THE PRESENT STATE OF DATA IN THE SAFETY ANALYSIS OF THE HTR

R.H. FLOWERS \ AERE, Harwell
J.B. SAYERS
M.S.T. PRICE \ Dragon Project
CONTENTS

1. Introduction
2. Release of fission products from coated particles
3. The interaction between fuel particle quality and in-reactor behaviour.
4. Matrix effects
5. Matrix-fuel tube interface
6. The fuel tube
7. The fate of fission products in the primary circuit
8. Conclusions.
TABLES

1. Diffusion coefficients of fission products in isotropic pyrocarbon at 1250°C.

2. Estimation of defective coatings from Sr release results.

3. Frequency distribution of breakages in fuel compacts for all measurements by Dragon Project.

4. Parameters affecting pressure failure statistics

5. Kernel porosity estimates.

6. Dimensional variations of the buffer layer

7. Coating data from fuel batches fabricated for the UKAEA Mk.III Reactor Physics Programme.

8. Between batch density variations in SiC and outer pyrocarbon layers.


10. Data used for particle endurance calculations.
FIGURES

1. Variation in release of Xe$^{133}$ as a function of burn-up for the Studsvik 17 test.

2. Variation in burn up at failure due to statistical variations in various particle parameters.

3. Variation in burn up versus fracture stress in SiC coat.

4. Decrease in particle life as a function of temperature transient.

5. Effect of matrix and fuel tube graphite characteristics on Cs$^{137}$ release.

6. Diffusion coefficients for fission product metals in matrix graphites.

7. Apparent partition factor for Cs$^{137}$ between natural graphite matrix and GILSCOCarbon fuel tube at the 1 $\mu$g/g level.

8. Diffusion coefficients for fission product metals in fuel tube graphite.

9. Vapour pressures of caesium and strontium over matrix and fuel tube graphites as a function of temperature at a characteristic loading of 0.1 $\mu$g/g.
1. **INTRODUCTION**

The use of a large number of small coated fuel particles, and the unusual possibility of knowing fission product diffusion rates in the canning materials, has led to the expectation of a much more quantitative analysis of fission product release from the HTR core than from any previous reactor type. This analysis has shown that in normal operation there must be a minute but predictable release, and at first sight this may appear to be a retrograde step in fuel design. It is only when we take into consideration the fact that the release is not only small and predictable, but also directly controllable by specification of fuel design, that we can appreciate that it represents a considerable step forward. The customary situation is that an essentially zero fission product release may be interrupted by large unpredictable releases due to can failure and activation products from in-core parasitic metals provide a significant and irregular hazard. In the HTR we have a core from which unforeseen release of fission products may be classified 'incredible' with a clear conscience and in which activated impurities will probably be at a much lower level due to the absence of metals in the core. These advantages are however dependent upon a reliable set of data relating to particle failure and fission product transport.

There is an understandable initial tendency for only the average properties to be designated in a reference fuel specification. The commercial considerations which restrict available data on fuel to mean values are paralleled by the simplifying assumptions of safety calculations which generally assume that the whole population of fuel is of exactly average quality. The tendency for initial scoping work is also to concentrate on average values.

It must be realised that the nature of HTR fuel and its fabrication methods are such that there is a statistical distribution of all its characteristics. Normally it is one tail of the distribution which is important. Realistic maximum proportions of the population having a lower quality than that defined by theoretical calculations or experimental results must be defined for each characteristic. The approach to this definition can be assisted by knowledge of the distribution presently found in manufacture.

Similarly as the HTR moves into the industrial phase there is an important need to characterise the distribution of the various parameters within the fuel produced. It is therefore necessary not only to measure within and between batch variations but also to estimate or guess the shape of the distribution otherwise statistically valid conclusions cannot be drawn. Distribution data within and between batches is thoroughly reliable only when production is under way. However, it is necessary to make estimates of within and between batch quality variations prior to production since these could affect the results of safety calculations.
Accidents are a key subject in any safety analysis. In particular we must consider:

(a) depressurisation faults,

(b) temperature transient due to channel blockage, circulator failure or control rod malfunction,

(c) boiler tube rupture.

In Paper No. 11 the point has been made that the HTR fuel element can withstand many hundreds of degrees excess temperature without structural failure; the important consequence of such a transient is therefore the possible breakage of particle coatings, since the reactor is likely to be designed to run as close as practicable to the point at which particle failure in the hottest and highest burn-up particles is imminent.

The design of the HTR fuel gives a valuable defence against the consequences of coating failure during overheating incidents both on account of the inevitable spread of failure temperatures amongst the tens of thousands particles at risk even in the most localised incident, and of the long delays involved in metal fission product diffusion in the fuel tubes. This should allow a procedure whereby failures are detected at an early stage relative to metal canned fuels, and also the identification and removal of elements long before they have caused any fuel or long-lived fission products to contaminate the reactor circuit. It is obvious these advantages are similarly dependent upon a good knowledge of particle endurance functions and of fission product diffusion and evaporation rates.

The depressurisation and water ingress accidents have to be analysed taking into account the fission products which have plated-out in the primary circuit during 30 years of operation. In this area there is considerable uncertainty in estimates of adsorption and desorption parameters, and pessimistic values must be used in current calculations. This is the situation with all types of gas cooled reactors, but in view of the closely controlled chemical composition of a helium coolant there is reason to expect advances in the quantitative treatment of these accidents in the HTR. The significance of water ingress would clearly be less with the adoption, at a later stage, of a direct cycle with a gas turbine.

This paper aims to discuss the present reliability of data relevant to the release of fission products under normal and defined accident conditions. It does not present complete calculations of fission product release since other papers at this symposium cover that aspect but it should indicate the probable accuracy of such calculations. Underlining what has been said earlier, it must be realised that data which is presently available generally derives from pilot or small scale fabrication. As a consequence of this and also because of the time required to complete and evaluate irradiation experiments, it can be inferred that present day data is pessimistic.
2. CALCULATION OF FISSION PRODUCT RELEASE FROM HTR FUEL ELEMENTS

Coated Particles

Our knowledge of fission product release from HTR fuel elements is based upon six years of observation of fuel elements in the Dragon reactor but also relies heavily upon measurements of diffusion and evaporation rates for non-gaseous species made in test reactor loops or in laboratories in many parts of Europe and the U.S.

The starting point for any HTR safety analysis must be the knowledge of the fission product source term and its distribution throughout the reactor. At the start of life this is made up from three sources which can be considered as uniformly distributed throughout the core.

