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A High Voltage Head-End Process for Waste Minimization and Reprocessing of Coated Particle Fuel for HTR

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A High Voltage Head-End Process for Waste Minimization and Reprocessing of Coated Particle Fuel for High Temperature Reactors

Executive summary

This paper describes results from experimental proof-of-principle tests to liberate coated particles and to fragment them. These tests were performed with surrogate fuel and coated particles in commercially available high voltage (HV) discharge installations. The product to be fragmented is poured into a vessel which is filled with water. An electrode delivering repetitive HV discharges under water is plunged into the vessel creating shock waves. Coated particles were liberated intact from their matrix and subsequently they were fragmented to sub-millimeter dimensions. The energy consumption is low and the installation is suitable for a hot cell environment and industrially relevant material streams. A process is proposed for the separation of the coated particles from the fuel element matrix. After further fragmentation of the coated particles, the fragmentation product can be directly fed into classical aqueous reprocessing.

Revisions

Rev.	Date	Short description	Author	Internal Review	Task Leader	WP Leader
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01	30/04/2010	First issue	Fuetterer, JRC			
02		2nd Issue				

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1 Introduction

Typical High Temperature Reactor (HTR) fuel consists of ∅0.5 mm oxide fuel kernels coated with successive layers of carbon and silicon carbide to form ∅1 mm so-called TRISO coated particles. Several thousand of these coated particles are embedded in a graphite matrix and brought to different macroscopic sizes and shapes. The most common are the fuel "pebble" (∅60 mm), cf. Fig. 1, Table I, and the fuel "compact" (a cylinder of typically ∅½ in × 2 in). Depending on fuel design, HTR fuel may therefore contain carbonaceous matter in excess of 95%. While the resulting thermal inertia yields excellent HTR safety performance, this fuel type also incurs large volumes of waste.

Reprocessing of coated particle fuel from HTR is useful in many cases to maximize the use of the fuel and/or for the separation of kernels and coatings in view of waste volume reduction. It is even mandatory for the U-Pu (fast reactor) and for the Th-U fuel cycle as well as for the incineration of minor actinides in these reactors. Coated particle fuel is characterized by its high resistance against mechanical impact and chemical attack, which makes it safe, rather proliferation resistant and suitable for direct disposal, yet difficult to reprocess. As a head-end process for reprocessing, there were attempts in the past to fragment coated particles, e.g. by mechanical grinding or pneumatic projection against a hard wall. However, these methods have a number of specific shortcomings.

- low efficiency and high energy consumption;
- possibly use of pressurized gases;
- production of toxic or explosive dust, safety problems;
- high level of noise or vibration;
- pollution of the matter to be fragmented by non-negligible quantities of abrasion products;
- high wear and tear from abrasion, limited lifetime, high capital and operation costs.

At the same time, HTR and other reactor types use large quantities of graphite, carbides or other ceramic material as matrix, moderator or reflector material. After its useful lifetime, this material is contaminated and activated, and, in particular due to its C-14 content, would consume excessive storage space if disposed of directly. Also, graphite of sufficient quality is a scarce resource, in particular when considering possible deployment of a relatively large number of HTR.

Two approaches can be envisaged:

1. Liberation of the coated particles from the graphite matrix and disposal in separate waste repositories. This makes sense and will reduce costs because matrix and particles have a different waste classification.
2. a) Liberation and separation of the coated particles from the graphite matrix, disposal of graphite. b) Subsequent fragmentation of the fuel particles to enable reprocessing.

This work successfully demonstrated feasibility of both the liberation and coated particle fragmentation process.

