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- Graphite and HTR compact fragmentation tests using pulsed currents -

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Document title

Graphite and HTR compact fragmentation tests using pulsed currents

Executive summary

This memo presents the initial results of a parametric study into the pulsed current fragmentation of carbonaceous materials. The tests were conducted using the PUMA pulsed current test facilities at CEA/Marcoule.

Pulsed current fragmentation is being considered as a head-end phase during the reprocessing of HTR fuels and fourth generation reactor fuels (gas FNR). Preliminary tests conducted at the Thalés site in Limours and at CEA/DAM in Moronvilliers have shown the potential of this process for fragmenting graphite and the coating on TRISO particles.

In order to minimise the number of tests involving HTR fuel compacts, this parametric study was conducted using graphite samples similar in geometry and mass to the compacts. The difference was the absence of TRISO particles.

The different parameters used in the study were:

- ✓ Pulse energy (350 J and 500 J)
- ✓ Frequency (maximum 10 Hz) and number of pulses
- ✓ Conductivity of the medium (industrial water vs. deionised water)
- ✓ Reactor load level (mass of material to be fragmented per test)

The results revealed two significant parameters - pulse energy and mass of material to be processed. The ideal situation is to conduct the process using the highest possible pulse energy in order to improve fragmentation performance. This is consistent with the results obtained using the CEA/Moronvilliers pulse generator (3.4 kJ).

If the reactor contains a sufficiently large load, the material will self-fragment. This is a known occurrence in traditional crushing techniques. However, when using a conductor material such as graphite, an increase in the mass to be processed has an effect on the conductivity of the suspension medium, and therefore on the discharge voltage and finally on the pulse energy. A compromise is therefore needed, based on the volume of the reactor.

At the same time, a method involving successive cycles of fragmentation/separation by screening can improve the fragmentation process in terms of fragmentation performance and minimising the production of fines.

After the parametric study using graphite samples to determine the most important parameters, tests were conducted using compacts. For compacts containing TRISO particles (20% by vol.), the fragmentation process required approximately half as much energy and the amount of fines produced was below 11%. The fragmentation occurred mainly at the graphite/particle interfaces. The TRISO particles remained whole with the exception of some whose coating was cracked and chipped. These results were promising for further tests.

Revisions

Rev.	Date	Short description	Author	Internal Review	Task Leader	WP Leader
01	11/11//2008	First issue	Cardinal CEA	Ph. DEHAUDT Chef du SDTC		

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Introduction

This memo presents the results of a parametric study into the pulsed current fragmentation of carbonaceous materials. The tests were conducted using the PUMA pulsed current test facility that was installed in mid-2008 at the G1 site in Marcoule.
Pulsed current fragmentation is being considered as a head-end phase during the reprocessing of fourth generation reactor fuels (gas FNR) and HTR compacts. Preliminary tests conducted at the Thalés site in Limours and at CEA/DAM in Moronvilliers have shown the potential of this process for fragmenting graphite and the coating on TRISO particles.
The main aim of these tests was to provide the necessary elements for understanding the processes that take place.

Materials and Method

The basic test involved the use of a high voltage generator fitted with a receptacle for the material to be processed (test reactor in a glove box (figure 1). The generation of high voltage pulses via electrical discharges was used first to disintegrate inactive materials, before being tried on uranium fuels.

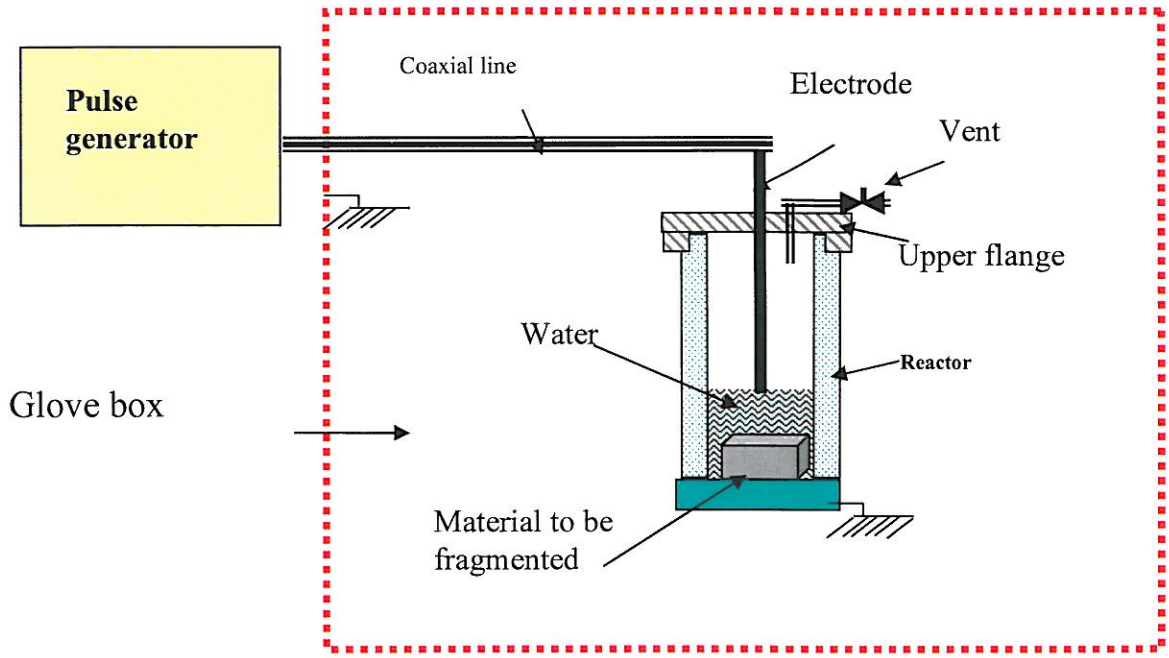

Figure 1. Skeleton diagram of the test reactor set-up

2.1 Test reactor

The test reactor was placed in a glove box. The working volume of the reactor was about 0.8 litres. A flat-point electrode system was used (fig. 1). The upper electrode was a 10 mm diameter rod with a rounded outer surface (demi-sphere). The lower electrode was a 60 mm diameter disc. This electrode carried the sample to be fragmented.

