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Research Article

Non-hydrothermal synthesis of robust and radiation-resistant sodalite microspheres for effective removal and in-situ immobilization of Cs⁺ and Sr²⁺

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Abstract: The safe management of radioactive ^{137}Cs and ^{90}Sr from nuclear wastewater requires not only efficient capture but also permanent immobilization, yet current strategies often treat these as separate processes, leading to incompatibility and complex operations. Herein, non-hydrothermal method based on geopolymer technology was developed to fabricate robust sodalite zeolite microspheres (GXU-SODs) for the integrated adsorption-immobilization of Cs^+ and Sr^{2+} . The synthesized GXU-SODs exhibited well-defined crystalline structure and spherical morphology with compressive strength of 13.88 MPa. Batch adsorption experiments revealed the maximum adsorption capacities (Q_m) of GXU-SODs were 56.67 and 59.64 $\text{mg}\cdot\text{g}^{-1}$ for Sr^{2+} and Cs^+ with rapid kinetics of 1.21 and 0.94 $\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$, respectively. The adsorption process followed pseudo-second order and Langmuir models, indicating monolayer homogeneous chemisorption. Meanwhile, GXU-SODs exhibited excellent radiation resistance, retaining >96 % structural integrity and adsorption performance after 500 kGy. Dynamic adsorption column tests confirmed excellent breakthrough performance and practical applicability in real seawater. The adsorbed GXU-SODs could permanently immobilize radionuclides via simple thermal treatment at 1100 °C without secondary additives, achieving 28-day leaching rates significantly below regulatory standard. Combining XRD, XPS, FT-IR, SEM-EDS and DFT calculations revealed the ion exchange, chemisorption and lattice incorporation within the sodalite cages were the primary mechanisms for the effective adsorption and long-term immobilization of Sr^{2+} and Cs^+ . This work presents a novel, scalable, and energy-efficient synthesis route for advanced sodalite-based materials and establishes an integrated adsorption-immobilization strategy for the treatment and safe disposal of radioactive wastes.

Keywords: Sodalite zeolite; geopolymer; radionuclides; dynamic adsorption; radiation resistance; in-situ immobilization

1 Introduction

Nuclear energy serves as a vital low-carbon power source to meet growing global energy demands, yet its utilization generates significant amounts of radioactive wastewater containing hazardous fission isotopes, notably ^{137}Cs and ^{90}Sr [1]. These radionuclides exist primarily as the soluble cations [2, 3], which pose persistent radiological risks owing to their high solubility and notable environmental mobility with half-lives of $t_{\text{Cs}}^{1/2}=30.17$ and $t_{\text{Sr}}^{1/2}=28.80$ years, respectively. Cs^+ readily enters ecological systems and bioaccumulates in the food chain due to its chemical similarity to K^+ , leading to chronic radiation exposure to human tissues [4]. Similarly, the selective uptake of Sr^{2+} is driven by its chemical behavior to Ca^{2+} , causing its incorporation into skeletal tissues and significantly increasing the risk of bone cancers and leukemia [5]. Therefore, developing efficient and advanced strategies that can effectively remove hazardous ions and simultaneously achieve safe immobilization poses a critical challenge for ensuring the environmental sustainability of nuclear energy.

Currently, various technologies have been developed for the removal of Cs^+ and Sr^{2+} including adsorption [6], ion exchange [7], membrane filtration [8], electrochemical treatment [9]. Among these, adsorption offers notable promise due to its cost-effectiveness, high removal efficiency and good adaptability to complex water matrices [10]. To date, COFs, MOFs, metal sulfides, clays and resin have been reported for adsorption Cs^+ and Sr^{2+} [11-14]. Compared to organic adsorbents, inorganic adsorbents are more favored for radioactive treatment due to excellent thermal and radiolytic stability [15]. However, efficient adsorption is merely the first step for the safe management of radioactive waste. Given the extremely hazardous issue of Cs^+ and Sr^{2+} , the secure isolation of these radionuclides from the biosphere demands not only effective capture but also permanent immobilization within durable matrices. Conventional immobilization strategies can physically isolate the contaminants, but it suffers from inherent drawbacks, such as the physicochemical incompatibility between the adsorbent and the matrix, which can lead to poor interfacial bonding and post-solidified body may cracked [16, 17]. Meanwhile, the durability of the final waste form relies heavily on the stability of the matrix

material introduced during this multi-step process, rather than on the adsorbent itself [18]. Therefore, it is urgent to develop robust and durable inorganic adsorbent, which not only efficiently adsorbs target ions in both static and dynamic adsorption but also possesses an intrinsic ability to be directly converted into a stable waste form via a simple treatment.

Geopolymers (GPs) are inorganic materials formed by alkali-activation of aluminosilicate precursors under ambient conditions, featuring a three-dimensional (3D) network structure with high mechanical strength [19], chemical durability [20] and excellent corrosion resistance [21]. GPs have emerged as potential candidates for adsorption and immobilization applications due to unique structural features and cost-effectiveness [22, 23]. However, the amorphous gel network of GPs exhibits limited adsorption capacity and selectivity for Cs^+ and Sr^{2+} , restricting their practical application in radioactive nuclide treatment. In contrast, zeolites possess well-defined crystalline frameworks with uniform pores, enabling efficient capture of Cs^+ and Sr^{2+} through size-selective and ion-exchange mechanisms [24-26]. Among zeolite members, sodalite (SOD) is particularly attractive as immobilization matrix, because its unique β -cage structure with structural simplicity and higher framework density provides strong binding sites interaction and excellent chemical stability [27-29]. However, conventional hydrothermal synthesis of SOD requires high pressure and organic templates, generating secondary waste and limiting scalable production of shaped bodies [30]. Given the structural homology between amorphous $[\text{SiO}_4]$ and $[\text{AlO}_4]$ tetrahedral units in geopolymer and their crystalline counterparts in zeolite, previous works have successfully developed green strategy for in-situ non-hydrothermal conversion into robust crystalline zeolite phases (GXU-NaAs, GXU-NaXs) by geopolymer technology with good dynamic adsorption [6, 31]. However, a critical but previously bottleneck exists that NaX and NaA zeolites exhibit superior specific surface areas and adsorption capacities, but their frameworks suffer from severe structural collapse at elevated temperatures ($> 800^\circ \text{C}$) due to the low framework density and large cage dimensions, which requires the addition of external glass or ceramic precursors for secondary sintering. Meanwhile, The ion-exchange mechanism in

GXU-NaA/NaX leaves radionuclides in exchangeable positions within the zeolite cages, resulting in relatively high leachability.

In recent years, the ceramization of geopolymers — i.e., the transformation of amorphous geopolymer networks into crystalline ceramic phases upon thermal treatment—has emerged as a promising strategy for radionuclide immobilization [7, 18]. For instance, Tian et al. demonstrated that Cs^+ can be incorporated into the nepheline structure when NaOH-activated geopolymers are calcined at 1000 °C, significantly reducing Cs leaching concentrations [7]. Shao et al. explored the in-situ immobilization of Sr^{2+} and Cs^+ by calcining pre-adsorbed geopolymer microspheres, achieving 28-day leaching rates that meet national standards [18]. Despite these advances, most reported integrated strategies still follow a sequential workflow of adsorption, separation, matrix mixing and sintering, which inevitably introduces interfacial incompatibility between the adsorbent and the ceramic matrix, complicates operational procedures, and generates secondary waste [16, 17]. Moreover, the immobilization bodies obtained in many cases remain predominantly amorphous or partially crystalline, limiting the ultimate chemical durability of the waste form [7, 18]. A few studies have attempted to use zeolite precursors as self-sintering hosts, but the phase evolution pathways and their correlation with nuclide speciation during thermal treatment have not been systematically elucidated, particularly for sodalite-based systems [28, 29]. Therefore, the key bottleneck for achieving an integrated adsorption-immobilization paradigm is not to pursue ever-higher adsorption capacity, but to identify a zeolitic framework that can combine efficient capture with intrinsic high-temperature self-consolidation. Our group develops robust, durable and excellent radiation-resistant SOD zeolite microspheres (GXU-SODs) using geopolymer technology for the first time. By controlling the proportions of Si, Na and Al, the geopolymer gel partially transforms into the SOD zeolite phase, while the remaining part serves as a strong matrix with good strength, thereby achieving the integrated adsorption-immobilization process for Cs^+ and Sr^{2+} . Meanwhile, further investigation is required

regarding the correlation between phase evolution during the thermal process and nuclide speciation, as well as the stability and underlying immobilization mechanism of the waste matrix.

Herein, this study innovatively proposes a new strategy of adsorption-immobilization integration for the safe management of Cs^+ and Sr^{2+} . Meanwhile, the static and dynamic adsorption performance in real seawater was investigated in order to achieve efficient purification of low-concentration nuclides in seawater. The atomic-level adsorption and calcination immobilization mechanism of Sr^{2+} and Cs^+ in the GXU-SODs cage was revealed via DFT method. The long-term chemical stability and radiation stability of GXU-SODs were assessed through leaching and irradiation experiments. Therefore, the present work represents a substantial advance beyond our prior studies, shifting the focus from mere contaminant removal to an all-in-one solution that simultaneously achieves efficient capture, radiation resistance, and permanent immobilization, significantly simplifying the radioactive waste management chain and reducing secondary waste generation.

2 Experimental section

2.1 Materials

All the detailed information of materials used in this experiment was shown in the supporting information (SI, S1).

2.2 Characterization

Detailed descriptions of the characterization can be found in the supplementary material (SI, S2).

2.3 Adsorption Experiments

The experimental conditions for static adsorption studies were summarized in Table S1. Adsorption data of Sr^{2+} and Cs^+ was performed according to Eqs. (1-14) (SI, S3-S8).

2.4 Normal Leaching Experiments and DFT Calculations

The normal leaching experiments, formula and DFT calculations were described in detail in Supporting Material (SI, S9-S10).

3 Results and Discussion

3.1 Effect of $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ on GXU-SODs

Fig. 1(a) depicts the synthesis route for GXU-SODs. 20 g of $\text{Al}_2\text{O}_3\text{-SiO}_2$ powder was mixed with a mount of liquid water glass in a beaker. The solid-liquid paste was initially homogenized manually using a glass rod, then mechanically dispersed at 2000 rpm for 2 min to achieve homogeneous geopolymer slurry. The well-mixed slurry was subsequently injected into ambient-temperature silicone oil at 1000 rpm, where spherical droplets formed due to interfacial tension of the silicone oil and solidified quickly during sedimentation. After curing at 85 °C, the microspheres were collected by vacuum filtration, washed, dried and programmed calcination at 500 °C for 3 h. The GXU-SODs were sieved to obtain uniform particles of 75-150 μm for subsequent experiments.

The molar ratio of $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ has a significant impact on zeolites crystal growth during the geopolymer polymerization process [32]. Fig. 1(b/b₁-g/g₁) presents Optical Microscope (OM) and Scanning Electron Microscope (SEM) images of GXU-SODs synthesized at different $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ ratios. The microspheres retained favorable sphericity with increasing $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ ratio shown in Fig. 1(b-g), whereas the surface morphology underwent distinct evolution in Fig. 1(b₁-g₁). The microsphere surface was characterized by well-developed cubic crystals with uniform size distribution at $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ ratio of 1.4 (Fig. 1(b/b₁)), the gel contains abundant Al-rich domains that facilitate the rapid nucleation of the NaA typical morphology [33]. As the ratio increased to 1.6 (Fig. 1(c/c₁)), the cubic crystals exhibited a marked reduction in size, indicating elevated alkalinity began to suppress NaA crystallization while promoting the dissolution of smaller crystallites. A further increase in the ratio to 1.8 resulted in the decrease of cubic crystals, while the fine spherical particulates began to form on the microsphere surface (Fig. 1(d/d₁)). The elevated OH⁻ concentration not only promotes the overall dissolution of the gel network, but also accentuates the differential dissolution rates between Si-rich and Al-rich regions. The microspheres exhibited a spherical morphology with uneven particle sizes at a ratio of 2 as shown in Fig. 1(e/e₁), which reflect

the homogenization of the local chemical potential across the gel matrix. as the system reaches a critical compositional threshold, the nucleation density increases while the growth rate becomes limited by the local supply of aluminosilicate precursors, resulting in uniformly sized spherical crystallites. As the ratio reached 2.2 in Fig. 1(f/f₁), the surface spherical particles became uniform in size with well-defined intergranular boundaries. However, the spherical particles showed irregular sizes and obvious bonding phenomenon at the ratio of 2.4 in Fig. 1(g/g₁) due to excessive dissolution-precipitation kinetics at high OH⁻ concentrations with Ostwald ripening and interparticle bonding [33].

