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# Detection of Strontium Present in Groundwater Using PHREEQC Simulation

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This paper focuses on modeling chemical species of strontium in groundwater of Wuyi Well, Beishan Area, Gansu Province, P.R. China on basis of thermodynamics data from JAEA (Japan Atomic Energy Agency) and research results from French National Lab of Thermodynamics. The result shows that the chemical species of strontium in groundwater are found mainly as forms of $Sr(C_2H_4NO_2)_2$ and $SrSO_4$. The form of $SrSO_4$ takes up a small part of the amount. pE has no effects on the distribution of chemical species of strontium. The concentration of ammonia-nitrogen has a significant effect on the distribution of chemical species of strontium. Therefore, ammonia-nitrogen should be avoided in the packing materials of radioactive wastes and geochemical engineering barrier.

**Keywords: Strontium species, Groundwater, Simulation.**

## INTRODUCTION

Nuclide strontium is highly radioactive waste chemically and a typical bone seeker. It is transmitted and enters human body by ways of water, soil, vegetables and animals. If nuclide strontium enters human body, it is mainly incorporated in inorganic parts of bones and it mainly radiates bone and bone marrow. Nuclide strontium is harmful to human beings and hard to get rid of. The nuclide strontium has a big effect on the environment and attracts wide attention.

There are numerous studies on nuclide strontium in different transmission medium (rock and soil mainly). In China, some scholars have built chemical equations of sorption properties of a few nuclides in purple soil [1]. Some scholars have published their papers. The research is about the average moving speed and retardation coefficient of $Sr^{85}$ in a substance of yellow soil [2]. Some scholars have published papers about the radio nuclides migration in aerated zone under artificial sprinkling. The research shows that strontium migrates in two directions chemically. The calculated distribution coefficients are from 400 to 800 and 30 to 60 mL/g respectively [3]. Some scholars studied on migration of Cs and Sr from Chernobyl fallout in Ukrainian, Belarusian and Russian soils [4]. Some have published an essay which studies the hydrothermal alteration and hydrothermal evolution of natural granite, geochemistry of uranium, thorium and rare earth elements and secondary mineral blocking behavior to the related radioactive nuclide compared with migration of nuclide in the process of disposal of high level waste [5].

Chemical substance exits in a particular state. One has to learn its occurrences and changes in a certain environmental situation before starting related researches in environmental crisis. The strontium content of the crust is up to 384 mg/Kg and the strontium content of ocean water is 8 mg/Kg. It has few natural forms. The study shows that Sr is found only as the forms of the sulfate mineral celestite($SrSO_4$) and the strontium carbonate ($SrCO_3$) [6] in the earth and $Sr^{2+}$ in ocean water [7]. Radioactive Sr produced in nuclear explosion is found as the form of $Sr^{2+}$ for it is highly oxidizable. It oxidizes and hydrolyzes in the water. Because of abundant $SO_4^{2-}$, there is a possibility of metal complex or $SrSO_4$ and $SrCO_3$, when $Sr^{2+}$ enters ground water.

One has to make sure the forms of $Sr^{2+}$ in ground water before analysis of its adsorption mechanism. The forms of strontium in the water are able to be measured by experiment. The experiment has its limitations as preparation before the experiment is too much complicated and uncertainty of instrumental analysis. In light of this, modeling chemical species distribution of the nuclide by computer is effective. Few have calculated the chemical species distribution of strontium in the water in thermodynamics in the field of modeling research.

PHREEQC is a computer program widely used to perform a variety of low-temperature aqueous geochemical reactions designed by U.S Geological Survey. Thermodynamics data for the modeling chemical species distribution of Sr in this essay are from the PHREEQC.

## EXPERIMENTAL

**Chemical composition:** The study focuses on the forms distribution of Sr and how pH and pE affect the form distribution of Sr in groundwater in Wuyi well, Beishan Area [8,9]. pH is 7.24 and pE is 5.8 in the groundwater. Detailed composition listed in Table-1.

