The Applicability of Distillation as an Alternative Nuclear Reprocessing Method

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Abstract— A customized two-stage model has been developed to simulate, analyse, and visualize distillation of actinides as a useful alternative low-pressure separation method in the nuclear recycling cases. Under the most optimal conditions of idealized thermodynamic equilibrium stages and under total reflux of distillate the investigated cases of chloride systems for the separation of such actinides are (A) UCl_4-CsCl-$PuCl_3$ and (B) $ThCl_4$-NaCl-$PuCl_3$. Simulatively, uranium tetrachloride in case A is successfully separated by distillation into a six-stage distillation column, and thorium tetrachloride from case B into an eight-stage distillation column. For this, a permissible mole fraction value of 1E-06 has been assumed for the residual impurification degree. With further separation effort of eleven to seventeen required separation stages, the monochlorides of plutonium trichloride from both systems A and B are simulatively shown to be separated as high pure distillation products.

Keywords—Conceptual design of a pyroprocessing unit, molten salt recovery, simulation of total-reflux distillation column, used nuclear fuel reprocessing.

I. INTRODUCTION

SINCE the beginning of the application of nuclear energy, despite the constant development of new reactor technology concepts in the past, the question of what to do with nuclear waste and how to get rid of it in the best possible way has remained open. Until today, after the classification of four reactor generations, the obsolete technology of second-generation reactors with solid fuel rods as fuel is still used [1]; a technology that can only use a very small fraction of the fuel at all, because the spent fuel components are not separable from the solid matrix and accumulate. The fourth-generation Molten Salt Reactor (MSR) already makes much more effective use of the fuel in the form of undiluted molten salt fuel as fluoride or chloride salts, as for example in thorium tetrachloride-based $ThCl_4$-NaCl-$PuCl_3$ chloride systems [2], [3], in which even the processing of the fuel by high-temperature distillation would already be potentially applicable. Another new reactor concept is the Dual Fluid Reactor of two liquid flow cycles: one undiluted molten fuel cycle with metallic uranium-chromium eutectic (DFRm version) or undiluted fuel material as chlorides from nuclear waste material (DFRs version) and another counter-current liquid lead cycle used for indirect cooling [4]-[6]. The idea of integration of a high temperature distillation unit in the DFRs version, for the separation of unburnt material components and feeding back into the reactor, makes it possible to completely recycle the nuclear waste until quite no disposal is required any more [7].

Liquid-liquid extraction as a developed separation method, such as that of the common PUREX process (plutonium uranium reduction extraction) [8], [9], is not suitable as a separation method, because a very large amount of numerous different solvents and extractants are required for material separation, and lots of separation units are required. In contrast to extraction, distillation as a separation technique is feasible as undiluted fuel and space-saving separation technology into a single separation device. The method of distillation of metal chlorides itself at higher temperatures has already been developed industrially in the 1940s in the so-called Kroll process [10] for the purification of titanium tetrachloride from titanium ores. Until then, several other practical studies on the distillation of metal chlorides have been developed such as the separation of chlorides MCl_2 with M of elements referred to the second main group of periodic table, from LiCl-KCl eutectic molten salts according to [11] and [12]. On an industrial scale, distillation has been successfully used for almost every product over the last century [13], [14]. So, in order to investigate distillation as a possible modern separation technology for nuclear applications, it is first useful to estimate the feasibility of distillation in the best-case scenario of substance separability using two representative substance system (A) UCl_4-CsCl-$PuCl_3$ and (B) $ThCl_4$-NaCl-$PuCl_3$ from [3], [7] with a highly simplified simulation model.

II. THEORY AND PROCEDURE ON DISTILLATION COLUMN MODELLING

For modelling and simulation of distillation columns, so-called equilibrium stage models are used as explained in [13], in which the column is composed of theoretical separation stages of thermodynamic equilibrium stages. Liquid is fed back to the column at the top of the column at a certain reflux ratio v to enable the counter-current principle of the column.