Fig. 1(h and i) depicts X-Ray Diffractometer (XRD) patterns, relative crystallinity and compressive strength of GXU-SODs with various n(Na₂O)/n(Al₂O₃) conditions. Fig. 1(h) shows as the n(Na₂O)/n(Al₂O₃) increased, the peak intensity of the NaA crystal type (PDF#39-0223, Na₉₆Al₉₆Si₉₆O₃₈₄) gradually decreased, while the peak intensity of sodalite (PDF#42-0216, Na₆[AlSiO₄]₆) gradually increased. This trend corroborates the morphological transition observed in Fig. 1(b/b₁-g/g₁), confirming that higher alkalinity promotes the conversion of the metastable NaA framework into the thermodynamically stable sodalite structure [32]. Upon reaching the n(Na₂O)/n(Al₂O₃) of 2.0, the NaA phase completely disappeared, leaving only the characteristic peak of sodalite, which indicated the completion of the phase transformation. Combined with Fig. 1(i), it can be seen that the relative crystallinity showed a trend of increasing first and then decreasing, reaching a relative high value of 87.46% at n(Na₂O)/n(Al₂O₃) of 2.2. This was because the excessively high OH⁻ concentration caused partial dissolution of the formed crystals and promoted the formation of defect-rich regions [31]. Meanwhile, the compressive strength decreased as the n(Na₂O)/n(Al₂O₃) increased, which was because higher OH⁻ dissolved in the solution to cause thinner slurry [32].

With respect to mechanical functionality, the gradual decline in compressive strength from 13.70 to 10.32 MPa as the n(Na₂O)/n(Al₂O₃) ratio increases from 1.4 to 2.4 is directly correlated with the progressive consumption of the amorphous geopolymer gel and the growth of discrete sodalite

crystallites. Even at the optimal sodalite crystallinity of 87.46% ($n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3) = 2.2$), the remaining of 12.5 wt% amorphous phase still maintains a compressive strength of 13.88 MPa, which is well above the 10 MPa threshold required for stable fixed-bed dynamic adsorption operations. This amorphous geopolymer binder acts as a tough continuous phase that prevents particle attrition, fragmentation and fines generation under continuous fluid scouring in column applications. Based on the above analysis, $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3) = 2.2$ was chosen as the optimal preparation condition.

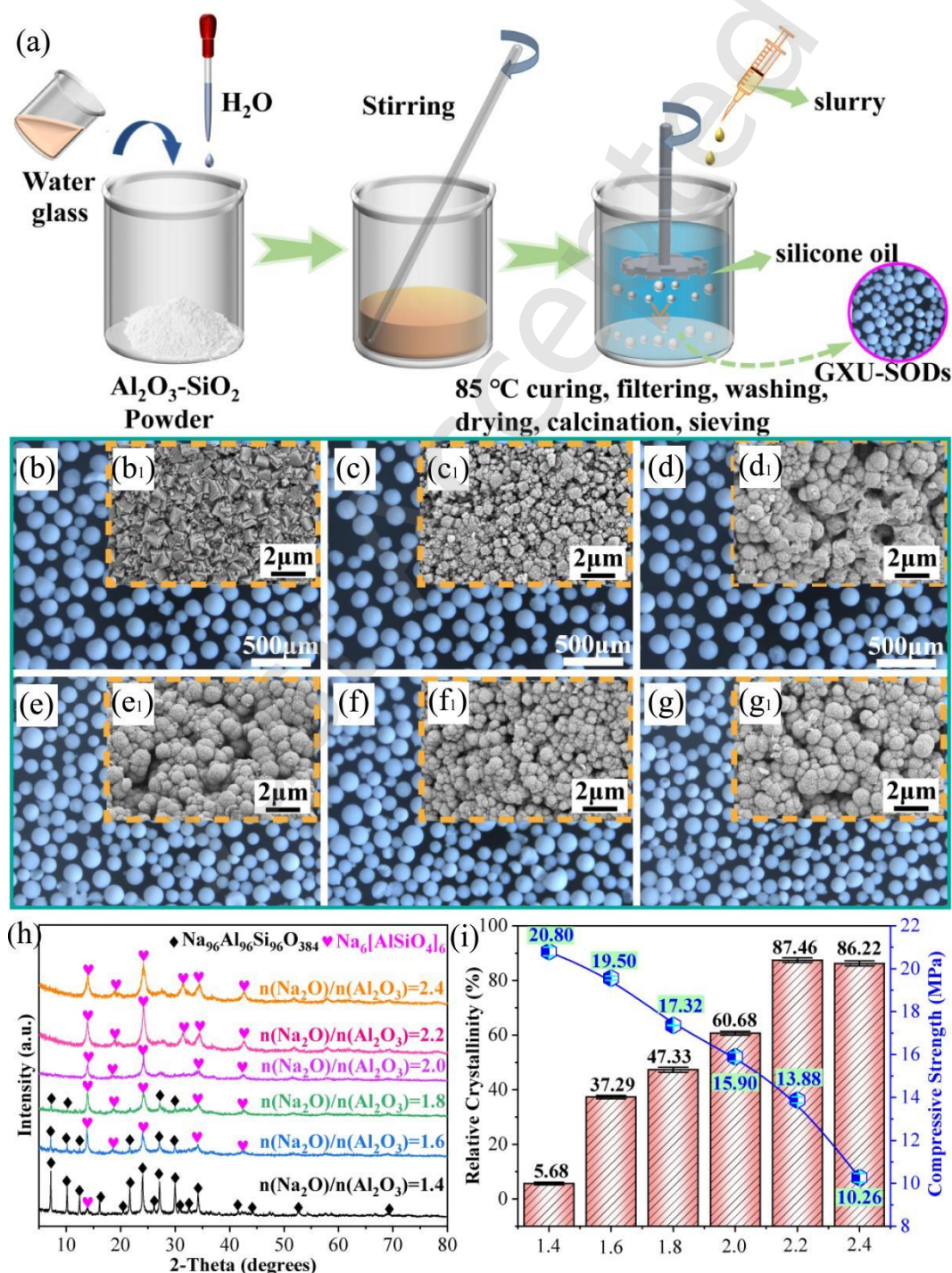


Fig 1. Flow chart of preparation of GXU-SODs (a), OM and SEM images of GXU-SODs with different

$n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ conditions (b/b₁-g/g₁: 1.4-2.4), XRD (h), relative crystallinity and compressive strength (i) of GXU-SODs with various $n(\text{Na}_2\text{O})/n(\text{Al}_2\text{O}_3)$ conditions.

3.2 Effect of curing time on GXU-SODs

The curing time presents a crucial parameter governing the crystallization process of zeolites [31]. Fig. 2 illustrates the effect of curing time on the OM, SEM, XRD, relative crystallinity and compressive strength of the as-synthesized GXU-SODs. The observed evolution of particle size and uniformity with curing time provides compelling evidence that crystallization is governed by the temporal evolution of local chemical environments, rather than a simple kinetic growth process. As observed in Fig. 2(a/a₁-c/c₁), the curing time exerted negligible effect on the microsphere sphericity, yet it significantly influenced the growth and ordered assembly of zeolite crystals. Spherical particles with non-uniform size were distributed on the surface of the microspheres at curing time of 6 h (Fig. 2(a/a₁)), indicating the process was in the nucleation and growth stage. As the curing time extended to 12 h, spherical particulates became relatively uniform in size shown in Fig. 2(b/b₁). The continuous diffusion and rearrangement of aluminate and silicate species within the amorphous gel, resulting in fostering synchronous nucleation and growth, thereby yielding the most uniform particle size distribution and the highest relative crystallinity. When it continued to increase to 24 h, adhesion phenomena occurred between the spherical particles on the microsphere surface (Fig. 2(c/c₁)). Fig. 2(d and e) depicts the intensity of the sodalite diffraction peaks and relative crystallinity initially increased with increasing curing time and subsequently decreased at the curing time of 24 h, which was because the alkaline environment caused crystal dissolution and lattice defects [6]. This alteration of the local chemical balance disrupts the previously established homogeneous growth conditions, leading to irregular adhesion and bonding of particles. Fig. 2(f) shows the compressive strength displayed a gradual decline trend as the time increased, which was due to the amount of amorphous gel decreased [34]. Notably, the maximum relative crystallinity was achieved at a curing time of 12 h, at which point the material

retained a compressive strength of 13.88 MPa. Therefore, choosing the curing time of 12 h was the optimal preparation condition.

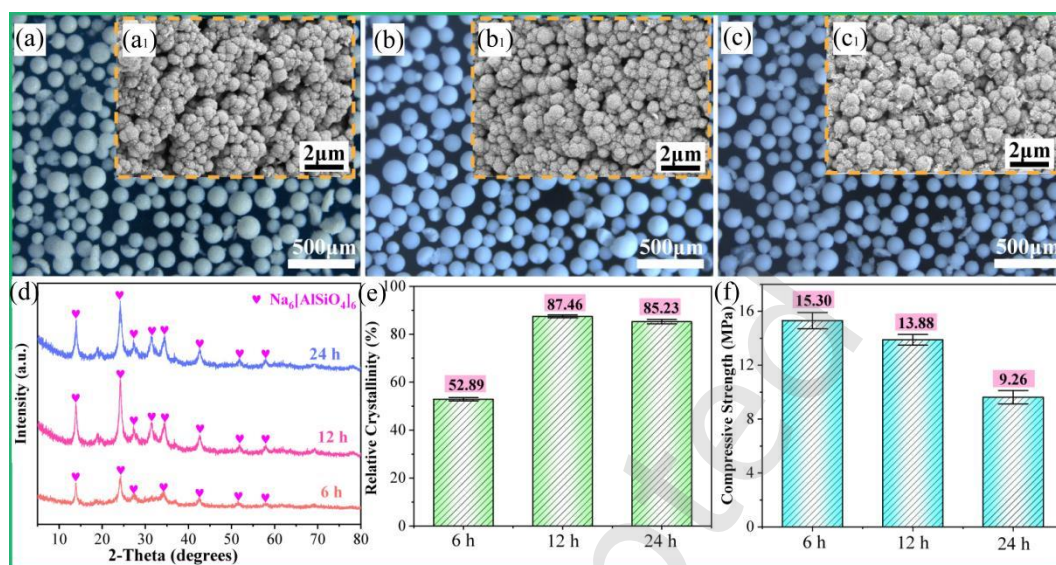


Fig 2. OM, SEM (a/a₁-c/c₁: 6-24 h), XRD (d), relative crystallinity (e) and compressive strength (f) of GXU-SODs with curing time.

3.3 SEM, cage structure, BET and pore volume of GXU-SODs

The morphology and pore structure of GXU-SODs synthesized under the optimized conditions were characterized with the results presented in Fig. 3. The SEM image of GXU-SODs showed well-defined spherical morphology with a distinctly porous surface in Fig. 3(a), which was advantageous for facilitating rapid diffusion and access of target ions to active sites. To gain atomic-level insights into the framework structure, DFT calculations were performed on the sodalite unit cell. As illustrated in Fig. 3(b), the sodalite framework was characterized by a cage-like topology constructed from six-membered rings with its fundamental elements being O, Al, Si, and Na. The average calculated distance between the farthest and closest Na and O atom was 2.74 Å, which was consistent with the unique configuration creates six-membered ring openings of the cages with a diameter of approximately 2.8 Å [35]. The porous characteristics of GXU-SODs were assessed by N₂ adsorption-desorption measurements in Fig. 3(c and d). The specific surface area (SSA) and pore volume of GXU-SODs were 46.98 m²·g⁻¹ and 0.15 cm³·g⁻¹, respectively. The N₂ adsorption-desorption isotherm results

exhibited a characteristic type IV isotherm [36], with the adsorption capacity of GXU-SODs significantly increased at relative pressure (p/p_0) between 0.9 and 1. Furthermore, a distinct H3-type hysteresis loop was observed within $p/p_0 = 0.7-0.95$, implying the existence of interconnected mesopore networks with high pore connectivity, which was favorable structural characteristics for the in-situ immobilization of radionuclides [37]. Fig. 3(d) shows that the abundant mesopores are mainly concentrated at about 2-5 nm, providing a large number of adsorption sites for subsequent adsorption.