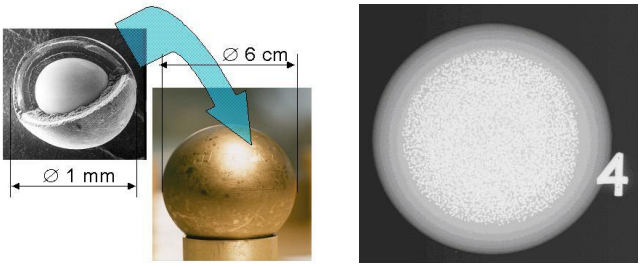

Fig. 1. Composition and X-ray of a typical HTR fuel pebble

TABLE I

Typical characteristics of coated particles and pebbles produced by NUKEM

Coated Particle	
Particle batch	HT 354-383
Kernel composition	UO_2
Kernel diameter [μm]	501
Enrichment [U-235 wt.%]	16.75
Thickness of coatings [μm]: buffer inner PyC SiC outer PyC	 92 38 33 41
Particle diameter [μm]	909
Pebble	
Heavy metal loading [g/pebble]	6.0
U-235 contents [g/pebble]	1.00 ± 1 %
Number of coated particles per pebble	9560
Volume packing fraction [%]	6.2
Defective SiC layers [U/U_{tot}]	7.8×10^{-6}
Matrix graphite grade	A3-3
Matrix density [kg/m³]	1750
Temp. at final heat treatment [°C]	1900

2 State of the art

Liberation of coated particles from fuel elements and recovery of the fuel has been attempted with various, mostly mechanical, techniques in the past [1], [2], [5], [6] and the issue is globally addressed in [3], where the authors state that mechanical methods are not feasible for this purpose although the authors of [5] had successfully tested the separation of particles from their matrix by brushing. The French Atomic Energy Commission CEA investigated how graphite fragmentation to grain sizes corresponding to coated particle fuel could be achieved using the pulsed HV discharge technique. Reference [3] Fig. 3 and reference [7] Fig. 2 show the results of tests with graphite where the grain size distribution is a function of the number of applied pulses. This was confirmed by our own work where in addition we have measured the energy consumption. The pulsed HV discharge technique can thus be recommended for fragmentation of reactor graphite used in moderator or reflector blocks as a head-end process before decontamination and recycling of this graphite to minimize graphite waste streams.

However, the aspect of separating the matrix material from the coated particles and the fragmentation of the coated particles with consecutive separation of coatings and fuel kernels by sieving was not addressed.

Contrary to the limited information given in [3], the patent [8] proposes a complete process for the separation of coated particles from their matrix material, for the fragmentation of the coated particles and for the separation of the fuel kernel from coatings and matrix material. The process parameters are given as well as the required electrical energy.

3 Experimental work

The principle of the used HV fragmentation method is described elsewhere (e.g. in [4]). The product to be fragmented is placed inside a container with electrically insulated side walls (Fig. 3, Fig. 4). The bottom of the container serves as the ground electrode. The container is filled with water, and another electrode is inserted into the water from the top. A HV discharge is sent across the water. The pathway of the discharge across the water creates a cavitation shock wave. At sufficiently short pulse rise times, the discharge preferentially crosses the material to be fragmented disintegrating it along the grain boundaries. Important process parameters that determine the feasibility of this method are discharge voltage, pulse rise time, electric arc length, reaction volume, electric conductivity of the water and number of discharges.

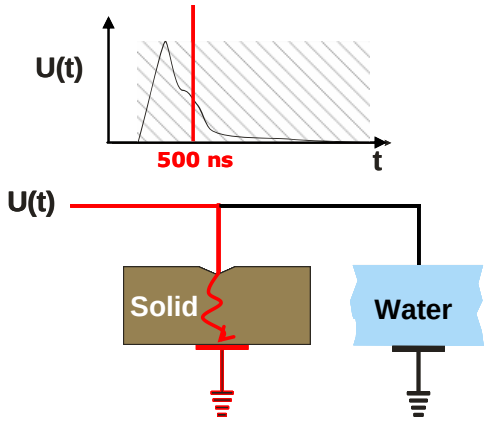

Fig. 2. Principle of HV discharge fragmentation

The discharge is created in a HV pulse generator (“Marx generator”) which is connected to a HV power supply. Pulse generator and process vessel are protected in a Faraday cage.

