



Original Article

Reprocessing of simulated voloxidized uranium–oxide SNF in the CARBEX process



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ABSTRACT

The concept of a new method, the CARBEX (CARBONate EXtraction) process, was proposed for reprocessing of spent uranium oxide fuel. The proposed process is based on use of water solutions of Na_2CO_3 or $(\text{NH}_4)_2\text{CO}_3$ and solvent extraction (SE) by the quaternary ammonium compounds for selective recovery and purification of U from the fission products (FPs). Applying of SE allows to reach high degree of purification of U from FPs. Carrying out the processes in poorly aggressive alkaline carbonate media leads to increasing safety of SNF's reprocessing and better selectivity of separation of lanthanides and actinides. Moreover carbonate reprocessing media allows to carry out a recycling and regeneration of reagents. We have been done laboratory scale experiments on the extraction components of simulated voloxidated spent fuel in the solutions of NaOH or $\text{Na}_2\text{CO}_3\text{--H}_2\text{O}_2$ and recovery of U from carbonate solutions by SE method using carbonate of methyltriethylammonium in toluene. It was shown that the purification factors of U from impurities of simulated FPs reached values $10^3\text{--}10^5$. The received results support our opinion that CARBEX after the further development can become more safe, simple and profitable method of spent fuel reprocessing.

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1. Introduction

The concept of the CARBEX process, a new hydrometallurgical method of spent nuclear fuel (SNF) reprocessing in carbonate media, was first published in 2008 [1]. It was based on the works of Japanese [2–5], American [6–9] and South Korean [10] researches of SNF reprocessing in carbonate media and our own works on the solvent extraction (SE) of U and other rare metals from carbonate solutions with quaternary ammonium compounds (QACs) [11]. Unlike the methods of spent fuel reprocessing described in the literature [4,8,10] which are based mainly on the precipitation techniques, CARBEX process is based on processes of solvent extraction. SE is a proven way to recover and effectively purify the U and Pu from fission products (FPs). In many ways the designed method is similar to the widely used PUREX process, but it differs from it in the choice of media and the extractant. Nitric acid is

replaced by sodium or ammonium carbonate and tri-*n*-butylphosphate (TBP) is replaced by the most radiation-resistant extractants – QACs. Replacing HNO_3 with Na_2CO_3 or $(\text{NH}_4)_2\text{CO}_3$ within the CARBEX process allows to reduce the danger of fires and explosions at the stage of SE reprocessing of highly radioactive solutions, their corrosive activity for equipment and the extractant. Moreover the use of CARBEX, could substantially reduce the volume of liquid radioactive waste, especially the highly radioactive one, at all stages of SNF reprocessing. The different solubility of lanthanides and actinides in carbonate solutions could help increase the selectivity of their separation during the dissolution of SNF and its subsequent SE reprocessing. A large amount of sodium could be recovered as NaHCO_3 by gaseous CO_2 aeration into NaOH and Na_2CO_3 solutions. Unspent $(\text{NH}_4)_2\text{CO}_3$ or NH_4HCO_3 could be further regenerated from the solution by distilling the gaseous NH_3 and CO_2 . Lower salts' concentrations in solutions simplifies subsequent reprocessing of liquid radioactive waste (LRW). These advantages could make SNF reprocessing technology safe, simple and more economically feasible due to the less amount of LRW produced.

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The concept of the CARBEX process includes the following sequence of technological operations [1]: 1) low-temperature volume oxidation (voloxidation) of the fuel composition; 2) oxidative dissolution of SNF in carbonate solutions in the presence of H_2O_2 ; 3) SE-based refining of U(VI) and Pu(VI) by carbonate of methyltriocylammonium ($(\text{MTOA})_2\text{CO}_3$) or carbonate of methyltri-C_{7–9}-alkylammonium ($(\text{MTAA})_2\text{CO}_3$) in a hydrocarbon diluents; 4) collective solid-phase re-extraction of carbonate compounds of U(VI) and Pu(VI); 5) fabrication of UO_2 and PuO_2 -based powders and pellets from carbonate compounds of U(VI) and Pu(VI).

