**Journal:** Journal of the Serbian Chemical Society
**Title:** The extraction of Sr2+ with dicyclohexano-18-crown-6 in conventional organic solvent and ionic liquid diluents
**Authors:** ZHENG WEI, YANG GAO\*, YU ZHOU, CAISHAN JIAO, MENG ZHANG, HONGGUO HOU, WEI LIU\*

Dear Prof. Ljiljana Damjanović Vasilić

 Thank you very much for your email on 19 Aug 2019. We would like to thank the reviewers for careful and thorough reading of this manuscript and for the thoughtful comments and constructive suggestions, which help to improve the quality of this manuscript. We have read the comments carefully, and the manuscript have been rechecked and provided in the revised manuscript according to these comments and suggestions. Attached please find our revised manuscript, and listed below are our point-by-point responses to the reviewer suggestions (the reviewer’s comments are in *italics*).

**Replies to Reviewer A:**

1. We thank the reviewer’s kindly suggestions that *“page 2. line 35. "90Sr and 137 Cs, whose half-lives are 28.8 and 30.2 years, respectively, have the characteristics of strong heat release and high radioactivity.". The statement should be clarified. The heat release primarily depends on activity.”.*

We have changed the corresponding sentence to “Two long-lived radionuclides 90Sr and 137Cs, whose half-lives are 28.8 and 30.2 years, respectively, have the characteristics of high radioactivity, and their specific activities are 3.2×105 Bq/μg and 5.09×106 Bq/μg, respectively. Therefore, they are also strong heat-release nuclides, which mainly contribute to the temperature elevation of HLLW.” on line 35of page 2.

2. We thank the reviewer’s careful suggestions that *“Page 2. Line 43. "Many chemical separation means" should be changed to "many separation procedures".”*.

In our revised manuscript, we have changed “Many chemical separation means” to “many separation procedures” on line 46 of page 2.

3. We thank the reviewer’s kindly suggestions that *“Page 2, line 52. "which was a kind of excellent extractant" should be changed to "which was an excellent extractant".”*.

We have made the corresponding revision in the update version on line 55 of page 2.

4. We thank the reviewer’s helpful suggestions that “*Page 4. line 97. "Considering the insufficient studies" should be changed to "considering the small number of studies*".”. In our revised manuscript, we have made the corresponding revision in the update version on line 100 of page 4.

5. We thank the reviewer’s helpful comments that “*Page 4. line 102. "better extraction" should be changed to "more efficient extraction"*.”.

In our revised manuscript, we have made the corresponding revision in the update version on line 105 of page 4.

6. According to the reviewer’s helpful comments that “*Page 6. line 151. "when small quantity of TCA was mixed with n-octanol" should be changed in "when small volume of TCA*"...”.

We have changed “when small quantity of TCA was mixed with n-octanol” to “when small volume of TCA was mixed with n-octanol” on line 154 of page 6.

7. We appreciate the reviewer’s careful suggestions that “*Page 6. line 151 "Besides, viscosity of mixture diluents dropped" should be changed in "Besides, the viscosity of the obtained mixture is lowered"*.”.

We have made the corresponding revision in the update version on line 154 of page 6.

8. We thank the reviewer’s kindly question that *“Page 6. "instability of TCA". What kind of instability? Please clarify!”*.

In our revised manuscript, we have changed the corresponding sentence to “Considering the irradiation instability of TCA which is easy to decompose and generates corrosive hydrochloric acid under irradiation conditions, mixture diluents should be mainly constituted of n-octanol” on line 157 of page6.

9. We thank the reviewer’s careful suggestions that “*Page 7. line 167 "The" >>"the"*”. We have made the corresponding revision in the update version on line171 of page 7.

10. We thank the reviewer’s helpful comments that “*Page 7. line 176. "extractant was more conducive" should be changed in "extractant is more convenient*".”.

We have made the corresponding revision in the update version on line 180 of page 7.

11. We appreciate the reviewer’s careful suggestions that “*Page 11, line 246. "Accordingly, it was concluded that the better DSr with the DCH18C6-CnmimNTf2 systems could be obtained as compared to that with the DCH18C6-70/30(v/v%) n-octanol/TCA system when HNO3 concentration in aqueous phase approached to zero." Please clarify statement.*”.

