy for hydration free energy (HFE), ion-oxygen distance (IOD) and coordination number (CN). For that reason, Mertz and co-workers [33] developed the 12-6-4 Lennard-Jones potential, which corrects the classical model with the inclusion of an extra attractive term which provides a more realistic representation of highly charged metal ions. For Sr^{2+} and Pb^{2+}, switching to the 12-6-4 model can improve accuracy in specific areas such as hydration energies and ion coordination, but the differences may not be as large as those observed with M^{3+} or M^{4+} ions. Therefore, given their availability, 12-6-4 LJ potentials were used.

The temporal evolution of the coordination between the metal ions (Sr^{2+} or Pb^{2+}) and the crown ether was studied using the PLUMED plugin to calculate the distance between the centers of mass (COM) of the crown ether and the metal ion. Fig. S1 in Supplementary Information indicate that during this 400 ns period, the Sr^{2+} ion does not approach the crown ether within the expected binding distance (~0.05 nm COM separation). This suggests that binding did not occur, likely due to insufficient sampling; the system may be trapped in high-energy states separated by large energy barriers. Given this lack of sampling, we concluded that unbiased simulations alone could not provide a converged picture of the Sr^{2+}-crown ether interaction. Metadynamics was therefore employed as a way to accelerate sampling of rare events and overcome high-energy barriers. By adding a history-dependent bias to the system, metadynamics allows us to explore configurations that are

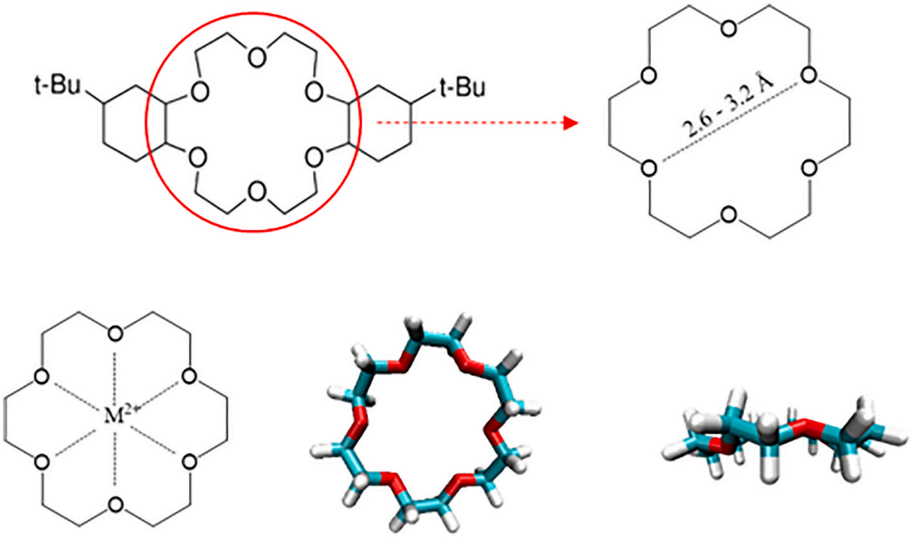


Fig. 1. Top panel, representation of the crown ether used in CE-PSresin, and the core crown ether used in the simulation. Middle panel interaction of the metal with the oxygen of the crown ether and the geometry of the 18-crown-6 from upper and side view.

rarely accessed in unbiased MD.

To have a complete exploration of the phase space of the system, we performed a 2 μs metadynamics simulation, based on the configurations obtained from the initial MD production run. To evaluate the affinity of each metal and the influence of the anions, we analyzed two collective variables: the distance between the centers of mass of the crown ether and the metal ion, and the coordination number between the metal ion and the two nitrate anions.