Fig. 1 shows a simplified and exemplary case of the stage model of a continuous distillation column with complete refeeding of distillate into the column from upper shown condenser and as well for the boiling product in evaporator at the bottom of the column. This column operation corresponds to the optimum case between a discontinuously operating column and a continuous one. In the discontinuous column, a mixture with molar composition $x_0 = x(t = t_0)$ is set at any position of the column, mostly preferred in the bottom, at the initial time t_0, whereas in the continuous column operation, continuous feed streams x_F are added to the column. Distillate

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products are removed at the top and bottom or as side streams of the column. Instead of pure numerical simulations based on triagonal matrix procedures as explained in [13], simulations based on graphical calculation methods for ternary mixtures and for the total reflux columns are chosen for the feasibility analysis in this article.
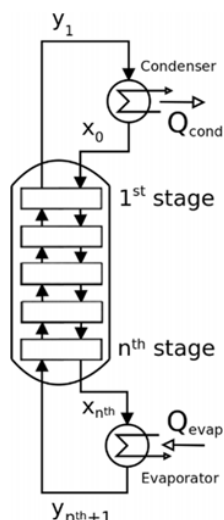

Fig. 1 View of the continuous total reflux distillation column scheme

A. Calculation Procedure for Simulations

Starting from ideal thermodynamic equilibria based on the Raoult's and Dalton's law, a two-stage simulative evaluation method of the total reflux distillation column for a ternary mixture of substances is performed. For thermodynamic equilibrium, the following simplified equation is used

$$x_{i,j}p_i^{LV}(T_j) = y_{i,j}p \quad (1)$$

Here x and y are the mole fraction values of liquid and vapor, depending on the substance component i and current stage number j, p^{LV} is the vapor pressure each pure substance component and each stage temperature T. The total pressure each separation stage is p with

$$p = \sum_{i=1}^{N} x_{i,j}p_i^{LV}(T_j) \quad (2)$$

Substitution of (2) into the equilibrium equations according to (1) follows to the commonly used evaluation formula

$$x_{ij,eq} = \frac{\left(\frac{p_i^{LV}(T_j)}{p_N^{LV}(T_j)}\right)^j \cdot x_{ij,0}}{1+\sum_{k=1}^{N-1}\left(\left(\frac{p_i^{LV}(T_j)}{p_N^{LV}(T_j)}\right)^j -1\right)x_{kj,0}} \quad (3)$$

In (3) the expressions $p_i^{LV}(T_j)/p_N^{LV}(T_j)$ are relative volatilities of the component i referring to the more non-volatile component N. These relative volatilities are important ratios as dimensionless numbers to describe the separability of these two components from each other at a given temperature T_j. The

main used evaluation principle is a two-stage mixed graphical and numerical estimation process: In the first evaluation part, the desired distillate compositions in the top and bottom are specified, which must be positioned on the same equilibrium line according to (3). Depending on the choice of the required number of separation stages with n_{th} exactly n_{th} evaluation points of the equilibrium line according to (3) are determined. According to [13], the Fenske-Underwood-Gilliland (FUG) method for multicomponent mixtures is suitable for estimating the theoretical required total number of separation stages. Secondly, the column height is estimable, assuming similar required column stage heights as it is for the titanium tetrachloride distillation from the distillative purification of titanium ores from given HETP-values (Height Equivalent value to Theoretical Plate) in [10]. The total column height H is easily estimated by:

$$H = HETP \cdot n_{th} \quad (4)$$

The temperature is chosen constant for all theoretical separation stages with a given composition x_F which is approximated by:

$$T_{j,0} = \sum_{i=1}^{N} x_{i,F}T_{B,i} \quad (5)$$

The separation stage positioning is determined iteratively until the position point, for example marked as red point in Fig. 2, is located on the distillation line within the constraints of fulfilling the product specifications.

The second evaluation phase starts at stage j_{F-1} in the direction of the condenser. Next, with given liquid mole fraction $x_{j_{F-1}}$ the vapor mole fraction $y_{j_{F-1}}$ is determined by (1). For the next separation stage by total reflux ratio the following condition must be fulfilled

$$x_{j_{F-2}} = y_{j_{F-1}} \quad (6)$$

According to (6), the temperature of the next column stage can be simplified estimated through the following averaging procedure for $j = j_{F-2}$

$$T_j = \sum_{i=1}^{N} x_{i,j}T_{B,i} \quad (7)$$

Then, for the next stage, the vapor mole fraction can be again estimated by (1) and the calculation process is repeated up to the condenser. Above the separation stage j_{F+1} in the direction to the evaporator, the iterative procedure changes. Interval bisection procedure with a given starting point at $T_{0,j_{F+1}}$ and $T_{B,max}$ is used to solve (1), numerically, under the condition where the solution $x_{j_{F+1}}$ mets the value $T_{j_{F+1}}$ by evaluating then (5) for the separation stage j_{F+1}. For the temperature estimation is used

$$T_{j0_{F+1}} = 1/2\left(T_{j_F} + T_{B,max}\right) \quad (8)$$

$$T_{B,max} = max\{T_{B,i}\}, i = 1, N \quad (9)$$

Similar, for the next calculation step $y_{j_{F+2}} = x_{j_{F+1}}$ is valid as condition to the total reflux column similar to (6) and the iterative procedure can be repeated until the evaporator stage is reached.

