Journal of Radioanalytical and Nuclear Chemistry (2025) 334:863–872
https://doi.org/10.1007/s10967-024-09838-3

Solubility, speciation and sorption calculation of americium in far-field and near-field Beishan groundwaters

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Received: 12 August 2024 / Accepted: 13 October 2024 / Published online: 23 October 2024
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Abstract
Knowledge of the speciation of americium in groundwaters is important for the safe disposal of the high-level radioactive waste. In this work, we investigated the solubility, speciation and sorption of americium in Beishan groundwaters with CHEMSPEC code. Our results showed that the solubility of Am in far-field and near-field Beishan groundwater is 1.57×10^{-7} and 2.3×10^{-8} mol/L, respectively. In addition, complexing inorganic anions can significantly affect the speciation of americium and the order of impact is as follows: $HCO_3^- > F^- > SO_4^{2-} > Si(OH)_{4(aq)}$. The sorption calculations indicated almost all americium in Beishan groundwaters would be sorbed on quartz.

Keywords Americium · Groundwater · Speciation · Sorption · Solubility

Introduction

Deep geological repository (DGR) based on "multi-barrier" concept has been considered as the unique strategy for the safety disposal of the high-level radioactive waste (HLW) [1–3]. The multi-barrier system typically consists of an engineered barrier and the natural barrier to isolate the waste from the biosphere. However, the retention, diffusion and sorption behaviors of radionuclides in multi-barrier systems are heavily dependent on the chemical speciation of radionuclides in the repository groundwater [4–6]. For the performance assessment of the DGR, it is important to investigate the chemical speciation of radionuclides based on the chemical components in repository groundwaters. Chemical speciation, *i.e.* the distribution of various elements among their chemical forms, is the calculated equilibrium composition based on the components, electric potential, temperature and pressure in the system [7]. Extensive studies were committed to exploring the species of radionuclides with advanced spectral analysis approaches, but the results are limited by the concentration of the radionuclides and preparation method of the samples [8–12]. Thus, the calculation method is still an effective and important way to figure out the species of radionuclides in repository groundwater.

In China, Beishan area has been selected as a candidate site for the HLW final deep geological repository, thus the solubility and speciation are critical for the reliable pre-safety assessment of the DGR [13, 14]. Actinides (*e.g.*, neptunium, plutonium, americium and curium) will make a major contribution to HLW radiotoxicity after storage periods more than ten thousand years. It is estimated that when China's proposed geological repository is open for loading, the country's HLW inventory will contain 9.58 tonnes of Am [15]. Americium, with an atomic number of 95, has around 30 isotopes or isomers, with mass numbers ranging from 232 to 247 and half-lives between 55 s to 7370 years [9]. Among these isotopes, Am-241 (half-lives $T_{1/2} = 432.2$ years) is the most important one because it would be the dominant radioactivity contributor in the buried wastes. Furthermore, americium is known in formal valence states of 0, III, IV, V, and VI, but only state III is stable in natural redox conditions.

Solubility and speciation data of americium in Beishan groundwaters are important to understand the mobility of americium in repository environments. *Kim* et al. studied the solubilities of actinides in a Ca-bentonite porewater

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with PHREEQC-2 code and the results suggested that the respective solubility of Am is 1.2×10^{-7} mol/L [16]. *Yang* et al. investigated the solubitlity of amecium in Beishan far-field and near-field groundwater by PHREEQC-3 code, while the data are calculated to be 6.3×10^{-7} mol/L and 2.4×10^{-6} mol/L, respectively [15]. However, these work didnot simulate the sorption curves of americium on mienrals based on calculated speciaiton of americium in groundwaters.

In the last few decades, surface complexation models (SCMs) have been developed to supply a more robust description of surface reactions. There have been some efforts in the past to do manual compilation of sorption data and development of SCM databases [17, 18]. The RES^{3}T (www.hzdr.de/res3t) is the most important and well-recognized open source thermodynamic sorption database, which collects numerous SCM parameters, such as mineral properties, surface complexation reactions, experimental conditions and extensive references [19]. At present, this database comprises information from 3531 literature references, including reactions between 149 minerals and 140 sorbing ligands, and a total of 7647 surface complexation reaction constants. A number of studies have employed this database to predict sorption data onto minerals or to develop a robust model [18, 20, 21]. Our previous work [22] also succsesfully simulated the sorption data with the RES^{3}T and the chemical speciation code CHEMSPEC, which is wrote by ourselves [7].

The objectives of this work were to investigate the solubility and speciation of americium in far-field and near-field Beishan groudwaters with CHEMSPEC code and study the influence of different complexing inorganic anions on Am speciation distribution. Furthermore, this work intented to predict the sorption behavior of americium on quartz based on aqeous speciaiton of americium in groundwaters with three different SCMs collected in the RES^{3}T database. Our work might help us understand the speication and moblity of amercium in Beishan groundwaters and provide valuable information for the performance assessment of the repository.