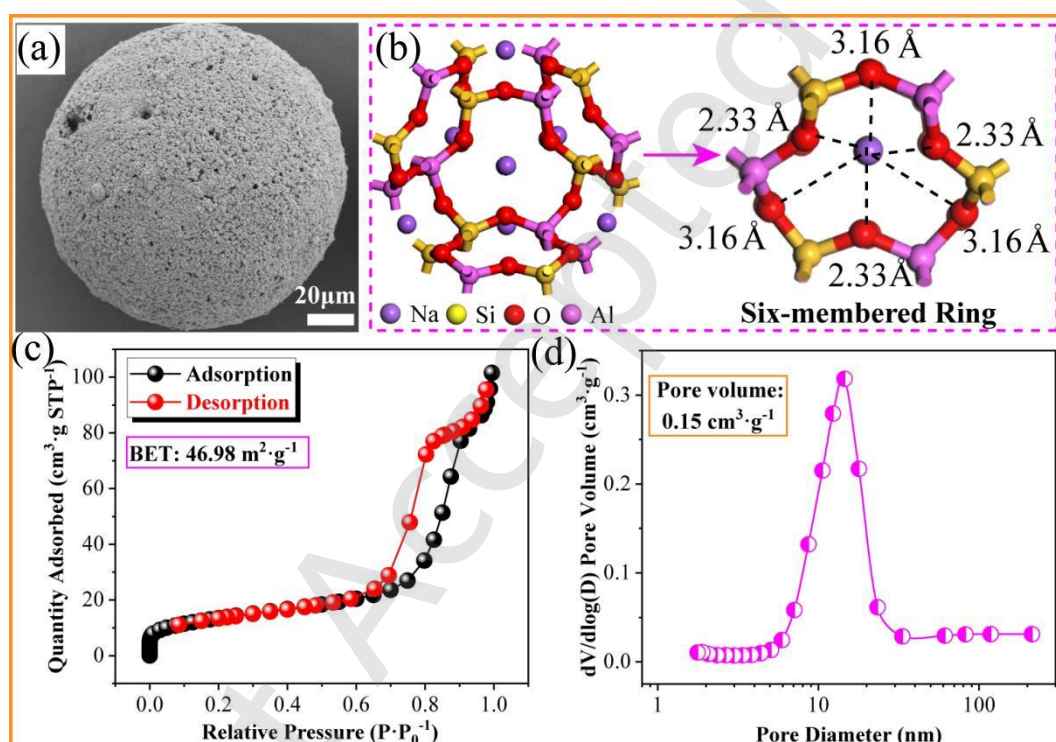


Fig 3. SEM (a), six-membered ring structure (b), N₂ adsorption-desorption isotherms (c) and pore volume (d) of GXU-SODs under the optimal preparation.

3.4 Static adsorption

3.4.1 Effect of pH, dosage and kinetics on adsorption of GXU-SODs for Cs⁺ and Sr²⁺

Fig. 4(a and b) shows with increasing adsorbent dosage, the adsorption capacity declined while the removal rate increased, reaching over 98% at 2.5 (Cs) and 3.0 (Sr) g·L⁻¹. This was because lower dosages provided insufficient active sites, whereas higher dosages led to an excess of sites and reduced capacity [34]. Therefore, 2.5 (Cs) and 3.0 (Sr) g·L⁻¹ was selected as the optimum condition. The surface charge characteristics of GXU-SODs are regulated by pH, and this change directly affects the

adsorption efficiency [38]. Fig. 4(c) shows the adsorption capacity of Cs^+ and Sr^{2+} increased with rising pH, and then stabilized at pH= 6-8. Zeta potential analysis further showed the zero potential point (pH_{PZC}) of GXU-SODs was about pH= 2.26, conforming that the surface protonation state was solution pH-dependent. Based on the optimal adsorption performance observed, a pH of 6 was selected for further experiments. To explore the adsorption kinetics of Sr^{2+} and Cs^+ by GXU-SODs, the experimental data was fitted with the pseudo-first-order (PFO), pseudo-second-order (PSO) and intraparticle diffusion (IPD) models (Table S2). Fig. 4(d) illustrates that with the prolongation of adsorption time, the adsorption capacity gradually increased and tended to reach saturation within 60/180 min for Cs^+ and Sr^{2+} , respectively. The kinetic behaviors of Sr^{2+} and Cs^+ adsorption onto could be satisfactorily described by the PSO model with remarkable correlation coefficients ($R_{\text{Sr}}^2 = 0.989$, $R_{\text{Cs}}^2 = 0.937$), suggesting that the overall adsorption rate is governed by chemical interactions between the adsorbate ions and the framework active sites. Meanwhile, the fitting curves of the IPD model in Fig. 4(e and f) did not pass through the origin, which suggested that IPD was not the dominant rate-limiting step of the entire adsorption process [39]. Notably, the PSO rate constant for Sr^{2+} and Cs^+ with kinetics of 1.21 and 0.94 $\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$. Given that the hydrated ionic radius of Sr^{2+} (4.12 Å) is considerably larger than that of Cs^+ (3.29 Å), the diffusion of Sr^{2+} through the narrow six-membered ring (6MR) apertures (~2.8 Å) of the sodalite cages requires more extensive dehydration and thus encounters a higher activation barrier. This interpretation is further supported by the IPD model: while both ions exhibit multi-linear plots, the first linear segment (representing boundary layer diffusion) is more pronounced for Sr^{2+} , indicating that surface film resistance and pore diffusion contribute significantly to the overall kinetics. Consequently, although the chemisorption rate constant is larger for Sr^{2+} , the combined resistance of external film diffusion and intraparticle mass transfer through the 6MR windows renders its overall adsorption kinetics slower than that of Cs^+ , which can access internal sites more readily due to its smaller hydrated size.

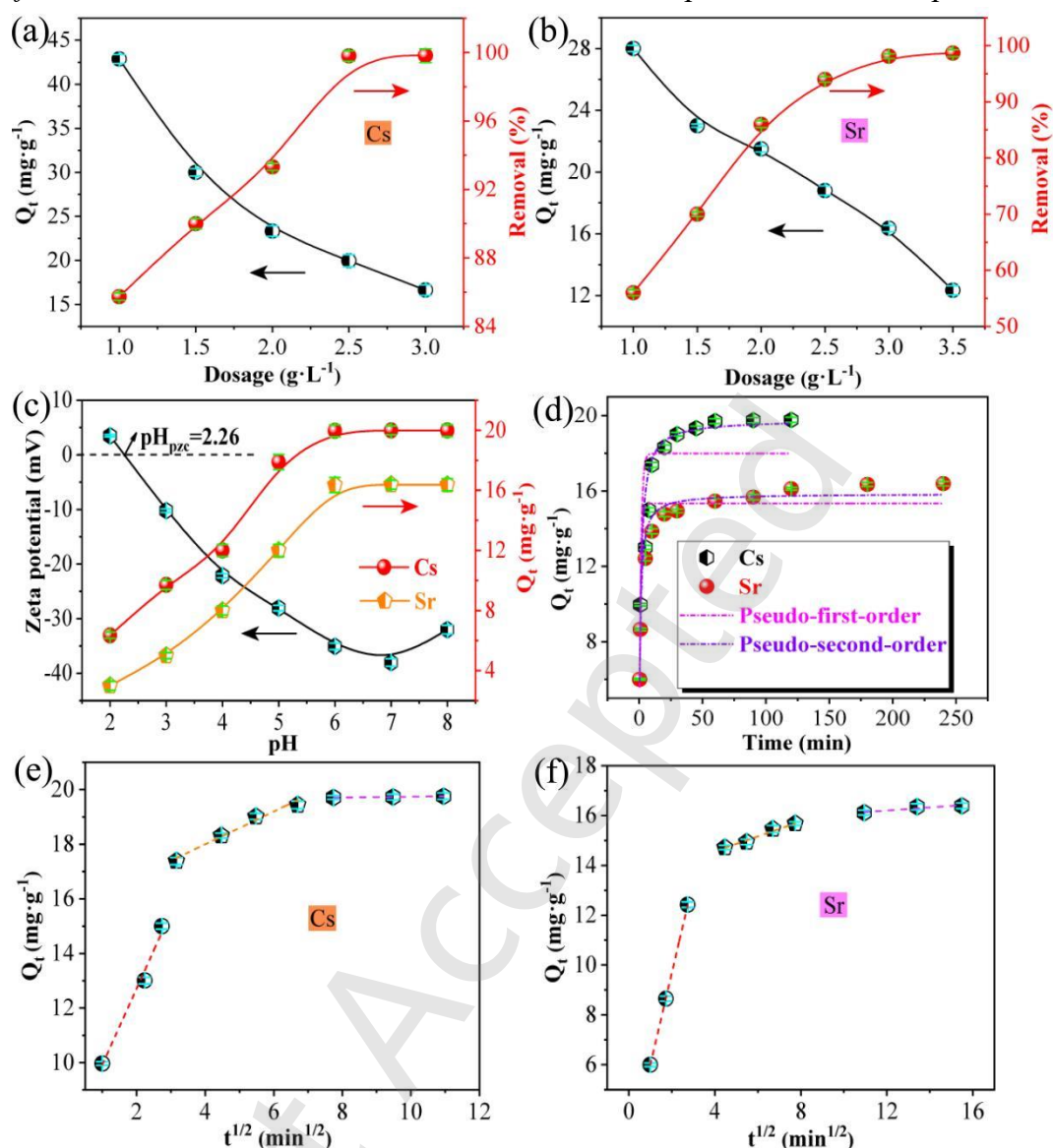


Fig 4. Effect of dosage on Cs^+ (a) and Sr^{2+} (b) removal of GXU-SODs, Effect of pH on the adsorption performance and the variation of surface zeta potential by GXU-SODs (c), PFO/PSO kinetic model fittings (d), IPD model fit plots of Cs^+ (e) and Sr^{2+} (f) removal by GXU-SODs.

3.4.2 Isothermal adsorption

To assess the adsorption capability of GXU-SODs, the adsorption isotherm data was fitted with the Langmuir, Freundlich, and Dubinin-Radushkevich (D-R) models (Table S3). As shown in Fig. 5(a and b), the saturated adsorption capacity of GXU-SODs for Sr^{2+} and Cs^+ increased with rising equilibrium concentration and then reached saturation, which was due to the effective active sites on the GXU-SODs have reached saturation [33]. The adsorption performance data of GXU-SODs for Sr^{2+}

and Cs^+ exhibited excellent consistency with the Langmuir isotherm model with $R_{\text{Sr}^{2+}} = 0.968/0.966/0.967$ and $R_{\text{Cs}^+} = 0.956/0.962/0.957$ at 298/318/338 K, respectively. The maximum adsorption capacities (Q_m) of GXU-SODs were determined to be 56.67 and 59.64 $\text{mg}\cdot\text{g}^{-1}$ for Sr^{2+} and Cs^+ , respectively, indicating a monolayer adsorption behavior with homogeneous distribution of binding sites [25]. Besides, the adsorption free energies were all below 8 $\text{kJ}\cdot\text{mol}^{-1}$ (Fig. 5(c and d)), which implied that physical adsorption was also involved in the whole adsorption process [40]. In the case of ion exchange on crystalline aluminosilicate frameworks, the relatively low E values arise primarily from the electrostatic attraction between the exchangeable cations and the negatively charged framework oxygen atoms. Meanwhile, the adsorption capacity decreased with increasing temperature, indicating that higher temperatures are unfavorable for the adsorption of Sr^{2+} and Cs^+ onto GXU-SODs. Thermodynamic parameters calculated using thermodynamic equations at different temperatures are provided in Table S4. The calculated ΔG° values of $\text{Sr}^{2+}/\text{Cs}^+$ at different temperatures were -5.79/-4.41, -3.32/-3.39 and -1.91/-4.02 $\text{kJ}\cdot\text{mol}^{-1}$ at 298, 318 and 338 K, respectively, confirming that the adsorption process was spontaneous. These results demonstrated that the lower temperatures enhanced the adsorption reaction process for Sr^{2+} and Cs^+ removal by GXU-SODs, which was consistent with the trend observed in Fig. 5. Based on the above comprehensive analysis, a unified mechanistic model indicates that the adsorption of Cs^+ and Sr^{2+} onto GXU-SODs is primarily governed by an ion-exchange process, in which electrostatic interactions between the hydrated cations and the negatively charged framework dominate the thermodynamic driving force (reflected by the low D-R energies), while the rate-controlling step involves chemical adsorption.