For binary mixtures, the evaluation is analogous. The exception to this is the determination of the graphical initial solution, which is determined for binary mixtures and temperature-dependent relative volatilities using the methods of McCabe Thiele [13]. From the desired position, the numbers of total required theoretical separation stages are obtained in both iteration directions to condenser and to the evaporator by continuing until the product specifications are reached. In the ternary mixture for comparison, the determination is carried out manually.

In order to verify the functional performance of own implemented code for the implemented two-stage simulation procedure in terms of its ability to represent the physical accuracy, a comparison is made by using a second simulation tool. Here, a free accessible ChemSep Lide version, a simulation tool for distillation and in this case for simulation of a total reflux distillation column is used for exemplary validation. Because more substance property data are required for such simulation another substance system is used. Only the desired column stage conditions with mole fractions are necessarily to be specified for this purpose

B. Required Substance Property Data

The melting and boiling points for all used chloride components are shown in Table I. Apart from plutonium trichloride and thorium tetrachloride, critical data are known or can be calculated approximately by [9], [15] and [16] (chapter 2 and 3). For uranium tetrachloride, the critical point is approximated according to [9] and is about 1598 K and 70.714 bar. For the monochlorides, the values of the critical points are assumed to be 3400 K and 355 bar for sodium chloride and 2462.1 K and 255 bar for caesium chloride. The chlorides used for simulation code validation are tin tetrachloride, antimony trichloride and cupric chloride. For all these chlorides all critical data are known, including other property data required for simulation with the ChemSep Lide version for total reflux distillation column simulations. The property data are available from secondary sources of [7]. All chlorides are assumed to be thermally stable.

TABLE I
MELTING AND BOILING POINTS OF PURE CHLORIDE COMPONENTS

Chloride	Melting point [K]	Boiling point [K]
CsCl	918	1569
NaCl	1073.8	1738
PuCl₃	1041	2000
ThCl₄	1043	1194
UCl₄	863	1068
SnCl₄	240	382
SbCl₃	346.45	496
CuCl₂	771	1266

Source: Adjusted secondary literature data from [7], [9]

In the two-stage simulation method, only the vapor pressure data and enthalpies of vaporization of the pure substances are required as temperature-dependent substance property data. The vapor pressures of the pure chlorides are given in Table II as a function of temperature. These are taken from various literature data with respect to [7], [9] and averaged to the following correlation equation with p[bar], T[K]

$$\log p = C_1 \frac{10^3}{T} + C_2 \log T + C_3 + C_4 T + C_5 T^2 \qquad (10)$$

TABLE II
LIQUID VAPOUR PRESSURE DATA FROM (10)

Chloride	C_1	C_2	C_3	C_4	C_5
CsCl	-8.835	0.0000	5.305	0.00E+00	0.00E+00
NaCl	-11.358	-4.2035	19.557	3.47E-04	-3.95E-12
PuCl₃	-12.587	-0.9495	9.428	0.00E+00	0.00E+00
ThCl₄	-10.137	-7.2000	30.414	0.00E+00	0.00E+00
UCl₄	-9.720	-6.6800	29.332	0.00E+00	0.00E+00
SnCl₄	-1.695	0.0000	4.436	0.00E+00	0.00E+00
SbCl₃	-3.644	-5.3400	21.741	0.00E+00	0.00E+00
CuCl₂	-2.573	0.0000	2.032	0.00E+00	0.00E+00

Source: Adjusted secondary literature data from [7],[9]