Calculation

Brief description of CHEMSPEC code

In general, CHEMSPEC code divides chemical materials into two categories in an aqueous solution, components and species. To be specific, components are reactants that can chemically react to form any species in a system. According to the input files, the code searches all possible species in database and outputs results after reaching convergence. In this context, the database, reaction model, calculation method and ionic strength coefficient represent the most crucial factors. The CHEMSPEC database used in this work were derived from the authoritative NAPSI_290502(260,802)(PSI) database, which encompasses 66 components and 448 species, respectively. The database covers thermodynamic data for Th, U, Np, Pu, Am and other high-yield and long-lived fission product elements that are of paramount importance for the disposal of high-level waste (HLW). In order to get reliable results, database is supposed to be precise, comprehensive and updated in time. CHEMSPEC can deal with single or complex systems involving acid–base, precipitation, redox and complexation reactions. With regard to the calculation method, the mass balance equation represents the core of the software. Moreover, in excess of four algorithms are employed to solve the equation, such as Newton–Raphson method, Levenberg–Marquardt algorithm and the steepest decent method. In comparison with other species analysis software such as PHREEQC, CHEMSPEC is more likely to reach convergence by automatically switching algorithm. Furthermore, among different ion strength formulae, CHEMSPEC is capable of selecting the optimal one when the concentration of ions changes. According to different values of the ionic strength, CHEMSPEC employs different models to calculate the activity coefficient of an ion in solution: (1) Extended Debye-Hückel model ($I<0.1$); (2) Davies model ($I<0.5$); (3) B-dot model ($I<1.0$); (4) SIT model ($1.0<I<2.0$). In this work, the ionic strength is less than 0.5, thus the Davies model and Extended Debye-Hückel model are used for calculation. Detailed information of the CHEMSPEC code can be referred to relative literature [7].

In addition, surface complexation models (SCMs) have been successfully incorporated into CHEMSPEC, which facilitates the simulation of radionuclide sorption on minerals. These models include non-electrostatic model (NEM), constant capacitance model (CCM), diffuse layer model (DLM), triple layer model (TLM) and basic stern model (BSM). The classification of SCM can be categorised according to different criteria, including the 1-pK and 2-pK model, 1-site and 2-sites model. The parameters required for each model vary considerably.

Far-field and near-field Beishan groundwaters

The recently drilled borehole BS28 is situated in close proximity to the Xingchang distrcit in Beishan, which is the location of the first China's underground research laboratory (URL) [14]. The water sample from BS28 at a depth of 500 m provides a representative illustration of the Beishan far-field groundwater [23]. Its chemical components are listed in Table 1. By in-situ measurements at a depth of 365–690 m, the pH and *Eh* values are reported to be 7.8 and 98 mV, respectively.

Table 1 Chemical compositions of groundwaters in the Beishan area (mmol/L)

	Na^+	Ca^{2+}	K^+	Mg^{2+}	Cl^-	HCO_3^-	SO_4^{2-}	F^-	NO_3^-	$SiO_{2(aq)}$	pH	*Eh*(mV)
near-field	32.3	0.342	0.116	0.119	17.9	2.31	0.115	none	0.367	0.0705	8.9	–182
far-field	45.2	4.58	0.409	2.08	36.2	2.15	9.18	0.106	0.228	0.0912	7.8	98

Table 2 Equilibrium and thermodynamic date for americium species

Species	Chemical reaction	log *K*
$AmOH^{2+}$	$Am^{3+} + H_2O = AmOH^{2+} + H^+$	−7.30
$Am(OH)_2^+$	$Am^{3+} + 2H_2O = Am(OH)_2^+ + 2H^+$	−15.2
$Am(OH)_3$	$Am^{3+} + 3H_2O = Am(OH)_3 + 3H^+$	−25.7
AmF^{2+}	$Am^{3+} + F^- = AmF^{2+}$	3.40
AmF_2^+	$Am^{3+} + 2F^- = AmF_2^+$	5.80
$AmCl^{2+}$	$Am^{3+} + Cl^- = AmCl^{2+}$	1.05
$AmSO_4^+$	$Am^{3+} + SO_4^{2-} = AmSO_4^+$	3.85
$Am(SO_4)_2^-$	$Am^{3+} + 2SO_4^{2-} = Am(SO_4)_2^-$	5.40
$AmNO_3^{2+}$	$Am^{3+} + NO_3^- = AmNO_3^{2+}$	1.33
$AmCO_3^+$	$Am^{3+} + HCO_3^- = AmCO_3^+ + H^+$	−2.53
$Am(CO_3)_2^-$	$Am^{3+} + 2HCO_3^- = Am(CO_3)_2^- + 2H^+$	−8.36
$Am(CO_3)_3^{3-}$	$Am^{3+} + 3HCO_3^- = Am(CO_3)_3^{3-} + 3H^+$	−15.8
$AmSiO(OH)_3^{2+}$	$Am^{3+} + Si(OH)_4 = AmSiO(CO_3)_3^{2+} + H^+$	−1.71
$Am(OH)_3$(cr)	$Am^{3+} + 3H_2O = Am(OH)_3$(cr) + $3H^+$	−15.2
$AmCO_3OH$(cr)	$Am^{3+} + H_2O + HCO_3^- = AmCO_3OH$(cr) + $2H^+$	−3.13