**Thermodynamic data of strontium:** Chemical species distribution calculation of nuclide is based on thermodynamics data. Complete and accurate data leads to a much more practical result. The data of the calculations were performed by reported research [10] and France National Lab of Thermodynamics (Table-2).

From Table-2, the thermodynamic data includes inorganic chemistry, thermodynamics of chemistry and reactions of phosphorus and strontium in the water.

## RESULTS AND DISCUSSION

**Chemical species of strontium:** It is measured that the concentration of strontium is 0.715 mg $L^{-1}$. Chemical species of strontium is calculated by using PHREEQC under 12 °C in groundwater. A minor tolerance exists in the composition part measured by ways of experiment. The minor tolerance will cause an imbalance of electric charges in aqueous solution. Computer modeling requires that electric charges should in the balanced state. Concentration of $Cl^-$ is relatively high. A minor adjustment of $Cl^-$, a charge balance ion, leads to a balanced state. As its weak ability to coordinate with strontium, it has little effect on chemical species of strontium.

From Table-3, strontium is mainly found as the forms of $Sr(C_2H_4NO_2)_2$ and $Sr^{2+}$ which accounts for 52.50 and 44.79 % of amount of the strontium, respectively. Strontium sulfate makes up a small fraction of the amount of strontium. Less than 1 % of the amount contains the rest forms of strontium. Acid radial of phosphorus does not affect species distribution of strontium.

**pH effects on species distribution of strontium:** The pH is a significant variable in groundwater and water-rock system. It varies from 4.3 to 9.0 in groundwater in Beishan Area. pH has an effect on species distribution of strontium.

TABLE-1
GROUNDWATER CHEMICAL COMPOSITION OF WUYI WELL IN BEISHAN AREA (mg/L)

| Component | Na | $SiO_2$ (aq) | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | Total Fe | $Cu^{2+}$ | $NH_4^+$ | $Mn^{2+}$ |
|---|---|---|---|---|---|---|---|---|---|
| Content | 47.83 | 12.91 | 73.88 | 8.88 | 8.98 | 0.86 | $10^{-4}$ | 0.12 | 0.022 |
| Component | $Li^+$ | $Sr^{2+}$ | $Al^{3+}$ | $SO_4^{2-}$ | $F^-$ | $Br^-$ | $NO_3^-$ | $Cl^-$ | $HCO_3^-$ |
| Content | 0.0112 | 0.715 | 0.06 | 161.8 | 0.26 | $10^{-4}$ | 10.42 | 61.35 | 103.7 |

TABLE-2
THERMODYNAMIC DATA FOR Sr USED FOR THE CALCULATIONS
(ONLY THE MOST IMPORTANT EQUILIBRIA ARE SHOWN) [Ref. 11]

