NEW METHODS FOR NUCLEAR WASTE TREATMENT OF THE DUAL FLUID REACTOR CONCEPT*

Dominik Böhm[a,b], Konrad Czerski[a,b], Armin Huke[a]
Jan-Christian Lewitz[a,c], Daniel Weissbach[a,b]
Stephan Gottlieb[a,d], Götz Ruprecht[a]

[a]Institute for Solid-State Nuclear Physics GmbH
Leistikowstr. 2, 14050 Berlin, Germany
[b]University of Szczecin, Wielkopolska 15, 70-451, Szczecin, Poland
[c]LTZ-Consulting GmbH, Tharandter Str. 12, 01159 Dresden, Germany
[d]Fachhochschule Dortmund — University of Applied Sciences and Arts
Sonnenstr. 96-100, 44139 Dortmund, Germany

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The Dual Fluid Reactor (DFR) is a new fast-neutron, high temperature, liquid fuel nuclear reactor concept which is able to utilize Used Nuclear Fuel (UNF) of previous nuclear reactor generations. The DFR features an integrated Pyroprocessing Unit (PPU) which separates transuranium elements as well as fission products when distillation of chlorinated liquid fuel is applied. Simplified calculations performed here show that the distillation process with maximum 12 stages is sufficient to obtain separation of corresponding chlorides to a very high precision level. Furthermore, insoluble metallic and non-chlorinated solid materials can be removed by simple crystallization techniques.

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1. Introduction

Since the beginning of commercial utilisation of nuclear energy, the problem of nuclear waste generation has still remained unsolved. Thus, one of the most important goals for developing of the Dual Fluid Reactor (DFR) concept was to propose a high efficient reactor which enables a drastic reduction of the Used Nuclear Fuel (UNF) volume by using it as fuel material. Furthermore, the DFR combines advantages of two different reactor concepts of the fourth generation: The Molten Salt Reactor (MSR), dealing with a liquid fuel and the Lead-Cooled Fast Reactor (LFR) using liquid metallic coolant. Differently to the MSR concept, the DFR works with a separate

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coolant and fuel loops [1, 2]. This allows for production of electricity, process heat and hydrogen at costs far below the current market prices [1–3]. The DFR is a fast neutron reactor in which transuranium elements are burned down effectively and long-lived fission products can be transmuted to short-lived ones. The fission products are on-line separated in a Pyroprocessing Unit (PPU) using distillation as a dry thermal separation procedure that utilizes differences between different boiling points of chemical compounds. The distillation process is performed on metal chlorides because of their low and well-separated boiling points, which is already commercially used in the chemical industry since the 1940s and known as Kroll process [4, 5]. Distillation is also a more advantageous reprocessing method compared to the old-fashioned liquid–liquid extraction process, called PUREX that is only able to separate single elements such as plutonium or uranium. Additionally, the distillation techniques do not need any further auxiliaries or solvents and are easily extended depending on the required separation accuracy of individual chemical compounds present as chlorides by extending the column stages. The PPU can also be used as a stand-alone unit on storage sites for UNF or directly at any reactor site [5].

2. Reactor design and integration of separation techniques

The Dual Fluid Reactor (DFR) is a heterogeneous, fast-neutron reactor with two separated liquids, one for undiluted molten fuel and the other liquid lead as a metallic coolant [1, 2, 6]. There are two variants of the DFR: one dealing with metal chlorides and another using metallic fuel [6]. In Fig. 1, a concept of the integrated PPU-DFR facility is presented.

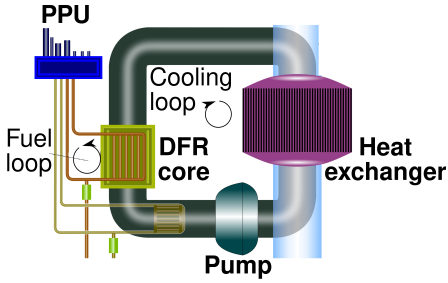

Fig. 1. The DFR fuel and cooling loops for a 1500 MWe DFR.