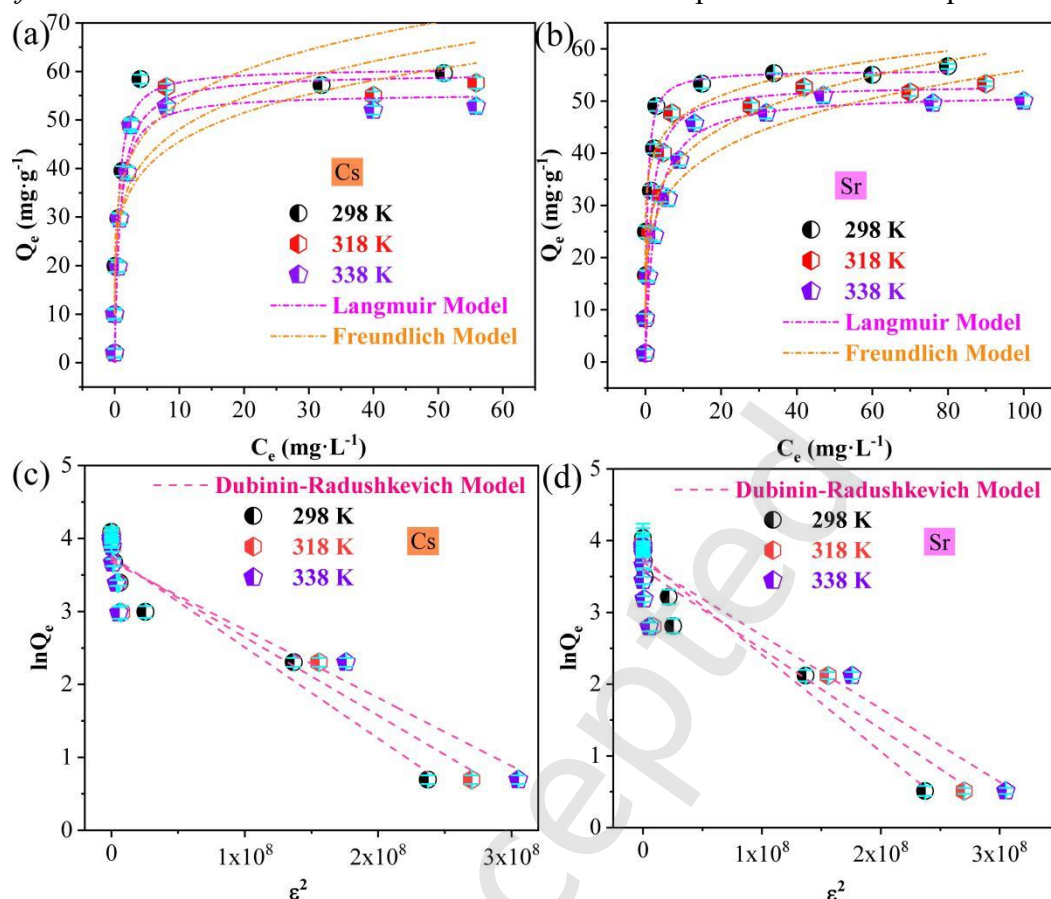


Fig 5. Langmuir (a), Freundlich (b) and D-R (c, d) models fitting by GXU-SODs for Cs⁺ and Sr²⁺ removal.

3.4.3 Influence of co-existing ions and real water on GXU-SODs for Cs⁺ and Sr²⁺ removal

Large amounts of various co-existing ions exist in real aqueous environments, which may exert a certain inhibitory effect on the adsorption performance of GXU-SODs for Sr²⁺ and Cs⁺. Fig. 6(a and b) illustrates the effects of coexisting ions (e.g., Na⁺, K⁺, Ca²⁺, and Mg²⁺) at different molar concentrations (0.01, 0.1, 0.2, and 0.5 M) on the adsorption of Sr²⁺ and Cs⁺ by GXU-SODs. With the increase in the concentration of various coexisting ions, the adsorption capacities of GXU-SODs for Sr²⁺ and Cs⁺ decreased, mainly due to the more intense competition for active sites on the adsorbent surface under the condition of high coexisting ion concentrations. Furthermore, the order of inhibitory effects of coexisting ions on the removal of Sr²⁺ and Cs⁺ by GXU-SODs was found to be Ca²⁺ > Mg²⁺ > K⁺ > Na⁺ and K⁺ > Na⁺ > Ca²⁺ > Mg²⁺, respectively. To quantitatively evaluate the selectivity, the distribution coefficients (K_d) for each competing ion system were calculated and summarized in Table S5 and S6. For Cs⁺ adsorption, the K_d values against monovalent cations (Na⁺ and K⁺) decreased from

1182 and 654 mL·g⁻¹ at 0.01 M to 593 and 219 mL·g⁻¹ at 0.5 M, respectively. For Sr²⁺ adsorption, the K_d values against Ca²⁺ and Mg²⁺ were 208 and 228 mL·g⁻¹ at 0.5 M, respectively. This is because Ca²⁺ exerted the greatest inhibitory effect on Sr²⁺ removal, which can be attributed to the fact that Ca²⁺ and Sr²⁺ had similar hydrogen ion radii [41]. K⁺ and Na⁺ showed stronger inhibitory effect on Cs⁺ adsorption due to their close hydrated ion radii (K⁺, Na⁺, Cs⁺: 3.31, 3.58, 3.29 Å) [41]. Fig. 6(c and d) illustrates the adsorption capacities of GXU-SODs in different real water bodies. The removal rates of Cs⁺ and Sr²⁺ by GXU-SODs in tapwater, seawater and natural surface water exceeded 95 % at dosage of 3.5 g·L⁻¹, confirming that GXU-SODs exhibited excellent application prospects for adsorption of Cs⁺ and Sr²⁺ in real aqueous environment.

Fig. S1(a, b) illustrates the effect of GXU-SODs dosage on the competitive adsorption of Cs⁺ and Sr²⁺ in binary equimolar solutions (initial total concentration: 50 mg·L⁻¹, Cs:Sr = 1:1, pH = 6.0, 25 °C). As the adsorbent dosage increased from 0.3 to 1.2 g·L⁻¹, the adsorption capacity of Sr²⁺ increased from 18.08 to 27.02 mg·g⁻¹, while that of Cs⁺ increased from 15.07 to 25.08 mg·g⁻¹. Notably, at each dosage level, Sr²⁺ consistently exhibited higher removal efficiency than Cs⁺ by approximately 4-7 percentage points, contrasting with the single-component system where Cs⁺ removal was superior. This reversal in selectivity under competitive conditions is attributed to the stronger electrostatic attraction between Sr²⁺ and the framework oxygen atoms, which thermodynamically favors Sr²⁺ occupancy at equilibrium despite the faster diffusion kinetics of Cs⁺. Compared with the single-component system, the adsorption capacities in the binary system were reduced by 55-75%, confirming that Cs⁺ and Sr²⁺ share common adsorption sites—primarily the exchangeable Na⁺ sites within the sodalite cages and surface defect sites—and compete strongly for these limited active centers. Fig. S1(c, d) shows the influence of contact time on the adsorption of Sr²⁺ and Cs⁺. The PSO model ($R^2 = 0.97/0.99$) fitted better than the PFO model ($R^2 = 0.91/0.98$), indicating that this model can characterize the adsorption behavior more accurately. Compared to the single-component system, the equilibrium times are prolonged, indicating that competitive adsorption slows down the adsorption kinetics of both ions. Therefore,

these results demonstrate that GXU-SODs can effectively co-adsorb Cs^+ and Sr^{2+} from binary solutions, confirming their practical viability for treating real wastewater containing mixed radionuclides.

To investigate the feasibility of GXU-SODs for continuous-flow treatment of radionuclide contaminated water, dynamic adsorption experiments were conducted using a fixed-bed column with the breakthrough curves for Cs^+ and Sr^{2+} presented in Fig. 6(e and f). The dynamic breakthrough curves for both Cs^+ and Sr^{2+} exhibited characteristic S-shaped profiles fitted by Thomas model, indicative of typical fixed-bed adsorption behavior [42]. Under the condition of 2 cm column height and $4 \text{ mL} \cdot \text{min}^{-1}$ flow rate, the start penetrating times of Cs^+ and Sr^{2+} were 100, 128 min, reaching 100 % penetration at 1200 and 560 min, respectively. When the flow rate increased to $8 \text{ mL} \cdot \text{min}^{-1}$, the corresponding penetration breakthrough times of Cs^+ and Sr^{2+} were 52, 46 min, while the exhaustion times were correspondingly shortened. The distinct breakthrough times observed for Cs^+ and Sr^{2+} arise from fundamental differences in their hydration properties and ion-exchange kinetics. The hydrated radii of Cs^+ and Sr^{2+} are 3.29 and 4.12 Å, respectively, whereas their corresponding ionic radii are 1.67 and 1.18 Å. The significantly larger hydrated radius of Sr^{2+} imposes a substantial steric hindrance during its diffusion through the sodalite six-membered ring aperture (~ 2.8 Å) and the interconnected micro-mesopore networks of GXU-SODs. As a result, Sr^{2+} exhibits slower mass transfer kinetics, which is consistent with its longer saturation time (180 min) compared to Cs^+ (60 min) in batch adsorption experiments (Fig. 4d). Beyond the size effect, the hydration energy plays a decisive role in governing the adsorption dynamics and breakthrough behavior. Sr^{2+} possesses a substantially higher hydration energy than Cs^+ due to its divalent charge and higher charge density. This strong hydration shell inhibits the close interaction between Sr^{2+} and the framework oxygen atoms of the sodalite cage. Conversely, Cs^+ with its lower hydration energy, can more readily shed its hydration shell and access the framework exchange sites, resulting in faster adsorption kinetics. Consequently, the excellent dynamic adsorption behavior of GXU-SODs towards Cs^+ and Sr^{2+} makes them highly advantages for decontaminating nuclear wastewater.

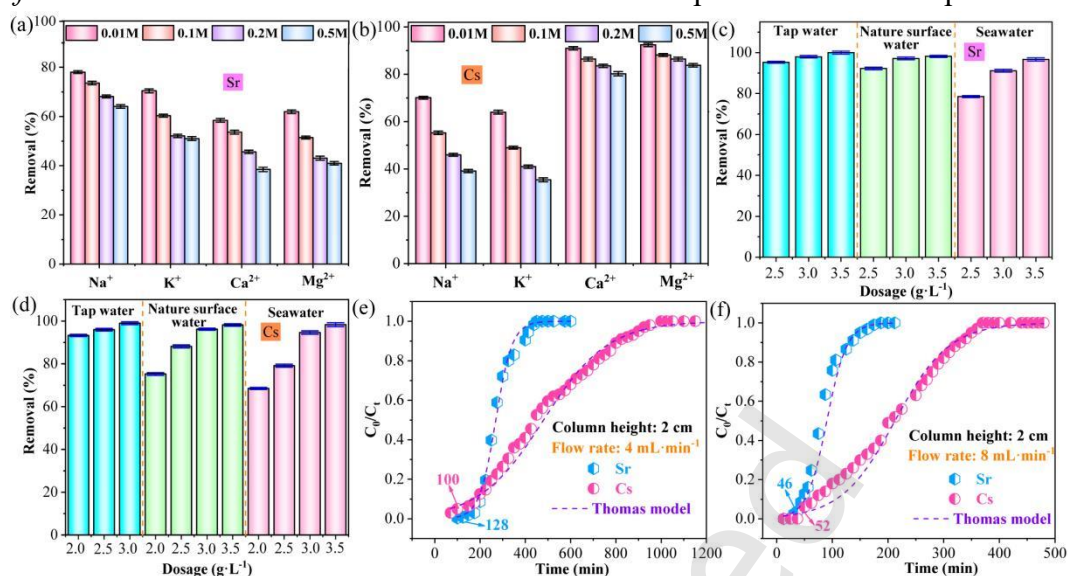


Fig 6. Effect of co-existing ions (a, b) and real water (c, d) on adsorption of Sr^{2+} and Cs^+ by GXU-SODs, Breakthrough curves and Thomas models of GXU-SODs for Sr^{2+} and Cs^+ with different flow rates in unfiltered seawater (e, f).

Table 1 and 2 compare the adsorption performance of GXU-SODs with various adsorbents for Cs^+ and Sr^{2+} removal. Compared to M/SZMs (20% slag) [38], $\text{NiSiO}@ \text{NiAlFe}$ LDHs [43], $\text{NaGP-lattice-10\%SLS}$ [45], the rate constant of GXU-SODs during the adsorption process for Cs^+ was 1/30, 1/30, 1/940 times shorter than that, respectively. In compared with Hydroxy Sodalite [44], IPA-MC [46] and $\text{NSC}@ \text{MS-4A}$ [47], the adsorption capacity of GXU-SODs for Cs^+ was 1.32-3.89 times higher than those above. Meanwhile, GXU-SODs also exhibited faster rate constant of Sr^{2+} than M/SZMs (20% slag) [38], Hydroxy Sodalite [44], CTS-SX [48] and SbS-1K [49]. Furthermore, GXU-SODs is characterized by a simple preparation process and higher adsorption capacity compared to PVA/GO/MnO_2 [50] and ZMPP-CFs [51]. Therefore, GXU-SODs demonstrates excellent faster adsorption rate and good adsorption performance for Sr^{2+} and Cs^+ .

Table 1 Comparison between GXU-SODs and other adsorbents for Cs^+ removal.

Adsorbent	pH	t (min)	T ($^{\circ}\text{C}$)	$Q_{m,\text{Cs}}$ ($\text{mg}\cdot\text{g}^{-1}$)	k_{Cs} ($\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$)	Ref.
M/SZMs (20% slag)	6	60	25	103.74	0.03	[38]

NiSiO@NiAlFe LDHs	10	20	25	61.50	0.03	[43]
Hydroxy Sodalite	7	24h	25	15	/	[44]
NaGP-lattice-10%SLS	6	180	25	80.10	0.001	[45]
IPA-MC	6	200	25	21.47	0.280	[46]
NSC@MS-4A	7	80	25	44.35	0.20	[47]
GXU-SODs	6	60	25	59.64	0.94	This study

Table 2 Comparison between GXU-SODs and other adsorbents for Sr^{2+} removal.

