

Graphite Dust Deflagration

*Ignition and Flame Propagation Data –
Supplement to EPRI Technical Report 1014797*

1015460

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Technical Update, October 2007

EPRI Project Manager

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PRODUCT DESCRIPTION

The graphite moderators of retired gas-cooled nuclear reactors present a difficult challenge during demolition activities. As part of the EPRI graphite initiative on the technical issues involved in the management and disposal of irradiated nuclear graphite, this report examines the international data on dust deflagration relevant to the decommissioning of graphite-moderated reactors. The report concludes that the risk of an explosion involving graphite dust during decommissioning is extremely low, and should not cause concern in planning a decommissioning strategy. Several international organizations, including EdF CIDEN (France), SoGIN (Italy), UKAEA (UK) and NUPEC (Japan), collaborated in this program by providing data.

Results

Section 4 of the original report described the principal experimental studies on dust deflagration to support reactor operation and decommissioning. It included individual descriptions of the identified research programs in Italy, Japan, the United Kingdom and France. The sections in this report repeat a brief description of the studies performed at Leeds University, UK, and CNPP/Les Renardieres, France, together with the data omitted from Appendices B and C in the earlier report.

This review concludes that the risk of a dust explosion involving graphite dust during reactor decommissioning is extremely low, and should not cause particular concern in planning a decommissioning strategy. In the course of designing the strategy, however, it is prudent to eliminate any potential risk by eliminating one or more of the essential conditions for deflagration that are identified in the report

Challenges and Objectives

- To collate all of the international dust deflagration work.
- To determine the conditions under which graphite can be safely handled in decommissioning without potentially causing dust deflagration events.

Applications, Values, and Use

Describe any possible future developments, potential upcoming changes, other business segments, and downstream applications. How do you see this market evolving?

EPRI Perspective

The EPRI graphite initiative started in 2005, with a review of available data (EPRI Report 1013091), which led to further work on three specific technical areas. The work described in this report provides an international consensus on the dust explosion issue. Utilities can use it where appropriate to justify and underpin the safe dismantling of graphite reactors in air. The report defines precautions necessary to avoid safety hazards. The next graphite project, which is in progress, reviews the existing data on the leaching of radioisotopes from graphite moderators, including their relevance to the dismantling of graphite structures. Another project is currently investigating graphite radioisotope separation technology, as there is a widespread view that disposal would be greatly facilitated if carbon-14 could be removed from the graphite blocks.

This project involves an engineering feasibility study of the most promising techniques for C-14 isotope separation.

Approach

This document is a supplement to the above report. It includes data used in the analysis, and originally intended for inclusion as Appendices, but which was not provided in the earlier report. Readers should refer to the original report for detailed discussion of these results.

Keywords

Low-level waste disposal

Graphite

Decommissioning

ABSTRACT

The graphite moderators of retired gas-cooled nuclear reactors present a difficult challenge during demolition activities. As part of the EPRI graphite initiative on the technical issues involved in the management and disposal of irradiated nuclear graphite, EPRI Report 1014797 *Graphite Dust Deflagration: A Review of International Data with Particular Reference to the Decommissioning of Graphite Moderated Reactors* (March 2007) examined the international data on dust deflagration relevant to the decommissioning of graphite-moderated reactors. Several international organizations, including EdF CIDEN (France), SoGIN (Italy), UKAEA (UK), and NUPEC (Japan) collaborated in this program by providing data, which are reviewed in the report. The report concluded that the risk of an explosion involving graphite dust during decommissioning is extremely low and should not cause concern in planning a decommissioning strategy. In the course of designing the strategy, however, it is prudent to eliminate any potential risk by eliminating one or more of the essential conditions for deflagration that were identified in that report.

This document is a supplement to the original report. It includes data that were used in the analysis and were originally intended to be included as appendices, but which were not provided in the earlier report. Reference should be made to the original report for the discussion of these results.

Section 4 of the original report described the principal experimental studies on dust deflagration to support reactor operation and decommissioning. The section included the individual descriptions of the identified research programs in Italy, Japan, the United Kingdom, and France. The current report repeats a brief description of the studies done at Leeds University, United Kingdom, and CNPP/Les Renardieres, France, together with the data that was omitted from Appendices B and C in the earlier report. Note that the figure numbers and references are those used in the original report, to which reference should be made for further details.

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1

LEEDS UNIVERSITY RESEARCH PROGRAM

The requirements of the program at the Energy and Resources Research Institute, University of Leeds, were very broadly defined initially, without assumptions about the ignition source or reliance on older information, as follows:

- Could graphite dust explode?
- Could the overpressures generated in an initial graphite dust explosion damage the graphite core (assuming that it took place within the FAZ either in the supposed void space of in fuel channels), generating further dust that could lead to a secondary explosion?
- Could an initial explosion which did not disrupt the core nevertheless escalate through secondary explosions in the connected volumes beyond the fuel channels and perhaps disrupt the reactor charge walls?

To address these broad questions, the following specific test program was agreed:

- Determine explosibility of graphite dust and hybrid methane/graphite mixtures in the standard 1 m³ ISO test rig (the use of methane as a driver with known explosion characteristics was considered certain to be necessary in order to obtain meaningful data on graphite dust alone)
- Determine flame speeds and temperatures
- Determine the ignition characteristics of graphite dust in the Hartmann apparatus
- Make a determination of likely overpressures in the reactor geometry in a scaled-down version, using methane
- Determine whether a secondary graphite dust explosion was possible in through interlinked volumes approximating to the reactor volume ratios
- Determine the extent of burnout of graphite particles passing through a flame front using the CBK8 model
- Determine the effect of likely contaminants on the behavior of the graphite dust (the likely contaminants in pile 1 being primarily lead (and oxides) and aluminium (from the fuel))
- Quantify approximately the agglomeration of graphite particles with time (*arising from observations during the experimental program*)

The work programme was devised by UKAEA in discussions with The University of Leeds and specialist members of the Windscale Projects Technical Committee, of which one of the present authors (AJW) is a member. The program was then managed by BNFL, who arranged for the graphite supply. Since no AGXP representative of the piles graphite was available, a consensus was reached that Pile Grade 'A' archived from the Calder Magnox reactors was an acceptable substitute.

Obtaining the specified granulometry proved surprisingly difficult. The supplied graphite was in blocks of dimensions 9" x 9" x 12", and these were broken by sledgehammer into small pieces, protecting the graphite first with plastic sheeting. A Retsch ZM100 milling machine was then employed but proved extremely unsatisfactory with the specified sized material, and a jaw crusher had to be used first to reduce the size of the fragments to millimetre-sized grains. The miller was then employed to produce a powder mainly < 60µm, with the first batches being discarded.

Whilst the product included material below 10µm (the specified upper size limit for the work), the average particle size produced by the Retsch machine was higher than specified with only 19% satisfying the < 10µm criterion. Accordingly, an external contractor was engaged to further grind and separate/classify the material before use in the tests. The result was a powder ground to < 30µm with 80% < 10µm, although separation was not possible because it was found that the particles rapidly agglomerated. This issue is discussed later. The resulting distribution shows a very strong peak at around 7µm and is shown in graphical form and as the standard Mastersizer output for comparison with other samples discussed in this review (Figure 1-1).

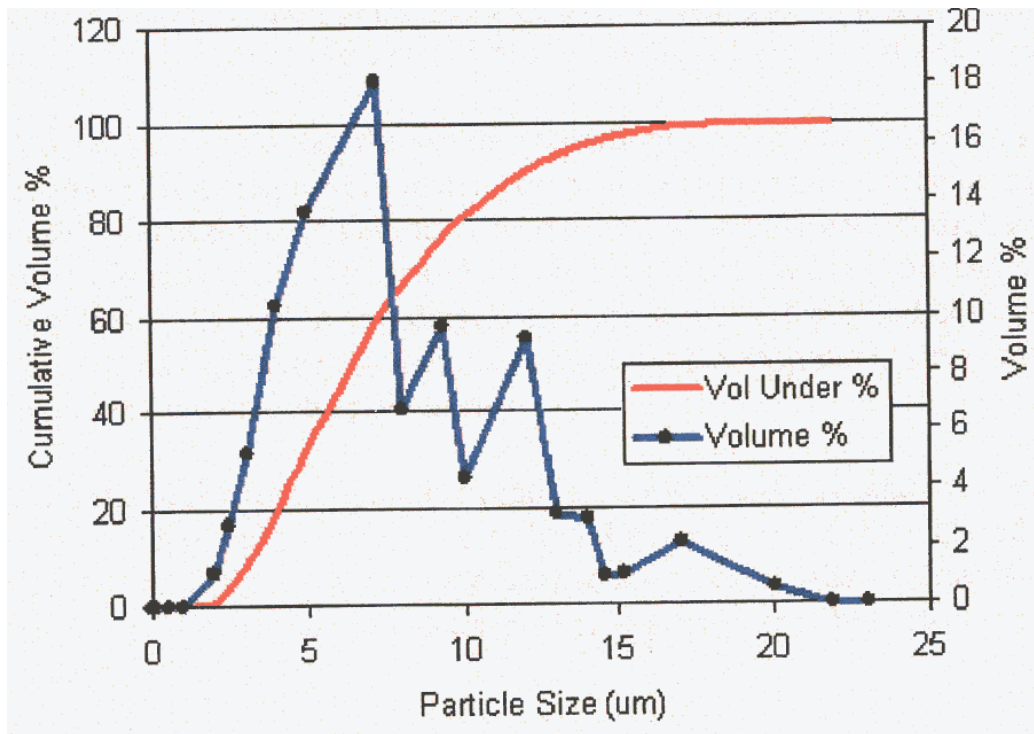


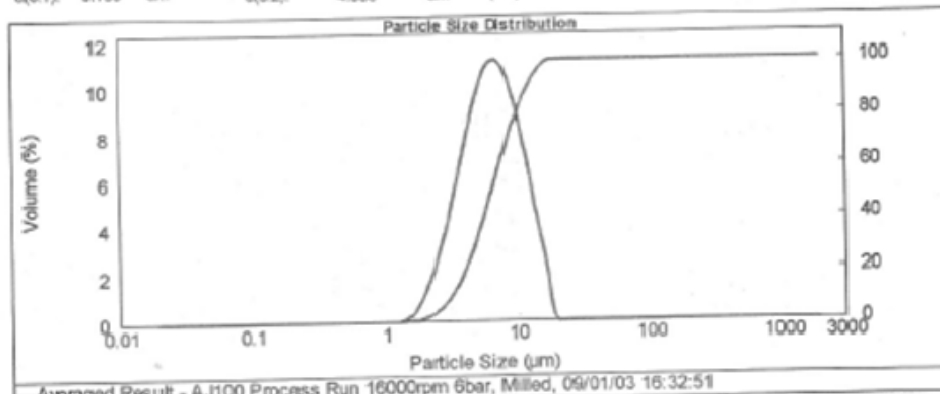
Figure 1-1
Size Distribution of PGA Graphite Dust used at The University of Leeds, UK

Result Analysis Report

Sample Name: Averaged Result - AJ100 Process Run 16000rpm 6bar, Milled
SOP Name: Graphite Wet IPA 0.1ab
Measured: 09/01/03 16:32:51
Sample Source & type: Factory = Leeds Uni
Measured by: lab
Analysed: 09/01/03 16:32:52
Sample bulk lot ref: TC1067
Result Source: Averaged

Particle Name: Graphite2
Accessory Name: Hydro 2000SM (A)
Obscuration: 13.90 %
Particle Rt: 2.420
Absorption: 0.1
Analysis model: General purpose
Dispersant Name: Propen-2-ol
Size range: 0.020 to 2000.000 um
Weighted Residual: 0.598 %
Dispersant Rt: 1.390
Result Emulation: Off

Concentration: 0.0107 %Vol
Vol. Weighted Mean D[4,3]: 7.068 um
Specific Surface Area: 1.09 m²/g
Span : 1.387
Uniformity: 0.427
Surface Weighted Mean D[3,2]: 5.518 um
Result units: Volume
d(0.1): 3.180 um
d(0.2): 4.035 um
d(0.5): 6.417 um
d(0.9): 12.081 um



| Size (µm) | Vol Under % | Size (µm) | Vol Under % | Size (µm) | Vol Under % | Size (µm) | Vol Under % |
|-----------|-------------|-----------|-------------|-----------|-------------|-----------|-------------|
| 0.581 | 0.00 | 12.000 | 89.72 | 30.000 | 100.00 | 120.724 | 100.00 |
| 1.034 | 0.00 | 13.000 | 92.85 | 32.000 | 100.00 | 125.000 | 100.00 |
| 2.000 | 1.18 | 14.000 | 95.23 | 33.500 | 100.00 | 145.000 | 100.00 |
| 2.500 | 3.95 | 14.500 | 96.19 | 35.620 | 100.00 | 154.104 | 100.00 |
| 3.101 | 9.19 | 15.157 | 97.26 | 40.244 | 100.00 | 196.714 | 100.00 |
| 4.000 | 19.57 | 17.000 | 99.43 | 45.469 | 100.00 | 250.000 | 100.00 |
| 5.053 | 33.18 | 20.000 | 100.00 | 51.371 | 100.00 | 300.000 | 100.00 |
| 7.267 | 59.35 | 21.860 | 100.00 | 58.041 | 100.00 | 350.000 | 100.00 |
| 8.000 | 66.96 | 23.000 | 100.00 | 65.575 | 100.00 | 420.000 | 100.00 |
| 9.302 | 78.17 | 24.000 | 100.00 | 75.000 | 100.00 | 500.000 | 100.00 |
| 10.000 | 80.53 | 25.000 | 100.00 | 108.852 | 100.00 | 600.000 | 100.00 |

Operator notes: Average of 2 measurements from Leeds Uni - Graphite

Figure 1-2
Mastersizer 2000 Report on Particle Size Distribution of PGA Graphite used at The University of Leeds, UK

This is a *very* much finer dust than employed continuous arc of energy 4J. The apparatus was proven with maize flour as the standard calibration material, for which deflagrations were obtained at concentrations above about 200 g.m^{-3} . These continued up to the maximum tested, 827 g.m^{-3} .