However, the chemcial compositions of near-field Beishan groundwater are complicted due to the effects of the interaction with backfill material, canister and HLW solidified material. Some work used the Beishan groudwater to conatct with Gaomiaozi bentonite (proposed backfill material) and low-carbon steel (proposed canister material), and the reacted groudwater was simulated to be the near-field repository groundwater (vault water) [15, 24, 25]. The reported chemical compositions are also listed in Table 1. One can see that the *Eh* of the near-field Beishan groundwater (− 182 mV) is much more reducing than that of the far-field Beishan groundwater (98 mV), while it might be caused by the iron in canisiter and the Fe(II)-bearing minerals in backfill materials, such as pyrite [26]. The temperature was maintained at 293 K, and the density of the solution was held constant at 1.0 g cm^{-3} throughout the calculations.

Thermodynamic data

Though americium can be found in several oxidation states in repository environments, only the III oxidation state is prevailing. Therefore, this study only take the aqueous and solid species of Am(III) into calculations. All used thermodynamic data of Am are listed in Table 2. Two crystalline solid species ($Am(OH)_3$(cr) and $AmCO_3OH$(cr)) are considered in the system. In addition to the hydrolysis reactions of Am(III), the complexation reaction of Am(III) with complexing inorganic anions such as bicarbonate, sulfate, fluoride and silicate is the most probable process responsible for Am speciation in Beishan groundwaters.

Results and discussion

The influence of pH value and total Am concentration

It is assumed that pH value is one of the most importatnt factors affecting Amecium speciation. We calculted the speciation distrbution of Amecium in Beishan far-field and near-field groundwaters for pH ranging from 3.0 to 11.0, as shown in Figs. 1 and 2. The different total concentration of Am for 10^{-6}, 10^{-7}, 10^{-8} and 10^{-9} mol/L in both Beishan groundwaters were investigated. The *Eh* value of the Beishan far-field groundwater was controlled to be 98 mV and kept constant throughout the calculations, while that of near-field groundwater was matained at −182 mV.

In Beishan far-field groundwater, when total Am concentration is 10^{-9} mol/L (shown in Fig. 1d), Am mainly existed in the species of $AmSO_4^+$, $Am(SO_4)_2^-$ and Am^{3+} for pH values inferior to 6.9, and the contents of the three species decreased slightly with the increase of pH. When 6.9 < pH < 10.4, the concentration of Am-carbonate and Am-silicate complexes gradually increase and then become the dominant species, such as $AmCO_3^+$, $Am(CO_3)_2^-$, $Am(CO_3)_3^{3-}$ and $AmSiO(OH)_3^{2+}$. When pH over 10.4, the concentration of OH^- in the solution gradually increases, leading to the formation of the crystalline precipitation $Am(OH)_{3(cr)}$, which is the solubility limiting solid phase. At pH 7.8, which is the measured pH value of the Beishan far-field groundwater, the domiante species are $AmCO_3^+$ (50.4%), $AmSiO(OH)_3^{2+}$ (34.6%) and $Am(CO_3)_2^-$ (7.9%). As the total concentration of Am increase to 10^{-8} and 10^{-7} mol/L in Beishan far-field groundwater, as shown in Fig. 1c and 1b. The species at pH 7.8 both only show a negliagible change, which consist of ~ 50% $AmCO_3^+$ and ~ 30% $AmSiO(OH)_3^{2+}$. However, in the case of 10^{-6} mol/L total Am concentration, the species at pH 7.8 show a significant change. The crstalline solid $AmCO_3OH_{(cr)}$ become to precipitate and present as the major speices, accounting for 84.3% of the Am. This result also indicates that the solubility of Am in Beishan far-field groundwater is 1.57×10^{-7} mol/L (*i.e.* mol/kg), which is in agreement with the experimental

