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Supplementary Materials

Radiation-Resistant Betaine Hydrochloride for Facile Ln/An Group Separation via Selective Dissolution

Zhuo-Fan Jin [1,2], He-Xiang Jia [1,2], Bing-Qing Wei [1,2], Qing-Gang Huang [1], De-Sheng Chen [1], Xiao-Jie Yin [1], Shi-Wei Cao [1], Zhi Qin [1] and Fang-Li Fan [1,*]

[1] Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China
[2] School of Nuclear Science and Technology, University of Chinese Academy of Sciences, Beijing 100000, China
* Correspondence: fanfangli@impcas.ac.cn

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1. Text S1: The XRD Pattern of SIMFUEL

The XRD results verify that only characteristic diffraction peaks corresponding to UO_2 are detected. No distinguishable diffraction peaks of Nd_2O_3 were observed, which can be attributed to its relatively low loading content in the SIMFUEL sample.

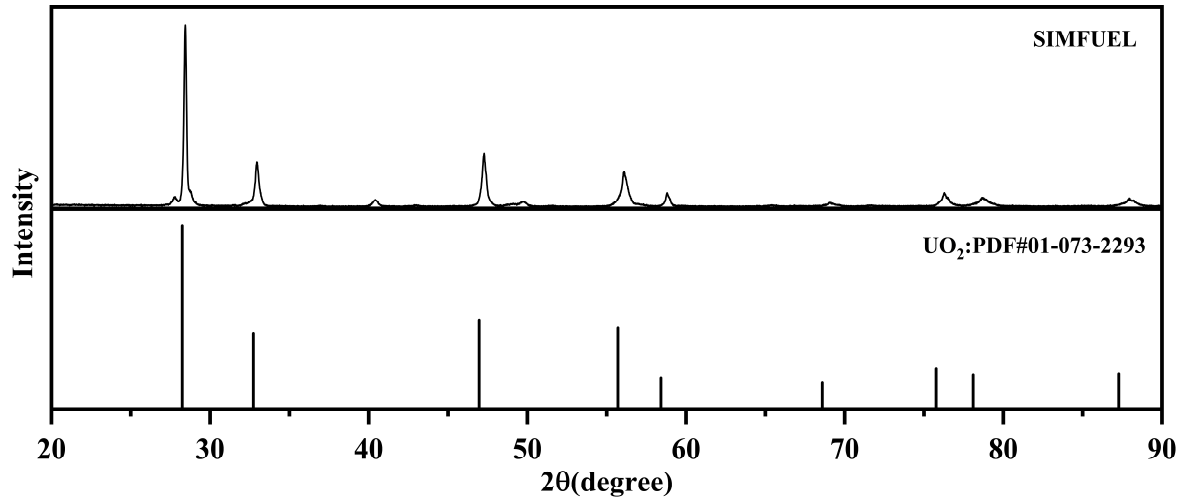


Figure S1. The XRD pattern of SIMFUEL.

2. Text S2: Solubility of Nd_2O_3, UO_2 and U_3O_8 in Betaine Hydrochloride (HbetCl) Solution

Selective dissolution experiments were initially performed using Nd_2O_3, UO_2 and U_3O_8 solid powder in betaine hydrochloride (HbetCl), respectively (Figure S2). For all dissolution experiments, a mixture of a metal oxide (25 mg) and HbetCl solution (2 mL) was heated to 40 °C for 1 h at the same stirring speed. The rare earth oxide, Nd_2O_3 was used on behalf of neutron poisons from the fission products, considering it be the biggest amount in the fission products. The dissolution behavior of Nd_2O_3, UO_2 and U_3O_8 metal oxides in HbetCl solution obtained within this work were similar to those previously reported using water-saturated [Hbet][Tf_2N] as dissolving media. This could be attributed to the same carboxyl group of $[Hbet]^+$ in [Hbet][Tf_2N] and HbetCl, which was the driving force for dissolution.