In the revised manuscript, we have added the statement “As seen in the top row, the DCH18C6-C*n*mimNTf2 systems shown unsatisfactory results. The difference of the *D*Sr values between the traditional and novel diluents is small from the 1 mol·L-1 HNO3 medium, and the distribution ratio even is lower in C4mimNTf2 and C6mimNTf2 solvents than in 70/30(v/v%) n-octanol/TCA solvent. It couldn’t meet the expectations in these ionic liquids in view of the current reports 26,31. However, it was found that much higher extraction efficiency of Sr2+ could be obtained at the DCH18C6-C*n*mimNTf2 systems when there was no HNO3 in aqueous phase, and the DCH18C6-70/30(v/v%) n-octanol/TCA system scarcely extracted Sr2+ from the aqueous phase without HNO3, as shown in the bottom row of Tab. 4.” on line 247 of page 11.

12. We thank the reviewer’s kindly suggestions that “*Page 12, line 267. "the lower acid aqueous" should be changed in "the lower pH of solution"*.”.

In the updated version, we have changed “the lower acid aqueous” to “the lower pH of solution” on line 282 of page 12.

13. We thank the reviewer’s kindly question that “Page 14, line 308. "*It was explained that shorter carbon chain benefited increasing Sr2+ extraction efficiency." Shorter chain of what?”*.

In our revised manuscript, we have changed the corresponding sentence to “It was explained that shorter carbon chain of the ILs cation benefited increasing Sr2+ extraction efficiency.” on line 323 of page 14.

14. We appreciate the reviewer’s careful suggestions that “*Page 14, line 310. "tremendous elevation" should be changed in "significant increase"*.”.

We have made the corresponding revision in the update version on line 325 of page 14.

**Technical issues:**

1. *The names of the authors must be typed in CAPITAL letters (see Instructions for authors*.

The names of the authors have been typed in CAPITAL letters

2. *The full name should be given for the first mentioning of the abbreviation HLLW (in Abstract)*.

The full name has been given for the first mentioning of the abbreviation HLLW (in Abstract).

3. *The figure 1. should be larger – the oxygen atoms are not clearly visible.*

The figure 1. has been larger.

4. *All units should be given in SI. If you still want to use milliliters/liters for volume units, please use them consistently. For example: “...equilibrating 1 ml (or 0.5 ml)... The DCH18C6 concentration in organic phase was* *0.01 mol·L-1...”*.

All volume units have been given in liters.

5. *The caption for the Figure 2. is wrong (the captions for the figures 2. and 3. are the same?*

The caption for the Figure 2 has been changed to “The effect of the initial HNO3 concentration in aqueous phase on the *D*Sr. Organic phase: 0.01 mol·L-1 DCH18C6.”.

6. *In references ref Yang et al. 27 (line 209) should be Yongqing et al.*

We have rechecked Ref. 27 and confirmed that Yang is the family name and Yongqing is the given name of the first author. Therefore, Yang *et al.* 30 on line 215 of page 9 is the right citation format.

7. *There are grammatical and typo errors trough the text.*

We have checked the whole manuscript carefully and tried our best to modify the grammatical and typo errors.

**Questions and remarks:**

1. *All experiments were conducted at temperature of 25°C. HLLW produces large amount of heat so it has considerably higher temperature than room temperature. Since* *the temperature has significant impact on distribution coefficients, authors should explain why they didn’t investigate this effect.*

We appreciate your insightful suggestions. Indeed, HLLW would be at high temperatures due to the radiation energy, and distribution ratio is closely dependent on temperature. However, in this work, the focus is put on the different extraction behaviors and extraction mechanisms of Sr2+ using DCH18C6 as extractant in n-octanol and tetrachloride acetylene mixture diluent and in ionic liquid diluents. In this paper, all experiments were conducted at temperature of 25°C, which is direct and convenient to compare the results in the two kinds of solvents. In the next work, we will investigate the influence of temperature on the distribution coefficient in detail.