The free energy landscape was reconstructed using RStudio with the MetadynMiner and MetadynMiner3 libraries [34]. The middle panel of

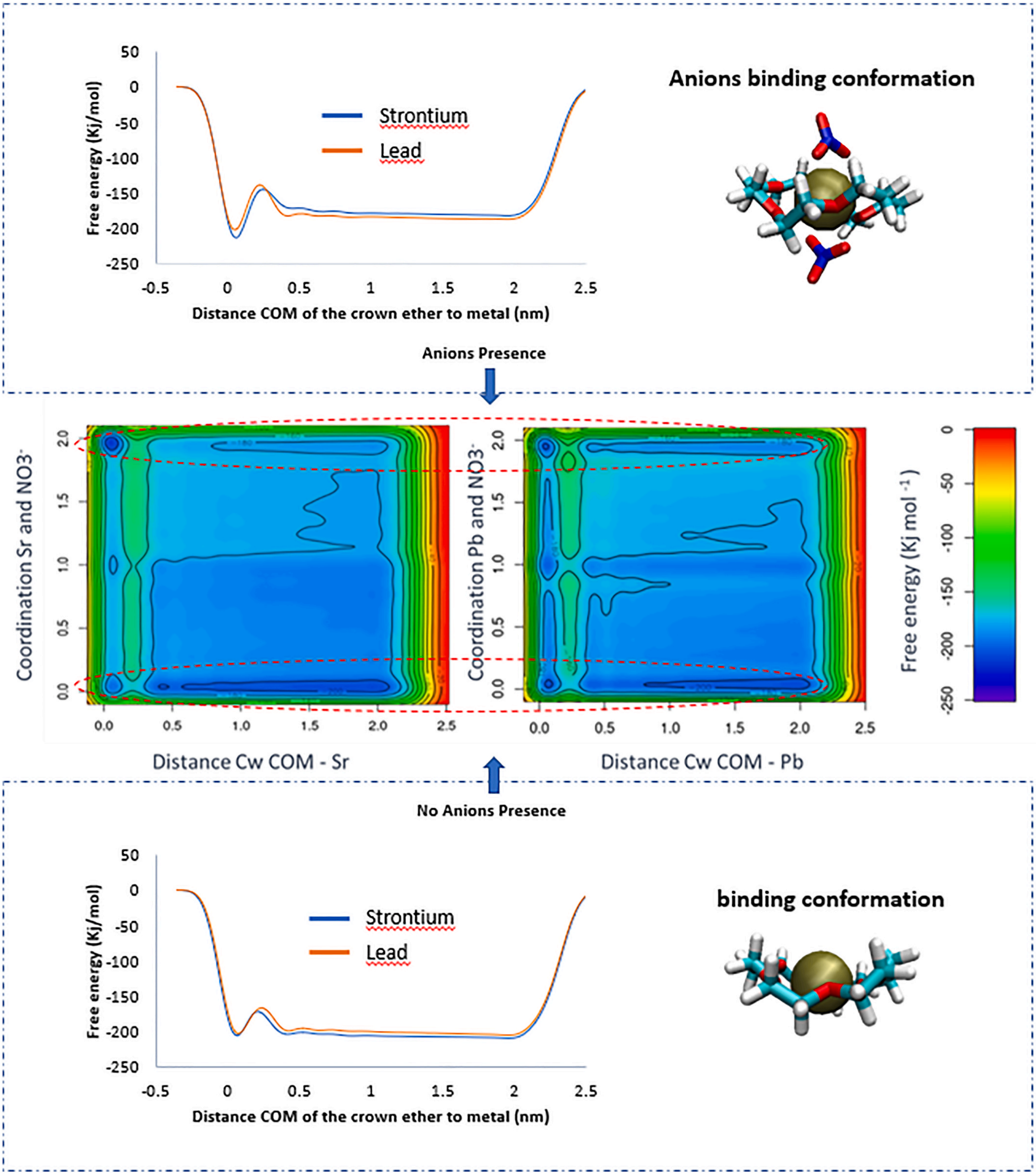


Fig. 2. Top panel, evolution of the free energy at presence of nitrates in the bonding of the metal with the 18-crown-6, along with the snapshot of the conformation of the complex in the bounding situation. Middle panel, free energy landscape for strontium and lead considering the coordination of the metal and the nitrates and the distance between the center of masses of the 18-crown-6 and the metal. Bottom panel, evolution of the free energy without the presence of nitrates in the bonding of the metal with the 18-crown-6.

Fig. 2 shows the free energy profiles obtained for strontium and lead, respectively. To provide a clearer interpretation, we extracted the data corresponding to two specific coordination states: coordination number 2 (presence of anions) and coordination number 0 (absence of anions). The resulting 1-D profiles are presented in the top panel (presence of anions) and bottom panel (absence of anions) of Fig. 2.