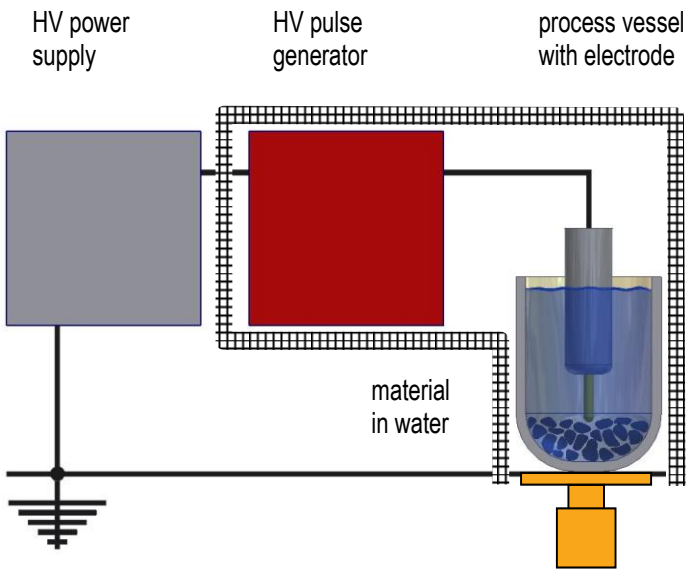

Fig. 3. Schematic of installation

Fig. 4. Fuel pebble (dummy) in reaction volume

A commercially available machine for this purpose is shown in Fig. 5.

Fig. 5. Commercially available fragmentation machine ("selFrag lab", height about 2 m)

The experiments described here were partly performed in an existing installation of a major European research center, partly in an experimental facility at selFrag AG, Switzerland.

The following materials were successfully fragmented: pieces of new reactor grade graphite of different types, shapes and sizes, surrogate fuel particles and fuel pebbles containing surrogate fuel particles. Real fuel and/or irradiated material were never used in our tests because of the required licenses and installations.

4 Results

The process proposed here envisages liberation of the coated particles from the fuel element (e.g. a "pebble") with consecutive fragmentation or complete removal of the coatings to make the fuel kernel accessible to chemical dissolution for further reprocessing. These process steps enabling full recovery of the fuel were experimentally verified.

1. Liberation of coated particles from the fuel element
2. Fragmentation of the coated particles

4.1 Step 1: Liberation of coated particles from the fuel element

First tests were performed with blocks of pure reactor grade graphite without coated particles. Later tests contained graphite pieces and coated particles together, and final tests fragmented fuel pebbles containing surrogate coated particles. The material was fed into the water-filled reactor of the volume V1 with a HV electrode on the top and a grounded sieve (hole diameter approx. 3 mm) on the bottom. The pulse generator repeatedly charged the electrode with voltages of 200 – 400 kV, the maximum discharge currents were of the order of 10 kA. The distance between the HV electrode and the sieve was chosen such that the electrode had a distance from the fragments of approx 1 cm and that discharges through the water were obtained. The discharges then caused electro-hydraulic shock waves in the water, which fragmented the matrix graphite. By correctly choosing volume V1, damage to the fuel particles could be avoided with these discharge parameters. For energy-optimized fragmentation, the hole diameter in the sieve was adapted to the size of the fuel particles. If the holes are too large, the fuel

particles remain in the matrix material, if they are too small, the matrix material is fragmented to unnecessarily fine grains. For the used test material, sieve holes of 3 mm were selected such that the maximum of the statistical fragment size distribution became approx. 1.5 mm. Thus, all fuel particles were separated from the matrix and were contained in the sieve fraction > 1mm, once the material had fully passed the sieve.

As a final test, the fuel element ("pebble") shown in Fig. 4 was submitted to HV discharges. Fig. 6 shows the result after only 3 seconds (15 pulses) where the pebble had broken up and the coated particles were visible.

Fig. 6. Crushed fuel pebble after 3 seconds (15 pulses)

After 1 minute of exposure (300 pulses) all coated particles were liberated. Owing to their mechanical resistance, all coated particles remained intact and could be separated by sieving.

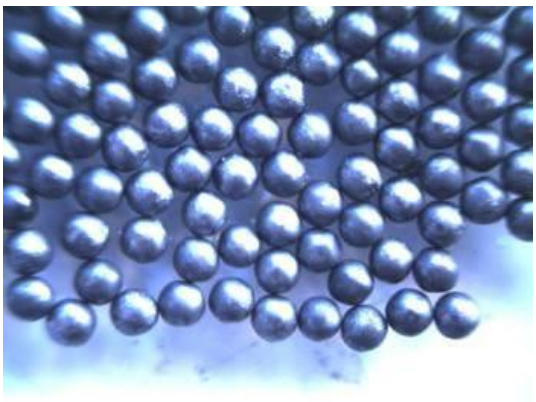

Fig. 7. Liberated coated particles (∅1 mm)

4.2 Step 2: Fragmentation of coated particles

Assuming that the fuel kernels (the central part of the coated particles) remain intact (to be confirmed with irradiated fuel), the following two options are possible before entering the next process step:

Option a): The process stream can be reduced by passing the product from process step 1 through a sieve, the hole diameter of which is slightly smaller than the fuel particles. The smaller fragments are thus separated from the particles. Only the particles will then be fed into process step 2.