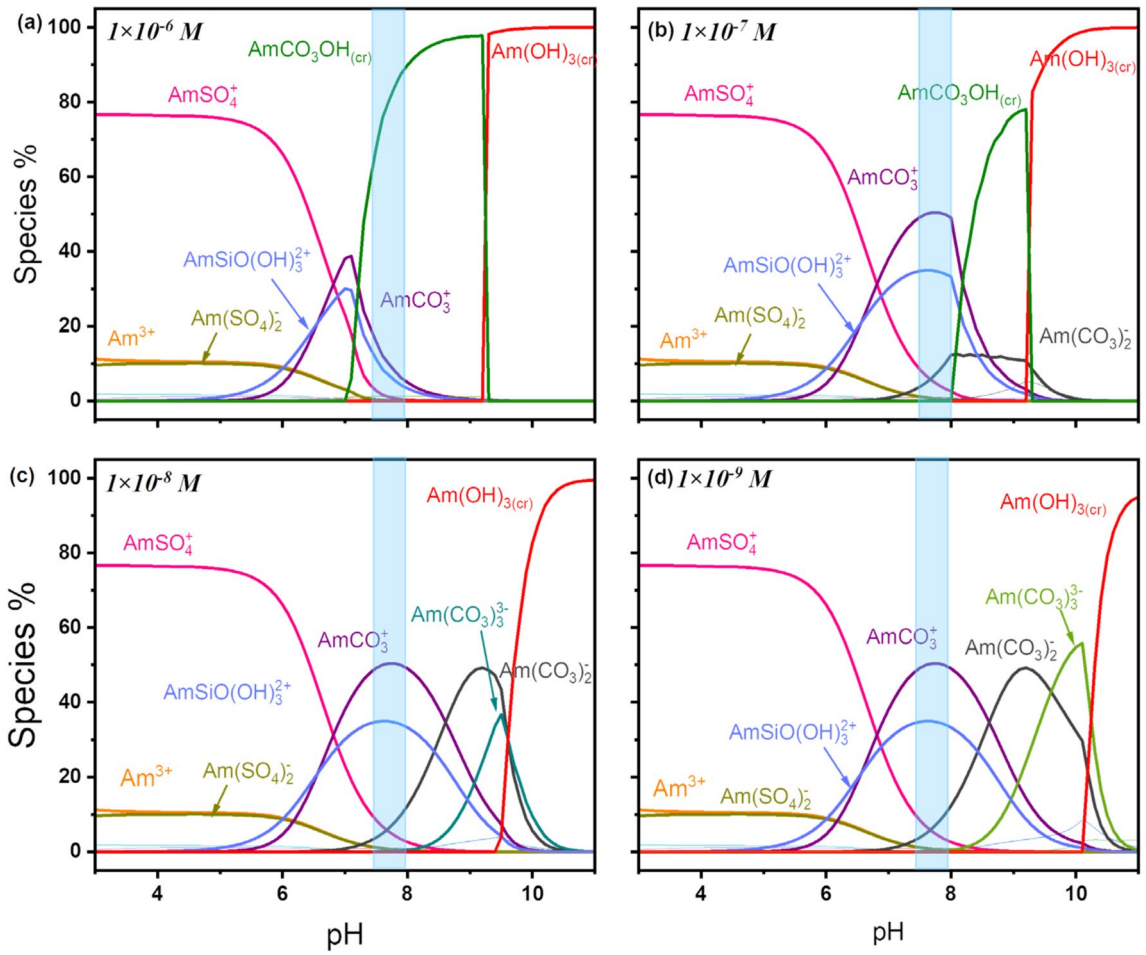


Fig. 1 Americium speciation distribution in Beishan far-field groundwater at different Am concentration. (blue area refers to the pH value of Beishan far-field groundwater)

observations (2×10^{-7} M) in synthesized groundwater [12] and the calculated resutls (1.2×10^{-7} M) in natrural gourndwater by PHREEQC software [16].

In Beishan near-field groundwater, when total Am concentration is 10^{-9} mol/L (shown in Fig. 2d), Am mainly existed as Am^{3+} (maximum 80%) at pH < 6.0. Compared to the far-field groundwater, the percentage of amercium monosulfate species decreases greatly under acidic conditions due to the low concentration of sulfate in the near-field groundwater. Concerning the pH range from 6.0 to 10.3, $AmCO_3^+$, $Am(CO_3)_2^-$ and $Am(CO_3)_3^{3-}$ species become the principal species. For the pH values greater than 10.3, the precipitate $Am(OH)_{3(cr)}$ appears and domniates the Am speciation. At pH 8.9, which is the measured pH value of the Beishan near-field groundwater, the main species are $Am(CO_3)_2^-$ (55.4%), $AmCO_3^+$ (24.9%), $Am(CO_3)_3^{3-}$ (9.1%) and $AmSiO(OH)_3^{2+}$ (8.0%). As the total Am concentration increase to 10^{-8} mol/L, the Am speciation show a minimal change. In contrast, when the total Am concentration increase to 10^{-7} and 10^{-6} mol/L in Beishan near-field groundwater, the crstalline solid $AmCO_3OH_{(cr)}$ appear and present as the main speices, which is similar to that in the far-field groundwater. The solubility of Am remain relatively constant, which is 2.3×10^{-8} mol/L in the near-field groundwater.

In addition, we found that the silicate might play a vital role in Am speciation distrubiton. Am and $Si(OH)_4$ can react to form $AmSiO(OH)_3^{2+}$, which can exist in large amounts in the pH from 6.0 to 9.0. Some reported groundwaters didnot include the concentration of silicate, thus the calculation based on those groundwaters might generate the unreliable results. For a better comparsion, we calculated the speciation distrubition without Am-silicate complex as functions of pH and Am concentration, as shown in Fig. S1. The main species of Am in Beishan far-field groundwater are $AmCO_3^+$ and $Am(CO_3)_2^-$, while crstalline precipitates appear when the amercium concentration is relatively high.