Adsorbent	pH	t (min)	T ($^{\circ}\text{C}$)	$Q_{m,\text{Sr}}$ ($\text{mg}\cdot\text{g}^{-1}$)	k_{Sr} ($\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$)	Ref.
M/SZMs (20% slag)	6	60	25	54.91	0.003	[38]
Hydroxy Sodalite	7	24h	25	43	/	[44]
CTS-SX	6	90	25	76.21	0.005	[48]
SbS-1K	6	20	25	61.12	0.109	[49]
PVA/GO/MnO ₂	7	8h	25	26.80	0.001	[50]
ZMPP-CFs	6	30	25	38.99	0.05	[51]
GXU-SODs	6	180	25	56.67	1.21	This study

3.5 Adsorption mechanism

To investigate the adsorption mechanisms of Sr^{2+} and Cs^{+} onto GXU-SODs, the microstructure and elemental mapping of the adsorbents were presented in Fig. 7(a-c). The GXU-SODs retained well-defined spherical morphology post adsorption, indicating that GXU-SODs possessed excellent mechanical stability. The uniform distribution of Sr and Cs elements on the spherical surface after adsorption confirmed that GXU-SODs had successfully adsorbed Sr^{2+} and Cs^{+} . XPS survey spectrum in Fig. 7(d) showed that a significant decline in the Na 1s peak, the Na content decreased from 8.92

at.% to 6.24 at.% after Cs⁺ uptake (Cs: 2.14 at.%), and to 5.88 at.% after Sr²⁺ uptake (Sr: 1.56 at.%), while the Si/Al ratio remained nearly constant (~1.06-1.08), confirming framework stability. The experimental ion-exchange capacities derived from Na loss were 0.43 mmol·g⁻¹ for Cs⁺ and 0.62 mmol·g⁻¹ for Sr²⁺ (1.24 meq·g⁻¹ on a charge-equivalent basis), which accounted for only 6.1% and 17.6%, respectively, of the theoretical IEC of sodalite (7.04 mmol·g⁻¹, based on Na₆[AlSiO₄]₆). This substantial discrepancy indicates that bulk ion exchange is not the sole adsorption mechanism. The remaining capacity arises from chemisorption at external surface sites. This combined mechanism is consistent with the Langmuir monolayer behavior, PSO kinetics, and D-R derived adsorption free energies, confirming a mixed physisorption-chemisorption nature. Fig. 7(e, f and g) depicts the characteristic peaks of Sr 3d_{3/2} and Sr 3d_{5/2} were observed at binding energies of 134.39 and 132.69 eV [51]. In addition, the typical peaks of Cs 3d appeared at 737.38 and 723.46 eV, which were assigned to Cs 3d_{3/2} and Cs 3d_{5/2}, respectively [51], and the typical peaks of Na 1s appeared at 1071.78 eV. To further elucidate the interaction mechanism between GXU-SODs and Sr²⁺ and Cs⁺, the pre and post-adsorption samples were characterized by FT-IR and XRD, with the results presented in Fig. 7(h and i). The peaks around 3433-3526 and 1648 cm⁻¹ were assigned to O-H stretching and bending vibrations of adsorbed water [6]. The peak at 1407-1456 cm⁻¹ arose from C-O stretching vibration in the air [33]. The band around 995 cm⁻¹ corresponded to Si/Al-O-Si stretching vibration [31]. The peak at 677 cm⁻¹ represented the symmetric stretching vibration of the Si(Al)-O-Si(Al) bonds in the zeolite framework [52]. The peak at 620 cm⁻¹ was attributed to the symmetric stretching of Al-O bonds [53]. The peak around 570 cm⁻¹ was a characteristic peak of the double six-membered ring bending vibration of Si-O-Al bonds [54]. The peak at 460 cm⁻¹ corresponded to the bending vibration of Si/Al-O bonds [30]. As shown in Fig. 7(i), XRD result shows that although the intensity of sodalite peak decreased after adsorption, the main characteristic peaks of sodalite zeolite (PDF#42-0216, Na₆[AlSiO₄]₆) remained well, suggesting the presence of ion exchange mechanism in the adsorption process.

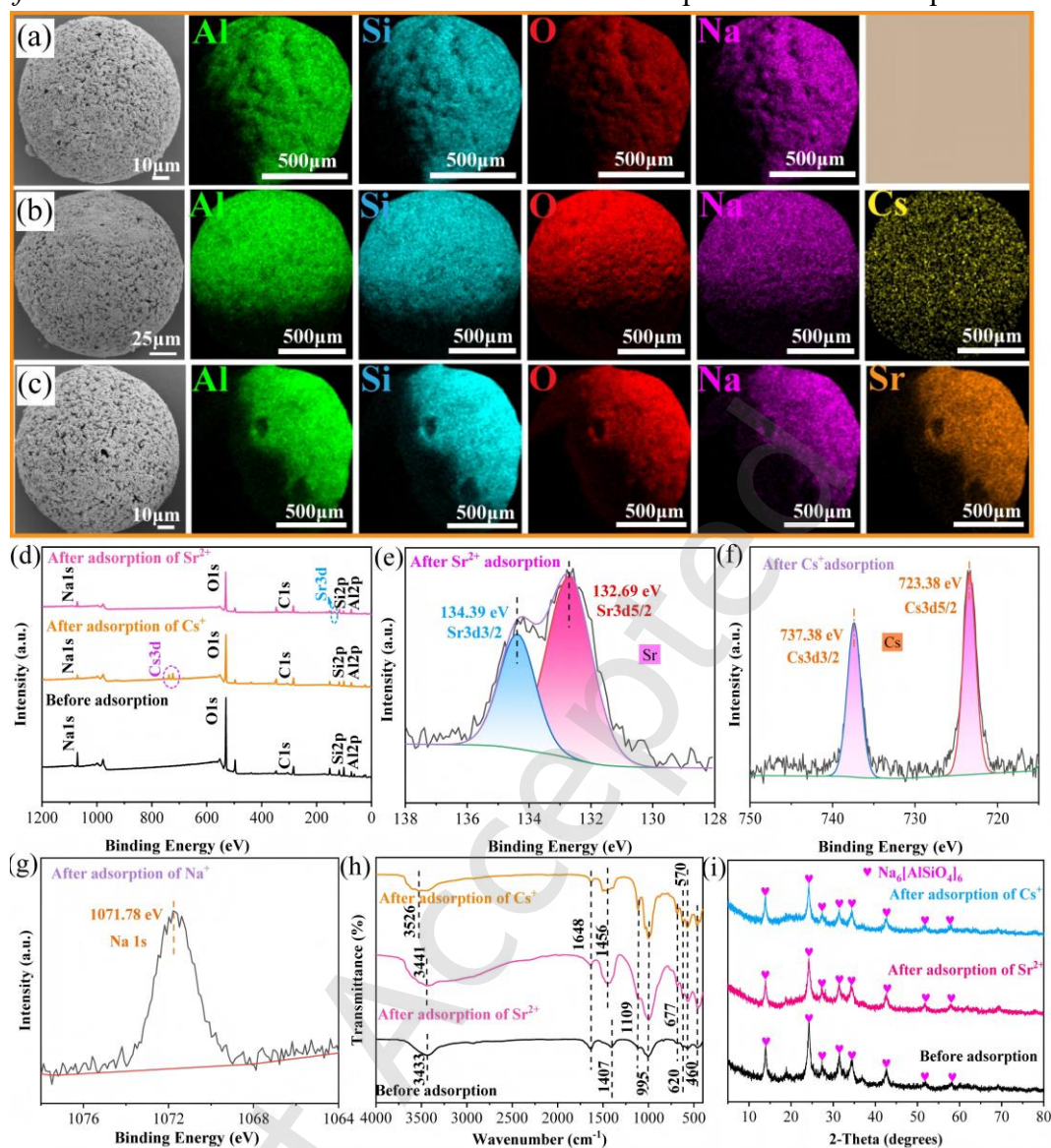


Fig 7. SEM/EDS of GXU-SODs before (a) and after (b: Cs, c: Sr) adsorption, XPS full spectrum (d) of GXU-SODs, XPS spectra of Sr 3d (e), Cs 3d (f) and Na 1s (g), FT-IR (h), XRD (i) image of GXU-SODs before and after adsorption.

To gain atomic-level insights into the adsorption mechanism, DFT calculations were performed to determine the optimal adsorption sites and adsorption energies (E_{ads}) of GXU-SODs for Sr²⁺ and Cs⁺ removal in Fig. 8(a-f). As illustrated in Fig. 8(a and b), GXU-SODs exhibited adsorption behavior towards Sr²⁺ and Cs⁺ at sites associated with the six-membered ring (6MR) apertures of the SOD cage, with stable configurations identified both inside and outside the 6MR, indicating 6MR serves as the primary gateway for cation exchange in SOD frameworks. The E_{ads} values for Sr²⁺/Cs⁺ were -3.90/-

1.71 (inside), -3.74/-1.84 (outside) eV, respectively. The more negative E_{ads} values for Sr^{2+} indicated a stronger binding affinity compared to Cs^+ . The DFT results showed Sr^{2+} had a substantially more negative adsorption energy than Cs^+ , implying a stronger binding affinity for Sr^{2+} . This apparent discrepancy can be rationalized by considering two complementary factors [55]. First, the adsorption capacity on a mass basis depended not only on the binding energy per cation but also on the number of accessible exchangeable sites per unit mass, which may differ for Sr^{2+} and Cs^+ due to their distinct hydrated ionic radii and dehydration energetics. Sr^{2+} had a much higher charge density and a significantly more negative hydration free energy, meaning that its entry into the narrow six-membered ring (~ 2.8 Å) requires a larger energy penalty to strip the hydration shell. Consequently, despite the stronger final framework binding, the higher activation barrier for dehydration may kinetically and thermodynamically limited the number of Sr^{2+} ions that can actually occupy the cage interior, whereas the more weakly hydrated Cs^+ can access a larger fraction of the available exchangeable sites, leading to a marginally higher overall Q_m . Second, the areal density of adsorbed species yields site-specific capacities of approximately 1.21 and 1.27 $\mu\text{mol}\cdot\text{m}^{-2}$ for Sr^{2+} and Cs^+ , respectively, which are indeed comparable, thereby rationalizing the comparable yet slightly lower macroscopic capacity. The closest distances between Na^+ and $\text{Sr}^{2+}/\text{Cs}^+$ in the optimal configurations were 2.45 and 2.89 Å, respectively (Fig. 8(c and d)), further confirming Sr^{2+} had stronger interaction to GXU-SODs. Meanwhile, Sr^{2+} and Cs^+ tended to be close to the oxygen atoms in 6MR, exhibiting strong electronic interactions thereby forming a more stable structure. The electron density difference maps revealed darker areas in this region indicated a more significant charge transfer between the $\text{Sr}^{2+}/\text{Cs}^+$ and the adjacent oxygen atoms (Fig. 8(e and f)), providing further insight into the adsorption mechanisms of GXU-SODs for Sr^{2+} and Cs^+ at the atomic level. Among the 0.1 M HCl, NaCl, NaOH, EDTA-2Na and H_2O solution, only 51-56 % of the Cs^+ and Sr^+ could be desorbed from the GXU-SODs by NaCl desorption agent in Fig. S2, indicating the remaining fraction could represent ions trapped within the β -cages versus those adsorbed on external surfaces in the desorption experiments.

This partial desorption behavior was rationalized by DFT calculations. The adsorption energies for Cs^+ and Sr^+ on the outer surface versus the inner cavity of the sodalite six-membered ring were not significantly different. However, the key distinction lies in the diffusion barrier: ions located inside the β -cage must overcome a significant energy barrier to pass through the narrow six-membered ring window ($\sim 2.8 \text{ \AA}$), whereas surface-bound ions are directly accessible to the surrounding solution. The hydrated radius of Cs^+ ($3.29 \text{ \AA}$) exceeds the 6MR aperture, necessitating partial or complete dehydration prior to penetration, which imposes a significant activation energy for internalization. This energetic bottleneck effectively retards the intra-cage diffusion process, manifesting as the slower secondary stage observed in the adsorption kinetics (Fig. 4d). Furthermore, the considerably more negative adsorption energy of Sr^{2+} inside the cage (-3.90 eV) compared to Cs^+ (-1.71 eV), combined with its larger hydrated radius ($\sim 4.12 \text{ \AA}$), results in an even higher energy for penetration. Therefore, the DFT-calculated energy not only rationalize the biphasic adsorption behavior of Cs^+ , but also quantitatively establish that the 6MR aperture functions as a kinetic gatekeeper in the sodalite framework. Only the surface-bound fraction can be readily desorbed, while the internally confined ions remain trapped due to the synergistic combination of strong ionic-covalent bonding and spatial restriction [56]. This partial desorption not only confirms that a substantial portion of radionuclides is securely encapsulated within the SOD cages but also provides a rational basis for subsequent in-situ immobilization studies.