2. *In HLLW, the radiation induced decomposition of both extraction agents and solvents is usually present in some extent, which can influence extraction efficiency. Authors should discuss this in manuscript.*

Thanks for your helpful suggestions. Indeed, during the extraction stage, the extractant and solvent suffer radiation and chemical degradation, resulting in the formation of a series of degradation products. Yuan *et al.* (ref. 33) studied the extraction of Sr2+ using irradiated C4mimPF6 in the presence of DCH18C6 and found that Sr2+ partitioning in irradiated C4mimPF6 decreases as the absorption dose increases, which proved that the radiation would affect the extraction behavior of Sr2+. We have discussed this in the update version on line 331 of page 15.

3. *The authors should discuss how the presence of* *other ions, which are usually present in HLLW, may affect separation of 90Sr2+ from HLLW*.

Thanks for your constructive suggestions. The HLLW really has complex composition, which comprises of minor actinides such as Am, Cm, and fission product elements including lanthanides, Cs, Sr, Zr, etc. The other ions in HLLW, such as K+, may affect separation of 90Sr by competitive extraction. We have discussed this in the update version on line 337 of page 14.

4. *The effect of* *initial Sr2+ concentration was investigated in quite narrow range - 0.4-2 mmol/dm3. Why did authors choose this range? Does this corresponds to the* *average concentration of 90Sr2+ in HLLW?*

Thanks a lot for your question. Just as you say, the studied initial Sr2+ concentration range corresponds to the average concentration of 90Sr2+ in HLLW. In fact, the Sr2+ concentration differs in HLLW from different reactor types. for example, the Sr2+ concentration is 0.34 mmol/L Sr2+ in pressurized heavy water reactor HLLW, 2.3 mmol/L in the pressurized water reactor HLLW in China, and 1 mmol/L in typical simulated Idaho National Engineering and Environmental Laboratory HLLW (refs. 4,5,14). According to the data from these references, the initial concentration of Sr2+ was set to 0.4-2 mmol/L in our paper.

5. *Why did authors chose* *Sr2+ concentration of 1.4 mmol/dm3 for the experiments?*

As mentioned above, the Sr2+ concentration is different in HLLW from different reactor types. The studied Sr2+ concentration of 1.14 mmol/L (*i.e*. 0.1 g/L) was in the middle of concentration range of 90Sr2+ in HLLW and it was adapted in some literatures. For example, Fan *et al.* (ref. 20) studied the extraction behavior of Sr2+ using DCH18C6 in 1,1,2,2-Tetrachloroethane as the extractant. The studied Sr2+ concentration was 1.14 mmol/L. Applying this concentration is helpful to compare the results with those in the references.

**Replies to Reviewer C:**

1. We thank the reviewer’s kindly suggestions that “*The authors should cite the literature* *about other methods for radioactive Sr2+ removal.*”.

In our revised manuscript, we have cited several literatures (Refs. 7-13) about other methods for radioactive Sr2+ removal.

2. We appreciate the reviewer’s insightful suggestions that *“The* *discussion about other methods and comparison with results presented in this manuscript would be very interesting.”.*

In the updated version, we have added the discussion about other methods and comparison with results presented in this manuscript on line 240 of page 11 “Furthermore, solvent extraction using DCH18C6 in the binary diluent as the extractant showed greater advantage compared to other methods aimed for the removal of Sr2+ from HLLW. For example, Zhang *et al.* 8 studied the coprecipitation method using zirconyl molybdopyrophosphate for the removal of 90Sr, and the result showed that *E*Sr only reached 92.96%.”.

3. We thank the reviewer’s helpful suggestions that “*The schematic depiction of the Sr2+ complex with crown ether would be very useful.*”.

We have added the schematic depiction of the Sr2+ complex with crown ether in our revision in Figure. 4.

In the end, we sincerely express our appreciation to you and the reviewer again. Your constructive comments help us to better revise our manuscript, and we would like to submit the revised manuscript to **Journal of the Serbian Chemical Society** for publication. We appreciate your taking care of our revision in advance.

Sincerely yours,

Yang Gao

Fundamental Science on Nuclear Safety and Simulation Technology Laboratory

Harbin Engineering University

Harbin 150001, China

Email: gaoyang@hrbeu.edu.cn

Tel: +86-451-8251-8913

Fax: +86-451-8256-9655