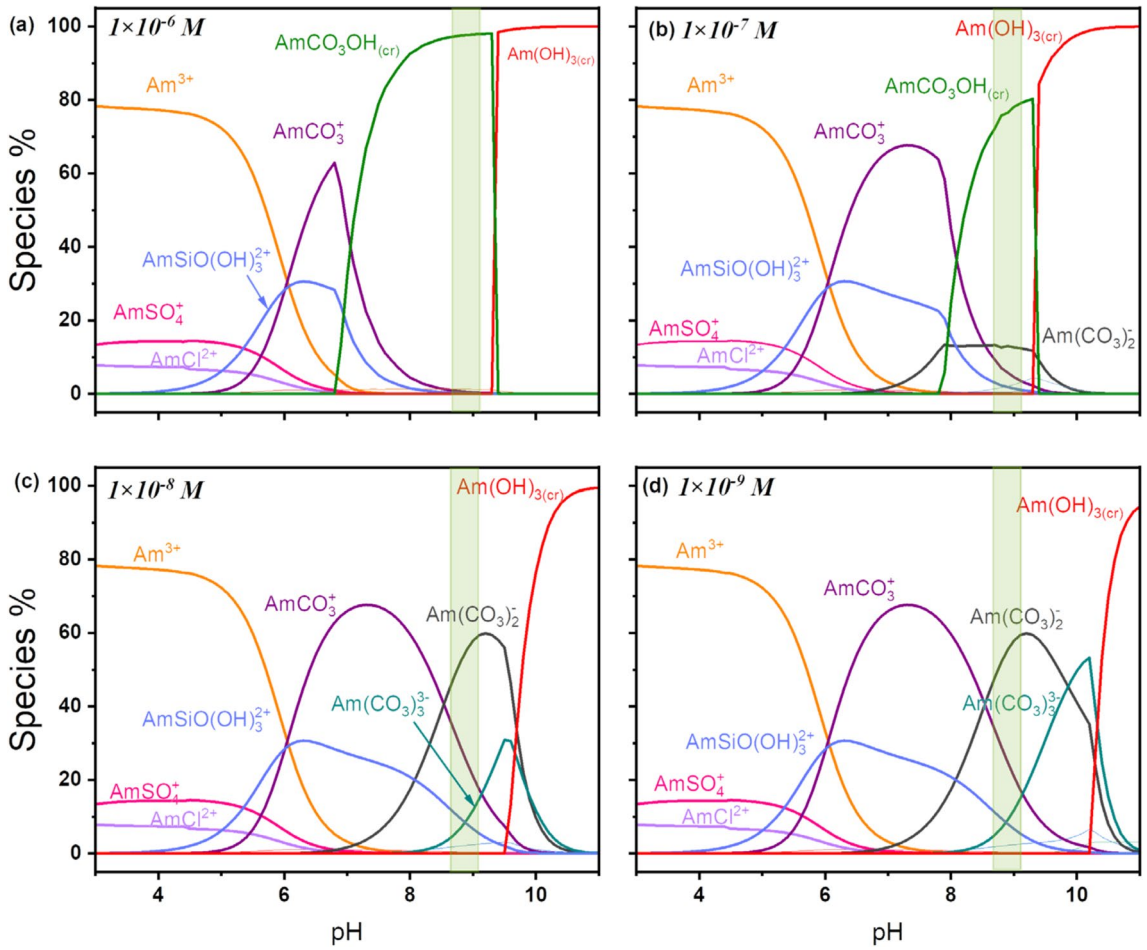


Fig. 2 Americium speciation distribution in Beishan near-field groundwater at different Am concentration. (green area refers to the pH value of Beishan near-field groundwater)

The influence of complexing inorganic anions

Both the near-field and far-field Beishan groundwaters contain varying concentrations of fluoride, sulfate, bicarbonate and silicate anions. The complexing inorganic anions can significantly affect the Am species due to the complexing reaction. For studying the extent of the influence of different anions on Am speciation distribution, we conducted speciation calculations in both groundwaters with varying concentrations of complexing inorganic anions. The results are shown in Fig. 3. The pH and *Eh* values were kept at their measured values throughout all calculations. Besides, the concentration of the corresponding complexing inorganic anion was altered while that of all other ions remained constant. The initial concentration of Am was 10^{-6} mol/L. Under this condition, Am mainly exists as a crstalline solid $AmCO_3OH_{(cr)}$ in both near-field and far-field Beishan groundwaters. Thus, it could directly show the influence of complexing inorganic anions on the solubility and speciation of americium.

In both near-field and far-field groundwater, as shown in Fig. 3, silicates have the weakest effect on Am speciation. The content of the crystalline solid $AmCO_3OH_{(cr)}$ stay constant (over 90%) with changes in silicate concentration. Apart from silicates, the fluorion also has a modest effect on Am speciation. Although the species AmF_2^+ increases sharply when the fluorion concentration is larger than $10^{-1.5}$ mol/L, the $AmCO_3OH_{(cr)}$ is still the primary species. Compared to fluorine and silicate ions, sulfate ions have a strong effect on Am speciation. The Am-sulfate complexes $AmSO_4^+$ and $Am(SO_4)^{2-}$ become the main speceis when the content of sulfate increaeses to $10^{-1.5}$ mol/L. However, bicarbonate anions have the strongest impact on Am speciation because the crystalline phase $AmCO_3OH_{(cr)}$ contains the contribution of bicarbonate. This solid becomes the main species only when the concentration of carbonate ions is larger than $10^{-3.8}$ mol/L. Otherwise, the primary Am species in groundwater is $AmSiO(OH)_3^{2+}$.