III. RESULTS AND DISCUSSION

For simulation purposes, the compositions of the examples to be recycled by distillation into one single distillation column are given in Table III, including the source.
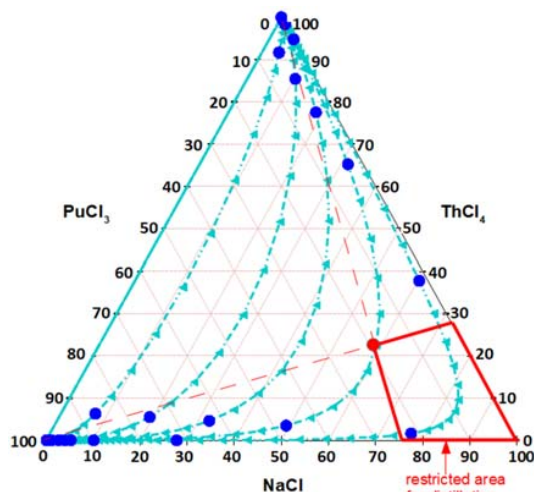
TABLE III
GIVEN COMPOSITIONS

Chloride	Melting point [K]	Boiling point [K]
CsCl	918	1569
NaCl	1073.8	1738
PuCl₃	1041	2000
ThCl₄	1043	1194
UCl₄	863	1068
SnCl₄	240	382
SbCl₃	346.45	496
CuCl₂	771	1266

Used compositions are assumed in mol-% and mean stage temperature following from (5). Source: [3], [7]
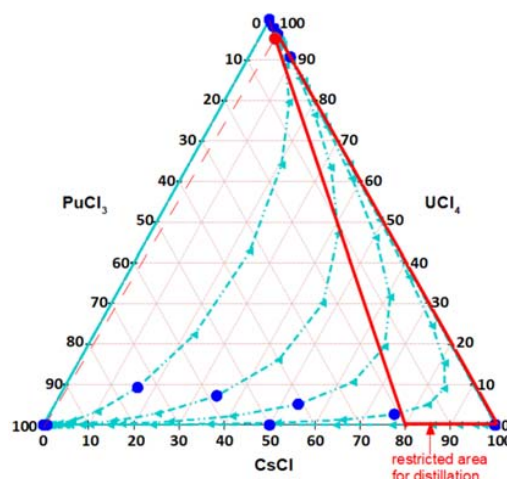
(a) View of ternary phase diagram for recycling case A, according to Table III

(b) View of ternary phase diagram for recycling case B, according to Table III

Fig. 2 View of ternary phase diagram as a trajectory distillation curve map for using continuous distillation columns.

The F position is marked as red dot following from given composition values from Table III. Trajectory curves at total reflux condition are the light blue, triangle marked curves, within dark blue dot marked estimated distillation stages. The red-marked enclosed area is restricted by any continuous distillation column operation, when assuming straight line equations for operating conditions. This restricted area is graphically determined by the red-dashed lines.

Fig. 2 shows the graphical results on the basis of the dark blue points, which represent one separation stage per distillation trajectory of a total reflux column. The distillation line on which the red dot is located corresponds to the specified separation cases A and B according to Table III. In both cases, many of the separation stages are located in positions close together near the high pure tetrachloride as the light boiler and the plutonium trichloride as the heavy boiler component. Therefore, the greatest separation effort is expected for the obtaining of separation accuracy of 1E-04-mol% according to the product specifications from [7]. Because the relative volatilities of the tetrachlorides compared to plutonium trichloride in separation case A are higher for uranium tetrachloride compared to thorium tetrachloride in case B, a higher separation effort for case B is predicted. This is also reflected to the higher density of located blue dots in the middle of the ternary diagrams from Fig. 2 in case B in combination with the shape of distillation curves. In the operation of a continuous distillation column with the feed position at location F and assuming idealized distillation stages, the distillation areas show up as enclosed areas between the distillation line of the red dot marked composition at feed position F from Table III and the red marked restricted area. For high purity separation of tetrachlorides, assuming straight lines for the operating line, it follows that no high purity separation of plutonium trichloride is directly possible into such single distillation column. For the solution, discontinuous column operation is then recommended, or a continuous second feed stream or sidestreams stripping the continuous column are required.
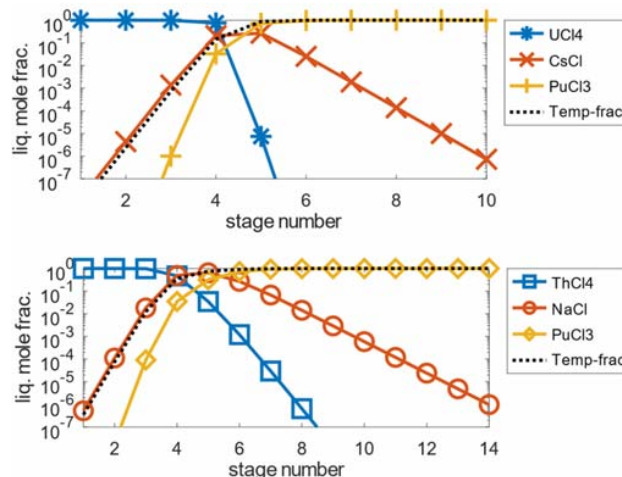