The process of SNF voloxidation is well studied and described in literature [12,13]. During the process of voloxidation the main component of SNF (UO_2) is completely transformed into U_3O_8 . The dissolution of U_3O_8 in Na_2CO_3 solutions in the presence of H_2O_2 leads to the formation of carbonate and mixed peroxo-carbonate complexes of U(VI): $[\text{UO}_2(\text{CO}_3)_3]^{4-}$, $[\text{UO}_2(\text{O}_2)(\text{CO}_3)_2]^{4-}$, $[(\text{UO}_2)_2(\text{O}_2)(\text{CO}_3)_4]^{6-}$ [14,15]. Dissolution of metallic U [16], UO_2 [5,6,17], (U,Gd) O_2 nuclear fuel scrap [18], simulated uranium-oxide SNF [19,20] and UO_2 -based spent fuel (ATM-105, ATM-106 and ATM-109) [21] in the solutions of $\text{M}_2\text{CO}_3\text{-H}_2\text{O}_2$ (where M is Li^+ , Na^+ , K^+ or NH_4^+) also lead to the formation of U(VI) species with similar structure.

During the SE separation of carbonate and mixed peroxo-carbonate species of U(VI) by $(\text{MTOA})_2\text{CO}_3$ and $(\text{MTAA})_2\text{CO}_3$ in toluene, the formation of complexes $(\text{R}_4\text{N})_4[\text{UO}_2(\text{O}_2)(\text{CO}_3)_2] \cdot n(\text{R}_4\text{N})_2\text{CO}_3$, $(\text{R}_4\text{N})_4[\text{UO}_2(\text{CO}_3)_3] \cdot n(\text{R}_4\text{N})_2\text{CO}_3$ ($n = 1-2$), $(\text{R}_4\text{N})_4[\text{UO}_2(\text{CO}_3)_3]$ and $(\text{R}_4\text{N})_4[\text{UO}_2(\text{O}_2)(\text{CO}_3)_2]$, where R_4N is MTOA or MTAA, was discovered [22]. Polinuclear complexes $(\text{R}_4\text{N})_6[(\text{UO}_2)_2(\text{O}_2)(\text{CO}_3)_4]$ and $(\text{R}_4\text{N})_6[(\text{UO}_2)_3(\text{O}_2)_2(\text{CO}_3)_4]$ form in the case of saturation of extractant [22]. Beside the SE using QACs [20,22–25] for extracting of U from carbonate solution, it was proposed to apply the sorption by amidoxime resins (functional groups: $\text{C}(\text{NH}_2) = \text{NOH}$) [26,27], the precipitation of $\text{UO}_2(\text{O}_2) \cdot n\text{H}_2\text{O}$ ($n = 2, 4$) after the neutralization solutions of U(VI) peroxo-carbonate solutions by 1.0 mol L^{-1} solution of HNO_3 (acidification) up to $\text{pH} = 3$ [28–30] and the electrolytic decarbonation in cell with cation exchange membrane [31,32].

The main goal of this research was to show feasibility of the main stages of CARBEX process (the oxidative dissolution and the SE refining of U) and to optimize the technological parameters of reprocessing of simulated voloxidized uranium-oxide SNF (SIMFUEL powder) at the laboratory scale.

2. Experimental

2.1. Materials and methods

Solid salts Na_2CO_3 , $(\text{NH}_4)_2\text{CO}_3$, NaOH and 35% solution of H_2O_2 of the analytical grade were used. $(\text{MTOA})_2\text{CO}_3$ was synthesized according to the work [33]. Toluene of the analytical grade was used for dissolving $(\text{MTOA})_2\text{CO}_3$.

We used SIMFUEL powder with the natural content of isotopes ^{235}U and ^{238}U , as well as simulated FPs in the form of oxides of stable isotopes: CeO_2 , MoO_3 , ZrO_2 , Sm_2O_3 , La_2O_3 , Nd_2O_3 , Sm_2O_3 , Y_2O_3 , SnO , SrO and carbonates: Cs_2CO_3 , BaCO_3 (see Table 1) of the

Table 1
The composition of SIMFUEL powder.