The impact of cation concentrations on the speciation of americium is generally weak because they do not form

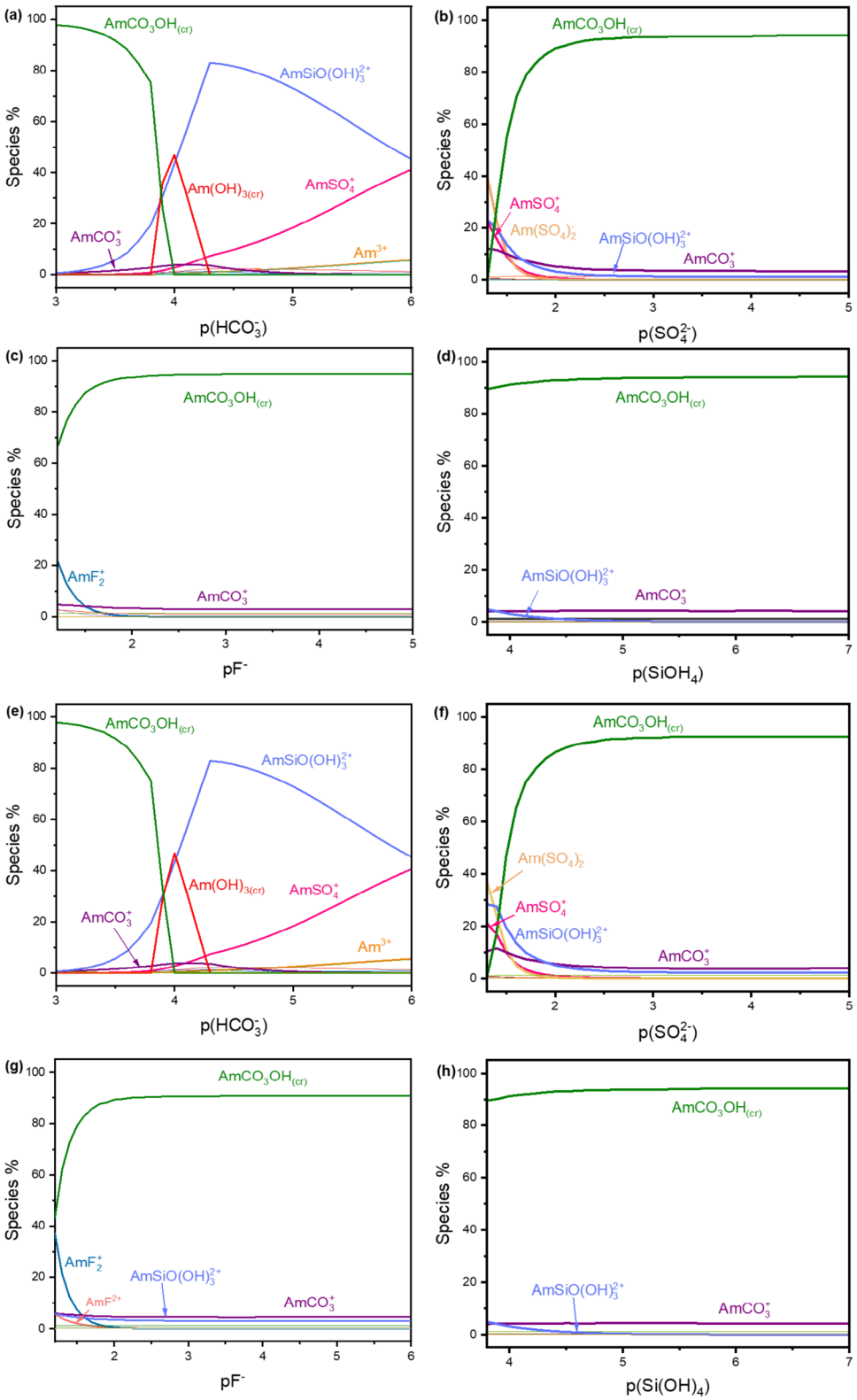


Fig. 3 The effect of anion concentrations on americium speciation distribution in Beishan far-field groundwater (**a**-**d**) and near-field groundwater (**e**–**h**)

complexes with americium. They affect the americium speciation through their effects on the ionic strength and competition for complexation reaction. The influence of chlorides on americium speciation in two groundwater is weak due to the calculations (Fig. S2).

In summary, an increase in the concentration of fluoride, silicate, and sulfate anions and a decrease in the concentration of carbonate anions, would result in an enlargement of Am solubility. The order of impact is as follows: $HCO_3^- > F^- > SO_4^{2-} > Si(OH)_{4(aq)}$.

Predictions of Am sorption on quartz

Generally, there are two kinds of colloids for actinides in groundwaters, *i.e.* intrinsic colloids and pseudo colloids. Both types of colloids have the capacity to transport through miles away. The excavation disturbance zone (EDZ) and surrouding rock of the HLW repository might cotains numerous fractures, which pose a great threat to the safty disposal. It is because that colloids moving through fractures may be more mobile than those in porous media due to the greater shear forces associated with fluid moving through a fracture.