Fig. 3 Simulation results of total reflux column for system A and B

The separation behaviour into a total reflux column according to the two-step simulation procedure on the other hand, gives the results shown in Fig. 3. It provides the results in the form of unsmoothed profile[1], in which a product specification of 1E-04 mol% has been set for the low-boiling component. Both results show impressively the high purity enrichment of the tetrachlorides into the first separation stage. In the bottom stage high-pure plutonium trichloride is accumulating. Exclusively the monochlorides caesium and sodium chloride in both separation cases A and B as medium boiling components in the global maxima of the fraction profiles from Fig. 3 are not separately enrichable from the tetrachloride component or plutonium trichloride. For a better separation, the operation of discontinuous distillation with bottom feed is recommended for an industrial scale distillation. In this option, the tetrachloride component is first separated with high purity at temperatures around the boiling point of the tetrachloride. After tetrachloride separation, the monochloride component can then be obtained at higher boiling temperature and for high reflux ratio referring to top product distillate. The result of the temperature follows a sigmoidal S-curve for all calculations and can be listed as dimensional liquid temperature fraction number with $T_{B,min}$ boiling point of low boiler component and $T_{B,max}$ boiling point of heavy boiler component from Fig. 3. The dimensional liquid temperature fraction number is calculated by

$$\frac{T - T_{B,min}}{T_{B,max} - T_{B,min}} \qquad (11)$$

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[1]Note that in all diagrams for the mole fraction in the logarithmic scaling, the results are only available at the discrete nodal points. The results in between are linearly interpolated for a logarithmic y-scaling view and only show the trend progression. Thus, the sum of all substances involved does not necessarily have to be 100-mol% at the iteration points between the discrete column stages (iteration nodes).

Under highly simplified assumptions that both distillation examples have similarly large HETP values as height equivalent values per column stage with 8 inches according to [10], an estimated column height of only 1.218 m follows for separation example A and 1.624 m for separation example B.

IV. COMPARISON WITH OTHER SOFTWARE CODES

To validate the two-step simulation code used in this article, a mixture example of a chlorinated tin alloy material was used named "Die casting alloy PY1815A per ASTM B-102" [17], which consists of 82 mass% tin, 13 mass% antimony and 5 mass-% copper. For this the chlorinated components and its properties from Table I and II were used with this same assumed composition as it is for the alloy. To specify the internal flows for the ChemSep model, one tenth of the annual production quantities of 9000 t/a according to [17] was set. Referring to achieve the separation accuracy of 1E-04-mol% of impurities in top of the total reflux distillation column, an eight-stage separation total reflux distillation column was simulated for both simulation models with ChemSep Lide total reflux column model and the two-stage simulation method. The composition above was set in the fifth separation stage.
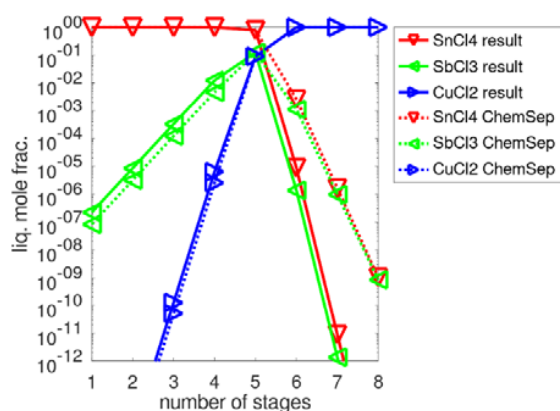

Fig. 4 Details of liquid mole fraction: Simulation results from the ChemSep Lide total reflux column model and the own implemented two-stage simulation method for the chlorinated material composition from "Die casting alloy PY1815A per ASTM B-102" Alloy [17]