Component	U_3O_8	CeO_2	MoO_3	Cs_2CO_3	ZrO_2	BaCO_3
Composition, %	95.94	0.31	0.41	0.76	0.77	0.22
Component	SrO	SnO	La_2O_3	Nd_2O_3	Sm_2O_3	Y_2O_3
Composition, %	0.10	0.05	0.47	0.73	0.13	0.11

analytical grade.

The concentrations of metals in solutions and solid samples were analyzed with the ICP-MS method by an $\text{iCAP}^{\text{TM}} \text{Q}$ (manufactured by Thermo Fisher Scientific, USA). The concentrations of Na_2CO_3 and $(\text{NH}_4)_2\text{CO}_3$ in solutions were established by the potentiometric titration of a liquid sample of $100.00 \text{ mmol L}^{-1}$ by solution of HCl , with a glass P13/BNC electrode using a digital pH/mV/temperature meter Elite pH Meter 3320. The electrode had been calibrated with 3 buffer solutions ($\text{pH} = 4.00, 7.00$ and 10.00). The concentration of H_2O_2 in solutions was established by titration with $20.00 \text{ mmol L}^{-1}$ potassium permanganate solution.

The specific area was found with the BET method using software QuadraWin (v. 5.02) based on the isotherm of adsorption of gaseous argon on the tested powder samples, on a Quadrasorb Kr/SI (manufactured by Quantachrome Instruments).

The phase content of the powders was established from the XRD of the samples. The XRD patterns was obtained with a D2 PHASER Bruker. The JCPDS-ICDD database and TOPAS structural refinement software were used to read the spectra and to determine the phase content.

2.2. Experimental approach

The SIMFUEL powder was prepared by mixing of the metal oxides and carbonates powders. Composition of the SIMFUEL powder was chosen according to the mass content of U and FPs in the SNF of the water-water energetic reactor VVER-1000, with the burnup rate of 60 MWd kgU^{-1} , after 3 years of cooling period. To prepare the simulator with an accurate composition, powders of metals' oxides and carbonates have been weighted with an accuracy of $\pm 0.0001 \text{ g}$, placed in a porcelain mortar and carefully grinded for 30 min. The grinded powder mixture was placed in a sealed metal container and shaken for 30 min. Obtained homogeneous SIMFUEL powder was heat-treated in air at $500 \text{ }^\circ\text{C}$ for 4 h and cooled to the room temperature. While reading the X-ray diffraction (XRD) pattern, the phases of U_3O_8 and CeO_2 were identified (Fig. 1). The specific area of heat-treated powder were determined by the BET method was $0.44 \text{ m}^2 \text{ g}^{-1}$.

The oxidative dissolution of the SIMFUEL powder was conducted in a glass reactor with the volume of 1000 mL equipped with a reflux condenser. A magnetic stirrer was used to stir the material with the speed of 500–1000 rpm. In order to intensify the oxidative dissolution and to speed up the process, a horn type ultrasonic waveguide, connected to an ultrasonic wave generator and the console of the apparatus Bulava-P UZAP-3/22-OP (manufactured by the Center of Ultrasonic Technologies, Russian Federation), was placed into the reactor. Ultrasound influence dissolution and leaching kinetics and reduce the time of the process. Acoustic cavitation in liquids, generated by ultrasonic waves, can accelerate chemical reactions rates. Presence of acoustic cavitation and

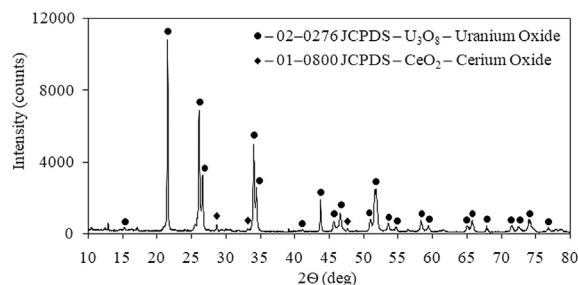


Fig. 1. XRD pattern of SIMFUEL powder after heat-treatment at $500 \text{ }^\circ\text{C}$ and 4 h processing time in an air atmosphere.

acoustic vortex streams in heterophasic reactions produce various mechanical effects: grinding of solid particles, increasing surface area of the reactants, accumulation of crystals' lattices defects, renewing (cleaning) and erosion of the surface of a solid reactant. All of this lead to the intensification of both the heat and mass transport. The ultrasonic treatment (UT) during the dissolution was conducted at the frequency (ν) of 22 ± 1.65 kHz and the intensity of (I) 10 W cm^{-2} .