The reactor design is very simple and consists of three important units: the reactor core where liquid metal chlorides are flowing through the SiC ceramic tubes and are cooled by surrounding liquid lead; the main heat exchanger used for the heat transfer and the PPU which separates the metal chloride compounds. Detailed information on the design can be found in Refs. [1, 2, 6].

3. The pyroprocessing unit (PPU)

The PPU consists of distillation columns for volatile, low and high boilers and crystallizers. In NuDest technical report [5], the PPU is augmented by a head-end process for the partitioning of UNF from nuclear power plants which can be deployed on site. Here, we present a study of the Pyroprocessing Unit for the treatment of UNF by P&T with the DFR. The corresponding flow-chart is depicted in Fig. 2.

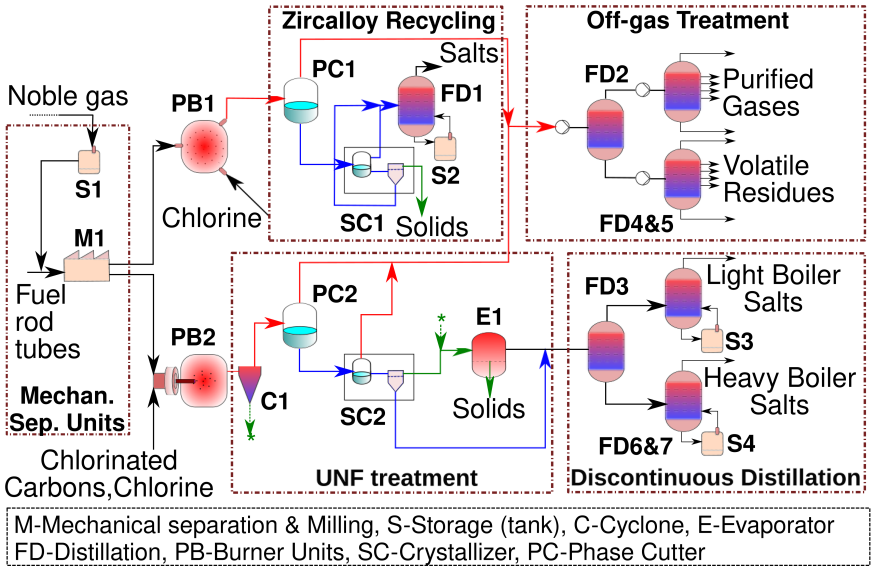

Fig. 2. Simplified flow chart of the Pyroprocessing Unit (PPU).

The calculations have been carried out for a facility reprocessing 1000 t/a of UNF which corresponds to about 1900 standard uranium fuel rod tubes of the Pressurized Water Reactor (PWR) type [7]. The separation accuracy should be a few ppm for most compounds. Besides the fuel rod content, the cladding material also has to be removed and recycled. If a 5 m long and 800 kg heavy fuel rod tube is assumed [7], a total mass of 505 t/a cladding material must be processed. From the point of view of process technology, all the process quantities and flows for separation are such small that discontinuous distillation operations are applied to achieve the assumed accuracy [8–10].

The entire separation process (Fig. 2) is divided into two separate subprocesses: first for the crushed UNF treatment (lower part of the diagram) and second for the zircalloy recycling (upper part). In both sub-processes, chlorination units are initially required for chlorination. The off-gas is separated in phase cutters and is recycled in an off-gas treatment module [5]. In

the following crystallizers of both sub-process cycles, non-chlorinated residual particles in the form of oxide ceramics as well as precious metals are filtered out of the melt.