In both scenarios, Sr^{2+} and Pb^{2+} exhibit similar affinities for 18-crown-6, consistent with their known strong retention in crown ether-based resins. Additionally, the presence of nitrate anions was found to enhance the affinity between the metal ions and the crown ether and do not compete with the crown ether for direct coordination with the metal ion. Instead, their presence serves to balance the charge and support the solvation environment [29,35]. Both systems showed similar complex conformations as depicted in the structures extracted from the simulation, with the metal ion positioned centrally within the crown ether and nitrates axially co-complexing the cation, proving the reported stable complex conformation. Concluding that metadynamics simulations, under the chosen description of the system, accurate describes the system in concordance with literature for what reported theoretically and experimentally regarding the complex conformation shape and stability in the presence of nitrate anions.

2.2.2. FES of metal-crown ether complexes using different counterions

Given the difficulties in separate Sr^{2+} and Pb^{2+} using nitrates as counterions and motivated to study the effects of the anion's presence, we analyzed the free-energy landscape of the cation-crown ether compounds with five more working media (chloride, perchlorate, acetate, formate and thiocyanate) for Sr^{2+} and Pb^{2+}, these ions were selected according to the literature, based on their capability of enhance the binding of Sr^{2+} or Pb^{2+} with crown ether, among those that present a reasonable cost, safety and their solutions are colourless [2–4].

Anions help to balance the positive charge of the metal ion, thereby reducing electrostatic repulsion within the complex, also, the size and polarizability of the anions can affect the solvation environment. Larger, more polarizable, anions can disrupt the solvation shell more effectively, and can enhance the complex's overall stability by lowering the free energy of the system. Additionally, geometric factors play a role in how anions interact with the metal ion-crown ether system. Finally, the geometry of anions, can influence the final complex as it affects the previously mentioned flexibility of the crown ether, thus affecting its binding affinity and selectivity [36,37].

Fig. 3 shows the 2-D profiles of the free energy landscape, for all the considered anions, focusing on the anion presence (the CN = 2 section). The left panel corresponds to the strontium system and the right panel to the lead system. For strontium the global minimum is found in all cases in the bonding situation (0.03 nm), while the lead system, only has a favorable binding with nitrate and formate. Given the obtained results, the separation of Sr^{2+} and Pb^{2+} could be achieved by retention of the Sr^{2+} and elution or directly not retention of Pb^{2+} in a crown ether-based resin in acetate medium.

2.3. Experimental validation

In order to test the effectiveness of the proposed theoretical procedure for the prediction of the interaction between a metal (or radionuclide) and an extractant (typically organic molecules) in different working media, we compared the agreement between the binding affinities predicted in simulations with the experimental ones, obtained through batch studies.

The free energy difference (ΔF) between the unbound and the bound situations, for each metal-crown ether complex in each anionic medium (NO_3^-, Cl^-, $HCOO^-$, CH_3COO^-, ClO_4^-, and SCN^-), was used to quantify the simulated binding affinity. A more negative ΔF value indicates a more favorable binding interaction. On the other hand, to get an experimental measure of the binding affinity, we performed batch studies for all the systems which were analyzed theoretically and we measured the distribution weight ratio (D_w). This experimental parameter is a measure of the affinity between the target cation (Sr^{2+} or Pb^{2+} in this case) and the crown ether within the studied medium. A high D_w value indicates strong affinity between the target metal and the extractant under specific conditions.

Fig. 4 presents a comparison between the computed free energy differences and the experimentally determined D_w values. A strong inverse correlation was observed for both metals: a negative ΔF (indicating a more favorable binding interaction) corresponds to a positive D_w indicating a strong affinity between the metal and the crown-ether in both cases. The overall trends observed in the simulations were consistently reflected in the experimental results, demonstrating the effectiveness of simulations of predicting the experimental behavior.

The single exception to this correlation was observed for the lead-crown ether complex using perchlorate as counterion, where simulations predicted an unfavorable binding interaction, but, experimental results indicated the presence of weak binding. Nevertheless, both values are small and discrepancy can be interpreted as a consequence of a weak interaction and the uncertainty of both determinations.