For graphite there was no reaction at any concentration between 42 and 1001 g.m^{-3} except that the conducting nature of the particles allowed some bridging of the arc, with small sparks developing. Drying the sample overnight made no difference with the 500 g.m^{-3} sample tested.

ISO Rig Test Data

The Leeds vessel (shown in Figure 1-3) is constructed to the general specifications of the ISO standard [21] but with some small deviations (which the standard permits) in order to facilitate the rapid turnaround of tests. The vessel volume is slightly oversize, at 1.138 m^3 , the external chamber is 4.5 litres and connected to the perforated C-ring within the vessel *via* a fast-acting pneumatic ball valve. The dust pot is initially pressurised to 20 bar and the main vessel pressure reduced to 923 mbar. Where a methane driver was employed, the methane was added to the main vessel volume using the technique of partial pressures. The ignition delay was 0.6 seconds to ensure that all dust had entered the vessel. Chemical igniters of energy between 5 and 15 kJ were mainly used, firing into a small hemispherical cup in the centre of the vessel to limit any problems resulting from directional ignition. A small number of later tests were conducted using igniters of 1 kJ, 2kJ, 3kJ and 4kJ. These confirmed that the threshold energy for initiating a deflagration in the graphite alone lay around 4 – 5 kJ.

Thermocouples are present at fixed positions within the vessel to allow measurements of flame speed and temperature, and there is full piezoelectric pressure instrumentation.

Figures 1-3 and 1-4 illustrate respectively the timescale of pressure changes within the vessel for a typical experimental run, and the relationship between maximum pressure attained and chemical igniter energy, which must be allowed for in evaluating the explosion parameters of the dusts.

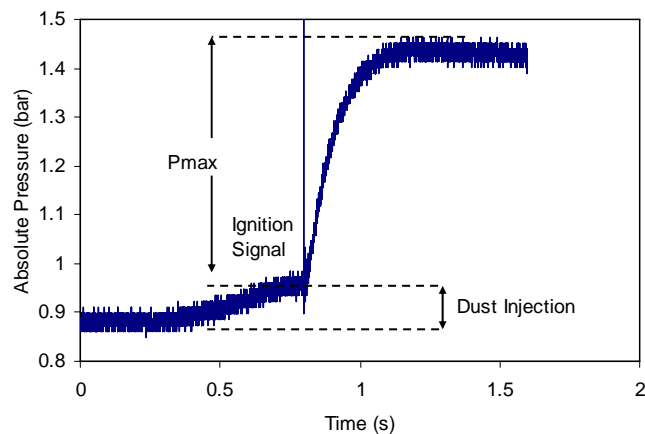


Figure 1-3
Pressure/Time Relationship for a Typical Deflagration in the Leeds Apparatus

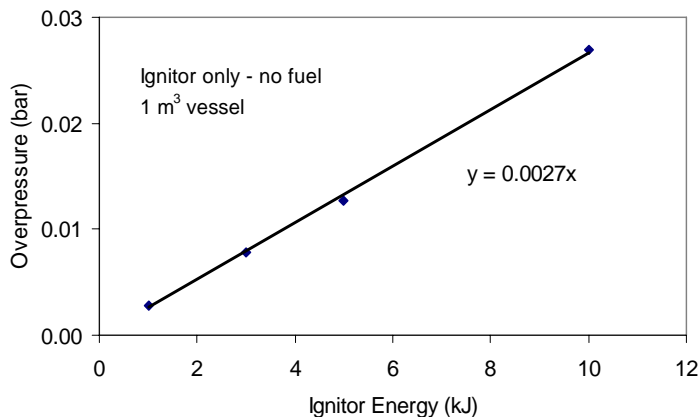


Figure 1-4
Overpressure as a Function of Igniter Energy

Following the negative results obtained in the Hartmann apparatus, there was concern that meaningful results could be obtained with graphite alone in the ISO rig. It was decided to conduct a lengthy series of tests were conducted using graphite in the presence of a methane ‘driver’, methane alone, and maize flour, before proceeding to tests with graphite alone. The full sequence of tests in the first phase of the work is listed in the next section.

The logic of this apparently complex approach to the problem is that graphite alone is clearly so weakly explosible that the pressure pulse and the apparent explosion parameters are completely dominated by the behavior of the chemical igniters and, in consequence, any data obtained are subject to significant uncertainties. In addition, the initial observations quickly established that, with the particular granulometry of dust used, a significant proportion remained unburned. In their first report to BNFL on behalf of UKAEA [43]¹, the Leeds authors describe a sophisticated methodology to evaluate the independent behavior of the graphite, deconvoluting the data from the effects of the chemical igniters by utilising 6% methane in the air within the vessel to provide a larger, more measurable effect since the lean methane/air mixture acts as a large flame front ignition source. The behavior of 6% methane in the absence of graphite was readily established with identical chemical detonators being used, and this in principle would allow the characteristics of the graphite to be established.

However, two other issues needed to be addressed. Firstly, this is not a standard methodology in the ISO specification, and so it was decided to validate the ‘methane-driver’ technique independently using maize flour, the standard high-reactivity material utilised in the industry for the calibration of test systems. To conform as closely as possible to the ISO specification, the maize was sieved to < 70µm, the normal requirement for dust-explosibility tests if specific particle sizes are not being addressed. An additional complication in the presence of methane is that the great majority, if not all, of the graphite participates in the deflagration because of the

¹ This report was independently peer reviewed [44] by a person with 25 years’ experience in the gas and dust explosion field, whose comments were included.

higher energy input available from the burning methane. An essential requirement before the analysis is fully complete is therefore to collect the residues from one or more methane-driven tests and assess the mass consumed and (ideally) the particle-size distribution of the residues.

It may also be noted that in tests involving methane/maize and maize alone, the maximum overpressures reached were similar for the optimum concentration of the maize dust indicating that the available oxygen was all consumed. The key data from tests involving maize, which are needed to interpret the graphite data, are given in Table 1-1.

**Table 1-1
Key Data from Tests Involving Maize**

| | 6% CH ₄ Only | Maize Only | 6% CH ₄ + Maize | Graphite Only | 6% CH ₄ + Graphite |
|-----------------------------------|----------------------------|--------------------------------|---|------------------|----------------------------------|
| K_{st} (bar.m.s ⁻¹) | 120 | 160 (@ 550 g.m ⁻³) | 330 (@ 350 g.m ⁻³ maize or 395 g.m ⁻³ total flammable content) | 14 | 200 |
| P_{max} (bar) | 6.1 | 8.5 (@ 700 g.m ⁻³) | 8.4 (@ 264 g.m ⁻³ maize or 309 g.m ⁻³ total flammable content | 0.5 | 7.7 |

The ratio of the concentrations at which the equivalent peak values of explosion pressure occur for maize and maize plus methane can be applied to the results for graphite. A ratio is also calculable from the values of K_{st} . The concentrations are all above the stoichiometric concentration for maize (220 g.m⁻³), indicating that even with this material, the largest particles are not fully burning out in the deflagration.

The interpretation of the hybrid tests works on the premise that the ratio of K_{st} values of methane and methane/maize will also apply to graphite: however there is some evidence that this ratio varies with K_{st} , increasing as K_{st} falls [45]. There is also some variability with gas concentration, which was however not varied in this series of tests. This could have been addressed to some extent by carrying out further tests with different concentrations of methane in air, but it was decided that the tests with graphite would proceed directly.

When 6% methane was combined with graphite, the behavior was similar to that observed with maize, as shown in Figure 1-5.

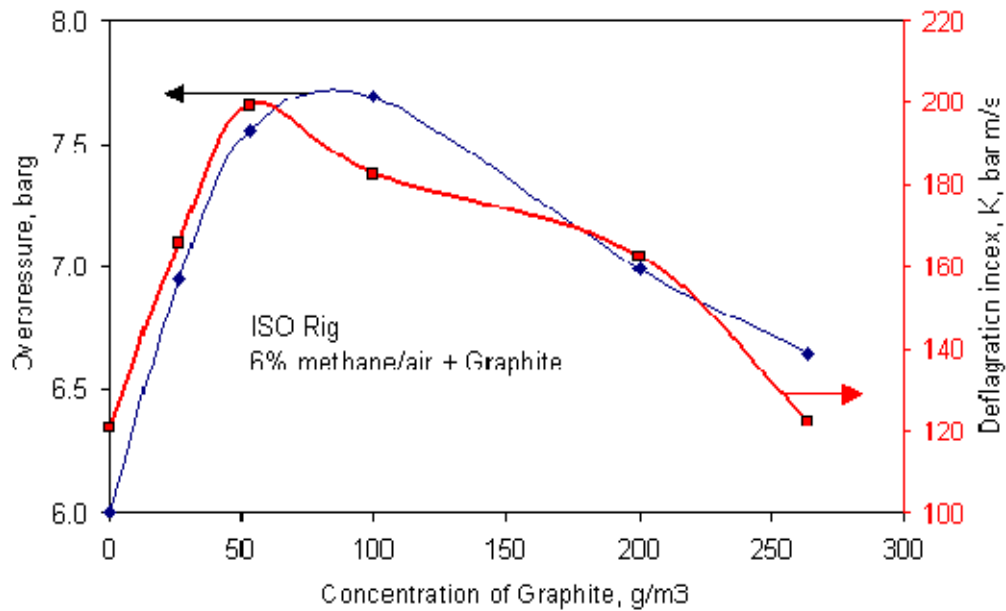


Figure 1-5
Maximum Overpressure and Deflagration Index as a Function of Graphite Concentration in Graphite/6% Methane Studies

The maximum pressure attained was 7.7 bar, 0.65 bar below that attained with the maize/methane mixture. The concentration at this maximum was 105 g.m^{-3} , close to the stoichiometric value for graphite alone but much richer than the stoichiometric value for graphite plus methane. The most likely explanation for the lower peak pressure is a lower flame temperature for carbon, possibly due to a greater proportion of the graphite dust not participating in the deflagration than for maize, although the temperatures attained in the two tests were different and this also contributes.

There is a ‘rule of thumb’ for estimating the flame temperature: the ratio of the peak absolute pressure to the initial absolute pressure, multiplied by the initial temperature in Kelvin is close to the peak flame temperature. This gives estimated peak flame temperatures of 2009K for methane/air, 2686K for methane/air/maize, 2727K for maize without methane, and 2497K for methane/air/graphite, assuming a reactant to product mole ratio of unity (which is precisely valid only if the ratio of post-reaction CO/CO_2 is the same in each case). The significance of these estimates is the estimate of an increase in flame temperature (above methane/air alone) due to graphite of 488K and due to maize of 677K. Making one further assumption, that the ratio of pressure and temperature rises due to maize and graphite will be proportional to their peak pressure and temperature ratios under deflagration conditions with no methane present, then one derives a peak pressure for graphite *alone* of 5.7 bar and a maximum flame temperature of 1931K. This is a significant lower value than for maize or typical hydrocarbons and is a principal reason why the deflagration in graphite is poor.