| Chemical reaction equations | $\log_{10} K$ | Inaccuracy |
|---|---|---|
| $Sr^{2+} = Sr^{2+}$ | 0.000 | 0.001 |
| $HCO_3^- + Sr^{2+} = SrHCO_3^+$ | 1.180 | 0.500 |
| $HCO_3^- + Sr^{2+} = SrCO_3 + H^+$ | -7.520 | 0.700 |
| $H_2PO_4^- + Sr^{2+} = SrHPO_4 + H^+$ | -4.700 | 0.500 |
| $H_2PO_4^- + Sr^{2+} = SrH_2PO_4^+$ | 0.830 | 0.400 |
| $H_2PO_4^- + Sr^{2+} = SrPO_4^- + 2H^+$ | -13.560 | 0.200 |
| $2H_2PO_4^- + Sr^{2+} = SrP_2O_7^{2-} + H_2O + 2H^+$ | -12.410 | 0.030 |
| $HCO_3^- + Sr^{2+} = Sr(CHO_2)^+ + 0.5O_2$ | -41.410 | 0.050 |
| $2HCO_3^- + H^+ + NH_3 + Sr^{2+} = Sr(C_2H_4NO_2)^+ + 1.5O_2 + H_2O$ | -116.980 | 0.150 |
| $2HCO_3^- + H^+ + Sr^{2+} = Sr(CH_3OCO_2)^+ + 1.5O_2$ | -119.850 | 0.150 |
| $2HCO_3^- + H^+ + Sr^{2+} = Sr(CH_3COO)^+ + 2O_2$ | 145.640 | 0.430 |
| $2HCO_3^- + Sr^{2+} = Sr(CHO_2)_2 + O_2$ | -83.36 | 0.200 |
| $3HCO_3^- + 2H^+ + Sr^{2+} = Sr(CH_3CH_2CO_2)^+ + 3.5O_2$ | -254.74 | 0.200 |
| $3HCO_3^- + 2H^+ + Sr^{2+} = Sr(CH_3CH_2OCO_2)^+ + 3O_2$ | -226.34 | 0.501 |
| $3HCO_3^- + 2H^+ + NH_3 + Sr^{2+} = Sr(C_3H_6NO_2)^+ + 3O_2 + H_2O$ | -224.80 | 0.400 |
| $4HCO_3^- + 2H^+ + Sr^{2+} = Sr(CH_3OCO_2)_2 + 3O_2$ | -240.040 | 0.600 |
| $4HCO_3^- + 2H^+ + Sr^{2+} = Sr(CH_3COO)_2 + 4O_2$ | -291.75 | 1.000 |
| $4HCO_3^- + 2H^+ + 2NH_3 + Sr^{2+} = Sr(C_2H_4NO_2)_2 + 3O_2 + 2H_2O$ | -13.290 | 0.301 |
| $4HCO_3^- + 3H^+ + Sr^{2+} = Sr(CH_3CH_2CH_2CO_2)^+ + 5O_2$ | -363.570 | 0.180 |
| $5HCO_3^- + 4H^+ + Sr^{2+} = Sr(CH_3CH_2CH_2CH_2CO_2)^+ + 6.5O_2$ | -472.540 | 0.775 |
| $6HCO_3^- + 4H^+ + 2NH_3 + Sr^{2+} = Sr(C_3H_6NO_2)_2 + 6O_2 + 2H_2O$ | -450.080 | 0.200 |
| $6HCO_3^- + 4H^+ + Sr^{2+} = Sr(CH_3CH_2OCO_2)_2 + 6O_2$ | -452.970 | 0.600 |
| $6HCO_3^- + 4H^+ + Sr^{2+} = Sr(CH_3CH_2CO_2)_2 + 7O_2$ | -509.820 | 0.900 |
| $8HCO_3^- + 6H^+ + Sr^{2+} = Sr(CH_3CH_2CH_2CO_2)_2 + 10O_2$ | -727.480 | 0.500 |
| $10HCO_3^- + 8H^+ + Sr^{2+} = Sr(CH_3CH_2CH_2CH_2CO_2)_2 + 13O_2$ | -945.360 | 0.100 |
| $SO_4^{2-} + Sr^{2+} = SrSO_4$ | 1.860 | 0.300 |
| $H_2O + Sr^{2+} = SrOH^+ + H^+$ | -13.290 | 0.200 |
| $NO_3^- + Sr^{2+} = SrNO_3^+$ | 0.800 | 0.200 |
| $PO_4^{3-} + Sr^{2+} = SrPO_4^-$ | 4.200 | 0.500 |
| $F^- + Sr^{2+} = SrF^+$ | -0.17 | 0.500 |
| $Cl^- + Sr^{2+} = SrCl^+$ | -0.23 | 0.500 |