(a) Uranium contamination in the outer PyC coat

(b) The proportion of particles without SiC or with defected SiC coating (described by the equivalent defective fraction $\phi'$).

(c) Proportion of particles having all coatings broken due for example to compaction (described by the equivalent broken fraction $\phi$).

The uranium contamination in the outer PyC coat in as-produced particles is measured routinely and is normally very low ($<10^{-6}$) (46).

The most important parameters in this calculation therefore concern the quality of the pyrocarbon and silicon carbide coatings on the fuel particles, and a few words on quality definition are necessary at this point.

It is broadly true to say that the release of rare gases and halogens through a high density isotropic pyrocarbon coating is zero for a fuel dwell time of 1000 days. Release has never been observed at any temperature except as a result of mechanical damage to the pyrocarbon by internal pressure or by 'amoeba' corrosion. This can be understood as a complete lack of the chemically bound surface states by means of which a fission product might move between crystallites. All metallic fission products, on the other hand, diffuse at a measurable rate through pyrocarbon, the rates reaching a maximum in the alkali earth metals at temperatures of interest (35) (Table 1).

<table>
<thead>
<tr>
<th>Species</th>
<th>$D_{(cm^2/s)}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe</td>
<td>$&lt;10^{-14}$</td>
<td>8, 9</td>
</tr>
<tr>
<td>I$_2$</td>
<td>$&lt;10^{-14}$</td>
<td>estimate</td>
</tr>
<tr>
<td>Cs</td>
<td>$8 \times 10^{-12}$</td>
<td>1, 2</td>
</tr>
<tr>
<td>Sr</td>
<td>$6 \times 10^{-9}$</td>
<td>4</td>
</tr>
<tr>
<td>U, Th</td>
<td>$8 \times 10^{13}$</td>
<td>6</td>
</tr>
</tbody>
</table>
For a dwell time of $10^8$ seconds and a pyrocarbon thickness of 70 $\mu\text{m}$ we can calculate that a diffusion coefficient of $10^{-14} \text{cm}^2/\text{s}$ or less will allow less than $10^{-4}$ fractional release of kernel contents, assuming a unit partition factor on both sides of the coating. The fact that all significant rare gas and iodine isotopes are of less than 10$^6$ seconds half-life makes it quite certain that there can be no diffusional release, and this is in accord with in-pile observations (7). At higher temperatures the situation is probably little changed, since the activation energy for rare gas diffusion is less than 40 kcal/mole (9) and the damage to coatings obscures any small diffusion release.

The situation with metal fission products in pyrocarbon coatings is quite different. There is a gradual increase in diffusion rate along the series Zr, U, Th, Ge, Ce, rare earths, Sr, Ba and it is necessary to evaluate the probable release of each metal at the temperature of interest. In fact, at 1250°C there would be a significant release of Sr, Ba, rare earths and Cs in $10^6$ seconds, and possibly even some U, Pu release. In current safety calculations no retention by pyrocarbon is assumed for Sr and Ba, and an allowance is made for some release of other metals.

Release of all fission products through pyrolytic SiC coatings is so small, and so dependent upon structure, that the measurement of diffusion coefficients is extremely difficult and unrewarding (11).

One suggested approach (10) to quantifying the quality of pyrocarbon and silicon carbide coatings, in a very large assembly of particles such as a fuel pin, is to assume that a perfect pyrocarbon coating releases no rare gas and that a perfect SiC coating releases no metals. Two quality indices, $\phi$ and $\phi'$, are then ascribed to the assembly to show the degree of imperfection in the two coatings. $\phi$, the equivalent broken particle fraction, defined as that fraction of uncoated kernels which would give the same steady-state rare gas release on the particle assembly in question, and $\phi'$, the equivalent defective particle fraction, is defined as the fraction of pyrocarbon coated particles which would give the same steady state Sr release as the particle assembly in question.

In making a core fission product release estimate the quantities $\phi$ and $\phi'$ are used as simple multipliers on the fission rate in each core zone in order to obtain a well defined source term. It follows that we must have an empirical knowledge of the rare gas and iodine release from our standard uncoated kernel, as a function of temperature and burn-up, and a similar knowledge of the release of each metal fission product from a pyrocarbon coated kernel, but the summation of all the individual coating defects and breakages is conveniently represented by just the two measurements on the actual assembly of coated particles. The $\phi$ and $\phi'$ values may increase with burn-up, due to coating deterioration, and this must be taken into account in the assessment, as discussed later.

There may be a small contribution to $\phi$ and $\phi'$ from uranium contamination in the outer PyC, but in fact Dragon Project data (46)
from a large number of coating runs show that the, provided a SiC interlayer is present, average 0 contamination fraction is only $1 \times 10^{-6}$ using current production practice. Therefore release from broken and defective particles will remain the dominant source term for fission products.

We should now enquire about the accuracy with which the components of this calculation are known at the present time.

(a) The assumption of zero gas and iodine release from an intact pyrocarbon coating is well supported by attempts at measuring D, and by R/B values from in-pile experiments where uranium contamination is often the sole cause of gas release even after over 100 days at power. Note that the use of $\phi$ takes care of cracks in the pyrocarbon but would not allow for an eventual breakthrough of a diffusion front. However, we can safely ignore that possibility at temperatures up to 1400°C.

(b) The assumption of a zero metal release from an intact SiC coating is justified by reference to the results of long irradiations in the Dragon reactor. For example, after 350 days at 1200 - 1250°C (12) reference triplex coatings on carbide kernels (which are known to represent a higher strontium metal diffusion potential than oxide kernels (13) released less than $10^{-4}$ of the kernel contents of 89 Sr, 90 Sr and 137 Cs. Moreover this small release was in the form of isolated spots in the matrix and fuel tube proving that even that had arisen from faults in isolated particles. A similar result was obtained at 1550°C for 80 days (14). We could reasonably conclude, therefore, that at temperatures up to 1250°C the release of any metal fission product through intact SiC will be less than $10^{-4}$ in a full dwell time, and that releases from defects in the SiC will be characteristic of release from a pyrocarbon coated kernel, with respect to temperature dependence and relative metal release rates.

(c) It is necessary to know $\phi$ and $\phi'$ for the coated particles to be loaded into the power reactor. Assuming that we require a fission product core release prediction accurate to within a factor of 5 then it will be necessary to guarantee $\phi$ and $\phi'$ values, at least for the hotter fuel, to within about a factor of 2 at the $10^{-4}$ level. At present it is known that routine acid leaching techniques can achieve a high accuracy in free uranium determination (15), and that this does correspond to $\phi$ within a factor of about two (10). Depending upon the sample sizes required to give the required confidence limit, other techniques may also be used for measuring $\phi$ and are capable of this factor 2 accuracy (10, 16).