As presented in Figure S2, the dissolution ability of these metal oxides is obvious different under the same condition and the concentration of HbetCl have no effect on the dissolution. The HbetCl solution was able to dissolve 100% of Nd_2O_3, whereas only a very little percentage of the same amount uranium oxides dissolved in this system as determined by ICP-OES. The reasons for this significant difference might come from the difference in the lattice energy or complexation ability with trivalent actinides or lanthanides. Moreover, U_3O_8 and UO_2

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displayed different solubility in the HbetCl solution, which indicated that metal speciation had a significant impact on the dissolution properties in our cases. Therefore, in order to decrease the solubility of uranium and ensure best separation effect between lanthanides and actinides, uranium species must be maintain as UO_2 in the sample before separation. Overall, HbetCl exhibited the unique dissolution selectivity for lanthanides over actinides. The drastic dissolution difference between Nd_2O_3 and UO_2 strongly support the potential use of HbetCl molecule for the separation of lanthanides and actinides from spent nuclear fuel.

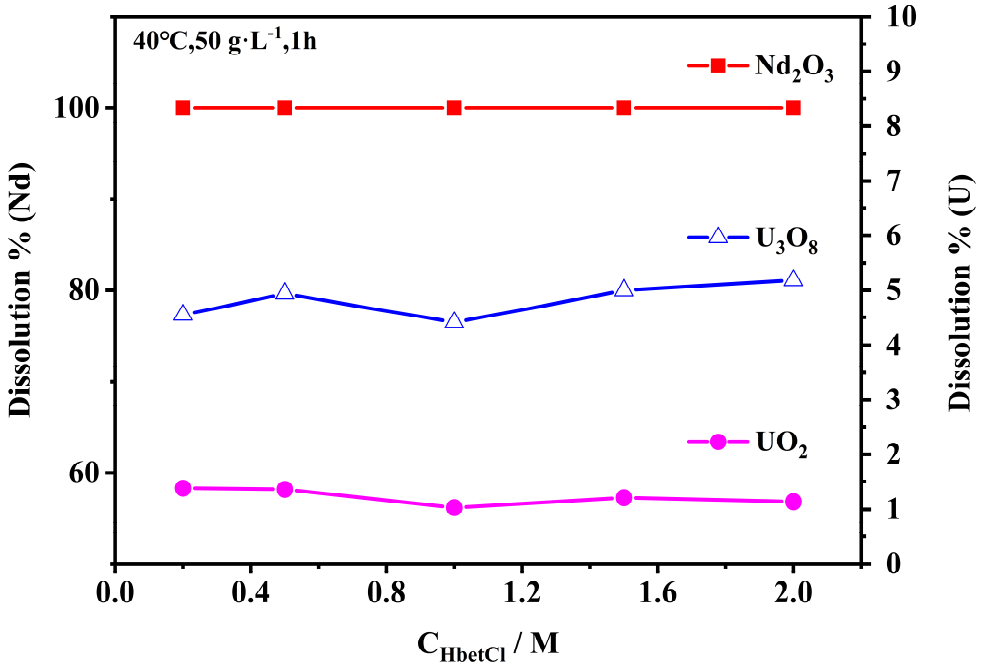


Figure S2. The dissolution of Nd_2O_3, UO_2 and U_3O_8 in HbetCl aqueous.

3. Text S3: Recover of Nd

X-ray diffraction (XRD) was carried out to confirm that the obtained oxides correspond to Nd_2O_3.