Another measure of reactivity is the flame speed, as determined from the thermocouples present within the ISO vessel. The average flame speeds in the centre of the vessel – between 20% - 80% of the radius and therefore isolated from any effects of the wall or ignition system – are shown in Figure 1-6 as a function of the graphite concentration:

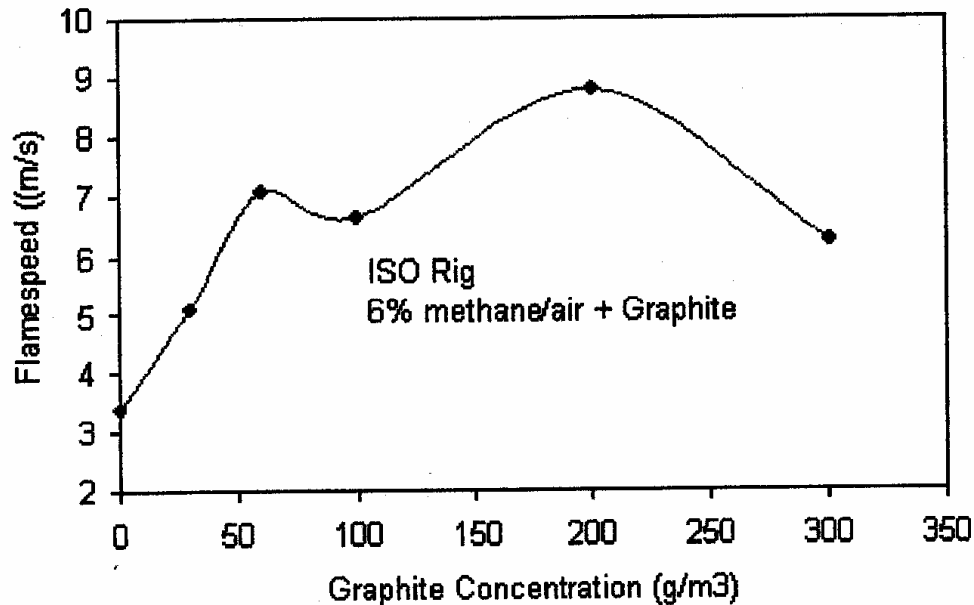


Figure 1-6
Average Flame Speeds in Hybrid Graphite/Methane/Air Mixtures

The presence of the graphite powder results in an almost three-fold increase in flame speed and suggests a maximum flame speed for graphite alone under the ISO conditions of 5.6 m.s^{-1} . The curve displays two peaks, and the first one, at around 55 g.m^{-3} , corresponds with the concentration at which the peak value of K_{st} was found. The authors of [43] were not able to explain the second peak.

On the basis of these assorted tests with hybrid mixtures and maize, the following results for graphite alone were predicted:

- A maximum K_{st} of 61 bar.m.s^{-1} at 86 g.m^{-3}
- A maximum peak pressure of 5.7 bar at a concentration of 278 g.m^{-3}
- A maximum constant volume flame temperature of 1931K

However, the flame speed results with their second peak suggest a different maximum reactive mixture (313 g.m^{-1}) and, since in these tests all of the graphite was consumed whereas it was suspected that different behavior would be found with graphite alone, the programme was moved on to tests with graphite alone.

In this situation, unsurprisingly, the severity of the deflagration was dramatically reduced. Fig 16 shows the maximum overpressures as a function of concentration of graphite, and they do not exceed 0.5 bar.

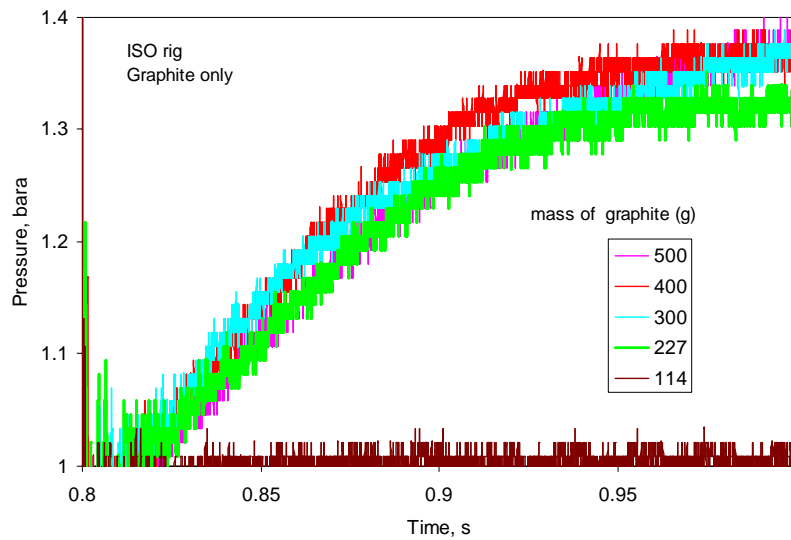


Figure 1-7
Maximum Overpressures for Graphite Alone in the Leeds ISO Apparatus

K_{st} also shows a large reduction compared with the results predicted from the hybrid results (Figure 1-8).

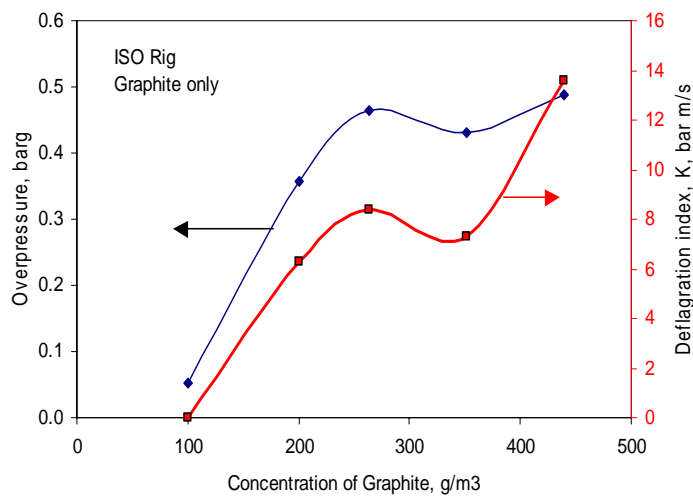


Figure 1-8
 K_{st} and Overpressure Data for Graphite Alone in the Leeds ISO Rig

A major difference from the hybrid tests was that as much as 60% of the graphite did not participate in the deflagration. The implication of this is that the 40% which did burn was the finest material, and therefore that less than about 3.5 μ m based upon the distribution in Figures 1-1 and 1-2.

There is however an alternative possibility, which is that the flame started to burn around the ignition source but then failed to propagate. This was however resolved by studies of the flame propagation utilising the thermocouple array in Phase 2 of the Leeds project [46]. The full sequence of these further tests conducted in the ISO vessel is given in the next section.

Figure 1-9 shows the flame arrival times for the maximum reactivity graphite/air explosion and Figure 1-10 shows the flame speed as a function of distance from the centre of the vessel.

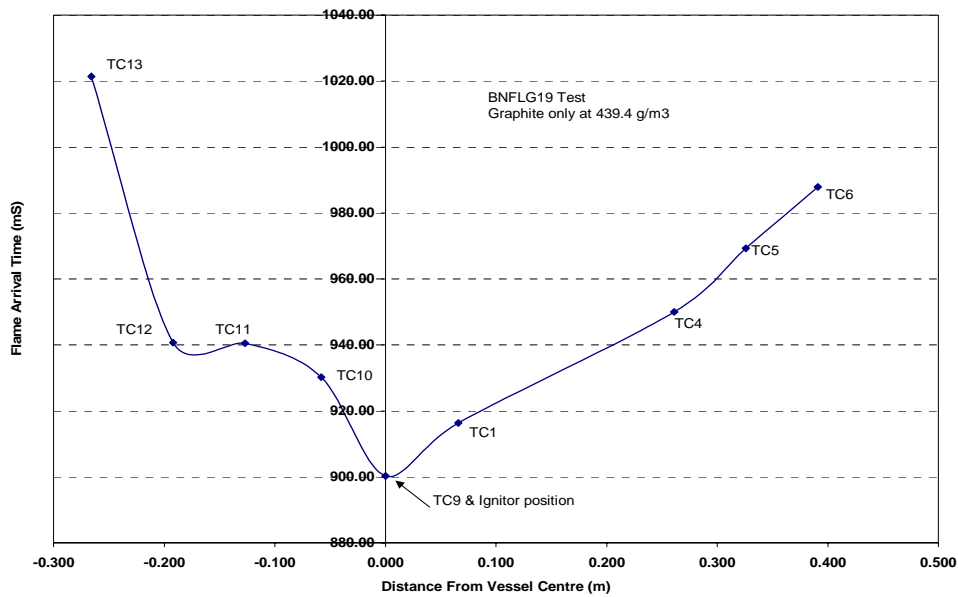


Figure 1-9
Flame Arrival Time for Graphite/Air Deflagration in the Leeds ISO Vessel

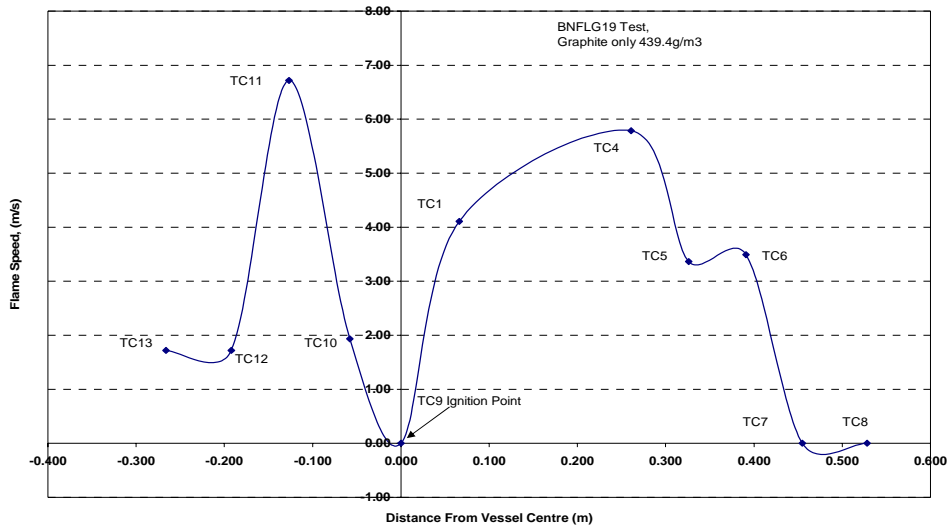


Figure 1-10
Flame Speed for Graphite/Air Deflagration in the Leeds ISO Vessel

The flame arrival records in Figure 1-9 show that the flame spreads to a diameter of at least 0.8m, which is in reasonable agreement with the size deduced from the pressure records. The flame speed records in Figure 1-10 indicate a flame propagation away from the ignition source, that later slows down with a possible identification of flame quenching. This evidence would support the explanation for the low overpressures based on flame quenching at around a 0.9m diameter flame size – in other words, evidence that the flame is self-extinguishing. Ideally, to prove this point, tests would be repeated in larger vessels. However, the French work ably demonstrates that this is the case.

There is lengthy debate in References 43 and 46 over the question of the unburnt material as to the relative importance of flame quenching and the failure of larger particles to participate in the deflagration. It remains essentially unresolved, but is highly likely that both explanations contribute to the residue of unburnt material and to the consequent low peak pressures. Reference 46 ventures that the maximum particle size which probably participates is 6 μm , but the evidence presented for this particular number is not entirely convincing.