The degree of extraction (α) of each element was calculated by the following equation:

$$\alpha(\%) = (W_M / W_{M,i}) \cdot 100 \quad (1)$$

where $W_{M,i}$ is the initial quantity of the metal and W_M is the quantity contained in the filtered solution after dissolution or leaching.

The SE of U(VI) from the oxidative dissolution carbonate solution was conducted in glass separating funnels with the volume of 50–250 mL, stirring intensely, at room temperature. The values of the purification factor (K_{PUR}) of U from impurities were calculated according to the equation:

$$K_{PUR} = [(W_U)/(W_M)] / [(W_{U,i})/(W_{M,i})] \quad (2)$$

where $W_{U,i}$ and $W_{M,i}$ are the masses of U and impurities in the initial SIMFUEL powder sample while W_U and W_M are the masses after the treatment. In case of oxidative dissolution of the SIMFUEL powder, the calculation of W_U and W_M was carried out using concentration of metal and volume of the carbonate solution after filtration.

Fig. 2 shows features of the operational scheme of the CARBEX process used to reprocess the SIMFUEL powder on a laboratory scale.

3. Results and discussion

3.1. Alkaline processing of the SIMFUEL powder

During the oxidative dissolution of SNF in solutions of $\text{Na}_2\text{CO}_3\text{--H}_2\text{O}_2$, along with U and Pu, the admixtures of Mo, Cs, Nd, Sm, Pr, Zr, Ce and Sr were dissolved [14]. Processing of SNF by solution of NaOH before oxidative dissolution resulted in selective separation of Mo and Cs. The removal of Mo and Cs will simplify subsequent SE processing of carbonate solutions obtained during the oxidative dissolution of SNF and increase the K_{PUR} of U and Pu from those admixtures.

MoO_3 is easily dissolved in aqueous solutions of NaOH and Na_2CO_3 with formation of Na_2MoO_4 , while metallic Mo dissolves only in the presence of an oxidant. When the SIMFUEL powder

(molybdenum in the form of MoO_3) was subjected to aqueous solutions of NaOH, the value $\alpha(\text{Mo})$ increased from 37% ($L:S = 2$) to 99% at a leaching stage, with an increase of the concentration of NaOH from 0.08 mol L^{-1} to 0.48 mol L^{-1} , temperature from 25°C to 75°C and mass ratio to liquid and solid phases ($L:S$) from 2 to 5 (see Fig. 3). After two stages of leaching, in 0.48 mol L^{-1} NaOH at 75°C , the value $\alpha(\text{Mo})$ was more than 99.9%. During each stage, equilibrium was achieved for 45–60 min.

In the studied system, Cs is less efficiently extracted. The maximum $\alpha(\text{Cs})$ attained for a leaching stage ($L:S = 5$) was 26%, and for 2 stages it was 53.2%. The value $\alpha(\text{Cs})$ also rises with an increase of concentration of NaOH and temperature (see Fig. 3).

The $\alpha(\text{U})$ into the alkaline solution practically does not depend on the conditions of the experiments and does not exceed 0.1–0.2%. The K_{PUR} values of purification of U from Mo and Cs, which were obtained after two stages of treatment by 0.48 mol L^{-1} NaOH at 75°C , $L:S = 5$, were 820 and 2.1, respectively. The K_{PUR} values of purification of U from Sn (1.2) and Sr (1.1) are smaller, their extraction was 13.9% and 7.7%, respectively. Purification from Zr, Ba or REEs does not occur for all practical purposes, their extraction is not over 0.2%.