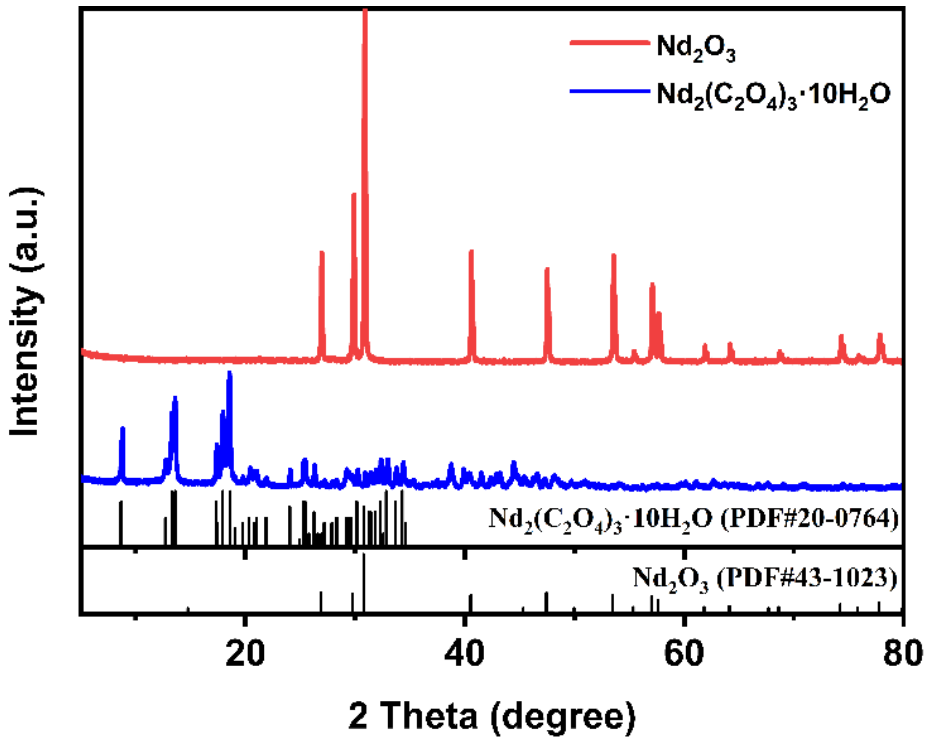


Figure S3. The XRD diagram of Nd_2O_3 and $Nd_2(C_2O_4)_3 \cdot 10H_2O$.

4. Text S4: Selective Dissolution of Nd_2O_3 from Simulated Spent Nuclear Fuel

To further examine the possibility of this separation system in the spent nuclear fuel, the dissolution performances of Nd_2O_3 and UO_2 in the samples with higher reduction temperature were investigated. As displayed in Figure S4, the dissolution of Nd_2O_3 is significantly affected by the reduction temperature, apparently because of the structure compactness or the chemical species in the samples after higher temperature treatment. Obviously, the dissolution efficiency of Nd_2O_3 decreases with increasing of treatment temperature. Besides, it can be seen that prolonging the dissolution time leads to a little increase of Nd_2O_3 dissolution, while the dissolution ratio of U increases slightly. Nevertheless, the maximal dissolution ratio of U is only about 3%, and the most UO_2 remains as solid form.