TABLE-3
CALCULATED SPECIES DISTRIBUTION
FOR Sr IN BEISHAN GROUNDWATER

| Species | Weight-molality (mol $L^{-1}$) | Activity (mol $L^{-1}$) | Percentage |
|---|---|---|---|
| $Sr(C_2H_4NO_2)_2$ | $4.29 \times 10^{-6}$ | $4.30 \times 10^{-6}$ | 52.50 |
| $Sr^{2+}$ | $3.66 \times 10^{-6}$ | $2.45 \times 10^{-6}$ | 44.79 |
| $SrSO_4$ | $1.74 \times 10^{-7}$ | $1.74 \times 10^{-7}$ | 2.13 |
| $SrHCO_3^+$ | $3.39 \times 10^{-8}$ | $3.07 \times 10^{-8}$ | 0.42 |
| $SrNO_3^+$ | $1.15 \times 10^{-8}$ | $1.04 \times 10^{-8}$ | 0.14 |
| $SrCl^+$ | 1.45 × 10-9 | 1.31 × 10-9 | 0.02 |
| $SrCO_3$ | $8.64 \times 10^{-10}$ | $8.66 \times 10^{-10}$ | 0.01 |
| $SrF^+$ | $2.03 \times 10^{-11}$ | $1.84 \times 10^{-11}$ | 0.00 |
| $SrOH^+$ | $2.42 \times 10^{-12}$ | $2.19 \times 10^{-12}$ | 0.00 |

Concentration of strontium is 0.715 mg $L^{-1}$. pH value varies from 4.3 to 9.5.

Table-4 and Fig. 1 indicate that pH has a little effect on the species distribution of strontium. Strontium presents as the forms of glycine metal complex and free ion, which depends on chemical property of strontium and pH value of the aqueous solution. Strontium exists as the only form of positive divalent. Its ionic radius is approximately 1.120 Å and its ionic potential is 1.785. Based on the divisions of Levison, it is in the boundary of active cation and inactive hydrolyzate elements. $Sr^{2+}$ obsess less power to combine with $O^{2-}$ than $H^+$ with pH scale in the water, which results in a complex situation of distribution of strontium.

Strontium is found in the form of glycine metal complex and free ion, which take up 97 % of the amount when pH value varies from 4.0 to 9.5. The percentage of $SrSO_4$, $SrNO_3^+$ and $SrCl^+$ remain nearly unchanged while $SrHCO_3^+$ and $SrCO_3$ change a lot.

Glycine $[C_2H_4NO_2)^+]$ is a non-polar and hydrophobic amino acid. It has poor ability to dissolve into water. Metallic ions ruin structures of glycine radical group after having combined with them. One and two glycine radical group leads to peptide bond reversible reaction. It increase productivity and dissolution of $Sr(C_2H_4NO_2)_2$. Therefore 52.5 % of the amount is $Sr(C_2H_4NO_2)_2$. There is a huge amount of $SO_4^{2-}$, which easily combines with metallic ion. In light of this, the volume of $SrSO_4$ is high.

When pH value varies from 4.5 to 7.0, carbonate in the water is found mainly as the form of $CO_2$ (aq). As the pH value turns greater, the water turns neutral. Concentration of $H^+$ decreases. Solubility of bicarbonate increases. $Sr^{2+}$ gains better ability to combine with $O^{2-}$. Percentage of $SrHCO_3^+$ grows higher. When pH value varies from 7.0 to 9.5, the water turns alkaline. $Sr^{2+}$ gain much more ability to combine with $O^{2-}$. Carbonate in the water is found mainly as the form of $CO_3^{2-}$ and $HCO_3^-$ instead of $CO_2$ (aq). Therefore $SrHCO_3^+$ and $SrCO_3$ exit. When pH value varies between 8 to 8.5, $HCO_3^-$ reaches its saturation point. $SrHCO_3^+$ reaches its max amount in the water, 0.46 %. As pH value becomes greater, $HCO_3^-$ and $OH^-$ react to $CO_3^{2-}$. When pH value is greater than 8.5, the amount of $SrHCO_3^+$ goes down while that of $SrCO_3$ goes up.