Our calculation results indicate that the solubility of Am in far-field and near-field Beishan groundwater is 1.57×10^{-7} and 2.3×10^{-8} mol/L, respectively. Therefore, amercium can easily precipitate when its aqueous concentration is larger than solubility. Moreover, the precipitates, which is the crstalline solid $AmCO_3OH_{(cr)}$ or $Am(OH)_{3(cr)}$, could polymerize to form radioactive intrinsic colloids, composed of Am oxides after aging. Particulate solids of size diameters ranging from 1 to 1000 nm are known as pseudo colloids or carrier colloids, including mineral colloids and humic acid colloids. Aqueous americium species are able to be sorbed on mineral surface, leading to the formation of the pseudo colloids. Both types of colloids should be studied scrupulously for a better safety assessment of the repository.

Hence, the americium sorption on quartz with three surface complexation models by CHEMSPEC in far-field groundwater and near-field Beishan groundwater was studied. All models come from the digitized thermodynamic sorption database (RES^{3}T). Detailed information of the models are shown in Table 3, including the specific surface area, site density and solid concentration of the sorbent, the themodynamics parameters of the americium surface species and the referred literature. For example, the specific surface area, site density and solid concentration of the sorbent are 0.51 m^2/g, 100 g/L and 5.0 sites/nm^2 when using *KFYNM99* model. The pK2 and the surface reactions were entered into the CHEMSPEC database prior to calculation. All thermodynamic data were taken from the relevant literature. All parameters were kept fixed during the calculation.

The DLM was chosen because of its simple parameters, broad applicability, solid theoretical basis and the ability to provide reliable predictive results. In addition, the DLM is less complex and more certain than other models when dealing with complex systems [21]. The sorption calculation also take into account aqueous chemical reacitons of Am(III) in solution (Talbe 2). The quartz is selected as the sorbent for this study because it is a major rock-forming mineral in many geological environments. It is also a major component in Beishan granite (surrounding rock) [1] and Gaomiaozi bentonite (backfill materials) [4].

The initial concentration of americium is set to 1.0×10^{-8} mol/L, which make it certain that all americium species are aqueous species under Beishan conditions. The calculation results are shown in Fig. 4. For the first model *KFYNM99*, as shown in Fig. 4a and 4b, one can see that surface complexes $> S_KOAm^{2+}$ become to form on quartz when pH > 3.0 both in Beishan far-field and near-filed groundwaters. In other words, Am begins to be sorbed on quartz surface at pH 3.0, while the sorption amount increase with the pH increase and reach the maximum (nearly 100%) at pH 4.0. Under Beishan pH condition, aqueous Am is totally sorbed on quartz surface, leading to the formation of the pseudo colloids, which enhance the mobility of americium. For the second model *DKM14*, the sorption edge curves and the data of the max sorption percentage of Am in both far-field and near-filed Beishan groundwaters are similar to those with *KFYNM99* model, as shown in Fig. 4c and 4d. This model also estimated that almost all americium is sorbed onto quartz at Beishan pH value. For the third model *WAT01b*, the sorption curves are different in Beishan far-field and near-filed groundwaters, as shown in Fig. 4e and 4f. In far-field groundwater, the surface species $> S_WOAm^{2+}$ appear at pH 5–10, become the primary species for the pH values between 6.0 and 9.0. At pH 7.8, almost 97.4% americium is sorbed on quartz surface. In near-field groundwater, the pH range that surface species exist as the principal species broaden significantly. Under Beishan pH condition, 98.0% americium present as the surface species.

To summarize, all three SCMs predicted that almost all americium would be sorbed onto quartz surface in two

Table 3 The surface complexation modelling parameters of Am adsorption on quartz

Model	Type	A_{sp} (m^2/g)	Solid/liquid raito (g/L)	Site density (sites/nm^2)	pK2	Surface Reactions	Reference
KFYNM99	DLM	0.51	100	5.0	6.57	$> S_kOH + Am^{3+} \rightleftharpoons > S_kOAm^{2+} + H^+$ log $K^{int} = 5.07$	[27]
DKM14	DLM	2.8	10	2.3	7.20	$> S_DOH + Am^{3+} \rightleftharpoons > S_DOAm^{2+} + H^+$ log $K^{int} = 5.07$	[28]
WAT01b	DLM	2.8	15.2	2.31	7.20	$> S_WOH + Am^{3+} \rightleftharpoons > S_WOAm^{2+} + H^+$ log $K^{int} = -0.23$	[29]