Option b): Full separation of the fuel from the matrix graphite and the coatings can be achieved by separating the entire product from process stream 1 in a sieve with a hole diameter which is slightly smaller than the kernel size (here 0.5 mm). Then the fuel remains fully on top of the sieve, together with some fraction of the matrix graphite.

The process streams obtained from Option a) or b) can be treated with HV discharges. There are again two options:

Option c): HV discharges over a closed cup or

Option d): HV discharges over a sieve with a hole diameter slightly smaller than the fuel kernels (here 0.4 mm);

The electric parameters can be identical to those mentioned above, however a reaction volume V2 was used, which is significantly smaller than V1. The electrode can plunge into the material fragments to be treated. The volume reduction yields an increased power density of the created shock waves, which make the coatings burst and which removes them from the fuel kernel. Contrary to the oxide kernel, the coating shells and possibly remaining matrix material are further fragmented. Once coatings and matrix material are sufficiently fragmented, they will drop through the sieve (option d) or, when using a closed cup (option c) can be sieved in a consecutive step.

Real irradiated nuclear material may behave differently than the used unirradiated surrogate particles. Therefore, a feasible default option would be to dissolve the product from option c) directly in nitric acid to maximize the recovery of nuclear material including the one that may have penetrated into the coating shells.

Fig. 8 shows several intact particles, while Fig. 9 and Fig. 10 are the result of process step 2.

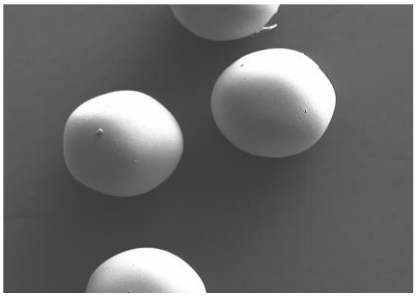

Fig. 8. Intact coated particles

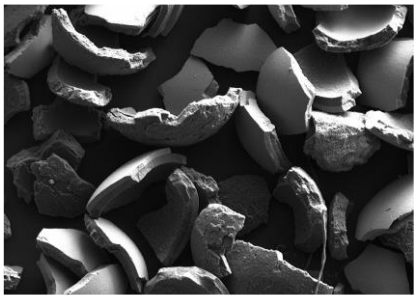

Fig. 9. Separated coating shells

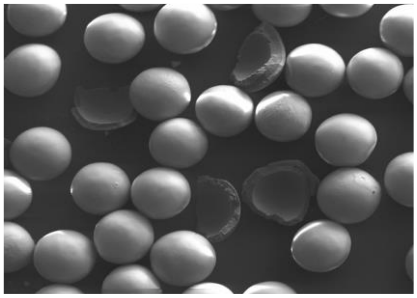

Fig. 10. Fuel kernels separated from their coatings

4.3 Energy requirements

In the used laboratory equipment, the required electric energy for Process Step 1 (liberation of the coated particles) is approx. 2000 – 5000 kWh/t depending on type and shape of the graphite pieces. The measured energy consumption for these tests is deemed conservative as the tests were run with laboratory installations and small amounts of material to be fragmented.

The electric energy required for Process Step 2 (fragmentation of coatings) is approx 8000 kWh/t.

A typical HTR fuel pebble has a mass of approx. 222.5 g and contains 9560 fuel particles with a total weight of approx. 28.8 g.

Assuming a burn-up of 10% FIMA of this fuel pebble, it will have produced 1200 kWhth heat and (assuming a 45% power conversion efficiency) 540 kWhel electricity.

As shown in Table 2, the electric energy for separation and fragmentation corresponds then to a fraction of only between 0.125 and 0.25% of a pebble's lifetime electricity output. In other words, energy consumption is no viability issue for fuel recovery with this technique.