After the removal of all gaseous and solid compounds (subsequently designated as head-end-pyro-processing in the PPU) from the melt, distillation is used as the main separation step in this pyrochemical separation process. For the crushed fuel head-end-pyro-processing, a cyclone and an evaporation unit can be used to separate most of the solids very early in the process. After crystallization, superficially adhesive residues of chlorides are present on the surface of the particles. Therefore, an evaporator is used for removal of any remaining chlorides on the surface. After head-end-pyro-processing, discontinuous distillation techniques are used to obtain pure molecular substances, especially of actinides and fission products. It is required for the nuclear fuel process to fractionate the mixture into two fractions of light volatiles and less volatile chemical components, because the control technology process is much simpler and requires smaller discontinuous distillation columns. It follows that a narrow temperature range of the two following distillation columns is more easily adjusted. In the case of the distillation of zirconium chlorides in the zircalloy recycling sub-process, only one single discontinuous distillation column is sufficient for desired product purity. The calculation results for the continuous distillation of the chlorinated fuel are presented in Fig. 3, assuming the validity of Raoult's law and ideal mixing behaviour of the liquid. The fractionating distillation column would contain

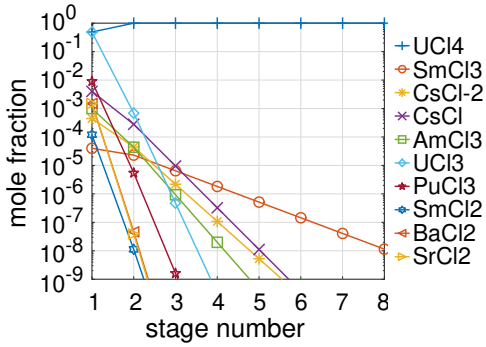

Fig. 3. Continuous fractionated distillation column results.

five stages. It can be seen that uranium tetrachloride is able to be decontaminated and can be drawn off nearly purely just after only 5 to 6 stages. The fine separation is performed in subsequent discontinuous distillation columns which requires less than 12 stages. Furthermore, the separation in the distillation columns as well as in crystallizers depends significantly on the yield of the chlorination process. Figure 4 shows the simulation results

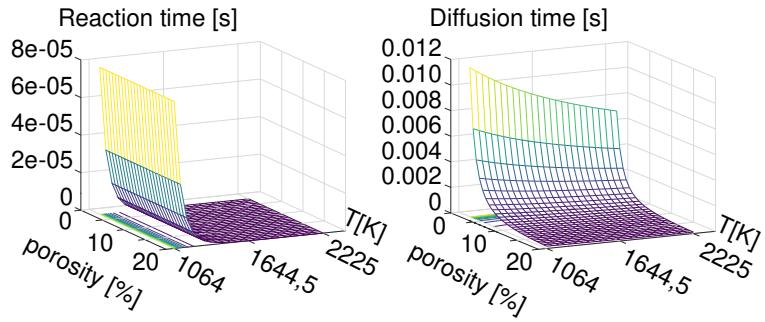

Fig. 4. Timescale: Simulation results for the chlorination of UNF on basis of uranium tetrachloride.

of the chlorination with carbon tetrachloride/chlorine mixtures for different temperatures where combinations of shrinking-core models and unified product-layer models [11–13] were used merely for a chemical reaction. Herewith the diffusion of carbon tetrachloride is the limiting time factor whereas reaction times are negligibly small. Depending on the porosity of the solid particles and on the necessity of oxygen desorption, which might be a problem especially for uranium dioxide [14, 15], high temperatures have to be chosen just about 2000 K and overpressure is required. Due to short reaction times (see Fig. 4), a burner with an electric plasma discharge flame to crack all the compounds and to recombine the elements is particularly suitable for chlorination procedure. Compared to chlorination in nuclear fuel processing, a smart chlorination in the zircalloy recycling is achieved in a smart burner unit as described in Refs. [5, 16] or, alternatively, in a fluidised bed reactor [17, 18]. For real process conditions, burner units are preferred. Since the PPU is an integral part of the DFR power plant, the DFR core can be simply connected to the thermal separation modules of the PPU (see Ref. [5]).

4. Conclusions

The results of simulation calculations show that on-line reprocessing of the nuclear fuel applying a simple pyroprocessing unit (PPU) combined with the Dual Fluid Reactor (DFR) is very effective and easy to handle. In addition, mixtures of different compositions are fractionated into the single molten salt compounds, and the separation precision is adjusted by the number of stages in the respective distillation column.

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