2.2 High voltage pulse generators

The pulsating current slowly stores up electrical energy (capacitive storage for PUMA), then releases it as a very quick surge, by commutation (compressed air switch).
A 300 kV generator was used:

- ✓ Rise time: 20-30 ns
- ✓ Pulse energy: 500 J
- ✓ Maximum frequency: 10 Hz

2.3 Method

The sample(s) were placed in the bottom of the reactor and covered with water. The area was inaccessible during the test. The current and voltage readings from the reactor were displayed on a fast TDS 5034 oscilloscope with segmented memory.
At the end of the test, the capacitive platform was fully discharged and short-circuited. The sample fragments were collected by filtration. They were then screened into different particle-size classes and weighed.

2.4 Test samples

The samples of carbonaceous materials were used:

- ✓ G2-type graphite: This graphite was used as a moderator in the G2 reactor
- ✓ Inert HTR compact: These products are manufactured by CERCA. Some contain about 20% by volume of TRISO particles, with a zirconium core (fig. 2).

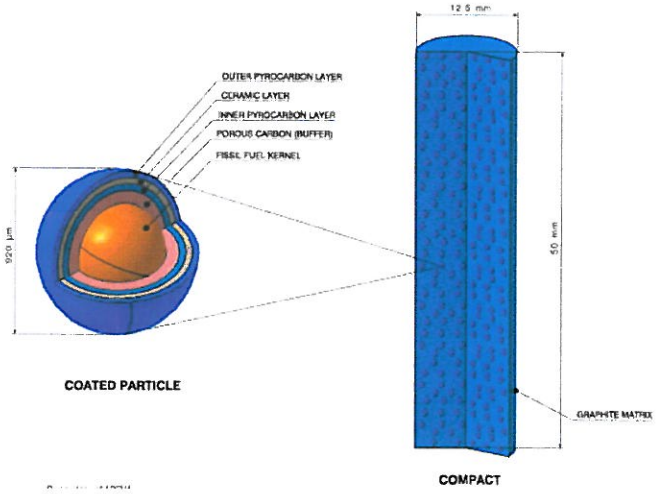

Figure 2. HTR compact

2.5 Test design

In order to keep the number of tests using HTR compacts to a minimum, given the small quantity available, the parametric study was conducted using G2 graphite samples.
The geometry and mass of the graphite samples were as close as possible to those of the compacts. The difference was the absence of TRISO particles.

The different parameters that were investigated during the parametric study were:

- ✓ Pulse energy (350 J and 500 J)
- ✓ Frequency (10 Hz maximum) and number of pulses
- ✓ Medium: For these tests the reactor always contained water, but with different levels of conductivity. The tests were conducted using industrial water (IW) and deionised water (DW).
- ✓ The mass of material to be fragmented per test.

The aim of these tests was to determine which parameters would affect the fragmentation of the graphite and, eventually, the HTR compacts. The diameter of the TRISO particles was approximately 700 µm.
The initial aim was to separate the graphite from the particles. The idea was to maximise the quantity of particles produced between 500 µm and 1 mm in size, so that during the first phase the quantity of fines would be limited and whole TRISO particles could be obtained. The second phase would involve further disintegration of the coating using pulsed currents.

2.6 Summary of test conditions

Test No.	Type of material	Number of samples	Type of liquid in the reactor	Energy/pulse in J	Frequency	No pulses/g material	Number of pulses
1	Graph G2	1	IW	500	10	10	158
2	Graph G2	1	IW	500	10	20	300
3	Graph G2	1	IW	500	10	50	837
4	Graph G2	1	IW	500	10	100	1504
5	Graph G2	5	IW	500	10	10	867
6	Graph G2	5	IW	500	10	5	405
7	Graph G2	1	DW	500	10	50	845
8	Graph G2	1	DW	500	10	20	354
9	Graph G2	1	DW	500	10	100	1843
10	Graph G2	10	IW	500	10	5	836
11	Graph G2	1	IW	500	5	20	340
12	Graph G2	5	IW	500	5	20	1869
13	Graph G2	>1mm Smpl 1	IW	500	10	10	145
14	Graph G2	>1mm Smpl 5	IW	500	10	10	562
15	Graph G2	>1mm Smpl 13	IW	500	10	10	132
16	Graph G2	>1mm Smpl 14	IW	500	10	10	413
17	compact	1	IW	500	10	15	152
18	compact	>1mm	IW	500	10	40	244

		Smpl 17					
19	Graph G2	1	IW	350	10	20	388
20	Graph G2	1	IW	350	10	100	1730
21	compact	>1mm Smpl 18	IW	500	10	40	147
22	Graph G2	1	IW	350	10	20	346
23	Graph G2	5	IW	350	10	10	904
24	Graph G2	1	DW	350	10	100	1866
25	Graph G2	5	DW	350	10	10	918
26	Graph G2	>1mm Smpl 19	IW	350	10	20	363
27	Graph G2	>1mm Smpl 22	IW	350	10	20	325
28	Graph G2	>1mm Smpl 25	DW	350	10	10	767
29	Graph G2	>1mm Smpl 19	IW	350	10	10	840
30	Graph G2	>1mm Smpl 26	IW	350	10	20	334
31	Graph G2	>1mm Smpl 27	DW	350	10	20	290
32	Graph G2	>1mm Smpl 23	IW	350	10	10	758
33	Graph G2	1	IW	500	10	15	249
34	Graph G2	1	IW	500	10	60	1045
35	Graph G2	5	IW	500	10	30	2335
36	Graph G2	1	IW	500	5	10	173
37	Graph G2	>1mm Smpl 10	IW	500	10	5	
38	Graph G2	>1mm Smpl 33	IW	500	10	40	

Results and interpretation

In order to be able to draw comparisons between the different fragmentation tests, a unit defined as the ratio between the number of pulses of electrical discharge (I) and the mass of the sample (g) was used - I/g.
Results are given in the form of histograms representing the mass of each particle size class as a percentage of the total mass of the sample.
Four particle size classes were used:

- ✓ Ultrafine particles: $\varnothing < 45$ µm
- ✓ Fine particles: 45 µm $< \varnothing <$ 500 µm
- ✓ Particles that meet the liberation criteria for TRISO pellets: 500 µm $< \varnothing <$ 1 mm
- ✓ Oversize particles (requiring further fragmentation): $\varnothing >$ 1 mm

After the fragmentation process, we assessed changes in and the distribution of the particle sizes that had been produced, in order to determine the success of the fragmentation.
The average matter loss during the test and the screening phase was approximately 0.5%.