Although a clear correlation was observed between the experimental and simulation results, the D_w values obtained in this study are relatively low compared to those of fully optimized resins, where a D_w value greater than 10^2 is indicative of high affinity.

Another important observation is that it is experimentally confirmed that the crown ether has the highest affinity for Sr^{2+} when acetate is used as counterion, while there are no evidences of experimental binding between the Pb^{2+} and the crown-ether confirming the lack of affinity.

Acetate could thus be the optimal medium in which Sr-selective retention can be achieved using a PSresins. It should be noted that this

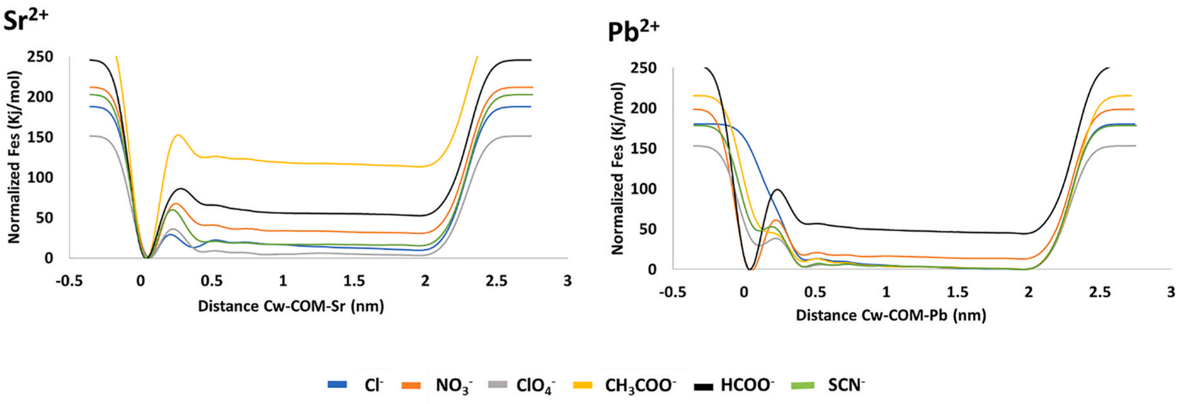
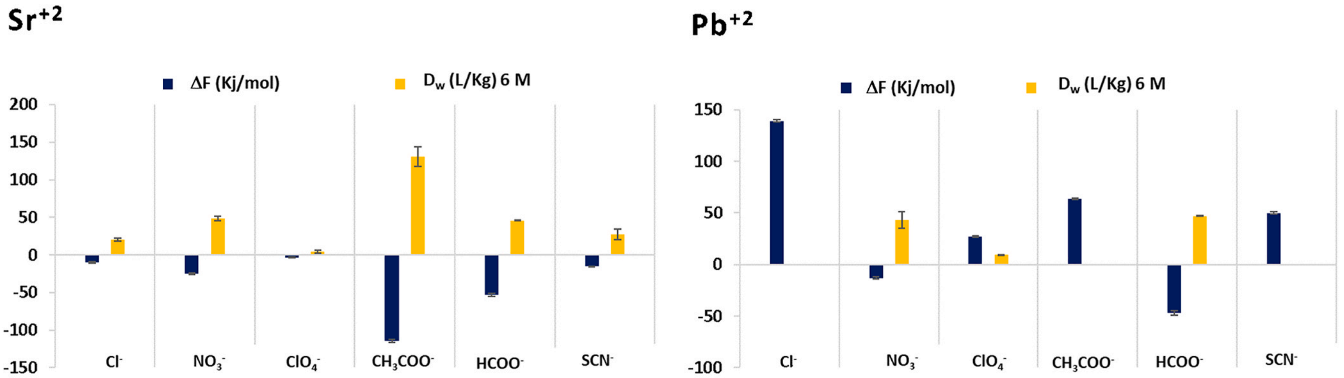


Fig. 3. Free energy profile for each system studied in presence of anions. Left panel refers to strontium and right panel to lead.