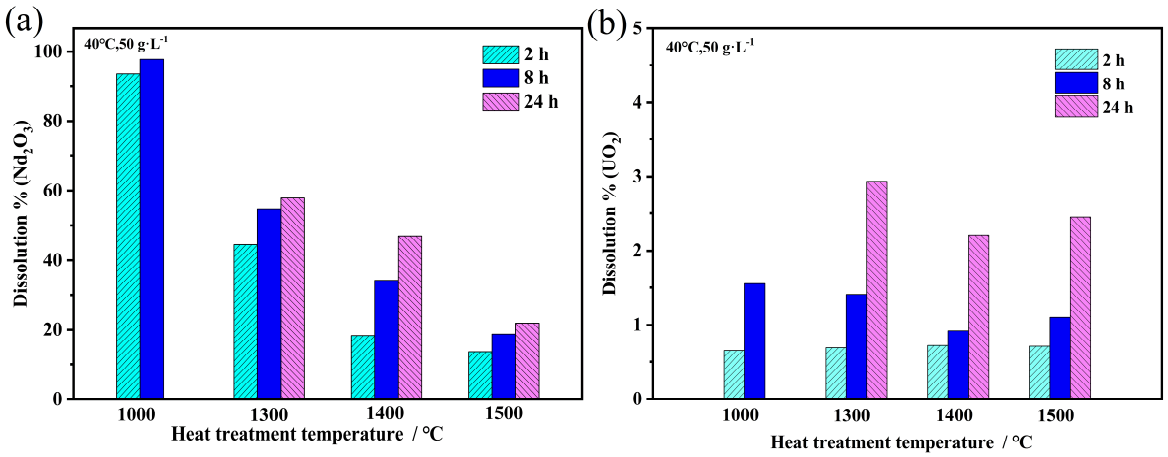


Figure S4. Effects of heat-treatment temperature and dissolution time on the dissolution efficiencies of (**a**) Nd_2O_3 and (**b**) UO_2 from SIMFUEL in 1 M aqueous HbetCl at 40 °C. The solid: liquid ratio is 50 mg/mL.

5. Text S5. Characterization of HBetCl

Radiolytic stability of the separation system is an important feature for applications in spent nuclear fuel reprocessing due to the highly radioactive environment. Though HBetCl are highly selective dissolution for lanthanides, their actual use for lanthanides separation requires testing their radiolytic stability. Different accumulative absorbed doses (100~1000 kGy) were applied to investigate radiolytic stability of HBetCl. No significant color change of the HBetCl solution or solid was observed before and after irradiation. Moreover, the solid HBetCl after irradiation keep the original state. However, this result is not consistent with the previous report where [Hbet][Tf_2N] solid changed into a pasty state at higher radiation dose. This difference may be attributed to the effect of anions in these two molecules.

In order to verify the observed radiolytic stability of HbetCl and no degradation under high irradiation dose, HbetCl sample was further measured by means of several analytical techniques. As shown in Figure S5, the comparison showed no discernible changes in any of the FTIR spectra, even those irradiated at a dose of 1000 kGy, suggesting that the excellent radiation resistance of HbetCl. The strong and sharp absorption band at 1730 cm^{-1} is mainly attributed to the stretching vibration of C=O of COOH group. Meanwhile, the maximal absorption wavelength (228 nm) of HbetCl solution did not change before and after irradiation in UV-Vis spectra (Figure S6). In addition, the maximal absorption intense increased with the increasing dose. Besides, a new broad shoulder around 303 nm for the sample of irradiated HbetCl was appeared. This result is similar to the previous studies due to the same cation $Hbet^+$. However, the comparison of ESI-MS spectra showed no new peaks in the irradiated HbetCl. Therefore, HbetCl exhibits excellent radiation resistance, where no degradation was observed after large dose γ irradiation.

Figure S7 shows the experimental results of Nd_2O_3 selective dissolution from simulated nuclear fuel using irradiated HbetCl solution. Here, 1 M HbetCl solution was irradiated directly. As can be seen, a high Nd_2O_3 dissolution efficiency of 93% for all irradiated HbetCl solution, suggesting that HbetCl is indeed an effective medium for selective dissolution separation of lanthanide. Moreover, the uranium dissolution is independent of the irradiation, which favors the recovery of UO_2 solid. Such an observation is an indication of the application of HBetCl in the spent nuclear fuel reprocessing.

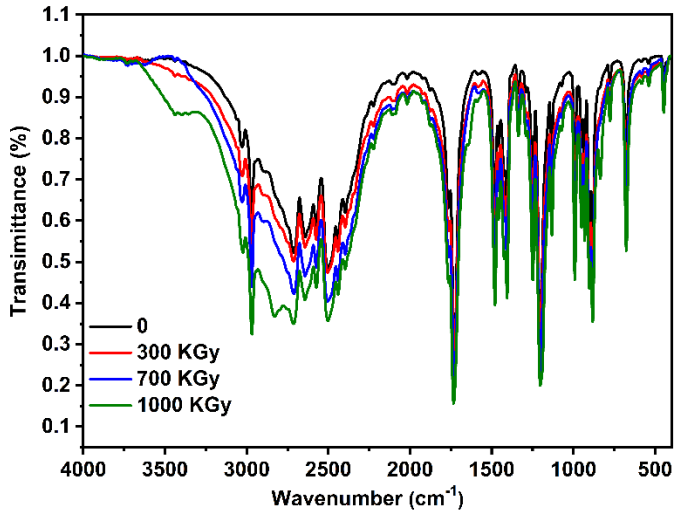


Figure S5. FT-IR spectra of HbetCl samples before and after γ-irradiation.