3.1 Parametric study

3.1.1 Influence of total energy

In this series of tests, the quantity of energy was defined using the ratio I/g, i.e. the number of pulses per gram of sample. This ratio (I/g) varied during the tests from 10 to 100. The energy of each pulse was 500 J.
The reactor contained one single sample of G2 graphite per test. The samples were all very similar in terms of geometry and mass. Figure 3 shows a breakdown by particle size class for the different energy values (I/g).

- ✓ The quantities of ultrafines and fines varied depending on the I/g ratio. For example, the percentage by mass of fines was 2% for 10 I/g, 7% for 20 I/g, 13% for 50 I/g, and 21% for 100 I/g.
- ✓ For the particle size class 500 µm $< \varnothing <$ 1 mm, the results were similar for energy levels ranging from 10 I/g to 50 I/g: 3.5% for 10 I/g, 7.9% for 20 I/g, and 14.6% for 50 I/g.

  For higher I/g values, the ratio between energy and mass for the particle size class 500 µm $< \varnothing <$ 1 mm fell. For 100 I/g, the percentage by mass was only 18.9%.

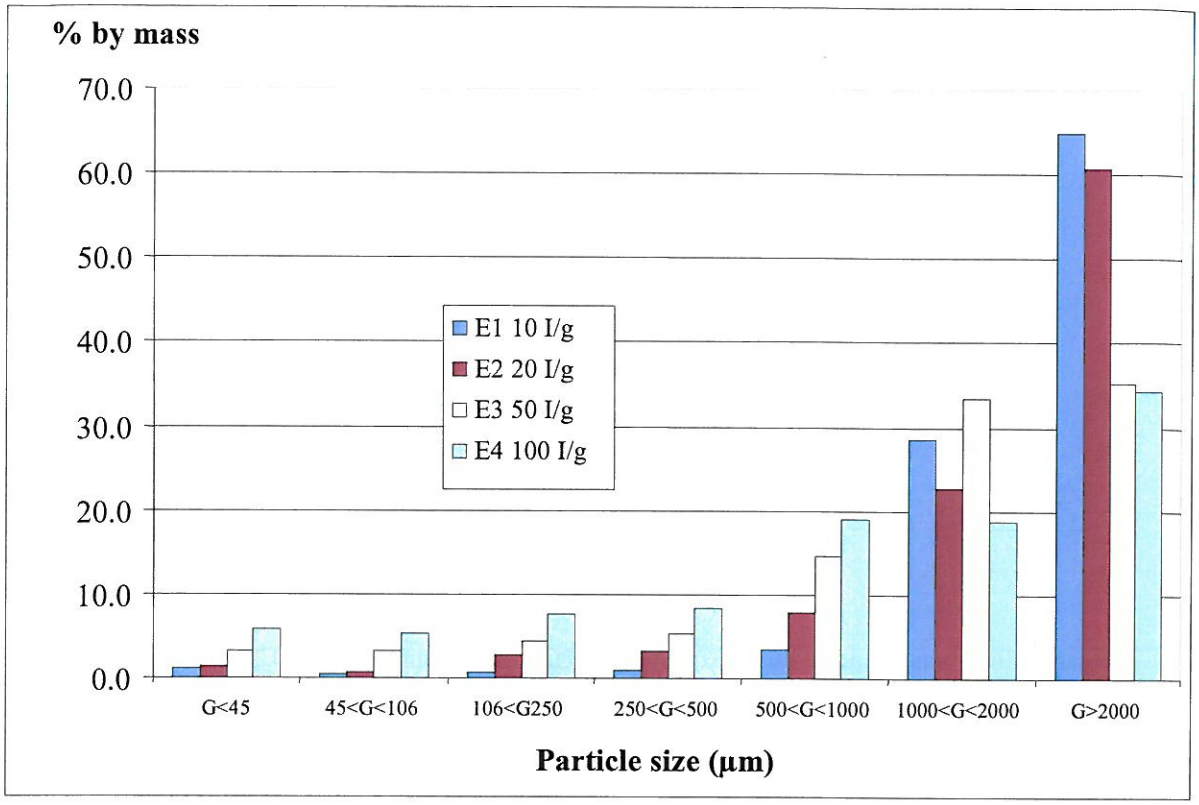

Figure 3. Influence of the I/g ratio

In order to maximise the proportion of particles 500 µm < ∅ < 1 mm compared to fines and ultrafines, the optimum I/g ratio seemed to fall at 50 I/g.
However, 60% of the sample (by mass) was larger than 1 mm and needed further fragmentation. One solution would be, after the screening and separation process, to repeat the fragmentation on the fraction larger than 1 mm. This is covered in point 3.2.

3.1.2 Influence of energy per pulse

Due to the equipment design it was only possible to test two pulse energy values, 350 J and 500 J.
In figure 4, the tests should be read in pairs: E4 and E20; E2 and E19; E5 and E23.
Logically, fragmentation performance should correlate to the amount of energy delivered by each pulse i.e. the higher the pulse intensity, the higher the mass of fragmented material.
However, fragmentation was not proportionate to the amount of energy discharged by a pulse. The samples were proportionately more damaged by high energy pulses. As regards the particle size class 500 µm < X < 1 mm, 19% of the sample by mass fell into this class at 500 J (E4), and 9% at 350 J (E20). When pulse energy was multiplied by a factor of 1.4, the quantity of fragmented material increased by 2.1. Graphite is a fragile ceramic material. The level of fragmentation was all the higher as the pulse energy increased.