3.2. Oxidative dissolution of SIMFUEL powder in carbonate solutions

The process of oxidative dissolution envisages the extraction of U from the SIMFUEL powder (in the form of U_3O_8) in $\text{Na}_2\text{CO}_3\text{--H}_2\text{O}_2$ solution. During the oxidative dissolution of U_3O_8 in 0.94 mol L^{-1} Na_2CO_3 in the presence of 0.34 mol L^{-1} H_2O_2 at 25°C and $L:S = 15$, carbonate solutions containing 40–60 gU L^{-1} were obtained for

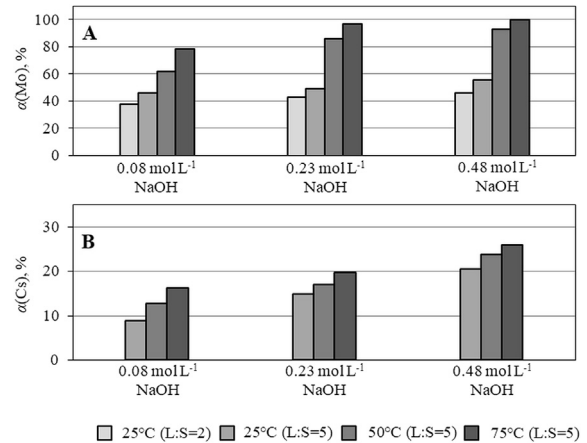


Fig. 3. Leaching Mo (A) and Cs (B) from the SIMFUEL powder by solutions of NaOH.

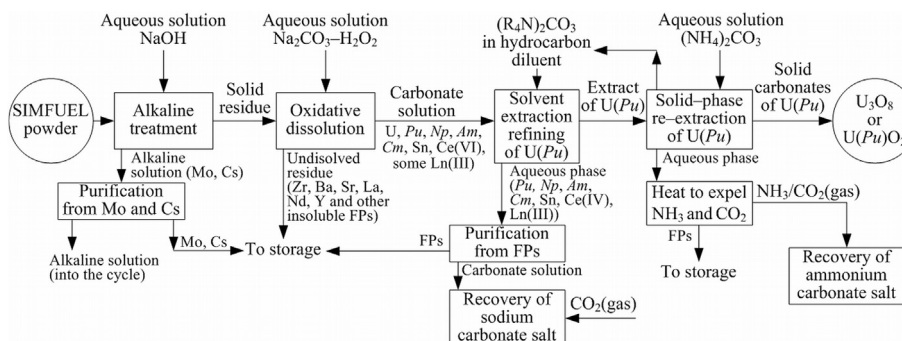


Fig. 2. Operational scheme of the CARBEX process for reprocessing SIMFUEL powder on a laboratory scale. TRU elements – estimated behavior.

6–9 h, with $\alpha(\text{U}_3\text{O}_8)$ equaling 99.9% [34]. With the UT applied, the time of complete dissolution decreases from 6–9 h to 45–90 min and the maximal concentration of U attained in the solution goes up to 140–160 gU L^{-1} [14,15].

The oxidative dissolution of the SIMFUEL powder after its alkaline treatment was conducted in solution $1.0 \text{ mol L}^{-1} \text{Na}_2\text{CO}_3 - 0.2 \text{ mol L}^{-1} \text{H}_2\text{O}_2$, with UT, 70°C , $L:S = 2.7$, $\tau(\text{leach.}) = 90 \text{ min}$ and H_2O_2 is supplied every 5 min. As a result of the oxidative dissolution we obtained a solution containing 142.5 gU L^{-1} , with $\alpha(\text{U}_3\text{O}_8) = 46\%$. The $\alpha(\text{Mo})$ and $\alpha(\text{Cs})$ into the carbonate solution was 92.7% and 90.1%. Due to low solubility of a number of oxides (Ce(IV), Ln(III), Zr(IV), Sr(II)), we observed purification from those metals in carbonate solutions already at the stage of oxidative dissolution of SIMFUEL powder. The concentrations of Y(III), Ln(III), Ce(IV) and Zr(IV) in the carbonate solution were in the range of 10–60 mg L^{-1} , and for Sn, Sr and Ba they were 5, 9 and 15 mg L^{-1} , respectively. The degree of extraction of these admixtures was not over 1–2%. The K_{PUR} values of purification of U from Ce(IV), Ln(III), Zr(IV), Sr and Ba were from 1.5 to 38.3 (see Table 2).