The comparison of the results from both models shows that both results are able to reproduce the separation behaviour quantitatively and qualitatively correctly. The simulation results from ChemSep and the two-step separation method are plotted together in Fig. 4 for the liquid mole fraction. Tin tetrachloride dominates within the first three separation stages. In the fourth and fifth separation stage, the low-boiling component antimony trichloride is accumulating as the middle boiling component. Copper chloride is dominant in separation stages six to eight. High-purity copper chloride can be simulatively enriched in the bottom of the column and high-purity tin tetrachloride in the top of the column under set product specifications. According to Table IV in the solution of the temperature profile and the comparison of the melting points according to Table I, it was simplified for modelling

purposes for comparison that no copper chloride compound crystallizes out.

TABLE IV
TEMPERATURE PROFILE OF SIMULATION RESULTS

Column stage No.	Temperatures from the two-stage model [K]	Temperatures from the ChemSep model [K]	Relative deviations mean value [%]
1	382.0	381.5	0.13
2	382.0	381.5	0.13
3	382.0	381.5	0.13
4	383.4	382.6	0.21
5	475.4	**391.1**	**19.46**
6	1266.0	**837.8**	**40.71**
7	1266.0	1268.8	0.21
8	1266.0	1270.6	0.36

Temperature profile and relative deviations from the simulation results between ChemSep Lide total reflux model and the own-implemented two-stage simulation method.

In the first four separation stages, a constant relative deviation in the logarithmic representation of the results between ChemSep Lide model and the two-stage simulation method is present for all chloride components. The following becomes clear in the comparison of the relative deviations of the logarithmic representation from Fig. 4 and Table IV: In the first separation stages, only a small relative deviation of the simulation values of mole fractions from one another are visible, whereas from the sixth separation stage to the eighth, the deviation within the representation is increasing linearly. In the fifth separation stage, the given default values about the composition of the mixture are set for both simulation models. Therefore, no deviations are present for mole fractions. However, the deviations for the temperature determination in this stage are significantly different as seen in Table IV. The relative deviation for the temperature stage estimation from Table IV is smaller than 1% in the first four and the last two column stages but are much more higher in the fifth and sixth separation stage with values about 20% and 40% in deviation range. Suspected was the deviation in the temperature averaging according to (5) and (7) into the two-stage simulation method compared to ChemSep and the following vapor pressure evaluation for example according to (10). Those deviations were also assumed for the separation cases A and B for the application of the graphical based two stage simulation method. Therefore, the following was suggested in the use of the two-stage simulation method for the separation analysis of the separation cases A and B referring to simulation accuracy: In the first separation stages, in which the tetrachloride component dominates, the numerical error should be very small. As soon as plutonium trichloride dominates in the higher separation stages, the error value increases linearly in the simulation progress up to higher column stages. In separation case B, compared to A, the boiling points are less equidistant to each other according to Table I, so it is to be expected that these error values are larger, especially in the high boiling range, but still smaller than in the validation example due to the even larger differences of the distances of the boiling points of tin tetrachloride and antimony trichloride to copper chloride. The

influence of the error on the required number of separation stages was determined to ± 1 required separation stages by extrapolation of an error of the same magnitude as in the validation example. This finally would have only little effect of the required column height approximation of about ± 0.2 m.

V. Conclusion and Future Aspects

Distillation as a separation technique for actinides in the form of chlorides was proven to be a very powerful usable separation process alternative to possible use of liquid-liquid extraction techniques. The usage for estimation itself from the validation of the two-stage simulation model was assumed to be suitable to all simulations of mixtures within similar equidistant boiling points of all the involved components.

Uranium tetrachloride from the nuclear waste recycling example was already completely obtained as high pure separation product as well as thorium tetrachloride. The column height is smaller than 2 meters which is still a very preeminent space-saving separation unit, compared to liquid-liquid extraction. Practically, it was recommended to perform the separations into a discontinuous operating distillation column for high-pure, fractional separation of all involved individual chloride components. For future investigations, small single-stage distillation experiments of non-actinide chlorides and later with uranium tetrachloride from natural uranium are to be carried out in order to investigate more closely the real separation effort and non-ideal thermodynamic separation behaviour. Following from these experiments, further investigations and then further practical separation experiments into a distillation column can be performed.

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