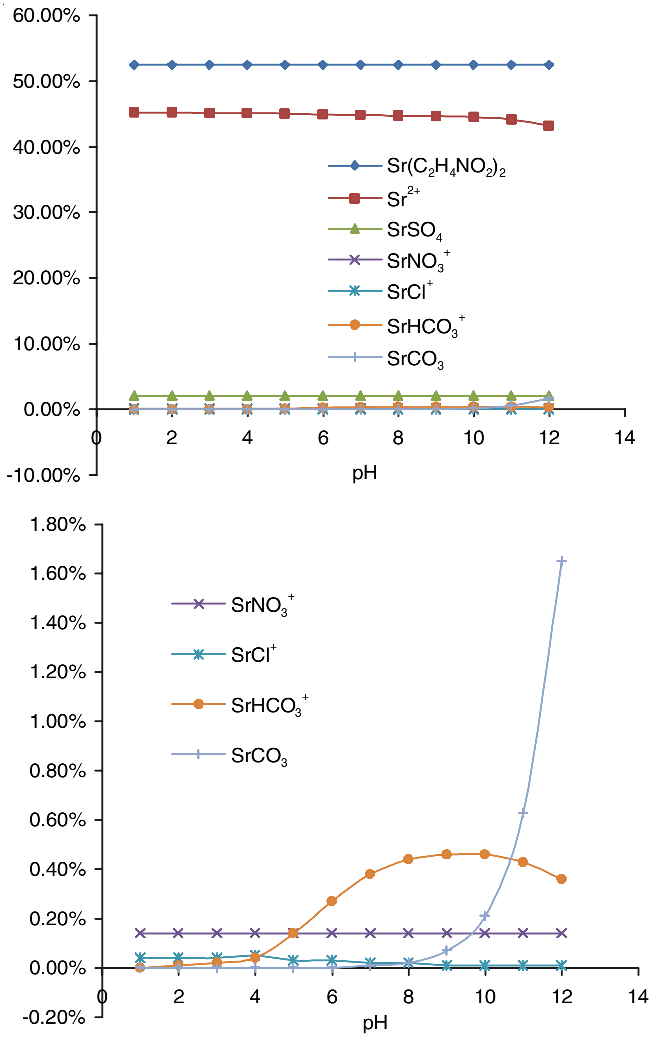


Fig. 1. Effects of pH on distribution of strontium species

TABLE-4
EFFECTS OF pH ON DISTRIBUTION OF STRONTIUM SPECIES

| pH | $Sr(C_2H_4NO_2)_2$ (%) | $Sr^{2+}$ (%) | $SrSO_4$ (%) | $SrNO_3^+$ (%) | $SrCl^+$ (%) | $SrHCO_3^+$ (%) | $SrCO_3$ (%) |
|---|---|---|---|---|---|---|---|
| 4 .0 | 52.50 | 45.20 | 2.12 | 0.14 | 0.04 | 0.00 | 0.00 |
| 4.5 | 52.50 | 45.19 | 2.13 | 0.14 | 0.04 | 0.01 | 0.00 |
| 5.0 | 52.50 | 45.17 | 2.13 | 0.14 | 0.04 | 0.02 | 0.00 |
| 5.5 | 52.50 | 45.14 | 2.13 | 0.14 | 0.05 | 0.04 | 0.00 |
| 6.0 | 52.50 | 45.06 | 2.13 | 0.14 | 0.03 | 0.14 | 0.00 |
| 6.5 | 52.50 | 44.94 | 2.13 | 0.14 | 0.03 | 0.27 | 0.00 |
| 7.0 | 52.50 | 44.83 | 2.13 | 0.14 | 0.02 | 0.38 | 0.01 |
| 7.5 | 52.50 | 44.76 | 2.13 | 0.14 | 0.02 | 0.44 | 0.02 |
| 8.0 | 52.50 | 44.70 | 2.13 | 0.14 | 0.01 | 0.46 | 0.07 |
| 8.5 | 52.50 | 44.56 | 2.12 | 0.14 | 0.01 | 0.46 | 0.21 |
| 9.0 | 52.50 | 44.18 | 2.12 | 0.14 | 0.01 | 0.43 | 0.63 |
| 9.5 | 52.50 | 43.25 | 2.10 | 0.14 | 0.01 | 0.36 | 1.65 |

**pE effects on species distribution of strontium:** Redox (reduction-oxidation) reaction varies. When pE is greater than 0, the groundwater is in oxidative environment. When pE is less than 0, the groundwater is in reductive environment. pE value in groundwater of Beishan area varies from -3.2 to 6.0 [11] (Table-5).

pH value has no effect electron on the distribution of the chemical species of strontium (atomic radius = 2.45 Å and ion radius = 1.12 Å). Its electron configuration is $1s^2\ 2s^2p^6\ 3s^2p^6d^{10}\ 4s^2p^6 5s^2$. When 2 electrons on the out layer are gone, the element radius is half the original strontium.