A chemical method has been applied by the Dragon Project to monitor all fresh fuels for irradiation in Charge IV, and so it is possible to look at the current achievement in $\phi$ values. Estimated values for the equivalent broken particle fraction are given in Table 3, which gives the results of measurements
<table>
<thead>
<tr>
<th>No. of Equivalent Breakages Per Compact</th>
<th>DRIVER FUEL BODIES</th>
<th>EXPERIMENTAL (UO₂) FUEL BODIES</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ANNULAR</td>
<td>TELEDIAL</td>
</tr>
<tr>
<td>No. of Compacts In Category</td>
<td>Total No. of Equivalent Breakages</td>
<td>No. of Compacts In Category</td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>80</td>
</tr>
<tr>
<td>1</td>
<td>118</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>63</td>
<td>126</td>
</tr>
<tr>
<td>3</td>
<td>18</td>
<td>54</td>
</tr>
<tr>
<td>4</td>
<td>7</td>
<td>28</td>
</tr>
<tr>
<td>5</td>
<td>4</td>
<td>20</td>
</tr>
<tr>
<td>6</td>
<td>8</td>
<td>48</td>
</tr>
<tr>
<td>7</td>
<td>4</td>
<td>28</td>
</tr>
<tr>
<td>8</td>
<td>3</td>
<td>24</td>
</tr>
<tr>
<td>9</td>
<td>3</td>
<td>27</td>
</tr>
<tr>
<td>11</td>
<td>3</td>
<td>33</td>
</tr>
<tr>
<td>12</td>
<td>2</td>
<td>24</td>
</tr>
<tr>
<td>14</td>
<td>2</td>
<td>28</td>
</tr>
<tr>
<td>15</td>
<td>3</td>
<td>45</td>
</tr>
<tr>
<td>17</td>
<td>1</td>
<td>17</td>
</tr>
<tr>
<td>18</td>
<td>1</td>
<td>18</td>
</tr>
<tr>
<td>19</td>
<td>3</td>
<td>57</td>
</tr>
<tr>
<td>39</td>
<td>1</td>
<td>39</td>
</tr>
<tr>
<td>43</td>
<td>1</td>
<td>43</td>
</tr>
<tr>
<td>49</td>
<td>1</td>
<td>49</td>
</tr>
<tr>
<td>58</td>
<td>1</td>
<td>58</td>
</tr>
<tr>
<td><strong>TOTALS</strong></td>
<td>648</td>
<td>884</td>
</tr>
</tbody>
</table>

**Average no. of Particles Per Compact**: 1 x 10^4

**Average Breakage Level**: 1.36 x 10^-4

**Overall Average Breakage Level**: = 1.2 x 10^-4
on all fuel bodies measured by the Project, excluding those seeded with bare kernels.

One might expect that the distribution of equivalent breakages would broadly follow a Poisson distribution and this has been reported by Audebeau (47) for Dragon driver fuel for 4 fuel elements manufactured over a limited time with the same conditions. The data given in Table 3 does not closely follow a Poisson distribution. This is to be expected since it is composed of a variety of different fuels manufactured under differing conditions. The simplest deduction is that no fuel body has been found by the Project to have more than 58 equivalent breakages in a sample population of 1371 compacts (containing a total of 1351 breakages).

The evidence points to a low probability of obtaining a high proportion of breakages. The adventitious intrusion of another type of breakage distribution although most unlikely cannot be excluded. It can be eliminated by either:

(i) development of the NDT methods mentioned above, or

(ii) reliance on the broken particle detection gear using techniques developed by the Project (50) to permit early removal from the reactor.

More disturbing from the point of view of control of production is the fact that the average breakage level is \(~ 1.2 \times 10^{-4}\). When the out-going quality is near to the specification limit a high rate of inspection is required. We can note, in passing, that the primary coolant activity predicted by Gulf General Atomic for Fort St. Vrain is based on a value of \(<1 \times 10^{-4}\) at the start of life (49).

It is evident that the achievable levels of broken particle fraction are so low that consequent effects on matrix dimensional behaviour can be neglected. Evidence from Peach Bottom and from special experiments in the Dragon reactor would indicate that a particle failure fraction of \(~0.5\) is required before the matrix dimensional behaviour is seriously affected.

The allowable tolerance in \(\phi^i\), or indeed the absolute magnitude of \(\phi^i\), have not recently been assessed. Under the assumption of zero 137 Cs release from pyrocarbon coatings, and ignoring silver isotopes, a maximum permissible value of \(~0.65\%\ was derived for \(\phi^i\) (17). There is little doubt that a revised value, taking the above factors properly into account would require that \(\phi^i\) be reduced to at most the \(10^{-3}\) level, and we should aim to guarantee it to within a factor two. At the present time there is no established method for providing this guarantee. One can only comment that Charge II fuel in Dragon showed over a period of 350 days (12), \(\phi^i\) values from \(3 \times 10^{-3}\) to \(10^{-2}\), and that work is in hand to obtain further values on reference particles.
Another direct measure of the total release of strontium from high burn up irradiated fuel particles from a small batch has recently been carried out by Horsley and Brown (20), the fuel particles being manufactured to reference Mk.III GCR parameters. The results in Table 2 below show that the average release from the whole batch is around $10^{-5}$, i.e. that the defects normally present in irradiated SiC are of less significance than broken particles unless the incident of failure from fabrication and in-service is reduced to the $10^{-5}$ level. Further it shows that the fraction of 'leaky' particles is only about 5%; the release from 95% of the particles correlating very well with the pre-irradiation measurement of $^{235}$U contamination.