Table 1-2
Summary of the ISO Rig Findings

| | Measured Peak (Graphite Alone) | Predicted Peak from Methane- Driven Tests | Predicted Concentration For Peak (G.M ⁻³) | Corrected Concentration Assuming 60% Unburnt (G.M ⁻³) | Measured Concentration For Peak (G.M ⁻³) |
|-----------------------------------|-----------------------------------|---|--|--|--|
| K_{st} (bar.m.s ⁻¹) | 14 | 61 | 86 | 215 | 450 |
| P_{max} (bar) | 0.5 | 5.7 | 278 | 695 | 450 |
| From Flame Speed Tests | | | 313 | 783 | |

The apparent wide inconsistency in this table derives largely from the attempts to improve upon the data using indirect means such as the methane driver. Whilst these efforts are to be commended, it is important to record explicitly the following conclusions:

- PGA graphite dust was weakly explosible with ignition energies of 5 kJ or higher;
- The graphite alone produced a very low maximum overpressure and K_{st} ;
- The flame front appears to be self-extinguishing within the volume of the vessel; *and*
- Only around 40% of the sample participated in the weak deflagration, and the inference is that this was the finest material, < 6µm.

Tests Specific to Windscale Pile 1

These results are of minor interest in the present context but are mentioned for completeness. A fully instrument approximate scale model of the connected volumes present in the reactor was constructed in order to assess the effects of overpressures and secondary explosions. These tests were *not* conducted with graphite because of its poor explosibility, but with quiescent methane/air mixtures in the range 6 – 10% methane backed up by initial quiescent tests in the ISO rig. The initiating event was established in an enclosure representing a theoretical void within the graphite core, and it was concluded that external structures would be unlikely to be subjected to an overpressure exceeding 0.2 bar.

Secondary explosions were assessed in the same linked geometry using 10% methane/air in the initiating chamber and either 0 or 5% in the rest of the geometry, with graphite powder laid in the linking pipe (representing a fuel channel within the graphite stack. These conditions were chosen as a ‘worst-case scenario’ in terms of induced velocities in the ‘channels’ and maximum turbulence.

The results showed no evidence of any secondary explosion – indeed the overpressures were *lowered* compared with the initial tests, showing that the induced graphite dust cloud behaved as a heat sink thereby partially suppressing the gas explosion in the initiating chamber.

Two issues were identified by the Leeds team which need to be borne in mind:

- The initiating gas explosion may have been over-driven, resulting in excessive turbulence which can result in flame quenching.
- The results may be influenced by agglomeration of the graphite powder (discussed in the following sub-section), whereas the agglomeration forces may have been overcome in the ISO test conditions. However, the agglomerated material is likely to be more characteristic of the reactor environment.

In general terms these are very encouraging results, albeit tempered by the somewhat artificial conditions employed.

Agglomeration and 'Ageing' of Samples

The phenomenon of agglomeration of particles, is illustrated through an apparent problem of repeatability/reproducibility is illustrated in Figure 1-11 where the overpressure is plotted as function of concentration. Although a consistent and expected trend is shown in terms of “single shot” tests at changing concentration, repeat tests at the same concentration (440 g.m⁻²) – identified as subset A in Figure 1-11 - show an apparently unacceptable variability. The tests in subset A were conducted under nominally “identical” conditions all using the same 5litre dust pot, same ignition source etc. The main difference between the tests is that they were carried out at different times over a 40 week period.

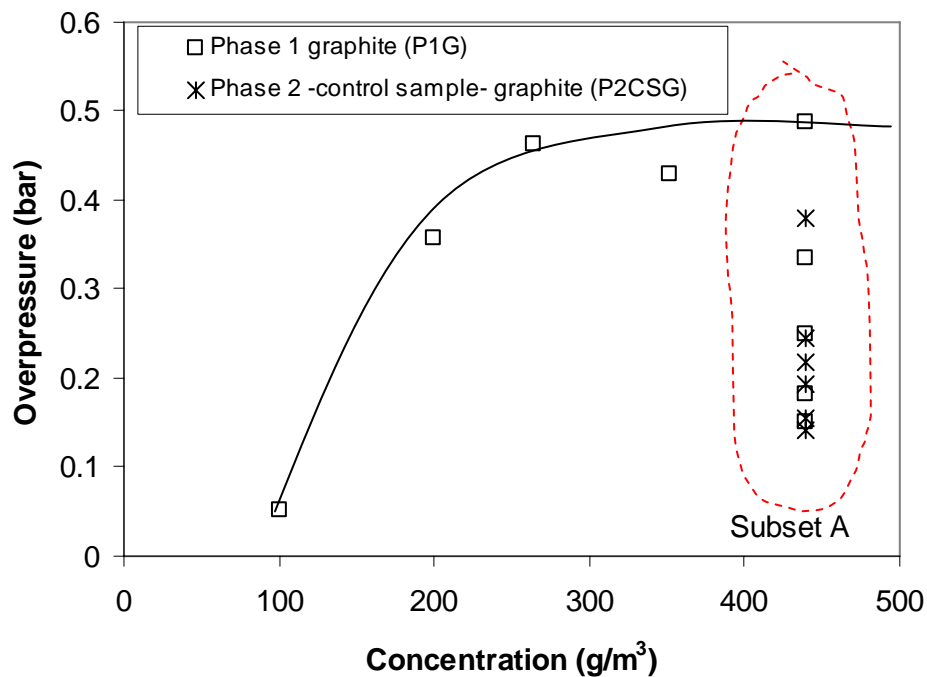


Figure 1-11
Systematic Reduction in Overpressure with Time since Preparation of Sample, Illustrated for Two Different Samples Over an Interval of 40 Weeks at Otherwise Identical Conditions

The supposition that the reduced overpressures arise because the smallest particles are no longer present is supported theoretically by a comprehensive mathematical treatment due to McDuff [47], which examines the energy of particle-particle interactions and concludes that it increases as the separation between the particles decreases. All adjacent particles have a van der Waals attraction which is brought about by a variety of forces, the principal of these being the interaction of dipoles of neighboring atoms and molecules modelled by a Lennard-Jones potential. When the separation distance is comparable to the particle radius, the particle-particle attraction increases as the inverse of the sixth power of the separation; when the radius of particles is significantly larger than their separation distance then the energy of interaction increases with compaction by approximately the square of the distance between particle edges, as verified experimentally by Rioux *et al.* in Reference 48, albeit on different materials. At *very* close approach, the Born repulsion of adjacent electron clouds becomes significant so that, in the absence of other influences, there is an energy minimum at an approach distance of approximately 1 nm.

At this point the question can be asked as to what has happened to the milled graphite dust during storage and drop-handling under normal gravity. If inter-particle separation were gradually decreasing by bulk weight and by compaction during drop-handling, and perhaps by slow particle migration processes, then particle-particle forces must be increasing with time contributing to, or in fact causing, particle ageing. It was shown that the dust-pot injection pressure necessary to achieve the same overpressure in the ISO vessel increased with the age of the sample, other parameters being approximately constant. Rioux *et al.* claim that many aggregates are of fractal nature. This means that where particle size growth is in unit fractal units the particles can grow relatively fast with complex fractal geometries. The fractal geometry of graphite dust was not investigated in the Leeds work, but the re-characterisation of the particle-size distribution has subsequently confirmed that the proportion of small

(< 5 μ m) particles is reduced whilst there is a modest increase in those in the 15 – 20 μ m range upon storage.

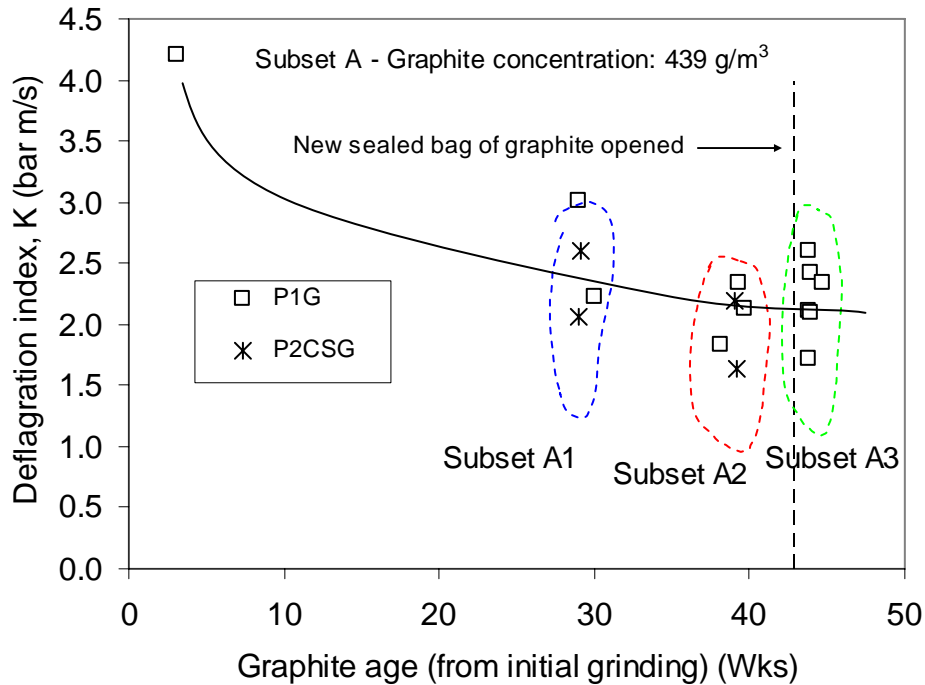


Figure 1-12
Confirmation of the Ageing Effect upon Deflagration Index with Additional Aged Samples

The problem with an explanation of a particle ageing effect being due to agglomeration of particles is that the ISO type vessel injection method uses high velocities in the jet air flow of the dust dispersion 'C' ring. It is considered by the Leeds team to be unlikely that the shear forces involved in this do not break up any agglomerated particles, unless the agglomerates are very firmly bound together. However, the size-distribution technique that is used is an aerodynamic sizer and the shear forces in this may also break weak agglomerates. Without a laser based in-situ particle size measurement it is very difficult to prove that the particle size distribution in the tested dust clouds is changing with age.

We may however conclude from this that, experimentally, the reduction in both overpressure and deflagration index with sample ageing is real and highly significant, since this will almost certainly apply to any samples encountered in decommissioning of graphite-moderated reactors.

The Effects of Additives

Because of the unique situation in Windscale Pile No. 1, the chosen impurities for this study were lead oxide and aluminium oxide, arising respectively from the melting, oxidation and possible dispersion of metallic lead used as spacing components, and from fuel-cladding debris created in the fire. Lead was employed as Pb_3O_4 in this study.

Seven tests were performed with graphite powder contaminated with lead oxide powder at a concentration of 1000 ppm lead (by mass) intimately mixed by opposed jet milling. The results are shown in Figures 1-13 and 1-14. Lead oxide appears to have no effect on the overpressure and reactivity K compared with graphite alone, as the results lie within the scatter of the results for pure graphite explosions at a particular age.

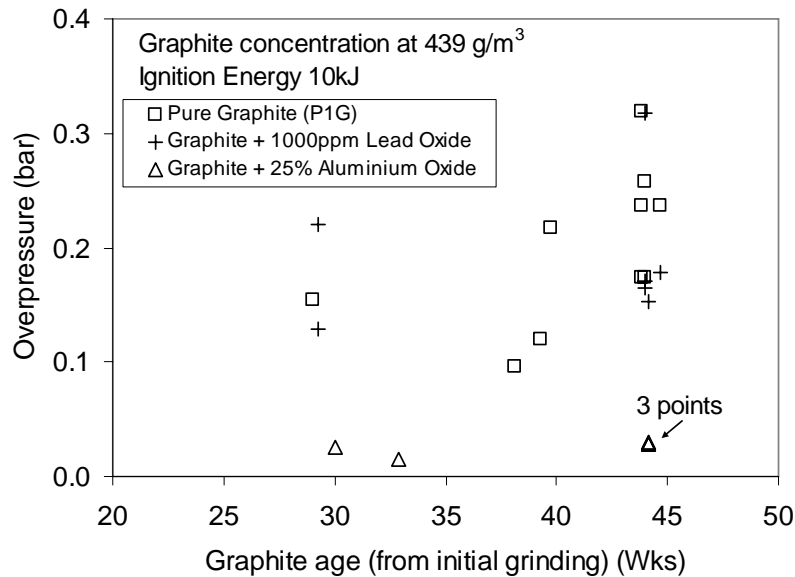


Figure 1-13
Illustrating the Lack of Effect of Catalysts on the Overpressure in the ISO Test, In Combination with the Ageing Effect

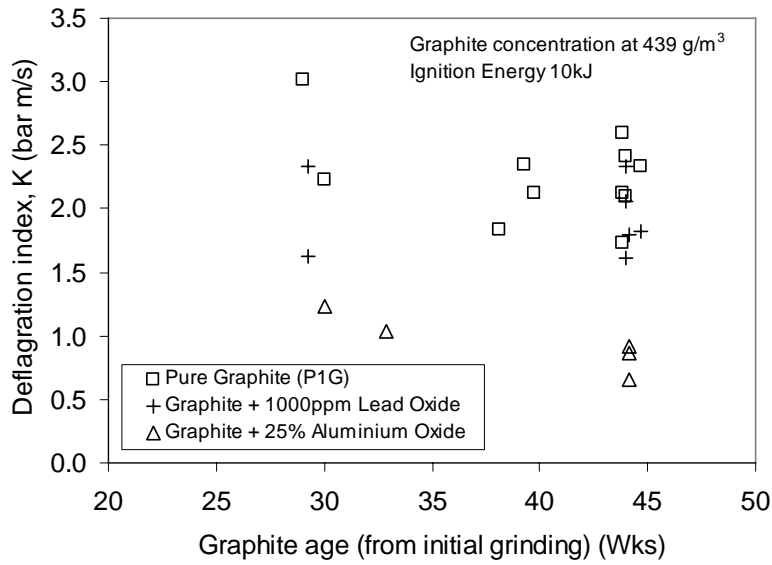


Figure 1-14
Illustrating the Lack of Effect of Catalysts on Deflagration Index, In Combination with the Ageing Effect

Lead oxide has potential catalytic effects, which may influence the ease of ignition of graphite. However, catalytic effects do not normally influence flame propagation in explosions as these are driven by kinetic reactions at high temperature in the flame front. Catalytic effects lower the temperature at which chemical reactions occur, but there is no low temperature basis on which explosion flames can propagate (apart from the cool flame regime in rich hydrocarbon explosions). As the pressure rise in an explosion is controlled by the temperature rise at the flame front and a catalyst cannot make this higher than thermodynamics will give, it may not be surprising that lead oxide has no influence on the explosion overpressure.