After theoretical calculation, nucleus of strontium obtains a powerful nuclear force to attract electron on the 3rd and 4th layer. Therefore, strontium exists in the state of positive 2. pH value has no effects on the number of strontium electrons and also does not affect distribution of strontium and its chemical species.

**Ammonia concentration effects on the species distribution of strontium:** The modeling results above shows that $Sr(C_2H_4NO_2)_2$ makes up 50 % of the amount of Sr. According to the chemical equation $4HCO_3^- + 2H^+ + 2NH_3 + Sr^{2+} = Sr(C_2H_4NO_2)_2 + 3O_2 + 2H_2O$ and chemical equilibrium theory, rich $HCO_3^-$ and $HCO_3^-$ effectively react to form $Sr(C_2H_4NO_2)_2$. The amount of $HCO_3^-$ is relatively higher than that of ammonia in Wuyi, Beishan Area. This calculation using PHREEQC software focuses on how ammonia concentration affects the ratio of $Sr(C_2H_4NO_2)_2$. The set values are: pH value is 7.24; pE value is 5.8; temperature is 12 °C; concentration of strontium is 0.715 mg $L^{-1}$. The results are presented in Fig. 2.

According to Fig. 2, during the period of ammonia concentration varies from 0.12 to 0.22 ppm, $Sr^{2+}$ concentration decreases while $Sr(C_2H_4NO_2)_2$ concentration increases sharply along with the increasing of the percentage of $Sr(C_2H_4NO_2)_2$. When ammonia concentration increases to 0.23 ppm, $Sr(C_2H_4NO_2)_2$ goes up to 100 % while the other species decrease to 0. Ammonia plays a very important role in distribution of strontium.

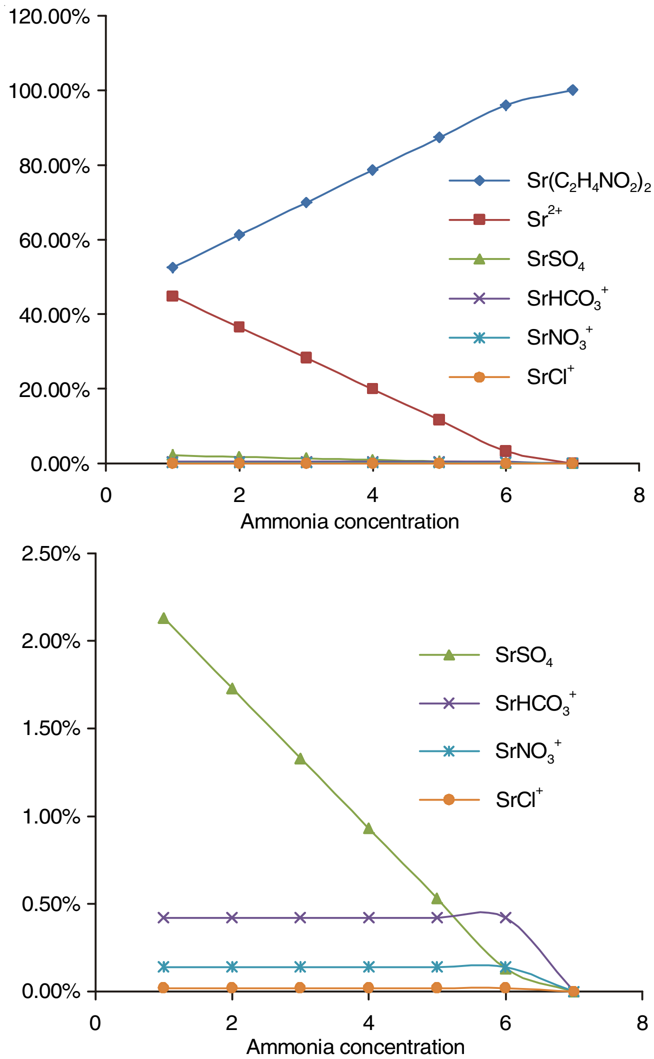


Fig. 2. Effects of the ammonia concentration on the distribution of strontium species