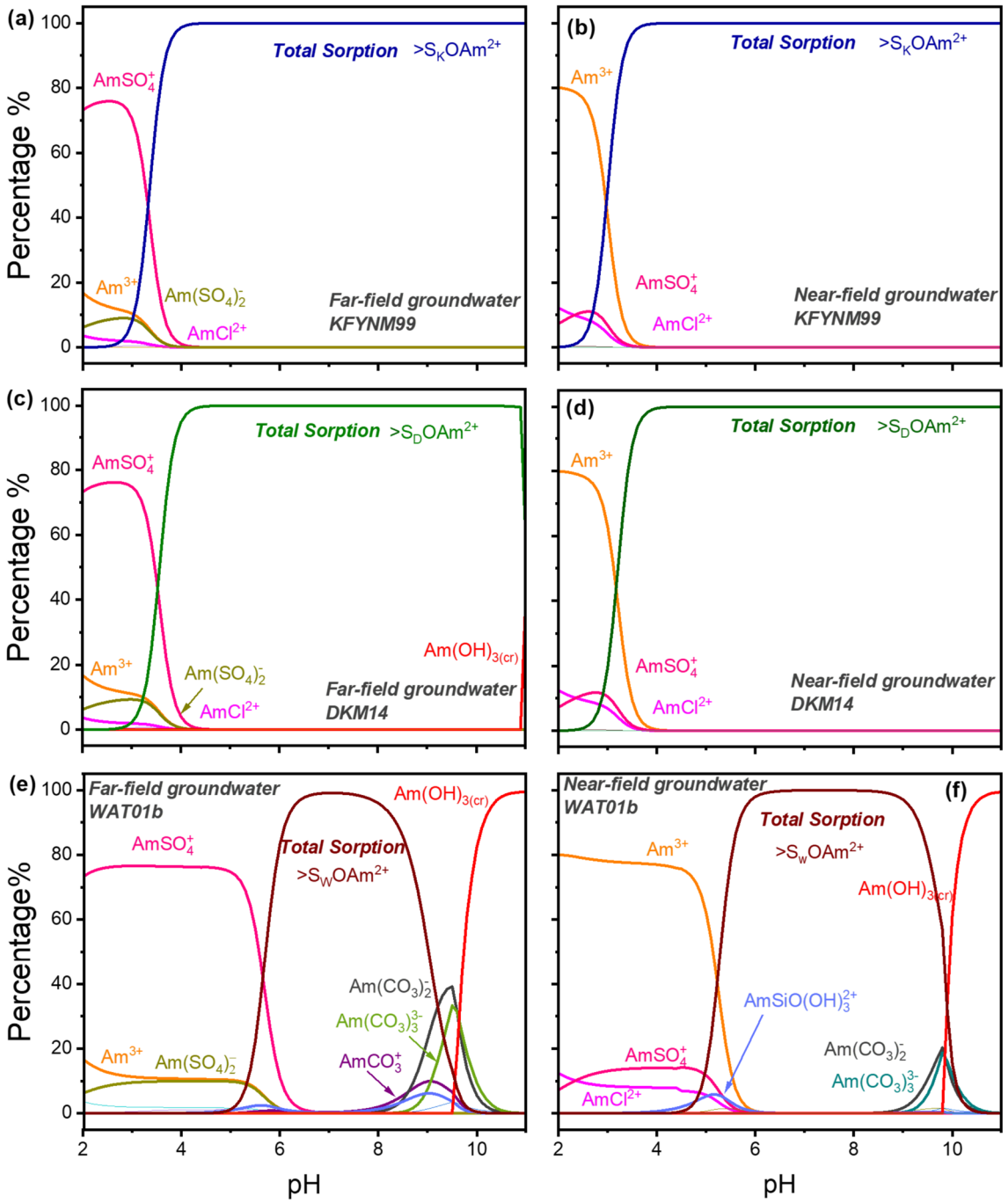


Fig. 4 The prediction of Am sorption on quartz in far-field groundwater (**a**, **c**, **e**) and near-field groundwater (**b**, **d**, **f**) with three different surface complexation models

groundwaters, thus the presentation of the quartz minerals could enhance the mobility of the americium through colloid-facilitated transport. Knowledge of the speciation and solubility of other actinides such as plutonium is important for the safe disposal of radioactive wastes. The stability constants for the species of plutonium are listed in Table S1. It can be estimated plutonium could form multivalent precipitates in groundwaters, such as $PuO_2(OH)_2{:}H_2O$(cr), $Pu(OH)_3$(cr), PuO_2(hyd_ag) and PuO_2CO_3(s). We would study the solubility, speciation and sorption of plutonium in Beishan groundwater in the future work.

Conclusions

In this work, the solubility, speciation and sorption on quartz of americium in far-field and near-field Beishan groundwaters were calculated with self-developed chemical speciation code CHEMSPEC. Our results indicated that the solubility of Am in far-field and near-field Beishan groundwater is 1.57×10^{-7} and 2.3×10^{-8} mol/L, respectively. An increase in the concentration of fluoride, silicate, and sulfate anions and a decrease in the concentration of carbonate anions, would result in an enlargement of Am solubility. Furthermore, the order of impact is as follows: $HCO_3^- > F^- > SO_4^{2-} > Si(OH)_{4(aq)}$. The sorption calculations with three SCMs all indicated americium would be sorbed on quartz under low concentrations, suggesting that quartz colloids might enhance the mobility of americium in repository environments through colloid-facilitated transport.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10967-024-09838-3.

Funding Funding for this research was provided by Talent Research Start-up Fund of Tongling University (No. 2022tlxyrc20) and National Natural Science Foundation of China (NSFC, No. 22276003).

Declarations

Conflict of 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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