To quantify the magnitude of electronic interaction and the bonding nature, Bader charge partitioning and projected density of states (PDOS) were performed on the optimized adsorption configurations in Fig. S3. Upon adsorption, an electron transfer of 0.62, 0.42 e from $\text{Sr}^{2+}/\text{Cs}^+$ to the surrounding framework oxygen atoms. The larger electron transfer for Sr^{2+} was fully consistent with its higher charge density and stronger electrostatic attraction to the negatively charged oxygen atoms in the six-membered ring. The PDOS results reveal both systems had similar electronic structures, where the deep energy level states ranging from -23 to -18 eV were mainly contributed by the s orbitals,

while the valence band region ranging from -11 to -3 eV was dominated by p orbitals, accompanied by a small amount of s and d orbital components. Considering the composition of the systems, the valence band mainly originated from the O-2p state and its hybridization with the Si and Al orbitals, indicating that the electronic structure is still controlled by the Si-O-Al framework. The peak positions of the occupied states of the Sr and Cs systems were basically the same, and no obvious isolated impurity energy levels are observed, suggesting that the introduction of both cations does not significantly damage the aluminum-silicate framework. For Sr^{2+} , the d-p hybridization is more pronounced than for Cs^+ , which primarily shows s-p mixing, consistent with the stronger chemical bonding of Sr^{2+} inferred from both Bader charges and adsorption energies. The above analysis demonstrated that the adsorption of both cations involved a synergistic effect of ionic (electrostatic) and covalent (orbital hybridization) contributions.

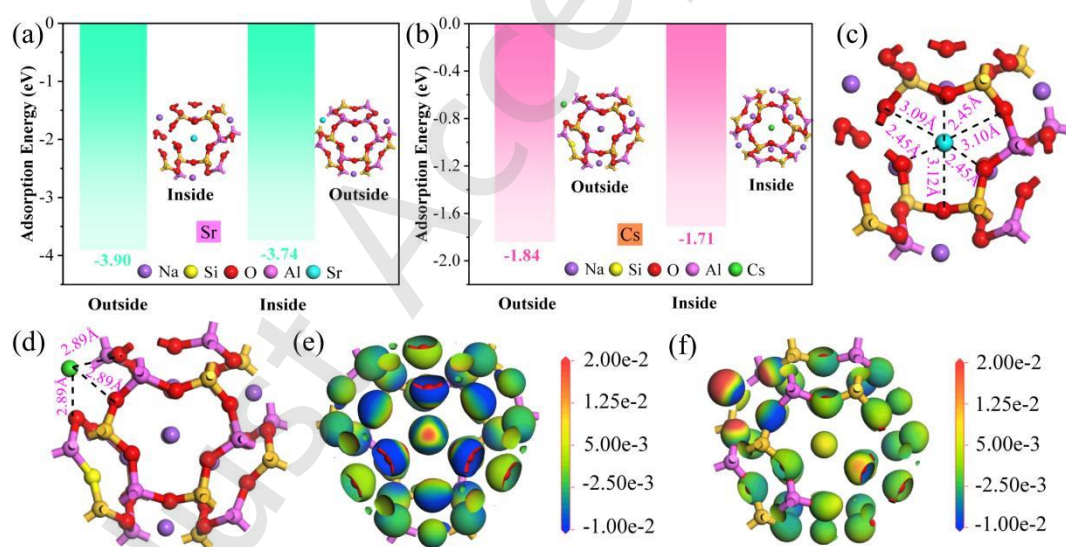


Fig 8. Adsorption energy and structural model diagrams of GXU-SODs by Sr^{2+} (a) and Cs^+ (b) removal, Optimal configuration and differential charge distribution of GXU-SODs removal for Sr^{2+} (c, e) and Cs^+ (d, f).

3.6 Radiation resistance of GXU-SODs

Given the intrinsic characteristics of gamma radiation in nuclear environments, evaluating the radiation resistance of materials is essential for assessing their practical applicability. In this study, to isolate the direct effects of gamma-ray photon interactions on the solid framework from the complex effects of aqueous radiolysis, the irradiation experiments were specifically conducted under solid-state

(dry) conditions. The SOD zeolite blocks cured for 280 d and GXU-SODs were placed in a gamma-ray space with a dose of 500 kGy, with the results presented in Fig. 9. Fig. 9(a) shows no obvious changes in either appearance or microscopic morphology of SOD zeolite blocks after exposure to 500 kGy. As illustrated in Fig. 9(b), the GXU-SODs still maintained structural integrity and excellent sphericity. Fig. 9(c) revealed the characteristic diffraction peaks corresponding to the SOD zeolite phase (PDF#42-0216, $\text{Na}_6[\text{AlSiO}_4]_6$) were well preserved after 500 kGy. Meanwhile, Fig. 9(d) showed typical SOD zeolite microstructure, which further confirmed GXU-SODs had excellent radiation resistance after 500 kGy. The compressive strength at 7 and 280 d after irradiation with 500 kGy was measured to be 11.20 ± 0.45 MPa and 13.70 ± 0.52 MPa, respectively, compared to the unirradiated counterparts of 13.88 ± 0.38 MPa and 15.65 ± 0.61 MPa as shown in Fig. 9(e). While these reductions are appreciable in percentage terms, the irradiated samples still maintain compressive strengths well above the minimum requirement of 10 MPa for practical fixed-bed column operations. The adsorption removal and capacity effects of 500 kGy radiation on Cs^+ and Sr^{2+} are shown in Fig. 9(f). Compared with the pristine sample, GXU-SODs remained excellent adsorption performance but slightly decreased capacity for cesium and strontium ions after radiation treatment. This suggests that the gamma irradiation impact on surface sites slightly outweighs any potential enhancement from defect-induced sites. This interpretation is further supported by the unchanged framework topology and the maintained ion-exchange selectivity after irradiation, confirming that the primary cation exchange capability is preserved within the cage structure.

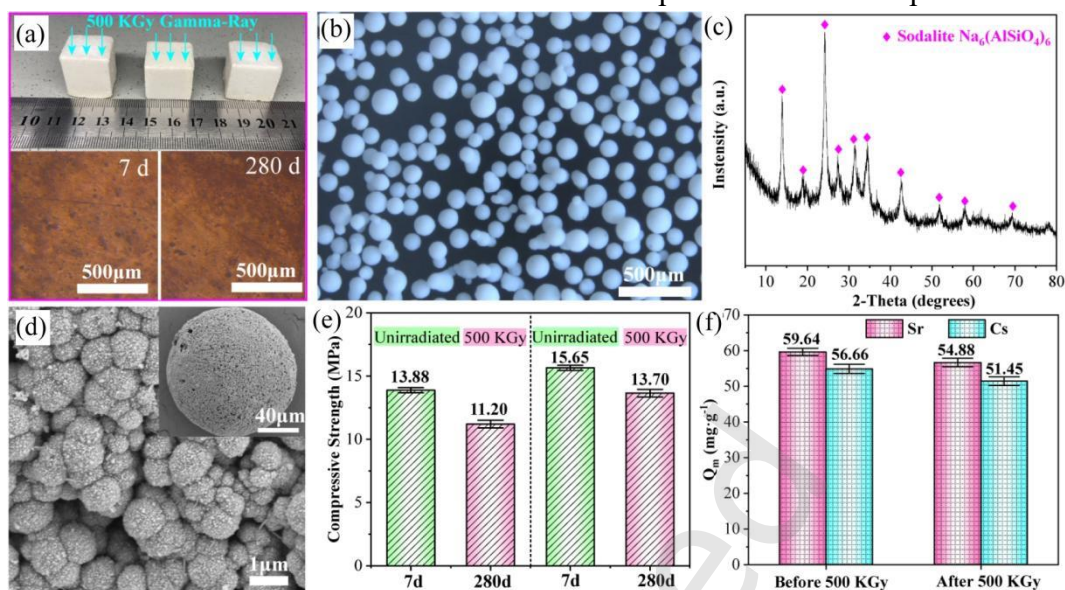


Fig 9. Appearance of SOD zeolite bulk post radiation (a), OM (b), XRD (c) and SEM (d) images of GXU-SODs after radiation, Compressive strength of SOD zeolite bulk (e), adsorption performance of GXU-SODs on Cs^+ and Sr^{2+} pre and post 500 kGy (f).

3.7 In-situ immobilization experiment for Cs^+ and Sr^{2+}

To clarify the thermally-driven phase transformation mechanism for the in-situ immobilization of radionuclides, TG-DSC curves and XRD patterns of Sr/Cs@GXU-SODs at the temperature ranges of 900 to 1200 °C were conducted, with the results comprehensively presented in Fig. 10. The TG-DSC curves revealed an initial weight loss below 200 °C corresponded to the desorption of adsorbed water (Fig. 10(a and b)) [57, 58]. Meanwhile, the total mass loss of Sr/Cs@GXU-SODs was 21.12 and 24.21 % due to the dehydration of sodalite framework [59]. A subsequent exothermic peak around 1100 °C corresponded to reconstruction of the sodalite framework into thermodynamically stable ceramic phases [60] with no endothermic melting signal, indicating that the phase transformation did not involve bulk liquid-phase sintering, which was consistent with solid-state reaction pathway [61]. The XRD patterns of Sr/Cs@GXU-SODs varied with temperature the range of 900-1200 °C shown in Fig. 10(c and d). Sr/Cs@GXU-SODs both showed amorphous peaks at 900 °C, but new diffraction peaks appeared at 1000-1200 °C, confirming the phase transformation occurred, which was consistent with the phenomenon shown in Fig. 10(a and b). The new characteristic diffraction peaks corresponded

to nepheline ($\text{Na}_{6.65}\text{Al}_{6.24}\text{Si}_{9.76}\text{O}_{32}$, PDF#83-2372), stronalsite ($\text{Na}_2\text{SrAl}_4(\text{SiO}_4)_4$, PDF#40-1451) and pollucite ($(\text{Cs}, \text{Na})\text{AlSi}_2\text{O}_6$, PDF#15-0317). The intensity of the diffraction peaks increased first and then decreased as the calcination temperature rose, and the peaks at 1100 °C had highest intensity than those of. Notably, no separate SrO or Cs_2O phases were detected, indicating that the radionuclides have been fully incorporated into the crystal lattice of the newly formed phases rather than merely physically encapsulated.

A critical safety consideration for the integrated adsorption-immobilization strategy is the potential volatilization of radionuclides—particularly Cs, which is known to exhibit appreciable volatility at elevated temperatures—during the calcination step. To address this concern, the TG-DSC analysis (Fig. 10(a and b)) showed no distinct mass loss events in the temperature range of 900–1200 °C beyond the dehydration-related weight loss below 200 °C, suggesting that Cs and Sr remain effectively retained within the solid matrix throughout the thermal treatment. This is consistent with the XRD observation that Cs is fully incorporated into the pollucite structure at 1100 °C, as pollucite formation is known to chemically stabilize Cs against volatilization. Nevertheless, for practical implementation, the calcination should be conducted under controlled atmosphere with appropriate off-gas treatment to capture any trace volatile species, following standard practices in nuclear waste vitrification.

The Rietveld refinement results for Cs/Sr@GXU-SODs calcined at 1100 °C revealed that the phase assemblage consists almost entirely of nepheline with only a trace amount of pollucite (0.48 wt.%), stronalsite (1.02 wt.%) and no detectable amorphous phase or Cs_2O /SrO impurity in Fig. S4. The refined nepheline lattice parameters ($a = b = 9.996 \text{ \AA}$, $c = 8.380(1) \text{ \AA}$) show a slight expansion compared to standard nepheline ($a = b = 9.982 \text{ \AA}$, $c = 8.368 \text{ \AA}$), providing crystallographic evidence for the incorporation of Sr^{2+} or Cs^+ ion into the Na^+ sites via substitutional solid solution, rather than physical encapsulation or phase separation. The theoretical maximum loadings of the solid solution end-members (28.2 wt.% for Sr, 55.6 wt.% for Cs) are substantially higher than the experimental values (6.58 wt.% Sr, 2.78 wt.% Cs), consistent with the low substitution ratios (~0.5–0.6 at.%)

required to accommodate the adsorbed radionuclides. The combined evidence confirms the lattice incorporation mechanism rather than physical encapsulation.