<table>
<thead>
<tr>
<th>Capsule</th>
<th>No. of particles</th>
<th>Av. temperature °C</th>
<th>Burn up %</th>
<th>Fast neutron dose n/cm²</th>
<th>Irradiation time (days)</th>
<th>Uranium contamination in outer PyC</th>
<th>Fraction of particles leaking %</th>
<th>Fractional release (strontium)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>733</td>
<td>1332</td>
<td>5.32</td>
<td>$\sim 10^{19}$</td>
<td>166</td>
<td>$4.3 \times 10^{-6}$</td>
<td>5.83</td>
<td>$2-3 \times 10^{-6}$</td>
</tr>
<tr>
<td>2</td>
<td>1750</td>
<td>1292</td>
<td>5.95</td>
<td>$\sim 10^{19}$</td>
<td>166</td>
<td>$4.3 \times 10^{-6}$</td>
<td>4.7</td>
<td>$2 \times 10^{-6}$</td>
</tr>
<tr>
<td>3</td>
<td>1450</td>
<td>1161</td>
<td>5.95</td>
<td>$\sim 10^{19}$</td>
<td>166</td>
<td>$4.3 \times 10^{-6}$</td>
<td>4.35</td>
<td>$5 \times 10^{-6}$</td>
</tr>
<tr>
<td>4</td>
<td>5.32</td>
<td>4.3 x 10^{-6}</td>
<td>5.95</td>
<td>$\sim 10^{19}$</td>
<td>166</td>
<td>$4.3 \times 10^{-6}$</td>
<td>5.45</td>
<td>$3.75 \times 10^{-6}$</td>
</tr>
</tbody>
</table>

Since the fractional release of $^{89}$Sr from a pyrocarbon coated particle would have been about 1% in these conditions we can see that $\beta^*$ at the end of life was around $\frac{10^{-5}}{10^{-2}} = 10^{-3}$.

(d) The relationship between $\beta$ and the steady state $^{R/B}$ of rare gases and iodine has been measured at Harwell (10) and in Dragon (18), using uncoated reference kernels. This relationship depends essentially
on the diffusion rate and the effective grain size within the porous UO₂, and at low burn up there is now agreement within a factor two on the R/B as a function of temperature for the important isotopes such as 131I, 85Kr, 85Kr, 90Kr, 137Xe. This is probably adequate for our purpose, but there is a greater uncertainty relating to the effect of burn up and of kernel reduction to UC₂. Although experiments are in hand on these topics, we can already look at the 133Xe release history of some long-irradiated Dragon elements(19) and infer that there is no systematic reduction in equivalent grain size with burn up. The effect of a fairly rapid reduction to UC₂, following coating failure, is unknown at present.

(e) In present safety calculations it is pessimistically assumed that the fractional release of long lived Cs, Sr and Ag is equal to \( \beta \), with no retention in bare kernels at all. This is correct enough at 1250°C(11), in a full dwell time, but definitely in error at the lowest fuel temperature of around 600°C (by a large factor) foroxide kernels.

(f) The relationship between \( \beta' \) and the release of the significant metals (\(^{137}\text{Cs}, \ 134\text{Cs}, \ 89\text{Sr}, \ 90\text{Sr}, \ 140\text{Ba}, \ 110\text{mAg}, \ \text{rare earths, transition metals} \) is by no means sufficiently established. The diffusion of Sr and Ba in pyrocarbon is so rapid that there can be little error in estimating their release as \( \beta' \) multiplied by the bare kernel release. As mentioned in (e), this bare kernel release is pessimistically taken as unity until further data becomes available. At the other extreme the release of Zr, Ru, may be safely set to zero regardless of \( \beta' \). For all metals inbetween, including U and Pu, we are unsure of the time dependent release through pyrocarbon as a function of temperature, and it has been usual to set it to zero in calculations on the grounds that matrix and fuel tube delays will very effectively take care of any release. This situation is not satisfactory for Cs isotopes, since they move relatively rapidly in matrix and fuel tube graphites; experiments are in progress to provide a direct measurement of Cs release from pyrocarbon coated reference kernels and this may be expected to lower the permissible \( \beta' \).

It would be very satisfactory if a number of other metals from this intermediate mobility range were similarly investigated. The emergence of Ag(18) as a significant \( \beta' \) source shows that there can still be surprises here. However the nature of Dragon heat exchanger deposits(32,33) does suggest that, apart from activation products, Ag is the last contender for serious attention.

In summary it is clear that we require measurement of the time dependent release of Cs and Ag from pyrocarbon coated kernels as a function of temperature, in order to specify the safe \( \beta' \) value within a factor two. In addition we require Sr, Ba releases at lower temperatures from pyrocarbon coated kernels in order to reduce the present pessimism.
3. THE INTERACTION BETWEEN FUEL PARTICLE QUALITY AND IN-REACTOR BEHAVIOUR

3.1 Introduction

Recent safety analyses of HTR's have included a progressive particle failure rate towards the end of life as a function of temperature and burn-up. In reality the particle endurance limit will depend not only on the statistical and random variations in operating parameters, but also the statistical spread in fabrication parameters; these time dependent $\phi$ and $\phi'$ functions are obviously key input data for a core fission product release calculation.

There is clear evidence from experimental observations of short lived fission gases from coated particle fuel tests that failure progressively increases after the critical burn-up for the weakest particle has been exceeded. Fig. 1 shows the increase in gas release from four separate capsules from the Studsvik 17 test. As the burn up and temperature for all the particles in each capsule are very similar the failure rate gives a good indication of the behaviour due to fabrication variations within a batch, although there has been no absolute correlation of these failures with the statistical variation in particle parameters. This is not entirely surprising since it is exceedingly difficult to measure all the parameters necessary for a complete assessment.

There are two major failure mechanisms in coated particle fuels, viz.

1) Pressure failure - either simultaneous failure of all coating layers, or failure following loss of outer PyC consequent on fast neutron failure or weakened by corrosion.

2) Amoeba failure.

The rate of amoeba attack in particles is considered to be more dependent on reactor operating conditions, i.e. temperature, temperature gradient across inside surfaces of the coating, time, and number of Pu fissions, than the fabrication variables, though that is not to say that variations in particle rating due to variations in U235, diameter of fuel kernel and stoichiometry are unimportant. However current designs aim to avoid failure by this mechanism by avoiding excessively high temperatures and temperature gradients. As a development it is hoped that the incorporation of oxygen getters in the fuel kernels will allow present limits to be relaxed.

In the pressure model, however, it is fairly clear that the statistical variations in fabrication parameters are as important as operational conditions. In this section of the paper the present state of acquisition of statistical data by the Dragon Project is summarised. Since we will see that we are lacking some of the data necessary to...
define the distribution of failure around the mean, in this paper the
effect of altering a number of the parameters by up to 3.6 standard
deviations (i.e. the 1 in $10^6$ particle) where known, is investigated
so that the relative importance of each parameter can be assessed.
Where this data is not available a 'reasonable' range has been assumed.
The important parameters and the type of data normally obtained is shown
in Table 4.4).