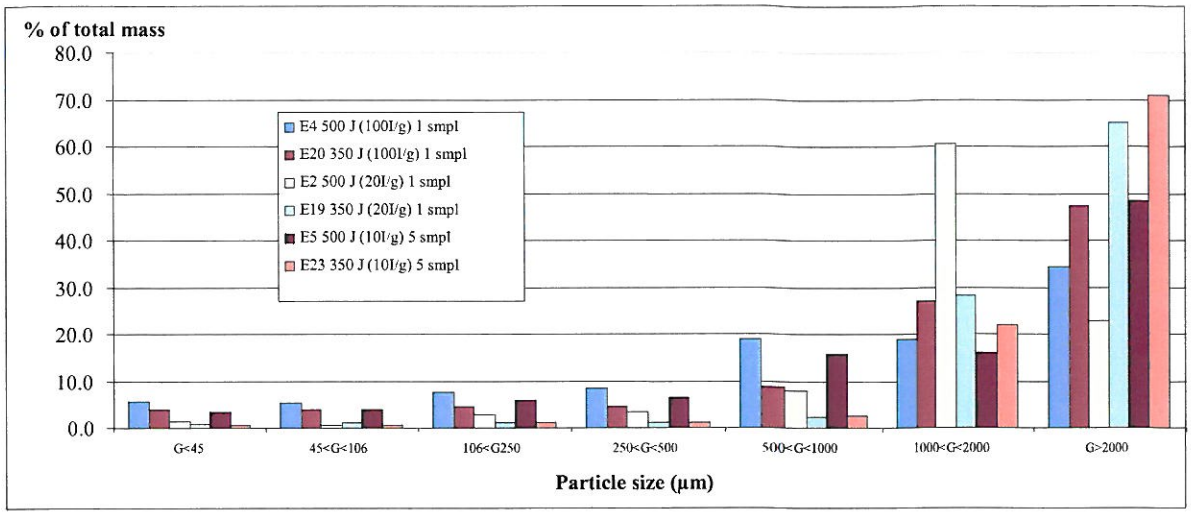

Figure 4. Influence of level of energy per pulse

Reactor load was found to be an extremely influential parameter (tests E2 and E5). This point will be discussed in more detail in point 3.1.5. An increase in the mass of material to be fragmented, coupled with a pulse of high energy, significantly increased fragmentation.
However, whatever the pulse energy, the quantity of fines and ultrafines that was produced remained proportionate to the quantity of material that fell into the particle size class 500 µm < X < 1 mm.
For the two particle size classes larger than 1 mm, significant variations were found between the tests. During this test phase we could find no explanation for this distribution which, it would appear, was independent of the percentage by mass.
In conclusion, the higher the pulse energy the higher the fragmentation yield. It would be interesting to determine the results produced by a high pulse energy, in line with the results obtained using the pulse generator at CEA/Moronvilliers (3.4 kJ) [1]. After ten pulses (3.4 kJ), 70% of the sample mass had been reduced to fragments smaller than a 1 mm mesh. In our tests, after approximately 1,500 pulses (500 J), 55 % of the sample mass had been reduced to fragments smaller than a 1 mm mesh.

3.1.3 Influence of pulse frequency

The reactor had no observation port and therefore it was not possible to see the electric arc or how the graphite behaved. When an pulse was discharged, the shock wave generated in the water created a suspension of the fragments of graphite inside the reactor.
The aim of this series of tests was to discover whether a fall in frequency allowed the graphite to decant inside the reactor between two pulses and settle near to the electrodes, ready for the following pulse.

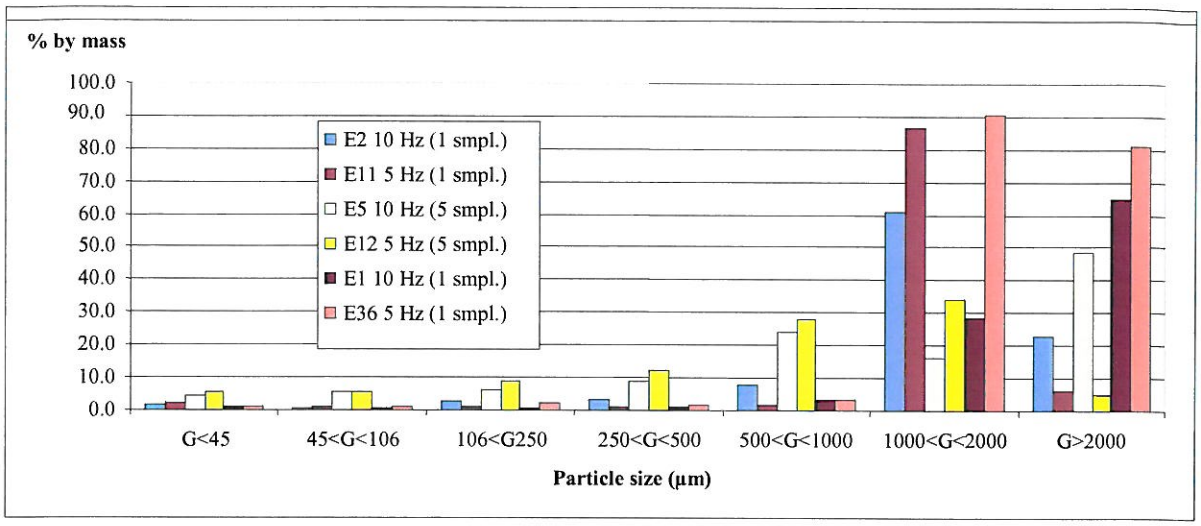

Figure 5. Influence of pulse frequency

The maximum operating frequency of the equipment was 10 Hz. Tests were conducted at 5 Hz and 10 Hz. The tests should be read in pairs: E2 and E11; E5 and E12; E1 and E36.
Our tests did not reveal any trends (fig. 5). In other words, either the time lapse between two pulses was not enough to allow the graphite to decant, or this parameter had no effect on our set-up since:

- ✓ The reactor's working volume was too small (0.8 litres)
- ✓ The reactor was cone-shaped, in order to collect the material on the flat electrode.

The material was therefore almost certain to be confined near to the electrodes.
Since there was no change in results, the maximum frequency of 10 Hz was used thereafter in order to minimise the fragmentation time.