Five deflagrations were undertaken with graphite powder mixed with 25 wt % alumina powder (Al_2O_3). These results are also shown as a function of the graphite age from milling in Figures 1-13 and 1-14. These results show that alumina mixed in at 25% by weight with the graphite gives significantly lower overpressures and reactivity K and the differences from graphite are well below the scatter of results for graphite alone for the same age. In Figures 1-13 and 1-14 the overpressure and reactivity at 44 weeks is reduced from an average for graphite of about 0.25 bar overpressure and a K_{st} of 2 bar m/s to a 0.02 bar overpressure and a K_{st} of 0.8 bar m/s. The very low overpressure is most significant as this indicates a reduction in overpressure due to the addition of 25% of alumina by a factor of 10. The overpressure results indicate that the duration of the initial mass burn rate was comparatively short and hence the flame was extinguished at a relatively small size. Alumina at this concentration is clearly an *inert*.

The mechanism of this overpressure reduction is due to the additional heat sink effect provided by the inert alumina. This is the same effect that explosion suppressant systems work on – for example, coal-dust explosions can be extinguished using injection of limestone dust ahead of flames. The mixing of limestone dust with coal dust is used experimentally to achieve a reduction in reactivity. For hydrocarbon flames the temperature has to be reduced below 1500K, by heat extraction, for the flame to be extinguished. Typically, for coal dust, 80% stone dust and 20% coal is 'just reactive'. A key factor is that the effect is not catalytic and there has to be a significant mass of inert dust relative to the mass of reactive dust for the inerting to be effective.

2

DETAILED DATA FROM THE UK LEEDS UNIVERISTY STUDIES EMPLOYING THE ISO RIG – NOT INCLUDED IN EPRI TECHNICAL 1014797

**Table 2-1
Sequence of Tests in Phase 1 of the Work**

| Test Report ID | Test Lab ID | Date of Test | Test Rig | % Gas (v/v) | Gas Type | Mass of Dust (g) | Dust Conc. (g/m ³) | Dust Type | Pmax (barg) | Dust Recovered (g) | Comments |
|----------------|-------------|--------------|----------|-------------|----------|------------------|--------------------------------|-----------|-------------|--------------------|-------------------------------|
| A1 | bnflg1 | 01/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 6.439 | N/A | 5kJ Chemical Igniter |
| A2 | bnflg14 | 11/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 5.75 | N/A | 5kJ Chemical Igniter |
| A3 | bnflg15 | 19/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 5.84 | N/A | 5kJ Chemical Igniter |
| A4 | bnflg16 | 19/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 5.77 | N/A | 5kJ Chemical Igniter |
| A5 | bnflg18 | 23/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 6.55 | N/A | 5kJ Chemical Igniter |
| A6 | bnflg21 | 26/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 6.22 | N/A | 5kJ Chemical Igniter |
| A7 | bnflg22 | 29/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | N/A | N/A | No File – Result Not Captured |
| A8 | bnflg23 | 30/09/2003 | ISO RIG | 6.0 | CH4 | 0.0 | 0.0 | N/A | 5.906 | N/A | |
| A9 | bnflg2 | 01/09/2003 | ISO RIG | 6.0 | CH4 | 100.0 | 87.9 | Maize | 7.22 | | 1 x 5kJ Chemical Igniter |
| A10 | bnflg3 | 01/09/2003 | ISO RIG | 6.0 | CH4 | 200.0 | 175.7 | Maize | 7.72 | | 1 x 5kJ Chemical Igniter |
| A11 | bnflg4 | 02/09/2003 | ISO RIG | 6.0 | CH4 | 300.0 | 263.6 | Maize | 8.36 | | 1 x 5kJ Chemical Igniter |
| A12 | bnflg5 | 02/09/2003 | ISO RIG | 6.0 | CH4 | 400.0 | 351.5 | Maize | 8.22 | | 1 x 5kJ Chemical Igniter |
| A13 | bnflg6 | 08/09/2003 | ISO RIG | 6.0 | CH4 | 500.0 | 439.4 | Maize | 8.14 | | 1 x 5kJ Chemical Igniter |
| A14 | bnflg28 | 09/10/2003 | ISO RIG | 6.0 | CH4 | 30.0 | 26.4 | Graphite | 6.949 | | 5kJ Chemical Igniter |
| A15 | bnflg26 | 08/10/2003 | ISO RIG | 6.0 | CH4 | 60.0 | 52.7 | Graphite | N/A | | No File – Result Not Captured |
| A16 | bnflg27 | 08/10/2003 | ISO RIG | 6.0 | CH4 | 60.0 | 52.7 | Graphite | 7.556 | | 5kJ Chemical Igniter |
| A17 | bnflg24 | 02/10/2003 | ISO RIG | 6.0 | CH4 | 113.9 | 100.1 | Graphite | 7.69 | | 5kJ Chemical Igniter |
| A18 | bnflg25 | 06/10/2003 | ISO RIG | 6.0 | CH4 | 227.6 | 200.0 | Graphite | 6.993 | | |
| A19 | bnflg17 | 19/09/2003 | ISO RIG | 6.0 | CH4 | 300.0 | 263.6 | Graphite | 6.65 | | 1 x 5kJ Chemical Igniter |
| A20 | bnflg20 | 25/09/2003 | ISO RIG | 0.0 | N/A | 0.0 | 0.0 | N/A | 0.00 | N/A | 2 x 5kJ Chemical Igniter |
| A21 | bnflg12 | 09/09/2003 | ISO RIG | 0.0 | N/A | 114.0 | 100.2 | Maize | 0.00 | | 1 x 5kJ Chemical Igniter |
| A22 | bnflg13 | 09/09/2003 | ISO RIG | 0.0 | N/A | 228.0 | 200.4 | Maize | 0.00 | | 1 x 5kJ Chemical Igniter |
| A23 | bnflg11 | 09/09/2003 | ISO RIG | 0.0 | N/A | 341.0 | 299.6 | Maize | 6.36 | | 1 x 5kJ Chemical Igniter |

Table 2-1 (continued)
Sequence of Tests in Phase 1 of the Work

| Test Report ID | Test Lab ID | Date of Test | Test Rig | % Gas (v/v) | Gas Type | Mass of Dust (g) | Dust Conc. (g/m ³) | Dust Type | Pmax (barg) | Dust Recovered (g) | Comments |
|----------------|-------------|--------------|----------|-------------|----------|------------------|--------------------------------|-----------|-------------|--------------------|---|
| A24 | bnflg7 | 08/09/2003 | ISO RIG | 0.0 | N/A | 567.0 | 498.2 | Maize | N/A | | 1 x 5kJ Chemical Igniter – No Ignition |
| A25 | bnflg10 | 09/09/2003 | ISO RIG | 0.0 | N/A | 570.0 | 500.9 | Maize | 8.28 | | 1 x 5kJ Chemical Igniter |
| A26 | bnflg9 | 09/09/2003 | ISO RIG | 0.0 | N/A | 797.0 | 700.4 | Maize | 8.5 | | 1 x 5kJ Chemical Igniter |
| A27 | bnflg8 | 08/09/2003 | ISO RIG | 0.0 | N/A | 1024.0 | 899.8 | Maize | 8.39 | | 1 x 5kJ Chemical Igniter |
| A28 | bnflg29 | 10/10/2003 | ISO RIG | 0.0 | N/A | 113.8 | 100.0 | Graphite | 0.052 | | 2 x 5kJ Chemical Igniter |
| A29 | bnflg30 | 13/10/2003 | ISO RIG | 0.0 | N/A | 227.6 | 200.0 | Graphite | 0.3566 | | 2 x 5kJ Chemical Igniter |
| A30 | bnflg33 | 16/10/2003 | ISO RIG | 0.0 | N/A | 300.0 | 263.6 | Graphite | 0.464 | | 3 x 5kJ Chemical Igniter |
| A31 | bnflg31 | 14/10/2003 | ISO RIG | 0.0 | N/A | 341.4 | 300.0 | Graphite | 0.1896 | | 3 x 5kJ Chemical Igniter, Faced Away from hemispherical Cup |
| A32 | bnflg32 | 15/10/2003 | ISO RIG | 0.0 | N/A | 400.0 | 351.5 | Graphite | 0.43 | | 3 x 5kJ Chemical Igniter |
| A33 | bnflg19 | 25/09/2003 | ISO RIG | 0.0 | N/A | 500.0 | 439.4 | Graphite | 0.48 | 300 | 2 x 5kJ Chemical Igniter |

**Table 2-2
Sequence of Tests in Phase 2 of the Work**

| Report Test ID | Lab Test ID | Type of Graphite /Powder | Post-Milling Graphite Age (Weeks) | Conc (G/M ³) | Pmax (Bar) Corrected | Dust Pot DP (Bar) | Total Ignitor Energy (Kj) | Kst, Bar M/S 25ms | Time Delay (Ms) | Pot Vol (L) | Comments |
|----------------|-------------|--------------------------|-----------------------------------|--------------------------|----------------------|-------------------|---------------------------|-------------------|-----------------|-------------|----------|
| 1 | bnflg29 | P1G | 5.3 | 100.0 | 0.025 | 19.540 | 10.0 | 1.729 | 597 | 5 | |
| 2 | bnflg30 | P1G | 5.7 | 200.0 | 0.330 | 19.720 | 10.0 | 3.887 | 597 | 5 | |
| 3 | bnflg33 | P1G | 6.1 | 263.6 | 0.424 | 19.830 | 15.0 | 4.535 | 597 | 5 | |
| 4 | bnflg32 | P1G | 6.0 | 351.5 | 0.390 | 19.490 | 15.0 | 4.662 | 597 | 5 | |
| 5 | bnflg19 | P1G | 3.1 | 439.4 | 0.461 | 19.550 | 10.0 | 4.212 | 597 | 5 | |
| 6 | bnflg49 | P1G | 29.0 | 439.4 | 0.154 | 15.960 | 10.0 | 3.013 | 597 | 5 | |
| 7 | bnflg53 | P1G | 30.0 | 439.4 | 0.279 | 22.550 | 10.0 | 2.230 | 697 | 5 | |
| 8 | bnflg57 | P1G | 38.1 | 439.4 | 0.096 | 20.390 | 10.0 | 1.834 | 697 | 5 | |
| 9 | bnflg62 | P1G | 39.3 | 439.4 | 0.120 | 19.170 | 10.0 | 2.344 | 597 | 5 | |
| 10 | bnflg63 | P1G | 39.7 | 439.4 | 0.217 | 20.950 | 10.0 | 2.126 | 697 | 5 | |
| 11 | bnflg82 | P1G | 43.9 | 439.4 | 0.319 | 19.147 | 10.0 | 2.597 | 697 | 5 | |
| 12 | bnflg84 | P1G | 43.9 | 439.4 | 0.237 | 19.225 | 10.0 | 2.117 | 697 | 5 | |
| 13 | bnflg85 | P1G | 43.9 | 439.4 | 0.173 | 19.805 | 10.0 | 1.725 | 697 | 5 | |
| 14 | bnflg86 | P1G | 44.0 | 439.4 | 0.174 | 19.909 | 10.0 | 2.095 | 697 | 5 | |
| 15 | bnflg87 | P1G | 44.0 | 439.4 | 0.258 | 19.724 | 10.0 | 2.418 | 697 | 5 | |
| 16 | bnflg99 | P1G | 44.7 | 439.4 | 0.237 | 19.915 | 10.0 | 2.335 | 697 | 5 | |
| 17 | bnflg64 | P1G | 42.0 | 439.4 | 0.070 | 20.316 | 5.0 | 1.647 | 697 | 5 | |
| 18 | bnflg65 | P1G | 42.1 | 439.4 | 0.002 | 19.603 | 2.0 | 0.503 | 697 | 5 | |
| 19 | bnflg66 | P1G | 42.1 | 439.4 | 0.000 | 20.165 | 1.0 | 0.000 | 697 | 5 | |