3.2 Kernels

The parameters of the kernel which affect safety analyses are:

- porosity
- diameter
- shape
- oxygen : uranium atom ratio

Kernel porosity

Porosity in the kernel of the Mk.III type of particle is the major
contributor to the available free space, although there are other
approaches which rely less on kernel porosity and more on porosity in the
buffer layer. Typically the average porosity is 20-25%. Between batch
variations can be held within fairly narrow limits but within batch
variations are virtually unknown.

Work at OSGAE Seibersdorf has produced some data on total free space
availability to correlate with PIE measurements of the same parameter.
Other laboratories are attempting to measure the porosity in individual
particles. However, a method which is sufficiently simple and cheap to
permit distribution functions to be measured and which will thereafter
do as a quality control is not available. An attempt has recently
been made by Lefevre and Thomas (53) following a suggestion by Audebeau (54)
to derive the standard deviation for the whole population using the
relationship

$$ s = \frac{s_p}{\sqrt{n}} $$

where $s$ = standard deviation between samples of size $n$ of the same
batch

$s_p$ = standard deviation for the whole population of the batch.

Two batches of kernels at either end of the range of interest were used
for an initial experiment and the results obtained are summarised in
Table 5. A comment on the measurement errors which are included in the
results is appended to Table 5. Taking the most pessimistic assumption
of no measurement errors then 1 in $10^6$ particles (3.6$s)$ will have a
porosity variation of greater than 7% from the batch average. These
TABLE 4
Parameters affecting Pressure Failure Statistics

<table>
<thead>
<tr>
<th>Particle Parameter or Environmental Variable</th>
<th>Type of Data available on Fabrication Variables</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total U content of kernel and enrichment</td>
<td>Average for batch</td>
</tr>
<tr>
<td>Stoichiometry of UO₂ kernel</td>
<td>Average for batch</td>
</tr>
<tr>
<td>Burn up a) total fissions</td>
<td>CO and fission</td>
</tr>
<tr>
<td></td>
<td>gas production</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Kernel volume</td>
<td></td>
</tr>
<tr>
<td>Kernel porosity</td>
<td></td>
</tr>
<tr>
<td>Buffer volume</td>
<td></td>
</tr>
<tr>
<td>Buffer porosity</td>
<td>Free volume</td>
</tr>
<tr>
<td>Swelling rate for fuel kernel</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total internal</td>
</tr>
<tr>
<td></td>
<td>Gas pressure</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Kernel surface temperature</td>
<td></td>
</tr>
<tr>
<td>% release of the fission product gases</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>UTS of SiC layer</td>
<td>SiC layer strength</td>
</tr>
<tr>
<td>SiC layer dimensions</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Statistical data for a few batches</td>
</tr>
<tr>
<td></td>
<td>Statistical data available.</td>
</tr>
<tr>
<td>Inner and outer PyC layer:</td>
<td></td>
</tr>
<tr>
<td>Dimensions</td>
<td></td>
</tr>
<tr>
<td>Crystallite Size</td>
<td>PyC shrinkage and creep behaviour.</td>
</tr>
<tr>
<td>Density</td>
<td></td>
</tr>
<tr>
<td>Microstructure</td>
<td>Prestressing of SiC</td>
</tr>
<tr>
<td>Anisotropy</td>
<td></td>
</tr>
<tr>
<td>Mean coating layer temperature</td>
<td></td>
</tr>
<tr>
<td>Fast neutron dose rate and total exposure</td>
<td></td>
</tr>
</tbody>
</table>
results are only strictly related to batches at the 19-25% porosity level, and are in contrast with value of $\sigma_p$ of 4% quoted to Aerts et al\(^{(55)}\) at the 90% density level.

**Kernel Diameter**

Whether made by a powder agglomeration method or a gel-precipitation route, with modern techniques a standard deviation of 25 $\mu$m within batch can be expected for 800 $\mu$m diameter kernels\(^{(56)}\).

**Kernel Shape**

Typically the ratio of maximum to minimum diameter of a kernel is specified to be less than 1.1. Aerts et al quote an average sphericity of 1.033 for powder agglomeration kernels and 1.032 for gel-precipitation process\(^{(55)}\), and a within batch standard deviation of 0.024\(^{(55)}\).

**Oxygen : Uranium atom ratio**

To minimise the carbon monoxide pressure during irradiation the kernels as manufactured should be as near stoichiometric as possible. With high temperature coatings deposited from methane the process almost guarantees a low O:U atom ratio otherwise the particles explode during manufacture. It is not difficult to achieve values of less than 2.005. Direct measurement of CO content at 1600°C from unirradiated particles produced by both Dragon Project and RFL Springfields has confirmed this, values equivalent to O/M ratios of UO$_2$.0005 being obtained for porous fuel kernels\(^{(57)}\). Aerts et al\(^{(56)}\) quote an average value of 2.005; on the other hand the standard deviation which they quote seems a little high at 0.005.

<table>
<thead>
<tr>
<th>Batch Identification</th>
<th>Porosity</th>
<th>Calculated</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean of 10 samples $\sigma_3$ (%)</td>
<td>$\sigma_p$ (%)</td>
</tr>
<tr>
<td>A</td>
<td>19.00 0.057</td>
<td>1.80</td>
</tr>
<tr>
<td>N6-175-R</td>
<td>25.95 0.063</td>
<td>1.99</td>
</tr>
</tbody>
</table>

**Note.** These results include measurement errors. In an attempt to evaluate these errors samples of steel and tungsten carbide spheres, 1000 and 800 $\mu$m diameter respectively, were measured. These results suggest that the reproducibility of the measurements with the steel and tungsten carbide spheres is of the same order as is quoted above for the UO$_2$ microspheres.
3.3 Coatings

The parameters of the coating which bear on safety calculations are:

- Thickness and contribution to free space availability of the buffer layer.
- Thickness of the structural layers
- Density of the structural layers
- Anisotropy of the structural layers
- Strength of the SiC layer

a) Thickness of the Buffer Layer and Contribution to Free Space Availability

The buffer layer is laid down at a much greater rate than any other part of the coating. It might be expected, therefore, that the variability in thickness of this layer would be somewhat larger than that of any other. Since the buffer layer contributes to the free space available for fission products it is important to quantify the buffer layer thickness variations together with a parameter which relates to the free space availability. As the layer cannot be distinguished in X-ray micro-radiography, its dimensions are therefore normally measured by ceramographic sectioning and typical results are given in Table 6.