3.1.4 Influence of the liquid medium inside the reactor

Two media were tested: industrial water (IW) and deionised water (DW). The aim was to estimate the influence of medium conductivity on the properties of the electrical discharge and ultimately on the success of the fragmentation.
Several comparative tests (IW vs. DW) were carried out, varying the I/g ratio. In general, the breakdown voltage in the reactor for the tests using deionised water was slightly higher. This was predictable since as the conductivity of the water fell, it was normal for the breakdown tension of the electrodes to increase.

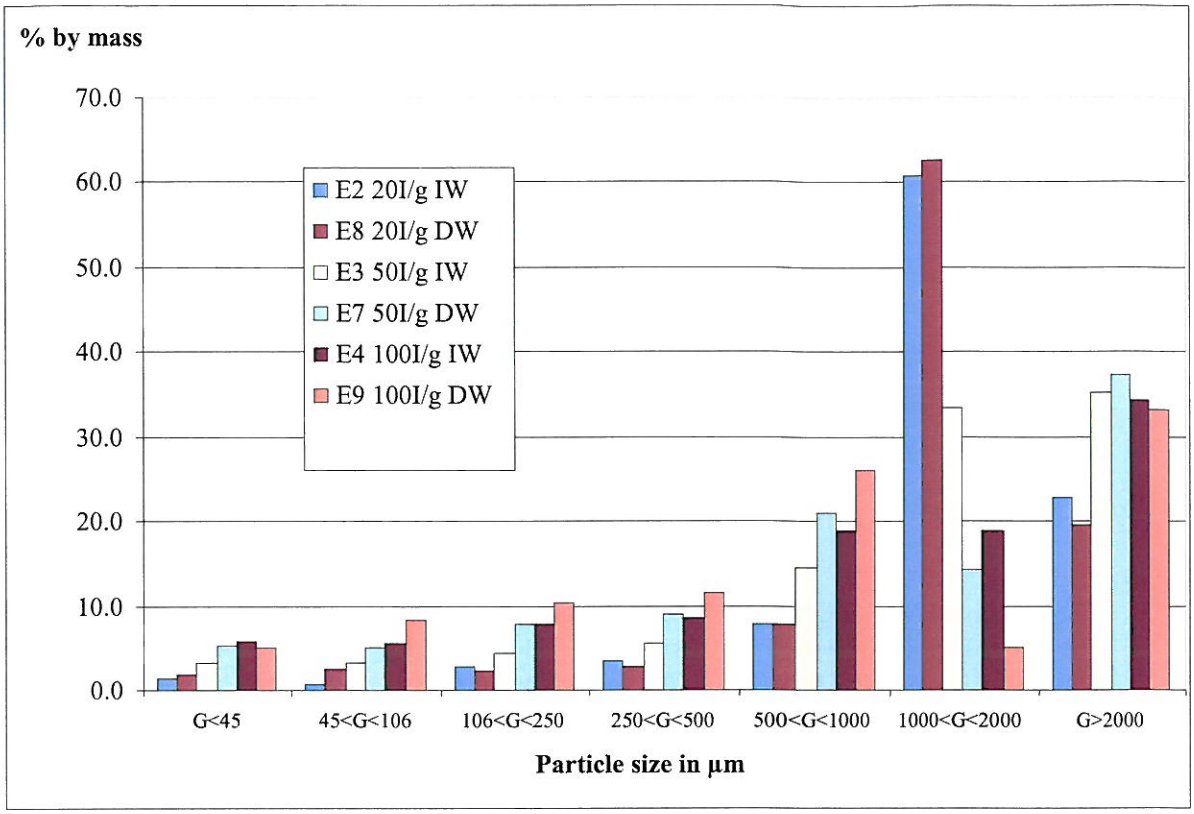

Figure 6. Influence of the liquid medium

Therefore, because the electrode breakdown voltage increased there was indeed an increase in pulse energy. For the same I/g ratio, the particle size class 500 µm < X < 1 mm was greater in terms of mass for the tests using deionised water than for the industrial water tests (fig. 6 - compare tests E3-E7 and E4-E9). The production of fines and ultrafines was still proportionate to the size by mass of the class 500 µm < X < 1 mm.
Deionised water significantly improved fragmentation yield. However, in order to retain the low conductivity the medium had to be changed after each test. In fact, the breakdown voltage tended to decrease throughout the tests. Having spoken with the University of Pau, their experiments have shown that discharges ionise the water and therefore lead to an increase in conductivity, thereby reducing the breakdown voltage.

3.1.5 Influence of reactor load

The graphite samples were placed around the bottom of the reactor. Figure 7 shows what proportion of each of the three loads (1, 5, and 10 samples - tests No. 1, 5, and 10) fell into each particle size class after fragmentation.
Test conditions were the same for each load:

- ✓ 500 J pulses
- ✓ I/g ratio of 10 I/g

The amount of material processed had a significant influence on fragmentation yield. The optimum load for our 0.8 litre reactor was approximately 5 samples. There are several hypotheses to explain this result.
As in a grinder, the quantity of material to be processed per batch affects the success of the grinding. With every pulse, the material gets moved around inside the reactor. Particles collide with each other

and self-fragment. An increase in sample mass has a positive effect. This is true for a load of 1 to 5 samples.
However, the phenomenon reverses for a load of 10 samples. The discharge or breakdown voltage measured between the reactor's electrodes during the 10-sample test was lower than for the 1- and 5-sample tests. The discharge tension was on average 200 kV for 1 sample, 140 kV for 5 samples and 60 kV for 10 samples.
One hypothesis relates to the fact that graphite is a conductor. The conductivity of the medium, comprising the water and the graphite in suspension, would certainly increase as the quantity of graphite increases. As the conductivity of the medium increases, the breakdown voltages inside the reactor will fall and therefore the quantity of energy per pulse will fall also.