ΔF (Kj/mol)	Cl^-	NO_3^-	ClO_4^-	CH_3COO^-	$HCOO^-$	SCN^-
Strontium	-10(1)	-25(1)	-3.5(0.9)	-114(2)	-53(2)	-15.4(0.9)
Lead	139.24(0.8)	-19.1(0.9)	26.9(0.9)	64(1)	-47(2)	50(1)

D_w (L/Kg)	Cl^-	NO_3^-	ClO_4^-	CH_3COO^-	$HCOO^-$	SCN^-
Strontium	20 (2)	47(3)	4(2)	131(13)	45.8(0.8)	27(7)
Lead	<0.5	43(8)	9.1(0.6)	<0.5	47.0(0.6)	<0.5

Fig. 4. Correlation between the differences in free energy and the experimental Dw for strontium (left panel) and lead (Right panel). Left table differences in free energy for strontium and lead. Right table Dw for Strontium and lead.

trend could be affected by the typical diluents and polymers used with the crown ethers in selective resins, and therefore, it has to be tested in a more realistic system.

2.4. *Thermodynamic analysis on the acetate system*

To understand why strong binding is observed with strontium when using acetate as the anion, but not with lead, we analyzed the free energy, internal energy and entropy of the systems. The thermodynamic parameters have been calculated for each system, taking as a reference point the non-bounding non-acetate situations, moving to the metal-crown ether binding in absence of acetate, and finally studying the effect of the acetate in the complex formation (Table 1).

The binding of the metal ion to the crown ether alone resulted in an unfavorable condition, characterized by a positive free energy difference. However, when acetate anions are introduced into the system, there is a noticeable internal energy penalty in both cases, accompanied by an increase in entropy.

In the strontium system, the increase in entropy compensates for the internal energy penalty, leading to a favorable overall free energy change. In contrast, in the lead system, the entropy gain is insufficient to overcome that penalty, resulting in an unfavorable situation. This explains why, in an acetate medium, lead does not bind to the resin, whereas strontium does.

The addition of acetate anions to the Sr^{2+}-crown ether complex leads to a favorable increase in entropy due to the release of water molecules from the solvation shells of strontium and the acetates. This entropy gain offsets the moderate internal energy penalty, resulting in a favorable free energy change. In the case of lead, the strong initial binding to the crown ether and the significant structural reorganization required to accommodate the acetates incur a large internal energy penalty. The entropy gain from desolvation is insufficient to compensate the internal energy cost, leading to an unfavorable overall free energy change. Therefore, the formation of a stable Sr^{2+}-crown ether-acetate complex is more favored due to the flexible coordination environment of strontium compared to the Pb^{2+}-crown ether-acetate complex, which is hindered by lead's substantial internal energy penalty [35,38]. These observations suggest that temperature plays a critical role in modulating the stability of these metal-crown ether complexes. For strontium, the favorable entropy gain indicates that higher temperatures could enhance the stability of the Sr^{2+}-crown ether-acetate complex, as increased temperature amplifies the entropic contribution. In contrast, for lead, the significant internal energy penalty remains dominant at elevated temperatures. Since the entropy change for the Pb^{2+}-crown ether-acetate complex is insufficient to offset the positive change, increasing the temperature will not improve its stability, resulting in either unfavorable or only weakly favorable complex formation.

Table 1

ΔF, ΔU and -T ΔS for strontium and lead, taking as a reference point the non-bounding non-acetate situations with the crown ether (Cw), moving to the metal-crown ether binding in absence of acetate, and to the metal-crown ether-acetate binding. In Kj/mol.

	No acetate			Acetate		
	ΔF	ΔU	-TΔS	ΔF	ΔU	-TΔS
Sr^{2+}-Cw	26 (3)	−33 (8)	59 (9)	−110 (2)	6 (3)	−116 (4)
Pb^{2+}-Cw	62 (2)	24 (7)	38 (7)	42 (3)	82 (5)	−40 (6)

Checking the metal-crown ether-acetate binding conformation, the Sr^{2+}-crown ether-acetate system showed an expected complex conformations as depicted in the structures extracted from the simulation (Left panel, Fig. 5), with the Sr^{2+} ion positioned centrally within the crown ether and acetates axially co-complexing the cation. However, in the case of lead, the acetates are in cis conformation (both acetates from the same side) co-complexing the cation (right panel, Fig. 5). This resulting binding conformation for Pb^{2+}-crown ether-acetate is thermodynamically less stable as it has been seen and this could be explained due to repulsion and steric hindrance effects between both acetates molecules not leading to a very stable compound.