The density is usually obtained by observing weight and density changes on proving runs and then it is assumed that, the deposition conditions remaining constant, the product remains constant. The density gradient column gives apparently erroneous results presumably due to penetration into the pore structure. A buffer layer density in the range 1.0-1.1 g/cm\(^3\) is often specified and limited Project data points to an average value near 1.0 g/cm\(^3\). Aerts et al\(^{56}\) quote an average value of 1.05 g/cm\(^3\) and a standard deviation between batches of 0.05 g/cm\(^3\).

A knowledge of the thickness and density of the buffer layer however does not in itself provide a value for available porosity. What evidence there is on this parameter is conflicting. Bildstein et al\(^{3}\) have reported reasonable agreement between the calculated and measured total porosity in fuel particles assuming that the volume represented by the difference between the theoretical density and measured density is available to the fission gases. More direct attempts at measuring the accessible porosity by the RFL workers\(^{59}\) have shown that only about 50% of the theoretical volume is available, and for conservatively it is this latter figure that is used in the endurance calculations.
### TABLE 6

**Dimensional Variations of the Buffer Layer (by ceramography)**

<table>
<thead>
<tr>
<th>Manufacturing Programme</th>
<th>Number of Batches Measured</th>
<th>Mean Buffer Layer Thickness (µm)</th>
<th>Mean Standard Deviation Within Batch (µm)</th>
<th>Mean Coefficient of Variation (%)</th>
<th>Mean Standard Deviation Between Batches (µm)</th>
<th>Mean Coefficient of Variation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dragon Driver D15 Batches 101-106</td>
<td>6</td>
<td>20.8</td>
<td>2.3</td>
<td>11.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dragon Driver D15 Batches 193-207</td>
<td>10</td>
<td>26.9</td>
<td></td>
<td>2.5</td>
<td>9.3</td>
<td></td>
</tr>
<tr>
<td>Matrix Fuel Type A</td>
<td>5</td>
<td>33.2</td>
<td>3.2</td>
<td>9.4</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

b) **Thickness of the Structural Layers**

The data position on the dimensions of coating layers is good. Inter-laboratory comparisons have been made by the Project and NUKE GmbH and separately between the Project and RFL Springfields covering specific batches of fuel the results of which show excellent agreement.

If a constant weight of kernels having the same mean density is loaded into the coating furnaces and if for each layer the integrated gas consumption and the deposition conditions are held as constant as possible by process controls, variations in average kernel diameter will lead to variations in average coat thickness such that the ratio

\[
\frac{\text{coat thickness}}{\text{kernel diameter}}
\]

is maintained relatively constant. To confirm this, Audebeau(47) has examined the data for the 142 batches coated as Type B fuel by the Dragon Project for the UKAEA Mk.III Reactor Physics Programme. He noted that a cyclic variation of both the mean and standard deviation of the kernel diameter was matched by a similar 'modulation' of the inner pyrocarbon layer thickness and subsequently confirmed a statistical correlation between coat thickness and kernel diameter. The auto-compensating effect of varying mean kernel diameter on coat thickness is less pronounced for the silicon carbide and outer layers. A summary of the within batch variations for the Type B fuel is given in Table 7 (47).
TABLE 7
Coating Data from the 142 Batches of Type B Fuel for the UKAEA Mk.III Reactor Physics Programme

<table>
<thead>
<tr>
<th>Layer Description</th>
<th>Mean (µm)</th>
<th>Mean Standard Deviation within batch (µm)</th>
<th>Coefficient of Variation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>All inner pyrocarbon layers</td>
<td>111</td>
<td>8.13</td>
<td>7.3</td>
</tr>
<tr>
<td>Silicon Carbide</td>
<td>31.3</td>
<td>2</td>
<td>6.4</td>
</tr>
<tr>
<td>Outer Pyrocarbon</td>
<td>47.6</td>
<td>4.3</td>
<td>9.0</td>
</tr>
<tr>
<td>Total Coating Thickness</td>
<td>190</td>
<td>10.1</td>
<td>5.3</td>
</tr>
</tbody>
</table>

Thomas (60) has recently reported that the scatter on total coat thickness for the sum of the inner pyrocarbon layers is typically ± 4 µm between batches of constant feed. His tabulated data, in fact, yields a standard deviation between batches of 2.0 µm and a coefficient of variation of 1.68%.

c) Density of the Structural Layers

The density specification for the pyrocarbon layers results from a consideration of the self-stressing effects during service due to their shrinkage characteristics. Voice and Lamb (75) have considered the requirements of the SiC layer and concluded that it should have a density of not less than 3.20 g/cm³.

When a single layer is deposited, its average density can be deduced from the weights and densities of the particles before and after the deposition, though the determination is not very accurate. Normally with multi-layer coated particles this cannot be carried out. Sometimes small discs are fluidised with the particles to enable the determination of the density of each layer, but the discs do not have the same hydrodynamic performance and spatial history as the particles and tend to have different deposits from those on the particles. Thomas (60) states that the density of pyrocarbon on discs is usually 0 - 0.03 g/cm³ higher than on the equivalent particles coated in a 5 inch reactor.

It is recognised that in practice there is no sharp transition between the seal and inner HDI layer, and a density gradient exists across these two layers.
It is obviously more satisfactory but most tedious to measure the density on fragments of the coating using a density gradient column (provided that no penetration of the pore structure occurs). Results of between batch variations of layer density measured on coating fragments are given in Table 8 for the silicon carbide and outer pyrocarbon layers.

| TABLE 3 |
|-----------------|-----------------|-----------------|
| Between Batch Density Variations in Silicon Carbide and Outer Pyrocarbon Layers |
| (a) Silicon Carbide (measured on one fragment per batch) |
| Manufacturing Programme | Layer Density (g/cm$^3$) | Number of Batches |
| | Mean | Minimum | Maximum | |
| Charge IV Core 2 driver | 3.196 | 3.188 | 3.208 | 75 |
| Charge IV Core 3 driver | 3.202 | 3.198 | 3.206 | 43 |
| Matrix Test | 3.20 | | | 9 |
| Colibri II | 3.19 | | | 26 |
| LEHPD 4 | 3.20 | | | 7 |
| (b) Outer Pyrocarbon (measured on one fragment per batch) |
| Experimental Programme | Origin of Pyrocarbon | Layer Density (g/cm$^3$) | Number of Batches |
| | | Mean | Minimum | Maximum | |
| Matrix Test | Methane | 1.75 | 1.69 | 1.86 | 15 |
| LEHPD 4 | Butane | 1.855 | 1.785 | 1.97 | 6 |

d) Anisotropy of the Structural Pyrocarbon Layers

It is important that the structural pyrocarbon layers be isotropic to achieve good dimensional stability under irradiation. A typical coating specification, according to Aerts et al (55) calls for the Bacon Anisotropy Factor (BAF) (62) to be in the range 1.00-1.08. The BAF can be measured using X-ray diffraction techniques on graphite discs fluidised at the same time as the particles (63). As pointed out above (under Density of Structural Layers) the deposit on the discs is unlikely to be the same as on the particles. This has led to the development of two optical methods of measuring anisotropy on ceramographic
sections of coatings. The first, termed OFTAF(64) was developed at KFA, Julich. The second, called OAF, was originated by OSGAE, Seibersdorf(65) and has been taken up by the UKAEA (RFL Springfields)(66) and the Dragon Project.