Table 2-2 (continued)
Sequence of Tests in Phase 2 of the Work

| Report Test ID | Lab Test ID | Type of Graphite /Powder | Post-Milling Graphite Age (Weeks) | Conc (G/M^3) | Pmax (Bar) Corrected | Dust Pot DP (Bar) | Total Ignitor Energy (Kj) | Kst, Bar M/S 25ms | Time Delay (Ms) | Pot Vol (L) | Comments |
|----------------|-------------|--------------------------|-----------------------------------|--------------|----------------------|-------------------|---------------------------|-------------------|-----------------|-------------|----------------|
| 20 | bnflg70 | P1G | 42.7 | 439.4 | 0.005 | 19.699 | 3.0 | 0.945 | 697 | 5 | |
| 21 | bnflg71 | P1G | 42.7 | 439.4 | 0.025 | 19.863 | 4.0 | 0.920 | 697 | 5 | |
| 22 | bnflg72 | P1G | 42.9 | 439.4 | 0.025 | 20.239 | 4.0 | 0.813 | 697 | 5 | |
| 23 | bnflg73 | P1G | 42.9 | 439.4 | 0.014 | 19.813 | 3.0 | 0.520 | 697 | 5 | New Bag opened |
| 24 | bnflg74 | P1G | 43.0 | 439.4 | 0.019 | 19.733 | 3.0 | 0.668 | 697 | 5 | |
| 25 | bnflg75 | P1G | 43.0 | 439.4 | 0.025 | 20.051 | 4.0 | 0.734 | 697 | 5 | |
| 26 | bnflg78 | P1G | 43.1 | 439.4 | 0.004 | 19.917 | 2.0 | 0.437 | 697 | 5 | |
| 27 | bnflg98 | P1G | 44.3 | 439.4 | 0.498 | 19.416 | 25.0 | 3.334 | 697 | 5 | |
| 28 | bnflg48 | P2CSG | 29.0 | 439.4 | 0.127 | 16.370 | 10.0 | 2.055 | 597 | 5 | |
| 29 | bnflg50 | P2CSG | 29.1 | 439.4 | 0.192 | 19.160 | 10.0 | 2.609 | 697 | 5 | |
| 30 | bnflg60 | P2CSG | 39.0 | 439.4 | 0.167 | 20.470 | 10.0 | 2.192 | 697 | 5 | |
| 31 | bnflg61 | P2CSG | 39.1 | 439.4 | 0.183 | 21.610 | 10.0 | 1.639 | 697 | 5 | |
| 32 | bnflg51 | LO-1000ppm mix | 29.3 | 439.4 | 0.220 | 20.340 | 10.0 | 2.329 | 697 | 5 | |
| 33 | bnflg52 | LO-1000ppm mix | 29.3 | 439.4 | 0.129 | 21.270 | 10.0 | 1.630 | 697 | 5 | |
| 34 | bnflg88 | LO-1000ppm mix | 44.0 | 439.4 | 0.165 | 19.808 | 10.0 | 1.617 | 697 | 5 | |
| 35 | bnflg89 | LO-1000ppm mix | 44.0 | 439.4 | 0.171 | 20.020 | 10.0 | 2.060 | 697 | 5 | |

Table 2-2 (continued)
Sequence of Tests in Phase 2 of the Work

| Report Test ID | Lab Test ID | Type of Graphite /Powder | Post-Milling Graphite Age (Weeks) | Conc (G/M^3) | Pmax (Bar) Corrected | Dust Pot DP (Bar) | Total Ignitor Energy (Kj) | Kst, Bar M/S 25ms | Time Delay (Ms) | Pot Vol (L) | Comments |
|----------------|-------------|--------------------------|-----------------------------------|--------------|----------------------|-------------------|---------------------------|-------------------|-----------------|-------------|--------------------------|
| 36 | bnflg90 | LO-1000ppm mix | 44.0 | 439.4 | 0.317 | 19.939 | 10.0 | 2.327 | 697 | 5 | Ignitor cup not scrubbed |
| 37 | bnflg91 | LO-1000ppm mix | 44.1 | 439.4 | 0.154 | 19.989 | 10.0 | 1.797 | 697 | 5 | |
| 38 | bnflg101 | LO-1000ppm mix | 44.7 | 439.4 | 0.178 | 19.548 | 10.0 | 1.818 | 697 | 5 | |
| 39 | bnflg76 | LO-1000ppm mix | 43.0 | 439.4 | 0.004 | 19.812 | 4.0 | 0.609 | 697 | 5 | |
| 40 | bnflg77 | LO-1000ppm mix | 43.1 | 439.4 | 0.016 | 19.701 | 4.0 | 0.853 | 697 | 5 | |
| 41 | bnflg79 | LO-1000ppm mix | 43.1 | 439.4 | 0.000 | 20.145 | 2.0 | 0.347 | 697 | 5 | |
| 42 | bnflg80 | LO-1000ppm mix | 43.1 | 439.4 | 0.000 | 19.899 | 2.0 | 0.482 | 697 | 5 | |
| 43 | bnflg54 | AO-25% mix | 30.0 | 330.0 | 0.026 | 21.340 | 10.0 | 1.232 | 697 | 5 | |
| 44 | bnflg55 | AO-25% mix | 32.9 | 439.4 | 0.015 | 23.020 | 10.0 | 1.038 | 697 | 5 | |
| 45 | bnflg92 | AO-25% mix | 44.1 | 439.4 | 0.029 | 19.915 | 10.0 | 0.916 | 697 | 5 | |
| 46 | bnflg93 | AO-25% mix | 44.1 | 439.4 | 0.028 | 20.216 | 10.0 | 0.870 | 697 | 5 | |

Table 2-2 (continued)
Sequence of Tests in Phase 2 of the Work

| Report Test ID | Lab Test ID | Type of Graphite /Powder | Post-Milling Graphite Age (Weeks) | Conc (G/M^3) | Pmax (Bar) Corrected | Dust Pot DP (Bar) | Total Ignitor Energy (Kj) | Kst, Bar M/S 25ms | Time Delay (Ms) | Pot Vol (L) | Comments |
|----------------|-------------|--------------------------|-----------------------------------|--------------|----------------------|-------------------|---------------------------|-------------------|-----------------|-------------|------------------------------|
| 47 | bnflg94 | AO-25% mix | 44.1 | 439.4 | 0.031 | 19.637 | 10.0 | 0.652 | 697 | 5 | |
| 48 | bnflg44 | P2CSG | 28.3 | 200.0 | 0.178 | 19.670 | 10.0 | 0.958 | 1361 | 10 | |
| 49 | bnflg43 | P2CSG | 28.1 | 400.0 | 0.240 | 19.470 | 10.0 | 0.835 | 1361 | 10 | |
| 50 | bnflg46 | P2CSG | 28.7 | 400.0 | 0.312 | 17.980 | 10.0 | 3.062 | 871 | 10 | |
| 51 | bnflg42 | P2CSG | 28.0 | 600.0 | 0.246 | 20.270 | 10.0 | 1.867 | 1361 | 10 | |
| 52 | bnflg47 | P2CSG | 28.9 | 600.0 | 0.167 | 19.040 | 10.0 | 2.186 | 1054 | 10 | |
| 53 | bnflg41 | P2CSG | 28.0 | 800.0 | 0.123 | 19.180 | 10.0 | 0.884 | 1361 | 10 | |
| 54 | bnflg95 | Ignitor only | N/A | 0.0 | 0.000 | N/A | 1.0 | N/A | 697 | 5 | Ignitor only- no fuel |
| 55 | bnflg96 | Ignitor only | N/A | 0.0 | 0.000 | N/A | 3.0 | N/A | 697 | 5 | Ignitor only- no fuel |
| 56 | bnflg97 | Ignitor only | N/A | 0.0 | -0.001 | N/A | 5.0 | N/A | 697 | 5 | Ignitor only- no fuel |

Table 2-2 (continued)
Sequence of Tests in Phase 2 of the Work

| Report Test ID | Lab Test ID | Type of Graphite /Powder | Post-Milling Graphite Age (Weeks) | Conc (G/M^3) | Pmax (Bar) Corrected | Dust Pot DP (Bar) | Total Ignitor Energy (Kj) | Kst, Bar M/S 25ms | Time Delay (Ms) | Pot Vol (L) | Comments |
|----------------|-------------|--------------------------|-----------------------------------|--------------|----------------------|-------------------|---------------------------|-------------------|-----------------|-------------|---|
| 57 | bnflg100 | Ignitor only | N/A | 0.0 | 0.000 | N/A | 10.0 | N/A | 697 | 5 | Ignitor only- no fuel |
| | | | | | | | | | | | |
| | | | | | | | | | | | |
| | | | | | | | | | | 5 | Slight discharge injection air and graphite at joint above injection valve, |
| | | | | | | | | | | | Discarded no trigger from comparator, |
| | | | | | | | | | | 10 | Discarded time delay not appropriate, |
| | | | | | | | | | | 5 | ignitor facing away from cup |
| | | | | | | | | | | | |

3

ELECTRICITE DE FRANCE PROGRAM IN ASSOCIATION WITH CNPP

The principal French studies into graphite dust deflagration are documented in reports from INERIS (Institut National de l'Environnement Industriel et des Risques) [49] and from CNPP (Centre National de Prévention et de Protection) [50, 51]. The initial INERIS studies were undertaken to support the proposed long-term storage of the graphite cores within the pressure vessels of the reactors: subsequently, the issue has been more comprehensively addressed with specific reference to decommissioning activities.

EdF-CIDEN made a commendable decision to explore the dust-deflagration issue further by seeking to replicate a real engineering situation with suspended graphite dust and a considerable ignition energy, with the objective of providing a convincing practical demonstration of the essentially benign nature of graphite dust. One of the present reviewers (AJW) was a consultant on the development of these final stages of this program, which was undertaken by the 'National Center for Protection and Prevention' (CNPP) in association with an EdF site at Moret-sur-Loing ("Les Renardières"), near Paris.

Phase 1

As a first step, CNPP conducted some additional basic studies [50] to confirm the initial INERIS results. The basic combustibility of graphite dusts was compared with that of graphite, utilising a simple ceramic 'boat' and a muffle furnace. The samples were as follows:

- Solid graphite
- Graphite dust in the range 100 – 400 μ m
- Graphite dust < 45 μ m
- Graphite dust < 15 μ m
- Graphite dust (70%) with iron dust (30%), all < 45 μ m

The last item was included to mimic the possibility of rupture of a powder-torch hose during cutting of components within the reactor.

In each case, the temperature of the furnace was raised from ambient to 900°C at a rate of 3 – 4 K.min⁻¹ and the extent of mass loss noted. As these conditions, in an essentially free supply of air, would be expected to result in significant oxidation of bulk graphite, let alone dusts, it is perhaps unsurprising that these materials all suffered significant mass loss, with some 32% loss from the solid graphite, and total oxidation of the samples with particle sizes of 45 μ m or less. Just 54% mass loss however occurred in the case of the mixture with iron powder, but this could result from total loss of the carbon if the iron is oxidised to Fe₂O₃.