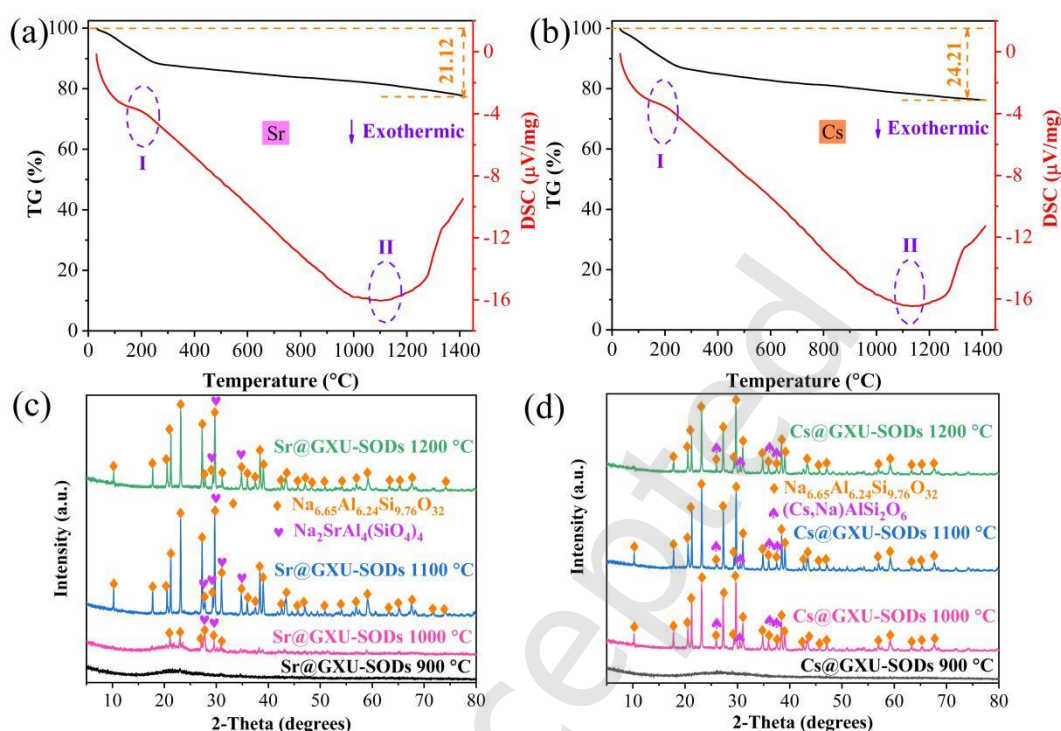


Fig 10. TG-DSC curves (a: Sr, b: Cs) and XRD pattern (c, d) of Sr/Cs@GXU-SODs at different calcination temperatures.

To investigate the morphological and thermal stability of Sr/Cs@GXU-SODs, OM images are shown in Fig. S5 with different calcination temperatures ranging from 900 to 1200 °C. Both Sr/Cs@GXU-SODs still maintained well-defined spherical morphology from 900 to 1100 °C (Fig. S5(a-c and e-g)). The above excellent structural integrity indicated that the phase transformation occurred primarily through internal lattice reconstruction without macroscopic particle sintering, which still preserved the original spherical geometry. However, the microspheres at 1200 °C transformed into molten mass with obvious pores appearing on the surface (Fig. S5(d and h)), which was attributed to exceeding the eutectic point of the aluminosilicate ceramic system [59]. Therefore, the calcination temperature of Sr/Cs@GXU-SODs should not exceed 1100 °C to achieve in-situ immobilization while preserving structural integrity. In order to investigate the leaching stability of GXU-SODs, the leaching concentrations of elements during the reaction process were measured in

Table S7. The leaching concentrations of Na, Al and Si were 0.28, 1.92 and 1.28 mg·L⁻¹, respectively.

The ion leaching concentrations were all lower than the threshold values determined by the Hazardous Waste Distinction Standard-Leaching Toxicity Distinction (GB5085.3-2007, China) and the National Environmental Quality Standard for Surface Water (GB3838-2002, China), indicating that the GXU-SODs did not pose any risk of secondary pollution.

To evaluate the effect of thermal treatment on the long-term chemical stability of Sr/Cs@GXU-SODs, normalized leaching rates were determined under non-sintered condition and after calcination at 900, 1000, and 1100 °C (Fig. 11(a-d)). The non-sintered Sr/Cs@GXU-SODs exhibited relatively high leaching rates during the 7-day period, followed by a gradual decline with 1.32×10^{-4} and $0.013 \text{ g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ for Sr²⁺ and Cs⁺ after 28 days, respectively shown in (Fig. 11(a)). This related to the DFT-derived diffusion barriers: Cs⁺ with its lower adsorption energy and closer hydrated radius to the 6MR aperture, is more weakly held and more easily leached from near-surface exchangeable sites, whereas Sr²⁺ is more deeply trapped inside the β -cage [60]. Fig. 11(b) shows that the leaching rate decreased compared to the non-sintering samples under the calcination at 900 °C, but the incomplete phase transformation left residual amorphous regions, which still allowed for slow ion diffusion. As the phase transformation progressed significantly, the leaching rate further decreased at 1000 °C (Fig. 11(c)). With the increasing temperature to 1100 °C, the leaching rate significantly decreased to 8.6×10^{-7} and $2.82 \times 10^{-6} \text{ g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ for Sr²⁺ and Cs⁺ after 28 days, respectively (Fig. 11(d)), which was close to the glass ceramic leaching rate (10^{-6} - 10^{-7}) and far below the regulatory standard EJ 1186-2005. This outstanding in-situ immobilization performance was attributed to the complete transformation into thermodynamically stable ceramic phases wherein radionuclides are immobilized via lattice incorporation rather than reversible ion exchange.

Based on the combined experimental evidence, schematic structural models are proposed in Fig. 11(e and f) to illustrate the in-situ immobilization mechanism. Upon thermal treatment at 1100 °C, the original SOD framework undergoes reconstructive phase transformation, wherein Sr²⁺ and Cs⁺ ions

originally confined within the β -cages become structurally incorporated into the newly formed aluminosilicate ceramic phases. In these dense crystal structures, Sr^{2+} and Cs^{+} are no longer exist as exchangeable cations within the β cages but are embedded in specific lattice sites, where Sr^{2+} and Cs^{+} are coordinated with framework oxygen atoms through strong ionic covalent bonds. Therefore, the outstanding in-situ immobilization performance observed at 1100 °C highlights the feasibility of the integrated adsorption-immobilization strategy as a viable approach for the safe treatment of radioactive waste.

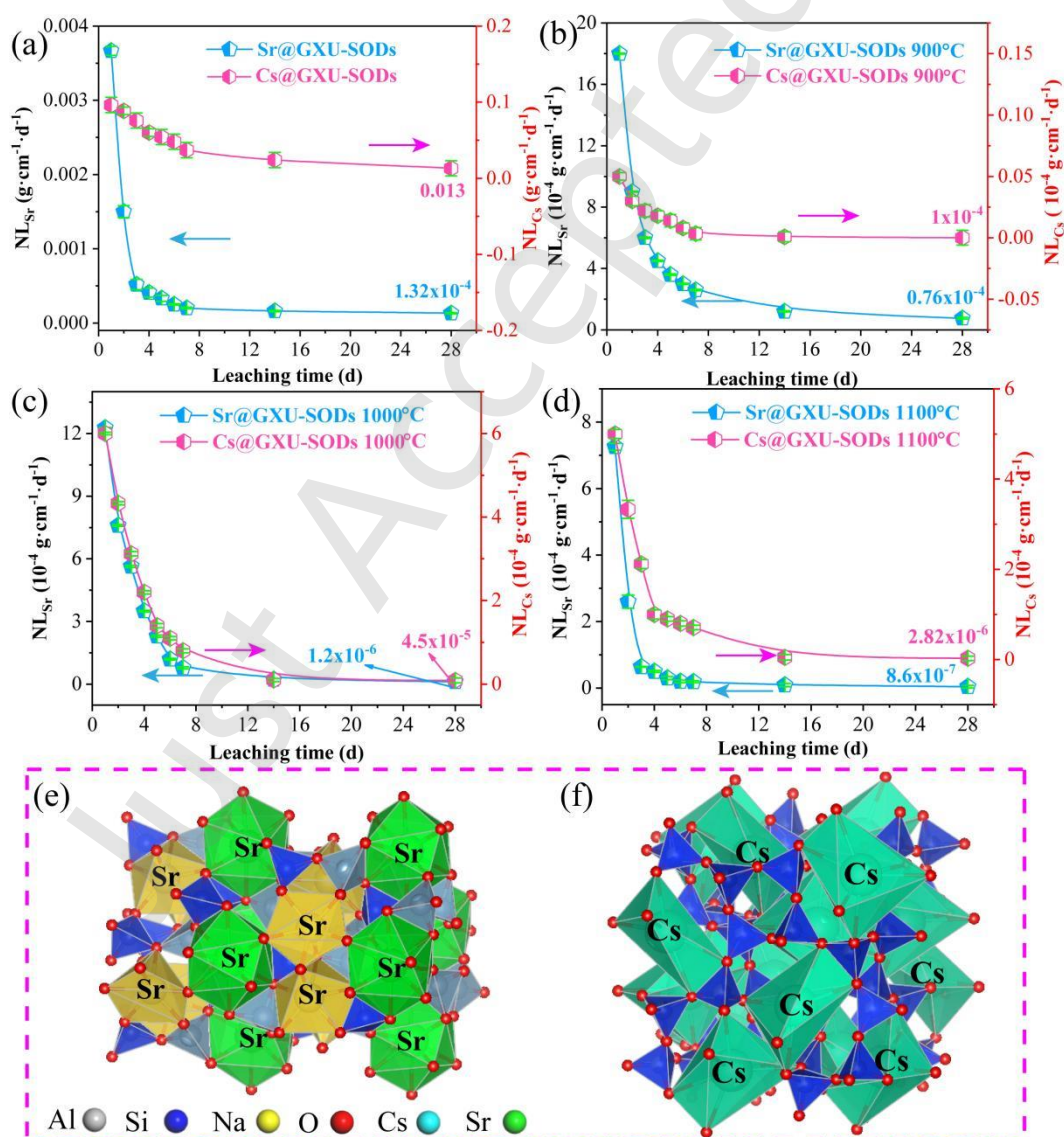


Fig 11. Normalized leaching rates of Sr/Cs@GXU-SODs under non-sintering (a) and sintering conditions (b-d: 900, 1000, 1100 °C), structural model (e: Sr, f: Cs) of Sr/Cs@GXU-SODs at 1100 °C.

It is noteworthy that the dramatic reduction in leaching rates after thermal treatment is not merely a consequence of physical densification or pore collapse [62, 63]. Rather, the phase transformation from the original sodalite framework to thermodynamically stable ceramic phases (nepheline, stromalolite, and pollucite) alters the chemical state of the confined radionuclides. In the original sodalite structure, Sr^{2+} and Cs^+ reside as extra-framework charge-balancing cations, remaining exchangeable and potentially releasable under aqueous conditions. Upon calcination at 1100 °C, however, the thermally induced phase transformation incorporates these cations into specific lattice positions, where they are coordinated by framework oxygen atoms through strong ionic/covalent bonds. This transition from exchangeable extra-framework species to lattice-incorporated constituents eliminates the primary leaching pathway, rendering the immobilization permanent and largely independent of the external ion environment. Therefore, the decisive factor for the ultralow leaching rates is the chemical fixation enabled by the newly formed crystalline phases, rather than physical encapsulation alone.

To elucidate the structural evolution and in-situ immobilization mechanism of radionuclides during thermal treatment, FT-IR spectroscopy was performed on Sr/Cs@GXU-SODs calcined at different temperatures (Fig. 12(a and b)). With rising calcination temperature, the intensity and peak position of each characteristic band evolve in a regular manner, reflecting the thermal evolution of framework topology, local coordination environment and bonding states at the molecular level, which is in good agreement with the XRD phase analysis results. The peaks around 3435cm^{-1} were assigned to O-H stretching and bending vibrations [6]. With the increase of calcination temperature, the intensity of this band decreased significantly, revealing that adsorbed water and structural hydroxyl groups inside the samples gradually eliminated under high temperature. The continuous improvement in matrix density can effectively reduce diffusion pathways for radionuclide leaching. The band around 991cm^{-1} corresponded to Si/Al-O-Si stretching vibration [31]. The peak at 686cm^{-1} represented the symmetric stretching vibration of the Si(Al)-O-Si(Al) bonds in the zeolite framework [52]. The peak at 611cm^{-1} was attributed to the symmetric stretching of Al-O bonds [53]. The peak around 570cm^{-1}

was a characteristic peak of the double six-membered ring bending vibration of Si-O-Al bonds [54].

The peak at 467 cm^{-1} corresponded to the bending vibration of Si/Al-O bonds [30]. In addition, a new peak at 1235 cm^{-1} was observed from Fig. 12(a) due to the vibration absorption of Sr-O bond in strontium aluminate silicate [18].