Concentrations of Pu and Np after the dissolution of PuO_2 and NpO_2 in the $\text{Na}_2\text{CO}_3 - \text{H}_2\text{O}_2$ solutions reached 0.01–0.07 gPu L^{-1} and 0.4 gNp L^{-1} respectively. At the stage of oxidation of Pu(IV) to Pu(VI) and Np(IV) to Np(VI) in the presence of persulphate, concentrations of Pu and Np reach 0.5–1.0 gPu L^{-1} and 1.5 gNp L^{-1} respectively. At the oxidative dissolution stage of the CARBEX process all Pu and Np would be transferred to the carbonate solution together with U.

3.3. Solvent extraction refining of uranium from carbonate solutions

The SE refining is the final stage of U purification before obtaining UO_2 powders. While developing the concept of the CARBEX process, the SE separation of U(VI) from carbonate solutions was supposed to be conducted by $(\text{MTOA})_2\text{CO}_3$ or $(\text{MTOA})_2\text{CO}_3$. During the development of the SE stage of the CARBEX process, two stages of SE is found to be necessary. The first stage is SE of U(VI) from carbonate solutions of oxidative dissolution of SNF, which results in co-extraction of FPs admixtures together with U(VI) into the organic phase. The second stage is the SE purification of U(VI) from the co-extracted admixtures, i.e. the SE refining. We suggested that the re-extraction in both stages should be conducted by $(\text{NH}_4)_2\text{CO}_3$ and NH_4HCO_3 solutions or their mixtures, with obtaining of crystalline powders $(\text{NH}_4)_4[\text{UO}_2(\text{CO}_3)_3]$ and with subsequent dissolution (after the first SE stage) and calcination with obtaining of U_3O_8 or UO_2 .

The carbonate solution containing 142.5 gU L^{-1} and the FPs obtained after the oxidative dissolution was used for conducting the first SE stage according to the operational scheme (see Fig. 4). Solvent extraction was carried out in counter-current mode. Optimal conditions of SE, washing and re-extraction (volume ratio of aqueous to organic phases (A:O), phase agitation time (equilibrium time) (τ), number of steps (n) and others) were established in our previous experiments. On the first stage of SE of $0.4 \text{ mol L}^{-1} (\text{MTOA})_2\text{CO}_3$ in toluene as an extractant was supplied. On the last

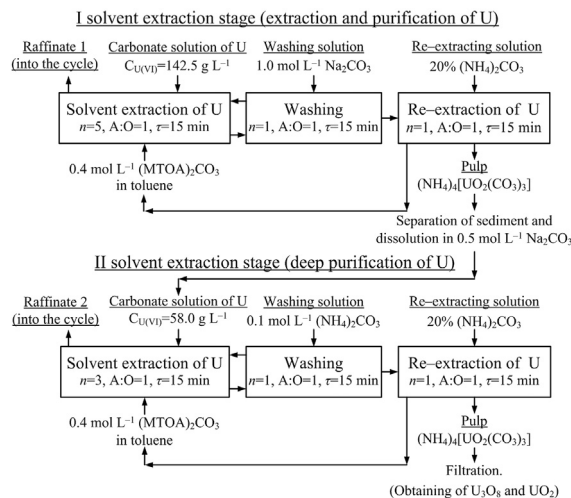


Fig. 4. Operational scheme of the first and second SE stages. A:O – volume ratio of aqueous to organic phases; τ – agitation time; n – number of steps.

stage of SE primary carbonate solution of U(VI) mixed with washing solution were supplied. After additional washing, extract from the last SE step was transferred to the subsequent stage of solid–phase re-extraction. Organic phase after re-extraction was again introduced to the cycle (to the first SE step).

After the solid–phase re-extraction, the carbonate sediment was dissolved in $0.5 \text{ mol L}^{-1} \text{Na}_2\text{CO}_3$, which lead to the formation of carbonate solution, containing 58.0 gU L^{-1} in the form of carbonate complex $\text{Na}_4[\text{UO}_2(\text{CO}_3)_3]$, which was required for the second SE stage.

Samples taken from the carbonate sediment obtained after re-extraction at the first and second SE stages, were dried and calcinated at 800°C for 2 h to form the U_3O_8 . It was found that the purification factors for all FPs admixtures reach the values of $10^3 - 10^5$ after two stages of SE (see Table 3). This fact gives the reason to consider SE refining to be an as-efficient method of purification of U from FPs, as the SE purification by tri-n-butyl phosphate from nitric acid solutions.