Overall, these test results are unsurprising. A significant oxidation rate was first observed with the dust at a temperature of around 580°C.

CNPP also conducted tests with graphite dust preheated to 850°C, following which they then utilised (i) an oxy-acetylene torch, and (ii) dropping molten metal. The first caused the dust to become suspended and ignite, which is essentially unsurprising, whilst the molten metal had no effect.

With dust at ambient temperature, a sample was then saturated in hydraulic oil following the French absorption standard NF T 90-361. It is to be noted that the reporting of this work in [50] is erroneous, and the correct ratios of oil to graphite are 173: 100 by weight for the graphite of 100 – 400µm granulometry and 215:100 for that of 45µm material. Thus, the dusts became aggregated with more than their own mass of oil under the specified conditions (essentially immersion for 20 minutes followed by draining for 30 minutes). The oil-soaked dusts of each of the specified granulometry were then each subjected to the flame of a “camping-gas” burner, and to particles of molten steel produced by a blowtorch, by arc welding, and by a grinder. Each of these processes, except for the grinder, produced some ignition of the oil-soaked material. With the camping-gas burner, applied for 30 seconds, it was slow: with the other methods producing molten metal, there was extinction in less than 10 seconds except for the intermediate particle size (< 45µm) which took up to one minute. The reason for this is not understood. Propagation of the flame beyond its initial area was negligible.

The final test done in this preliminary CNPP work was to inject dusts into a vertical cylinder containing an open flame at the base, in order to compare the behaviour of graphite dusts of various granulometries with that of iron dust and the standard maize flour. The dusts were blown from a 100g heap in small ‘puffs’ into a cylindrical aperture in the side of the bottom of the cylinder using an air gun. The initial flame height was approximately one third of the height of the cylinder, which was a 60 cm long sleeve with a triangular opening at the base.

There were three thermocouples at different heights within the cylinder. Only the lowest of these recorded changes of significance. From an initial temperature of 670°C, graphite of all three granulometries, and the graphite/iron mixture produced a temperature increase to 750°C, whilst iron powder alone produced 800°C and maize flour 900°C. Flames were emitted from the top of the cylinder only with the iron powder and, more significantly, with the flour: graphite/iron – approximately 30 cm in height above the top of the cylinder, iron alone approximately one metre, and with maize flour 1.2 m.

Whilst these initial CNPP tests do not constitute a suitable matrix of tests to provide an envelope for assessing the behaviour of the graphite dusts comprehensively, they are in accord with the observations from other laboratories. Reference 50 concludes that graphite dusts of fine granulometry may be inflamed under certain conditions (*e.g.* oil soaking) and that suspended dust at a concentration of at least 60 g.m⁻³ may be inflamed if a sufficient energy source is present. Nevertheless, the dusts are low in reactivity with air in comparison with iron powder and maize flour.

Phase 2

This second phase of the CNPP work, documented in detail in [51], was itself conducted in two stages. The first covered the development of an opacity methodology for the determination of dust concentration in air, and the second the utilisation of high-speed photography to study the propagation of flame fronts in a ‘semi-confined environment’ - a transparent pipe of size comparable to realistic situations likely to be encountered in decommissioning a commercial reactor – essentially with the objective of proving that, even if a major ignition source were present and a suspension of fine graphite dusts occurred, a flame front moving along a pipe of typical plant dimensions would be self extinguishing. It is with this final study that one of us (AJW) was engaged², and it was developed with the recognition that routine studies in equipment such as the ISO vessel, although important, were not directly representative of the conditions to be encountered in reactor decommissioning.

The development of the opacimeter was also conducted in a plexiglass tube, vertically mounted and with a capacity of 200 litre.



Figure 3-1
CNPP Apparatus for the Determination of the Relationship between Opacimeters and Graphite Dust Concentration

² It is felt important to note here that this consultancy was limited to the general design of the tests and not with specific operational decisions such as the use of specific graphite concentrations or the removal of the opacimeters, discussed later.

The vertical tube was mounted on a closed plexiglass base and the upper end was closed by a further transparent plate. Graphite dust was introduced by a pulse of air delivered to a reservoir of graphite dust such that it was discharged around a circular tube at the base of the plexiglass tube. The injection system was extremely efficient and the average concentration of dust in the vessel could be calculated accurately. The tube was fitted with an upper and a lower opacimeter, each consisting of opposed light emitters and receivers linked to a simple electronic measurement system. The test allowed the determination of the relationship between the dust concentration and the light transmitted, which is the difference from 100% of the opacity value.

For the opacimeter studies, UCAR graphite grade GS45E³ was used to prepare a powder of which 90% was < 49.53µm (30% lay between 25 and 45µm, 31% between 10 and 25µm and 29% < 10µm). The density of the settled but unpacked powder was 0.34 g.cm⁻³.

After 'zeroing' the output of the opacimeter, the valve was adjusted to blow in the dust, and the gas pressure was maintained for 10 seconds to ensure the best possible mixing of the dust with the air inside the tube. Numerous injections were made, exploring the best separation of the emitters and receivers, which was eventually set at 10 cm for the formal calibration tests. Some systematic differences were observed between the readings of the upper and lower opacimeters, the lower one consistently showing a greater reduction in light transmission than the upper one, indicative of a slightly higher dust concentration in the lower part of the tube, explained in [51] in terms of the mode of dust injection. The maximum recorded relative difference was 27% at a mean dust concentration of 25 g.m⁻³. There was also a modest scatter in all data which was, however, not sufficient to obscure this systematic difference. A 'global' average of all measurements, including both detectors, was utilised in forming the relationship between concentration and opacity.

The formal calibration utilised dust concentrations of 25, 50, 100 and 150 g.m⁻³. At this highest concentration it was clear that the instrumentation was approaching saturation, and this represents an upper concentration limit for this detector system unless it is possible to place emitter and receiver much closer together. The relationship in Figure 3-2 was obtained:

³ This grade of graphite is not utilised in any French reactor system, nor indeed, in any of the others facing decommissioning in the other nations participating in this comparison. However, this is not regarded as significant in terms of the performance of the dust.

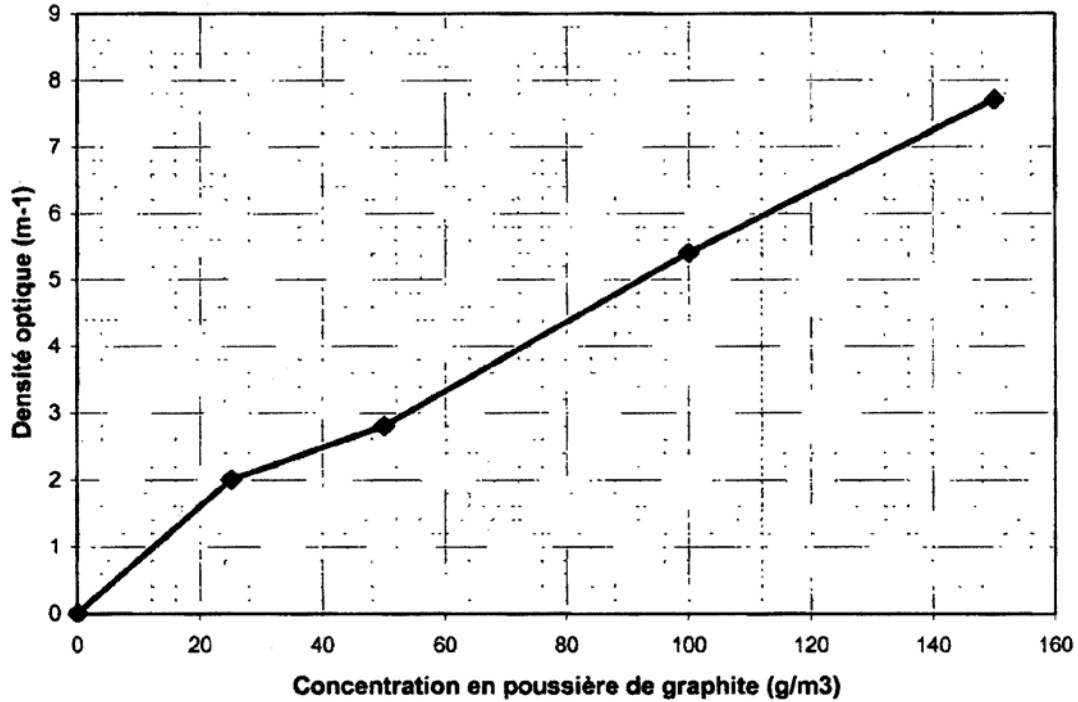


Figure 3-2
Opacimeters with 10-cm Separation between Emitter and Receiver (*Relationship between Graphite Dust Concentration and Optical Density*)

Two opacimeters were incorporated into the design of the large horizontal plexiglass tube used for the major part of the experimental program.

This tube was a little over 4 metres in length, approximately 50 cm diameter and with an internal volume of 742 litres. In this case, a two-section dust-injection system was employed to ensure an even distribution of material throughout the tube. In each half of the tube, a metal channel was placed containing evenly-distributed graphite dust. Across the top of each channel passed a perforated pipe, the air pulse emitted from it blowing downwards on to the channel of graphite dust, rapidly raising a quasi-uniform dust cloud. At one end of the tube was the ignition system, which is discussed below. At the opposite end of the horizontal tube, a pressure-relieving filter system was installed.

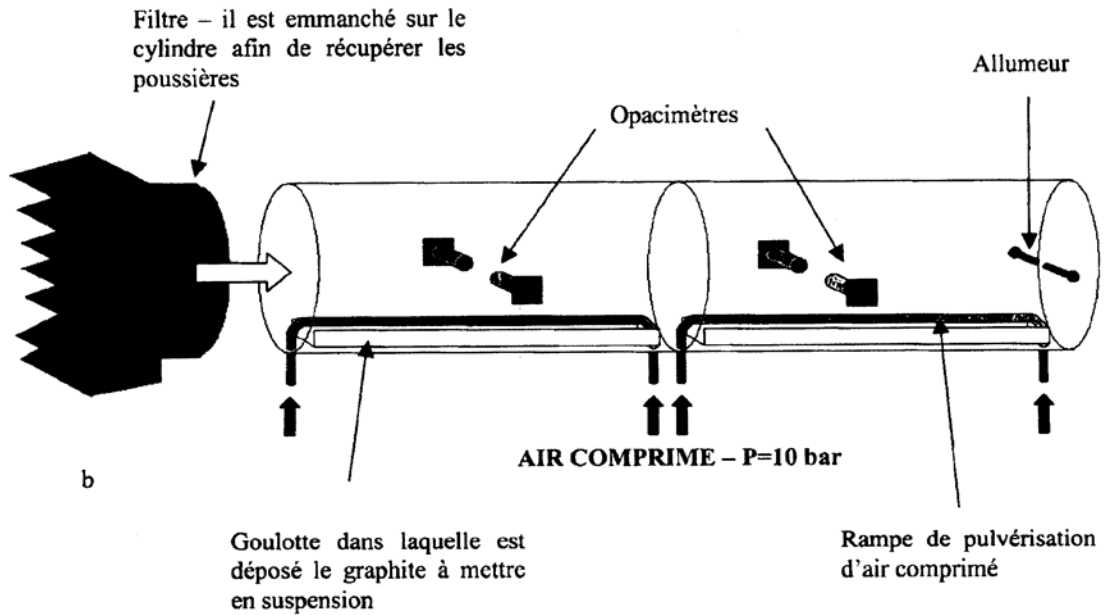


Figure 3-3
The CNPP Flame-Propagation Apparatus, showing the Position of the Igniters ('Allumeur'), Graphite-Dust Channels ('Goulettes'), Air-Injection System, Pressure-Relieving Filter, and Opacimeters



Figure 3-4
Detail of Gas-Injection System and Graphite-Dust Channel

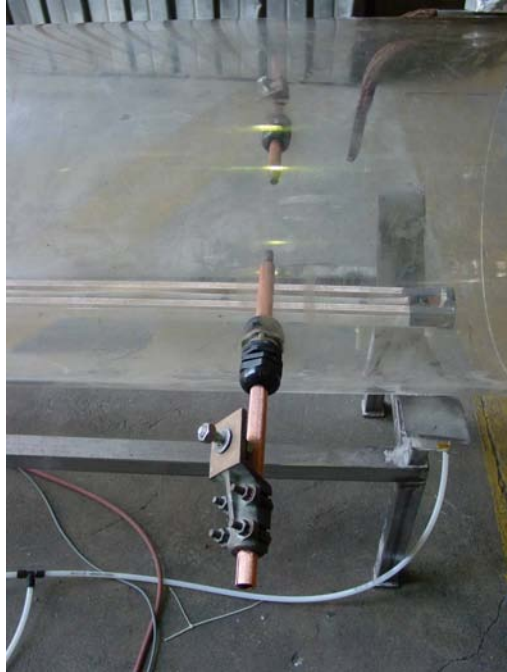


Figure 3-5
Detail of Electrodes used for Ignition



Figure 3-6
General View of CNPP Apparatus in Position at Renardières

The electrodes consisted of graphite covered with copper at a spacing of 16 cm, disposed 35 cms from the closed end of the plexiglass tube. The arc was initiated between the electrodes with the aid of a fusible silver filament. The arc was driven from a 430V AC source at 14 kA, for durations of 10, 20 or 30 milliseconds, corresponding to an input of 60, 120 and 180 kJ. Clearly, these energies are much larger than those utilised in, for example, the standard ISO test, but were chosen such that the range encompasses three possible accident scenarios: (i) an electrical breakdown in a robotic handling device operating at 410V and 30 kW; (ii) the same in a remote platform at 75 kW; and (iii) a plasma torch operating at 720 kW and 230V.