From the coordination point of view, strontium ions typically prefer higher coordination numbers, usually around 7 to 8, which allows for a more flexible coordination environment [39]. This flexibility allows the interaction of the metal both with the crown ether and with the acetate anions without significant disruption of the overall complex. Additionally, the strontium and acetate interactions are mainly ionic; reason why they can integrate into the coordination sphere without causing major distortions or coordination competition [40,41]. In contrast, lead ions generally prefer lower coordination numbers, around 4 to 6, even though can get to larger coordination numbers [42–44], and can have a more covalent interactions than strontium, due to the inert pair effect. This preference might result into a less flexible coordination sphere. When acetates attempt to coordinate with lead, they can disrupt the Pb^{2+}-crown ether interactions, introducing significant steric and electronic strain. In addition, lead ions can have a stronger interaction with acetates, which can thus compete directly with the interactions between lead and the crown ether [45–48], not leading to a stable conformation.

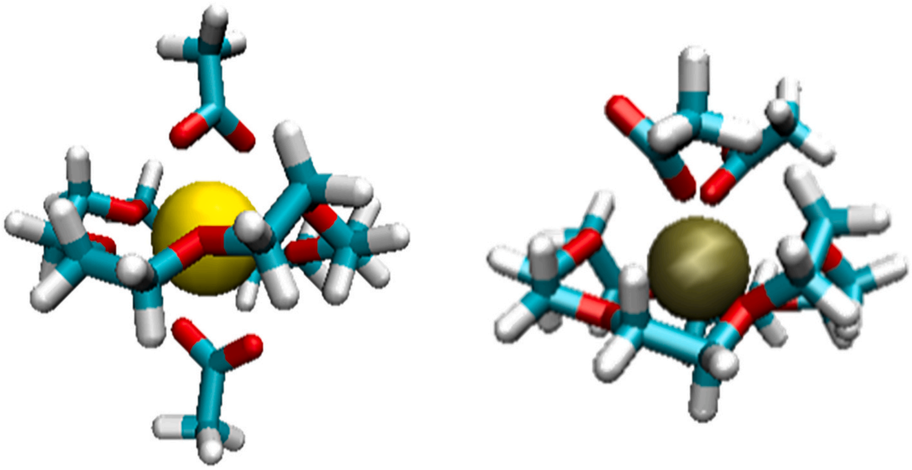


Fig. 5. Snapshots in binding situation in presence of acetate. Left-panel for strontium and right panel for lead.

3. Conclusions

The use of all-atoms MD simulations provides a useful support in understanding the medium effect on the stability of the metal-crown ether complex. Metadynamics, in particular, allows the identification of a suitable working medium where the highest affinity between the target element and extractant is achieved.

The correlation between experimental and simulation results confirms that molecular dynamics is an effective tool for predicting experimental binding affinities. Compared to other simulation strategies, the MD is more realistic as it includes the water molecules, allowing to consider the effect of solvation they caused.

The results obtained are also in agreement with literature in what regards nitric acid, the medium in which both cations are complexed with the crown-ether. Regarding the other anions studied, all present similar behavior for both cations, except in acetic medium. In the system formed by Sr^{2+} or Pb^{2+} with 18-crown-6, when using acetate as counterions, the Sr^{2+}-crown ether complex has the highest stability while the Pb^{2+}-crown ether compound is not stable. This result suggests the possibility to use acetate as medium to remove Pb^{2+} from crown-ether PSresins making possible the measurement of ^{90}Sr in the presence of ^{210}Pb in the sample.

The established methodology is a straightforward strategy that considers the main parameters affecting the affinity between a target element and extractant with low statistical errors. It can be used as a screening tool in the preliminary selection of promising extractant candidates and separation media likely to meet the separation of radionuclides, enabling a future faster development of scintillating and non-scintillating resins with reduced time and economic investment.

CRediT authorship contribution statement

I. Giménez: Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. **G. Sormani:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **A. Rodríguez:** Writing – review & editing, Supervision, Project administration, Methodology, Investigation, Conceptualization. **A. Hassanali:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **H. Bagán:** Writing – review & editing, Supervision, Funding acquisition. **A. Tarancón:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aca.2025.344047.

Data availability

Data will be made available on request.

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