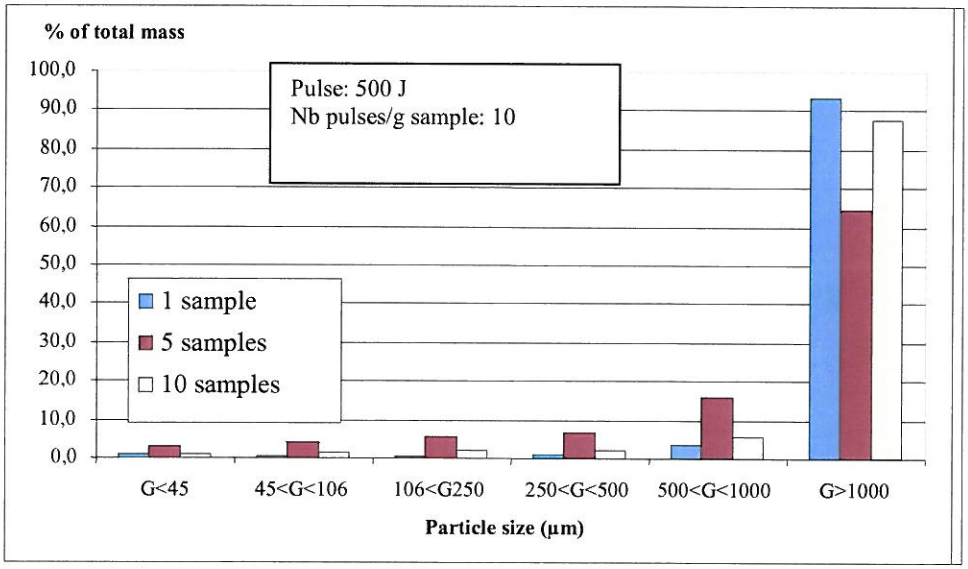

Figure 7: Influence of the mass to be fragmented

Reactor load is therefore a very important parameter in fragmentation. It must be adjusted for each shape of reactor.

3.2 Fragmentation/Screening cycles

For this series of tests, the material was screened in between each fragmentation stage. Each sample underwent several cycles of fragmentation/screening. The aim was to remove any pieces of the material that met the dimensional requirements as well as any fines before the next fragmentation stage, thereby limiting any over-grinding of the samples.
Figures 8 and 9 show the results obtained from each of the two test set-ups: 500 J with 5 samples and 350 J with 1 sample. The masses are given as a cumulative percentage.

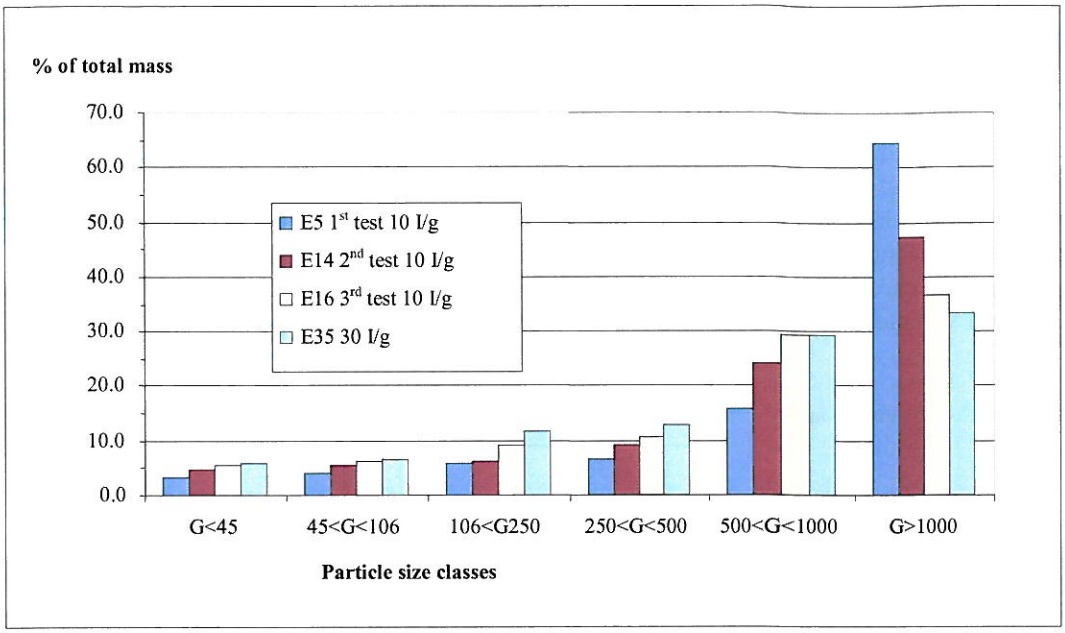

Figure 8: Comparison between fragmentation/screening cycles and direct fragmentation. 500 J - 5 samples