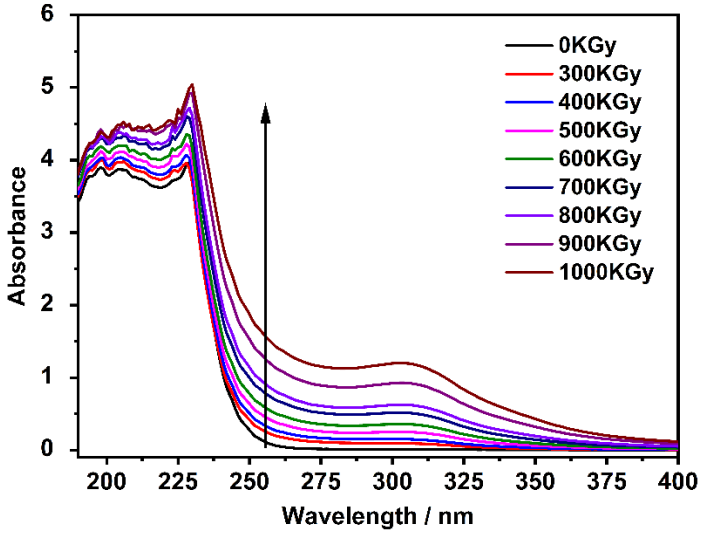


Figure S6. UV-Vis spectra of HbetCl solution before and after irradiation.

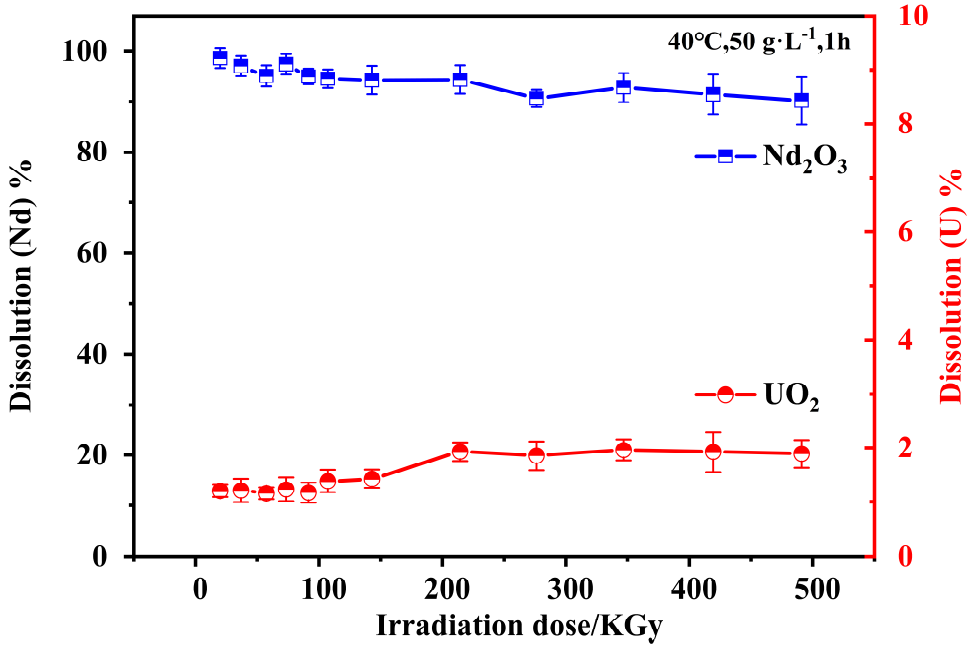


Figure S7. Influence of HbetCl with different accumulative absorbed doses on the dissolution of Nd_2O_3 and UO_2 from simulated nuclear fuel. The solid: liquid ratio is 50 mg/mL. 1 M HbetCl solution was irradiated directly.