Indicative measurements of temperature and pressure within the apparatus were effected in order to allow comparison of the nature of the pressure and temperature waves under different conditions.

The progress of each test was recorded using a high-speed camera system operating at 250 images per second: this has enabled some remarkable and convincing images of the relative performance of different dusts to be recorded.

A strict protocol was followed between tests. After each one, the apparatus was thoroughly cleaned and wiped out, with removal of all residual powder. The channels were then carefully re-filled with the appropriate amount of powder for the next test. For tests with graphite this was generally 480 g. A new silver filament was attached to the electrodes, and the length of the tube beyond the electrodes was shielded with a polyester plaque. The filter was replaced if necessary. The opacimeter trajectory was checked to be 10 cm and the zero of optical density re-set.

Next, the voltage, intensity (current) and duration of the required electrical arc were set, the time of air injection to suspend the powder and the interval before ignition were pre-set and the camera pre-trigger set, such that it would commence recording a few milliseconds before the ignition arc was struck. For the principle tests, 3.5 seconds were allowed for suspension before the arc was triggered, and the air injection was maintained for a further 3.5 seconds.

The apparatus was initially calibrated with a fine wheat flour, 'Francine Fluide', using a 330V arc, 5 kA current, a 10 ms arc, four seconds of suspension before striking the arc with the air injection maintained for a further three seconds. 1460 g flour was used (slightly under 2000 g.m^{-3} if all was suspended). In accordance with expectations, the discharge caused ignition of the flour, with propagation along the length of the tube with a pressure pulse sufficient to damage the filter system slightly. A comprehensive video record of this exists to illustrate that the apparatus responds positively to a significant deflagration and propagation of the flame, in any strongly-explosible substance (see Figure 3-7)



Figure 3-7
Propagation of Flame Front in Wheat Flour in the CNPP/Les Renardières Test, Illustrating the Expected Performance of the Test Rig with a ‘Highly Explosible’ Substance. Here the Flame Approaches the End of the Tube having Travelled a Distance of Approximately Four Metres

A point of significance regarding the behaviour of the wheat flour was that the flame gained in intensity as it approached the ‘open’ end of the tube, indicating that it was receiving a better supply of oxygen at that time and, hence, that there was some oxygen deficiency within the tube during the ignition of the flour. A period of latency took place after the brightness of initial heat subsided : with graphite this ended with complete extinction, whereas in flour tests it was followed by a renewal of the combustion. This latency might be associated with a progressive deepening of oxidation within each particle, which was prevented in graphite by high thermal conductivity and lack of volatiles.

The principal results obtained using graphite powder are recorded in Table 3-1. In each case, a corresponding test utilising the same ignition parameters was recorded with air alone. This was necessary for comparison purposes because the discharge at the electrodes was sufficient to vaporise some graphite, admixed with copper, to give a visible ball of flame in the vicinity of the electrodes. In the video records of which an example is given below, each image is therefore a composite showing the system at the same elapsed time with and without graphite.

At this stage of the studies, the graphite source was crushed unirradiated fuel-sleeve graphite from batches intended for either the Chinon or St. Laurent reactors.

Table 3-1
Principal Results using Graphite Powder

| Test | Medium | Arc Energy (kJ) | Duration of Arc (ms) | Duration of Incandescence (ms) | Maximum Distance Traveled by Flame (m) |
|------|----------|-----------------|----------------------|--------------------------------|--|
| 914 | Air Only | 59.5 | 9.9 | 80 | 0.90 |
| 915 | Air Only | 62.5 | 9.8 | 76 | 0.83 |
| 3006 | Graphite | 65.2 | 10.0 | 156 | 1.04 |
| 3007 | Graphite | 70.2 | 10.2 | 104 | 1.16 |
| 909 | Air Only | 113 | 19.3 | 156 | 1.28 |
| 911 | Air Only | 106 | 19.3 | 144 | 1.61 |
| 3004 | Graphite | 110 | 19.2 | 156 | 1.40 |
| 3005 | Graphite | 112 | 19.3 | 168 | 1.17 |
| 907 | Air Only | 170 | 29.4 | 200 | 1.35 |
| 908 | Air Only | 165 | 29.4 | 216 | 1.66 |
| 3002 | Graphite | ND | 29.4 | 508 | 1.91 |
| 3003 | Graphite | 164 | 29.4 | 500 | 2.25 |

As may be seen, the results are grouped by ignition energy, and shown alongside the corresponding results for air alone. This is an important comparison because the electrodes alone produce significant incandescence due to evaporation and oxidation of the graphite and copper, which moves along the tube. Thus, the results for graphite need to be viewed in comparison with this ‘graphite-free’ equivalent, and three sets of time-lapse photographs are shown below.

A further point of interpretation of this table is the final column – maximum distance travelled by flame. Generally, the observed flame travels a little further in the presence of the graphite. Since the graphite is oxidisable, and significant energy is initially transferred to the suspended material close to the electrodes, this is not surprising. However, inspection of the visual record then shows clearly that the flame front is slowed, and eventually extinguished with termination of oxidation, in contrast to the behaviour of the wheat flour illustrated earlier.

A final significant point is that the standard concentration of graphite used in the tests – equivalent to 647 g.m^{-3} if suspended with 100% efficiency, which is unlikely – greatly exceeds the measurable range of the opacimeters, from which no usable data were therefore recovered. Indeed, [51] records that the opacimeters were also an obstacle to combustion and retarded the propagation of the flame, so it was decided to remove them altogether once their unflinching saturation (guaranteeing a concentration of graphite dust in excess of 150 g.m^{-3} and the general homogeneity of the dust suspension had been confirmed. By definition, this ‘confirmation’ must have been conducted with a lesser amount of graphite dust. The specific graphite concentration chosen is not adequately explained in [51]. It lies on the shoulder of the ‘peak’ identified by the INERIS study, but not at a sufficiently high level to cover the apparently anomalous high result at 1250 g.m^{-3} which we have already challenged. It is assumed that the chosen concentration

provided the best visual records, and this is important: the great value of this test is the absolute confirmation in a ‘real’ situation that a large-scale graphite-dust deflagration is self-extinguishing whereas a ‘strongly explosible’ material like wheat flour clearly produced a ‘positive’ result sufficient to blow out the filter⁴ and cause (according to anecdotal reports) ‘a number of little fires’ on the floor area surrounding the apparatus.

It was also noted that the pressure signal was extremely weak, with changes of only a few millibars. Anecdotal remarks indicate a maximum pressure increase of 6.4 mbar for a 115 kJ ignition, accompanied by a sound ‘approximating to 55 Hz’ – in other words, a low-pitched ‘*whoomph*’ rather than a sharp report as from a detonation. Whilst this behaviour would be expected for a deflagration, there is reason to believe that this sound was associated with the destruction of fragments of the electrodes since it occurred in the absence of graphite, this being corroborated by the photographic evidence of the behaviour of graphite dusts within the tube and by its resonance being correlatable with the length of the tube. It was also noted anecdotally that there was no significant temperature change recorded at the surface of the tube in any of the tests.

Three sets of stills from the high-speed photography were obtained, each corresponding to one zone of ignition energy from the above Table 3-1. The first compares runs 914 and 3007, the second 911 and 3005 and the final set 907 and 3002.

In each case, the evidence for the extinguishing of the flame front in the presence of graphite is unequivocal. The stills comparing runs 914 (air only, upper images) and 3007 (with graphite, lower images) are given as an example in Section 4.

The conclusions of [51] in regard to this interpretation of this work may be summarised as follows:

- The zone of incandescence produced from the *electrodes* is of the same size in the reference tests with air alone as in the presence of graphite for the same energy input
- In the presence of a graphite dust suspension, the graphite particles are initially heated by the electric arc and may reach temperatures higher than 1000°C: in opposition to this effect, the presence of the graphite may also suppress somewhat the visible emissions from the electrical arc
- The incandescence within the graphite dust then expands in volume as a result of the heating (some surface oxidation of the hottest particles also occurs, making a further exothermic contribution, and this contributes to an extension of the duration of the visible incandescence)
- The incandescence within the graphite dust suspension does not produce energy sufficient to compensate for losses through conduction, convection and radiation: in consequence the dust suspension cools and the incandescence is extinguished – this despite modest blowing of fresh air from beneath
- Under the conditions of this experiment, there was no deflagration in the graphite dust

⁴ In a number of cases the filter was also blown out in the tests with graphite, presumably through the pressure wave produced from the electrical arc.

Whilst this experiment, designed to allow reproducible testing conditions, was far from a 'perfect' replication of a realistic situation in decommissioning, the inability of such significant energy inputs to create a deflagration gives a large degree of support to the view of the present authors that there is no significant risk of encountering dust-deflagration issues in the course of decommissioning a graphite-moderated reactor.

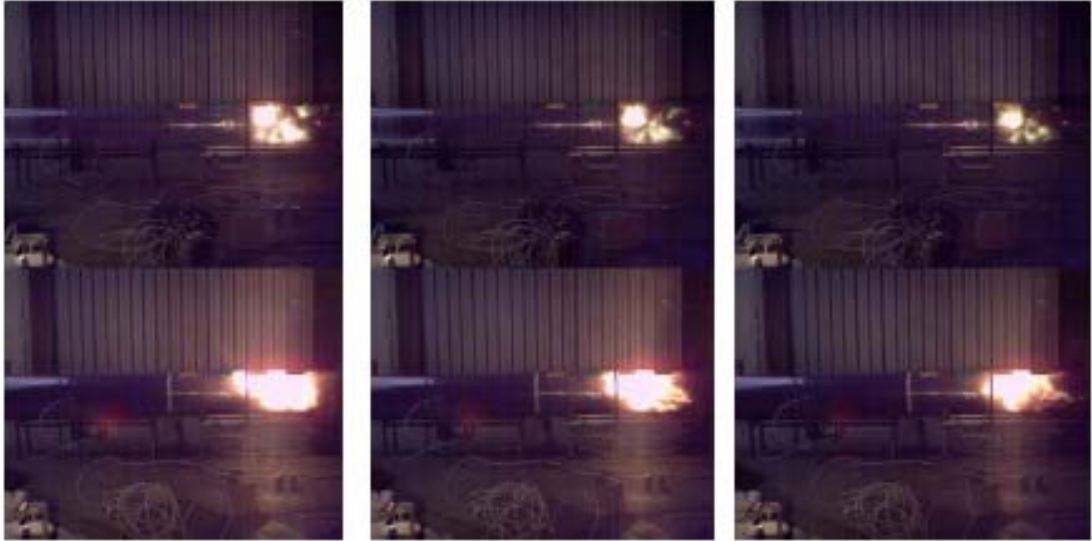
4

STILLS FROM THE HIGH-SPEED PHOTOGRAPHY EMPLOYED DURING THE CNPP/EDF FLAME- PROPAGATION STUDY AT MORET-SUR-LOING (“LES RENARDIÈRES”)

Tests 914 and 3007

For these tests, the ignition energies were 59.5 kJ (914, air only) and 70.2 kJ (3007, with graphite). In each case the air-only result is the upper photograph. Images were obtained at intervals of 4 ms.







5

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