It should be noted that Pu(IV) is poorly extracted from carbonate solutions by $(\text{MTOA})_2\text{CO}_3$ so it could be separated from U(VI) at this

Table 3

The K_{PUR} values of purification of U from some FPs in the CARBEX process.

Stage of SE	K_{PUR}					
	La	Zr	Sr	Nd	Y	Ba
1st	20.7	$5.3 \cdot 10^3$	26.3	13	40.5	35.8
2nd	$2.5 \cdot 10^3$	$5.3 \cdot 10^5$	$3.2 \cdot 10^3$	$1.3 \cdot 10^3$	$4.1 \cdot 10^3$	$4.8 \cdot 10^3$
Stage of SE	K_{PUR}					
	Ce	Sn	Mo	Sm	Cs	
1st	254	1.1	128	14.5	52.7	
2nd	$2.4 \cdot 10^4$	$6.1 \cdot 10^2$	$1.3 \cdot 10^5$	$1.5 \cdot 10^3$	$4.5 \cdot 10^5$	

Table 2

The K_{PUR} values after alkaline treatment (2nd stage) and oxidative dissolution of SIMFUEL powder in $1.0 \text{ mol L}^{-1} \text{Na}_2\text{CO}_3 - 0.2 \text{ mol L}^{-1} \text{H}_2\text{O}_2$, with UT, 70°C , $L:S = 10$, $\tau(\text{leach.}) = 90 \text{ min}$.

Stage	K_{PUR}											
	U	Ce	Mo	Cs	Zr	Ba	Sr	Sn	La	Nd	Sm	Y
Alkaline treatment (2nd stage)	–	1	820	2.1	1	1	1.1	1.2	1	1	1	1
Oxidative dissolution	–	2.8	1.1	1.1	20	38.3	32.9	1.3	24.5	12.9	1.5	4.5

stage. However, in the studied systems, the extraction yield of Pu(VI) is comparable with the one of U(VI). This fact allows achieving either collective, or the separate purification of U and Pu from FPs in the CARBEX process.

4. Conclusions

In the present study using simulated voloxidized uranium–oxide spent fuel, it was established that a new SNF reprocessing method, named CARBEX is effectively working. This reprocessing method, consists of two main stages, the oxidative dissolution of voloxidized SNF by water solutions of Na_2CO_3 or $(\text{NH}_4)_2\text{CO}_3$ and solvent extraction refining of uranium from FPs by the quaternary ammonium compounds.

Obtained data has shown that the treatment of heat–treated sample (at 500 °C and 4 h processing time in an air atmosphere) of SIMFUEL powder containing oxides of U, Ce, Mo, Zr, Sr, Sn, La, Nd, Sm, Y and carbonates of Cs, Ba with an aqueous solution of NaOH results in extraction of more than 99.9% of Mo and 53.2% of Cs. Applying ultrasonic treatment reduces time of U_3O_8 leaching from SIMFUEL powder in $\text{Na}_2\text{CO}_3\text{--H}_2\text{O}_2$ solutions from 6–9 h to 45–90 min and increases concentration of uranium from 40 to 60 gU L^{-1} to 140–160 gU L^{-1} . At the oxidative dissolution stage, uranium is purified from Zr, Ba, Sr and REEs. Achieved value of the coefficient of purification of U from impurities of simulated FPs after performing two solvent extraction stages was $10^3\text{--}10^5$. Obtained results allow us to continue designing and developing an efficient method of recovery and purification of uranium from impurities in carbonate solutions used in the CARBEX process.

However further development of CARBEX process requires understanding of behavior of plutonium, neptunium, other transuranium elements and also some FPs at main stages of this process. Another major development point is the optimization of solvent extraction refining stage that could help to increase values of purification factors of uranium from FPs to the ones reached in conventional PUREX process ($10^6\text{--}10^7$) or even up to 10^8 in the perspective. Final CARBEX's development phase would be testing and evaluation of all the process's stages on the real samples of spent fuel. After the further development and optimization, CARBEX process could become a good alternative to widely used PUREX process, making SNF reprocessing technology more safe, simple and economically feasible.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.net.2019.05.020>.

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