The results are usually reported as equivalent BAF measurements. A theoretical relationship between the optical anisotropy and the BAF has been proposed by the workers at KFA Julich. Moore and Alexander(66) have however concluded that no straightforward relationship has been established between OAF and BAF results.

A programme of OAF measurements on behalf of the Dragon Project has been carried out by OSGAE. Some typical results are given in Table 9. These are average values for particles which have been predetermined as of minimum, mean and maximum coat thickness. They show a reduction of BAF with increasing coat thickness within the same batch.

Control of structural variations within a layer is likely to be the key to extending the performance of the structural pyrocarbon layers. Thus Luhleich et al(67) cite two types of particle of very similar average properties:

<table>
<thead>
<tr>
<th></th>
<th>BAF</th>
<th>Layer Density (g/cm²)</th>
<th>Crystallite Size Lc (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type 1</td>
<td>1.03</td>
<td>1.89</td>
<td>140</td>
</tr>
<tr>
<td>Type 2</td>
<td>1.06</td>
<td>1.94</td>
<td>165</td>
</tr>
</tbody>
</table>

where Type 1 survived a severe irradiation test to $1.6 \times 10^{22}$ n/cm² at 1300°C, while Type 2 did not. They used a 'cold oxidation' technique to eliminate the effects of polishing and to attack preferentially the less crystalline regions. In this way they were able to show that the Type 2 particle coating had a markedly layered 'onion type' structure whereas in the Type 1 particles this appearance was much less pronounced.

In the Dragon Project, examination of the Metallurgical Series I of irradiations is proceeding and tests on unirradiated particles in this series are being brought into line with current practice. The interim conclusion of Mayr(68) is that radial gradients of OAF in the outer pyrocarbon correlate with outer pyrocarbon failure.

It is our opinion that the subject of anisotropy in the structural pyrocarbon layers is still developing at a considerable rate. The feedback from the irradiation tests will undoubtedly lead to improved performance.

e) Silicon Carbide Strength

Work by Evans, Padgett and Davidge(69) on the fracture stress of pyrolytic SiC coatings extracted from particles has shown that for standard SiC (Type 1, Fig 3) there is a very wide variation of strengths within a batch with a value for the 1 in $10^4$ particle of about 30 MN/m², compared to the batch average of 100 MN/m². By careful control of the deposition conditions these figures can be raised to 90 MN/m² and 200 MN/m² respectively (Type 2) with potential for even further improvements.
TABLE 9
Anisotropy Measurements on Outer Pyrocarbon Layers

<table>
<thead>
<tr>
<th>Manufacturing Programme</th>
<th>Average Coat Thickness (µm)</th>
<th>BAF (measured by SGAE using OAF)</th>
<th>No. of Batches</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Average BAF at Minimum of Layer Thickness</td>
<td>Average BAF at Mean of Layer Thickness</td>
<td>Average BAF at Maximum Layer Thickness</td>
</tr>
<tr>
<td>Natural UO₂ - NFD</td>
<td>67</td>
<td>1.15 (1.04 - 1.22)</td>
<td>1.10 (1.08 - 1.26)</td>
</tr>
<tr>
<td>Natural UO₂ (DPID/123)</td>
<td>37</td>
<td>1.05 (1.00 - 1.26)</td>
<td>1.03 (1.00 - 1.14)</td>
</tr>
<tr>
<td>SNAM</td>
<td>37</td>
<td>1.02 (1.00 - 1.04)</td>
<td>1.02 (1.01 - 1.03)</td>
</tr>
<tr>
<td>LE Fuels</td>
<td>35</td>
<td>1.12 (1.01 - 1.34)</td>
<td>1.09 (1.00 - 1.22)</td>
</tr>
</tbody>
</table>

3.4 Particle Endurance Limits

A graph showing the calculated dependence of burn up at failure with the statistical variations in particle parameters taken one at a time has been constructed by Martin(70) using the simple Williamson and Horner model(71) is given in Fig. 2 using the parameters listed in Table 10. The figures used for one standard deviation for the kernel porosity and diameter, and the thicknesses of the coating layers are those reported in the previous section.

The graph shows that variations in porosity affect the endurance life more than variations in kernel diameter or individual coating thicknesses. Even though the SiC layer is well controlled (σ = 2 µm) the effect is very comparable to the somewhat larger variations in the PyC coats; this results from putting a realistic breaking stress for Type 2 SiC (90 MN/m²) into the model rather than assuming it breaks immediately it goes into tension. Since, as would be expected, the mean standard deviation of total coating thickness is considerably less than the sum of the mean standard deviations of individual layers, i.e. 10 µ compared to 14 µ, variations in total thickness will still be less important than kernel porosity.
### Table 10
Data used for Particle Endurance Calculations

<table>
<thead>
<tr>
<th>Normal particle parameters:</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter</td>
<td>800 µm</td>
</tr>
<tr>
<td>Kernel porosity</td>
<td>20-25%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Layer thicknesses (µm):</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Buffer</td>
<td>35</td>
</tr>
<tr>
<td>Seal</td>
<td>20</td>
</tr>
<tr>
<td>Inner HDI PyC</td>
<td>45</td>
</tr>
<tr>
<td>SiC</td>
<td>35</td>
</tr>
<tr>
<td>Outer HDI PyC</td>
<td>55</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>PyC densities (g/cm³):</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Seal</td>
<td>1.6</td>
</tr>
<tr>
<td>Inner PyC</td>
<td>1.7</td>
</tr>
<tr>
<td>Outer PyC</td>
<td>1.8</td>
</tr>
</tbody>
</table>

Fission product release + CO = 90% of total fission gas production.

Swelling due to solid fp's = 0.5% ΔV/

SiC fracture stress = 90 MN/m² (for 1 in 10⁴ particle).