## Conclusion

1) PHREEQC is good for calculating the species distribution of strontium. Strontium is mainly found as the forms of $Sr(C_2H_4NO_2)_2$ and $Sr^{2+}$ which accounts for 52.50 and 44.79 % of the amount of strontium, respectively. $SrSO_4$ accounts for a

TABLE-5
EFFECTS OF pE ON SPECIES DISTRIBUTION OF STRONTIUM UNDER DIFFERENT pH VALUES

| pH | pE | $Sr(C_2H_4NO_2)_2$ (%) | $Sr^{2+}$ (%) | $SrSO_4$ (%) | $SrHCO_3^+$ (%) | $SrNO_3^+$ (%) | $SrCl^+$ (%) |
|---|---|---|---|---|---|---|---|
| 6.2 | -3.2 | 52.50 | 45.01 | 2.13 | 0.19 | 0.14 | 0.03 |
| 6.2 | -2 | 52.50 | 45.01 | 2.13 | 0.19 | 0.14 | 0.03 |
| 6.2 | -0 | 52.50 | 45.01 | 2.13 | 0.19 | 0.14 | 0.03 |
| 6.2 | 2 | 52.50 | 45.01 | 2.13 | 0.19 | 0.14 | 0.03 |
| 6.2 | 4 | 52.50 | 45.01 | 2.13 | 0.19 | 0.14 | 0.03 |
| 6.2 | 6 | 52.50 | 45.01 | 2.13 | 0.19 | 0.14 | 0.03 |
| 7.24 | -3.2 | 52.50 | 44.81 | 2.12 | 0.41 | 0.14 | 0.02 |
| 7.24 | -2 | 52.50 | 44.81 | 2.12 | 0.41 | 0.14 | 0.02 |
| 7.24 | -0 | 52.50 | 44.81 | 2.12 | 0.41 | 0.14 | 0.02 |
| 7.24 | 2 | 52.50 | 44.81 | 2.12 | 0.41 | 0.14 | 0.02 |
| 7.24 | 4 | 52.50 | 44.81 | 2.12 | 0.41 | 0.14 | 0.02 |
| 7.24 | 6 | 52.50 | 44.81 | 2.12 | 0.41 | 0.14 | 0.02 |
| 8.41 | -3.2 | 52.50 | 44.60 | 2.12 | 0.46 | 0.17 | 0.14 |
| 8.41 | -2 | 52.50 | 44.60 | 2.12 | 0.46 | 0.17 | 0.14 |
| 8.41 | -0 | 52.50 | 44.60 | 2.12 | 0.46 | 0.17 | 0.14 |
| 8.41 | 2 | 52.50 | 44.60 | 2.12 | 0.46 | 0.17 | 0.14 |
| 8.41 | 4 | 52.50 | 44.60 | 2.12 | 0.46 | 0.17 | 0.14 |
| 8.41 | 6 | 52.50 | 44.60 | 2.12 | 0.46 | 0.17 | 0.14 |

Temperature is 12 °C, concentration of Sr is 0.715 mg $L^{-1}$

small fraction of the amount of strontium. Less than 1 % of the amount contains the rest forms of strontium.

2) pH value has an effect on the chemical species of strontium while pH does not. As pH value increases, the amount of $SrHCO_3^+$ and $SrCO_3$ changes.

3) When pH values varies and pE value varies between negative three and positive five. The pE has little effect on the species distribution of strontium.

4) Concentration of ammonia -nitrogen affect profoundly the species distribution of strontium. A little increase on concentration of ammonia-nitrogen affect greatly the species distribution of strontium. Therefore, ammonia-nitrogen should be avoided in the production of package materials and geochemical engineer block barrier.

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