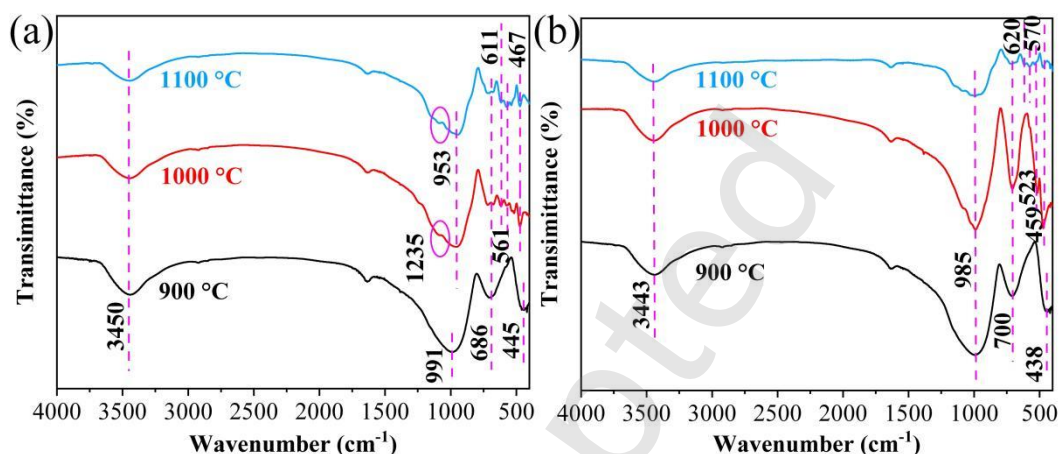


Fig 12. FT-IR images (a, b) of Sr/Cs@GXU-SODs at different calcination temperatures.

To reveal the calcination temperature on the in-situ immobilization performance of Sr@GXU-SODs, SEM-EDS spot scanning was employed to characterize the micromorphology and micro-area elemental distribution of samples calcined at 900, 1000 and 1100 °C in Fig. 13(a-c). Fig. 13(a) indicates that the sample calcined at 900 °C possesses abundant pore structures, with Sr mass fractions varying from 4.60 to 11.20 wt.% among different micro-regions, showing remarkable compositional heterogeneity. Fig. 13(b) shows numerous irregular pores remain inside. The maximum difference in Sr content between micro-zones reaches 5.17 wt.%, demonstrating insufficient sintering mass transfer and atomic diffusion at this temperature, which inhibits homogeneous immobilization. Fig. 13(c) depicts the internal structure maintains dense accompanied by a remarkable decline in porosity. The Sr mass fractions at three random detection points stabilize within the range of 6.25-6.91 wt.% with only a content difference of 0.66 wt.%, reflecting a substantial improvement in elemental distribution homogeneity. In summary, calcination at 1100 °C simultaneously realizes sintering densification of materials and homogeneous lattice immobilization of strontium, endowing the solidified matrix with outstanding structural stability and radionuclide retention capacity. Therefore, 1100 °C was

determined as the optimal process parameter for in-situ calcination immobilization of strontium after adsorption.

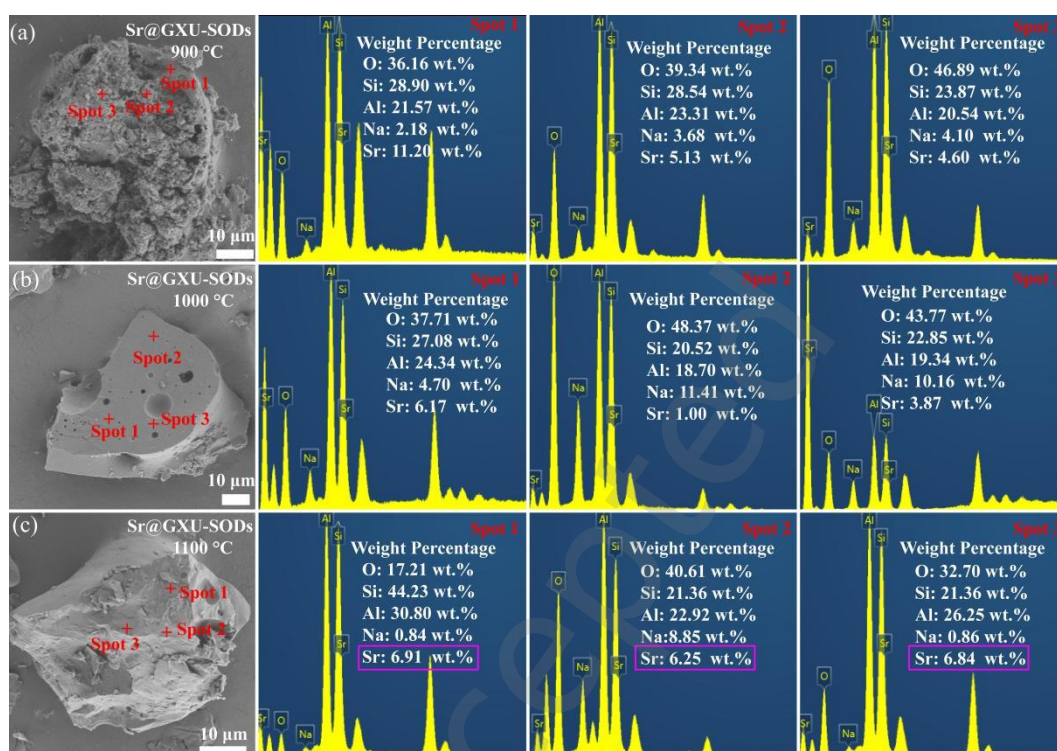


Fig 13. Internal surface and spot scanning images of Sr@GXU-SODs at different calcination temperatures (a-c: 900, 1000, 1100 °C).

In-situ immobilization performance of Cs@GXU-SODs at different temperatures is also measured by spot scanning calcined at 900, 1000 and 1100 °C in Fig. 14(a-c). The results demonstrate that the sample calcined at 900 °C exists as loosely aggregated fine particles without obvious sintering fusion between grains in Fig. 14(a). The mass fractions of Cs in different micro-regions range from 3.25 to 6.99 wt.%, exhibiting evident local enrichment. This indicates that cesium is merely loaded on particle surfaces in a surface-adsorbed state without effective bulk diffusion at this temperature, leading to weak bonding strength between Cs and the matrix and insufficient immobilization stability. Fig. 14(b) shows that the sample calcined at 1000 °C still retains abundant internal pores. The maximum difference in Cs content among micro-zones reaches 9.15 wt.%, reflecting that although thermal migration of Cs atoms occurs at this temperature, sintering mass transfer and lattice diffusion are inadequate [54]. Fig. 14(c) shows after calcination at 1100 °C, the sintering densification degree

greatly improves. Meanwhile, the Cs mass fractions at three random detection points stabilize within 2.55-3.00 wt.% with an extremely low content difference of only 0.45 wt.%, achieving highly homogeneous bulk distribution of cesium. The above results verify that high temperature of 1100 °C simultaneously promotes matrix sintering densification and uniform bulk diffusion of cesium atoms, enabling the transformation of cesium from surface-adsorbed state to stable bulk solid-solution state [64]. While the overall process remains predominantly solid-state, thereby enabling efficient lattice incorporation of radionuclides. Therefore, 1100 °C was identified as the optimal process parameter for in-situ calcination immobilization after cesium adsorption.

While the present 1100 °C thermal treatment affords exceptional radionuclide retention (with 28-day leaching rates of 8.6×10^{-7} and $2.82 \times 10^{-6} \text{ g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ for Sr^{2+} and Cs^{+} , respectively) through reconstructive phase transformation into thermodynamically stable ceramic phases, alternative low-energy immobilization routes—hydrothermal treatment, mechanochemical conversion, and chemical hardening—are worth considering for their operational simplicity, yet each entails inherent performance trade-offs. Hydrothermal conversion (110-200 °C) enables pollucite/sodalite formation but yields leaching rates substantially higher (10^{-3} - $10^{-4} \text{ g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$) because immobilization relies predominantly on ion exchange within zeolitic cages rather than the strong ionic-covalent lattice incorporation achieved by high-temperature sintering. Mechanochemical ball milling operates at ambient temperature via defect-mediated incorporation, but its long-term durability under geological disposal conditions remains inadequately validated and scalability presents practical challenges. Chemical hardening in cementitious/geopolymer matrices, while practically convenient, primarily offers physical encapsulation with limited chemical binding, resulting in leaching rates orders of magnitude higher (10^{-2} - $10^{-3} \text{ g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$) and suffers from matrix-adsorbent incompatibility and susceptibility to degradation. Thus, although high-temperature sintering incurs a greater energy penalty, it delivers two to four orders of magnitude superior retention performance, radiation resistance, and long-term chemical durability—making it the preferred choice for high-hazard wastes where

permanent isolation is paramount; for low- to intermediate-level wastes, hydrothermal or cementitious routes may suffice if regulatory leaching criteria are met. Future efforts could explore intermediate-temperature or microwave-assisted sintering to reduce energy consumption while preserving the beneficial lattice-incorporation mechanism characteristic of ceramic waste forms.

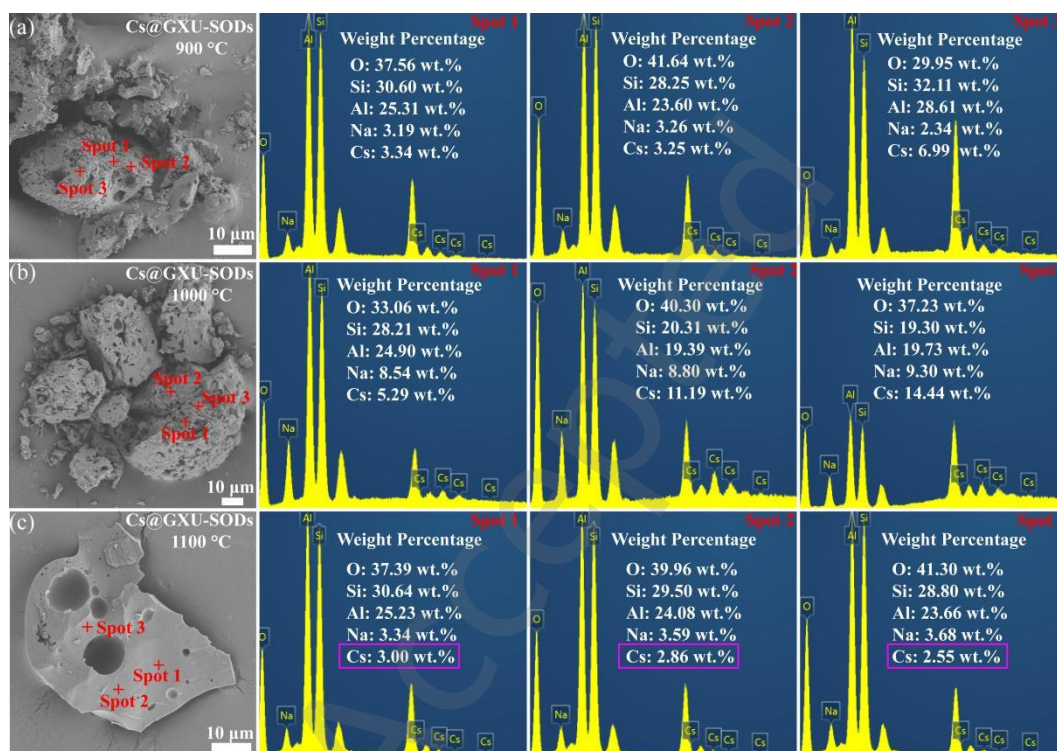


Fig 14. Internal surface and spot scanning images of Cs@GXU-SODs at different calcination temperatures (a-c: 900, 1000, 1100 °C).

4 Conclusion

This study fabricated a novel one-pot non-hydrothermal approach for the fabrication of robust sodalite zeolite microspheres (GXU-SODs) via geopolymer technology, enabling an integrated adsorption-immobilization strategy for the efficient removal and permanent stabilization of Cs^+ and Sr^{2+} from radioactive wastewater. The optimized GXU-SODs exhibited well-defined spherical morphology, high crystallinity, and favorable textural properties, achieving maximum adsorption capacities of $59.64 \text{ mg} \cdot \text{g}^{-1}$ for Cs^+ and $56.67 \text{ mg} \cdot \text{g}^{-1}$ for Sr^{2+} , with rapid kinetics governed by monolayer chemisorption. The material demonstrated exceptional radiation resistance, retaining over 96% of its structural integrity and adsorption performance after exposure to 500 KGy. Dynamic column

experiments further confirmed its practical applicability for continuous-flow treatment in real seawater matrices. Mechanistic investigations combining spectroscopic characterizations and DFT calculations revealed that the adsorption process was primarily driven by ion exchange with the six-membered ring apertures of the sodalite cage and surface chemisorption on external sites. Moreover, the spent adsorbents could be directly converted into durable ceramic waste forms via simple thermal treatment at 1100 °C without the need for secondary additives. The Sr/Cs@GXU-SODs at 1100 °C exhibited significantly reduced 28-day leaching rates of 8.6×10^{-7} and $2.82 \times 10^{-6} \text{ g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ for Sr^{2+} and Cs^{+} well below the regulatory limits, confirming the in-situ immobilization of radionuclides through lattice incorporation rather than reversible physical confinement. This work not only provides a scalable and template-free synthesis route for advanced sodalite-based materials but also establishes a streamlined adsorption-immobilization paradigm that minimizes process complexity, reduces secondary waste generation, and significantly enhances the safety and sustainability of radioactive waste management.

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Author contributions

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Availability of data and materials

Not applicable.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

Electronic Supplementary Material (ESM)

Supplementary materials are available in the online version of this article.

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