TABLE II

Energy requirements for fragmentation

	Electric energy required
Energy required to separate particles from matrix of a pebble [kWh_{el}]	0.445 – 1.1125
Energy required to fragment all particles of a pebble [kWh_{el}]	0.23
Energy produced by a pebble [kWh_{el}]	540
Energy for reprocessing [kWh_{el}]	0.675 – 1.3425
Energy fraction for reprocessing [%]	0.125 – 0.249

5 Conclusions and future work

This paper shortly described results from experimental proof-of-principle tests to fragment fuel elements and coated particles. They were performed in modified HV discharge installations. A process was tested and patented and is proposed for the separation of ceramic nuclear fuel from its matrix material. Surrogate fuel elements, fuel particles and graphite pieces were fragmented to sub-millimeter dimensions. The energy consumption is low and, despite the need for technical adaptations, the installation seems suitable for a hot cell environment and industrially relevant material streams. A process is proposed for the separation of the particles from the fuel element matrix (e.g. pebbles or compacts). After further fragmentation of the fuel particles, the fragmentation product can be directly fed into classical aqueous reprocessing. The graphite (matrix or moderator) in turn can also be further fragmented to facilitate decontamination and to obtain a powder as the starting product for possible re-fabrication of reactor graphite.

In the experiments performed with surrogate particles, the kernel remained intact. If this can be confirmed also with irradiated material, it would enable full separation of the coatings and

matrix material from the kernels and thus a desirable reduction of process streams. However, this separation of kernels and coatings is not even mandatory as the fuel is usually dissolved chemically, so that a large fracture in the coating is deemed sufficient for this purpose. The recovery fraction of irradiated fuel from fragmented coated particles is yet to be verified experimentally. Because of the higher density of real fuel kernels compared to the used dummies, it can be expected that this method

will also work with nuclear material. Due to the weakening of materials under irradiation, it is probable that the HV discharge techniques works even better with irradiated fuel.

Compared to mechanical methods, the pulsed HV discharge technique features a number of advantages: no dust and low production of undesired fines, high speed, high yield, low energy consumption, easy process control, suitability for relevant material streams (order of tons per hour) and capability to move from batch to continuous operation.

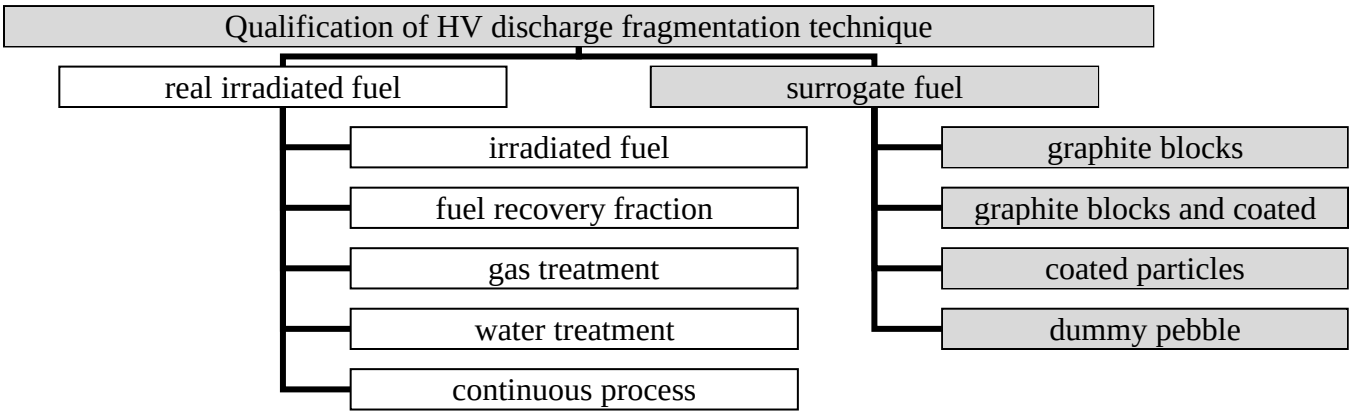

Fig. 11. Schematic of performed (shaded) and future work

For establishing full viability of the process, further tests will have to include use of irradiated fuel. The fuel recovery rate from the fragmentation product (dissolution in acid) has to be measured and optimized, the equipment has to be adapted to hot cell conditions and radioactive gas and process water treatment has to be integrated into the system design. A schematic indicating already performed and future work is shown in Fig. 11.

The presented results demonstrated the proof of principle and can be considered a significant step towards practical feasibility of HTR with symbiotic or closed fuel cycles and for minimization of their waste streams.

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