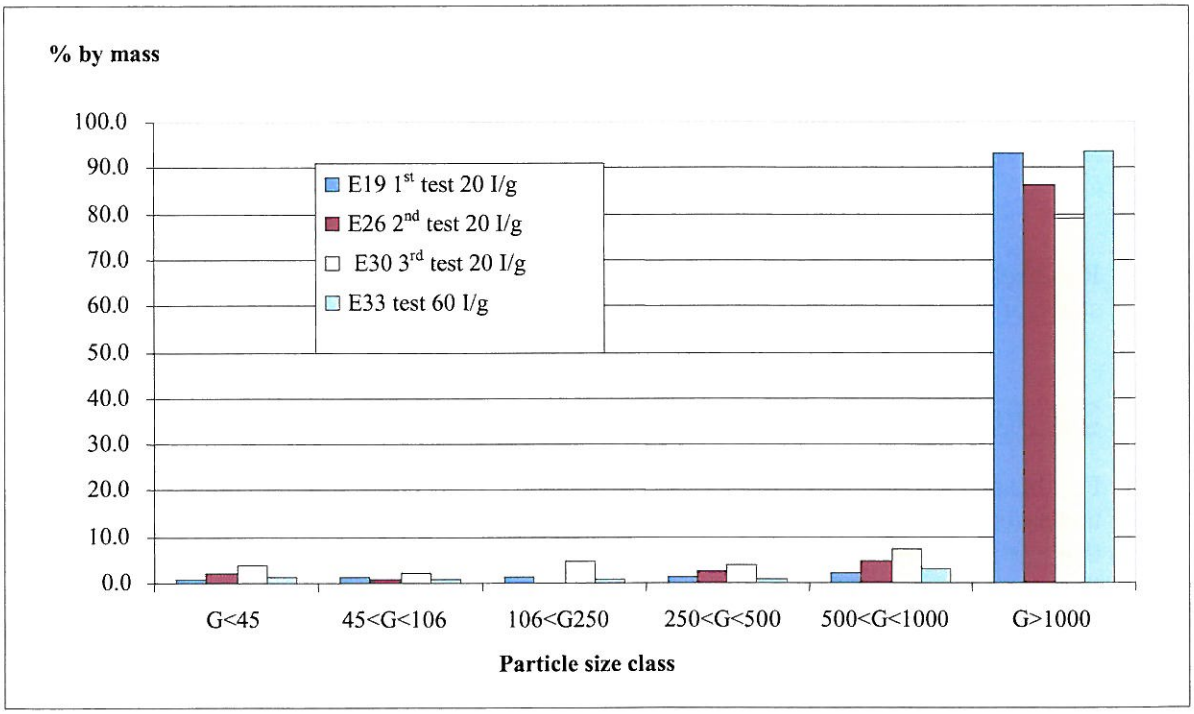

Figure 9: Comparison between fragmentation/screening cycles and direct fragmentation. 350 J - 1 sample

Tests E5, E14 and E16 were the three successive cycles of fragmentation/screening. Test E35 was a parallel test using the same total fragmentation energy, but without the intermediate screening stages. Figure 8 shows a similarity between tests E19, E26 and E30 on the one hand, and E33 on the other.

The main results were as follows:

- ✓ As regards the particle size class 500 µm<X<1 mm for tests E16-E35 (fig. 8) and E30-E33 (fig. 9), with each cycle the proportionate mass of this fraction increased compared to the other particle size classes. This is less evident in figure 7.

  However, we note that after each cycle, the quantity of material to be treated in the following cycle fell since the screened fraction smaller than 1 mm had been removed. The previous tests involving reactor load (section 3.1.5) proved the importance of this parameter. This phenomenon would have been more pronounced for this test (fig. 8), since from an initial load of 5 samples, the variation in mass after each test was significant compared to the test in figure 8. The loads were 87 g for E5, 55 g for E14 and 40 g for E16. Taking for example the particle size class 500 µm<X<1 mm, 15.8% by weight of the sample fell into this class after the first test, 8.3% after the second, and 5.1% after the third. Fragmentation yield fell considerably. This fall is very certainly linked to the fall in mass of the sample being treated.
  Compensating for this loss in mass by adding a new sample would ensure optimal reactor load at all times.
  If we take the first test, E5, as a reference, we see that 15.8% of this sample by mass was 500 µm < X < 1 mm. By the third cycle, by compensating for the loss in mass, the fraction by mass for particle class 500 µm < X <1 mm could be about 45%. This would be clearly higher than the 29% obtained from the direct cycle (fig. 7, E16 500<G<1000).

- ✓ The quantity of fines (X < 500 µm) produced in the cyclic tests was approximately 6% smaller for an equivalent fraction by mass of the particle class 500 µm <X<1 mm (see fig. 7) i.e. for E16 (X<500 µm), approximately 31% and E35 (X<500 µm) approximately 37%.

In conclusion, the fragmentation/screening cycle method can improve the fragmentation process in terms of fragmentation performance and minimising the production of fines.
However, a lot of handling is required which could cause problems in high-activity units.

3.3 HTR compacts

Tests were conducted using both compacts containing 20% by volume of inert TRISO particles, and compacts without particles.
In order to limit the number of compacts used, tests were conducted using only one compact per test. As we have already seen, this means that reactor load was not optimal. These tests were used to compare the results from compacts and from graphite samples.

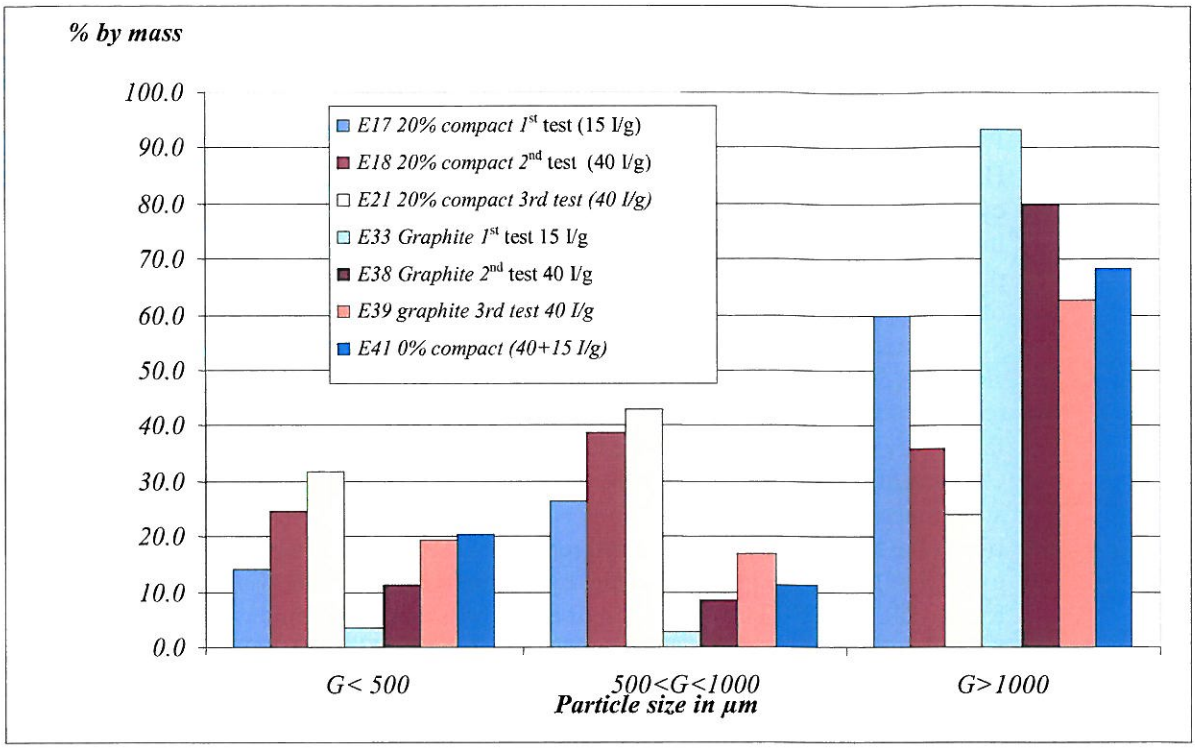

Figure 10: Comparison between compacts with 20% TRISO and graphite

In figure 10, the particles smaller than 500 µm have been combined into a single particle size class in order to make reading the histogram easier.
The main results were as follows:

1. Comparison between G2 graphite and particle-free compacts

The particle-size curves are fairly similar for samples E38 (G2 graphite) and E41 (particle-free compact) for the same l/g ratio. For example, the percentages by mass that fell into particle class 500 µm<X<1 mm were respectively:

- ✓ 8.5% for graphite G2
- ✓ 11% for particle-free compacts

The use of G2 graphite samples instead of particle-free compacts could be continued.

2. Comparison between a compact containing 20% by volume of TRISO particles (fig. 9, E18) and a particle-free compact (fig. 9, E41)

For a similar l/g ratio, the particle size class 500 µm<X<1 mm contained 39% by weight of the 20%-particle compact, and 11% by weight of the particle-free compact. The oversize fraction, i.e. particle class X>1 mm, was twice as large for the particle compact. When looked at in proportion to the particle size class 500 µm<X<1 mm, fewer fines were produced by the fragmentation process from the particle-free compact (20% by weight) than from the particle compact (25% by weight).
The presence of particles promotes fragmentation. These results confirm the preliminary tests [1]. Half as much energy was needed to fragment a compact containing 20% particles than a piece of graphite or a particle-free compact.

By reducing the mass being processed during each fragmentation/screening cycle, the fragmentation yield also fell. This was similar to the phenomenon described in point 3.2.

The macrographic and micrographic analyses carried out by DTEC/SDTC/LEMA confirmed there was good separation between the particles and the graphite. The average diameter of particles was 800 μm. Based on this finding, it would seem that the particles remained largely intact. The layers of the TRISO coating were still there. The fragmentation occurred mainly at the graphite/particle interfaces (see fig. 10).

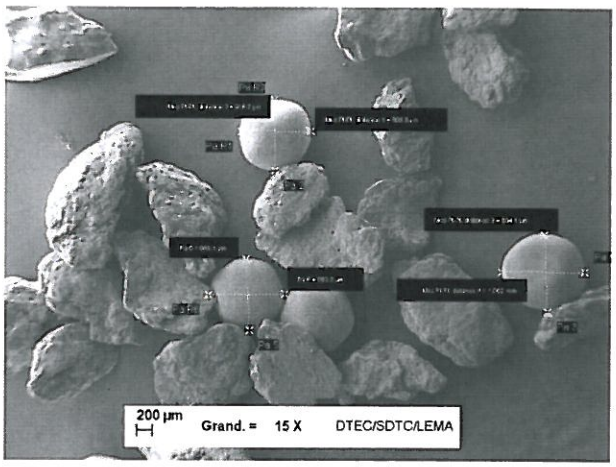

Figure 11. Sample E21 (compact containing 20% TRISO particles)

However, some particles did have cracks on the surface of the coating, and even a total lack of coating in some parts (see fig. 11). The EDS analysis together with the microscopy showed that the ZrO_2 core of the particle had been exposed (see fig. 11).

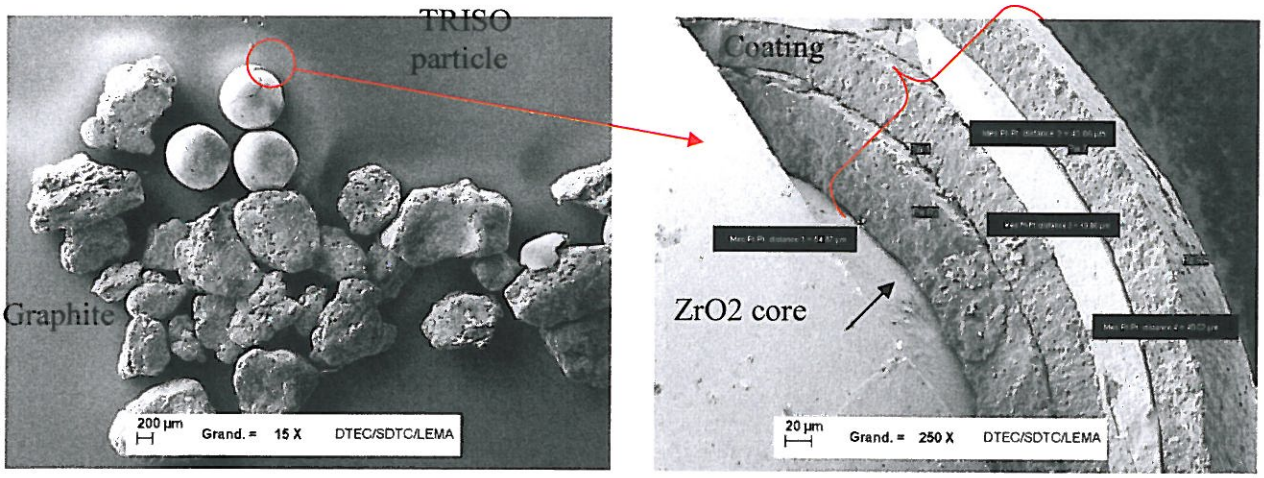

Figure 12. Sample E21 (compact containing 20% TRISO particles)

In conclusion, the presence of TRISO particles promotes fragmentation.

- ✓ Fewer fines are produced since the fragmentation occurs mainly at the graphite/particle interface
- ✓ The amount of energy needed to fragment a compact containing particles is half as much as that needed for a particle-free compact.

4 Bibliography

[1] NT DTEC/SDTC/2007/11 *essais 2006 de déstructuration par courants pulsés de combustibles pour les systèmes du futur*, Ph. Regnard, A. Beziat