Therefore, although from the quality control standpoint it is desirable to measure dimensional variations (since in any case it is one of the easier parameters to measure on a routine basis), it is even more important to be able to control the kernel porosity within the limits we have assumed here.

In a standard particle, if it is assumed that half the theoretical voidage in the buffer layer is available to fission gases, this layer contributes approximately 25% to the total initial free space. It is therefore apparent that better data on this parameter is also needed. For instance, if all the porosity were available this is calculated to be worth an extra 3% burn up. It is of course at the end of life that the maximum available porosity is needed. While there are many post irradiation measurements of the total gas content from particles broken at operating temperature or above (58, 59) and also measurements of fission gas pressure at room temperature, as yet there is no reported direct measurements of voidage on irradiated particles.

---

- 467 -
Pyrocarbon density and Anisotropy

Both these parameters feed directly into the endurance model, the shrinkage of the PyC maintaining the SiC under compression for a long period of its life. The magnitude of this compressive stress depends on the shrinkage rate of the PyC which is dependent on both density and anisotropy. For the calculations performed in this paper, the shrinkage curves recommended by Manzel(72) have been used. Although in the computer models it is quite easy to examine the effects of small variations in density and anisotropy, there is currently insufficient confidence in the shrinkage data to warrant such an exercise, especially since there is virtually no data on variations within a batch on these parameters. Clearly this is an area that requires further study.

SiC Strength

The effect of increasing the fracture stress from 30 MN/m² for the 1 in 10⁴ particle to 90 MN/m² has been computed using the Walther model(70,73); this results in a dramatic increase in calculated burn up from 6.8% to 11.5% (Fig. 3). Even further increases in endurance could be obtained by increasing the SiC coat thickness. However, the more reliance that is placed on the SiC coat, the more the testing that will be required to ensure that the quality is controlled.

Effect of temperature transients

Transient behaviour can be very simply simulated in models and even more fortunately in practice by post irradiation heating. For instance, it is quite clear that the failures obtained in the Studsvik 17 irradiation experiment were due to pressure since some particles failed on heating to temperatures only marginally higher than the irradiation temperature(57). Others, presumably the upper end of the distribution of endurance, could be heated 300°C above the irradiation temperature without shattering. The theoretical relationship between temperature transient and decrease in particle burn up using the Walther model is shown in Fig. 4 for a particle with Type 2 'high strength' SiC, which has a calculated endurance for the mean particle of 11.5%; e.g. a transient of 200°C any time in the last 1.5% burn up, or a 100°C transient in the last 0.75% of its burn up can be seen to result in fracture.
4. MATRIX EFFECTS

The matrix material has an influence on fission product release in two respects; it has an absorptive capacity for metals and it can exert a diffusion rate delay on their movement to the fuel tubes or fuel free graphite layer. The influence of these effects, and of analogous diffusion and evaporation processes in the fuel tube, is calculated by means of finite difference methods which are available as computer programmes(21,22,23).

In Fig. 5 is shown the calculated effect of a ten-fold variation in diffusion coefficient of $^{137}$Cs in matrix graphite on the overall fractional release from a peak temperature position of a tubular interacting pin. Also shown is the effect of a factor 25 reduction in absorptive capacity - that is, a change in partition coefficient between matrix and fuel-tube. Bearing in mind our supposed requirement for a factor of five accuracy in overall release it appears necessary to know the partition factor within a factor 5 and the diffusion coefficient within a factor 100, since these levels of uncertainty correspond to at most a factor two in fractional release with a fixed $\phi$ value.

Matrix graphites are not currently selected for their fission product retention properties, for the simple reason that there are more basic questions of cost, availability and mechanical stability at stake, but it is an area for potential future development. In particular, a directly-cooled element would rely on diffusion rates in matrix graphite.

The other question to be answered concerns the accuracy with which we know the diffusion and partition coefficients of the matrix materials in use. Fig. 6 shows the results of Harwell work on diffusion coefficients for Cs, Sr and Ag, the most important metals. There are no data for other metals, nor for $I_2$ and rare gases, but it is a reasonable judgement that these are respectively too slow and too fast to have any bearing on safety calculations with the present fuel temperatures.

Partition factors expressing the equilibrium distribution of metals between matrix and fuel tube graphite have been measured by Rowland(24) and by Hooper(25). Typical results for Sr show a value of 1.0 with a factor two uncertainty, at 1000-1200°C and a realistic loading of around 1 $\mu g/g$. Results for other metals are not known, and in particular the value for $^{137}$Cs is the subject of much discussion. Although Hooper measured a factor (with the higher concentration in the matrix) of around 100 at 1000°C (Fig. 7) there are reasons for doubting the validity of using such a factor in calculations. In particular the partition factor tends to fall with increasing time, for reasons which are connected with the complex diffusion mechanisms of Cs in fuel tube graphite. At the present time it is considered that the best estimate for this partition factor is 25 independent of temperature, with an uncertainty of a factor ten.

This analysis is obviously incomplete without some correlation with actual matrix behaviour in, for example, the Dragon reactor. Unfortunately it is too early yet to report on the behaviour of experimental fuels with a known $\phi$ and $\phi'$ but a number of such experiments is in hand.

In summary, we require more accurate data on the relevant partition factor to be used for Cs and Ag, and we require an experimental check of the predicted quantities of Cs, Sr and Ag leaving the matrix as a function of time and temperature. Our knowledge of diffusion rates in matrix graphite is already very adequate, except that in a directly-cooled element it would be prudent to investigate also the rare-earth and heavy metal diffusion rates.
5. **THE MATRIX FUEL-TUBE INTERFACE**

In a fuel element design such as at Fort St. Vrain\(^{(26)}\) or as in the Oldbury B study\(^{(27)}\), there is a more or less well defined helium gap of about 1 mm between fuelled matrix and fuel tube. Post irradiation examination of Dragon fuel\(^{(28)}\) has shown that contact between the surfaces greatly accelerates diffusion of metals to the fuel tube, and in analogous calculations Abbey and Faircloth\(^{(29)}\) have indeed predicted a virtually complete retention of \(^{137}\)Cs in a matrix surrounded by such a gap. However, in real fuel elements there is usually good contact on enough of the matrix surface to reduce this benefit to at most a factor two. The helium gap is now a routine component of FIPDIG runs\(^{(23)}\) on species other than \(^{12}\) or rare gases.

The phenomenon results from the very low vapour pressure of metals adsorbed on graphite, and is insensitive to the present data uncertainties.

6. **THE FUEL TUBE**

The fuel tube, or the fuel free zone in a directly-cooled fuel element, influences the release of fission products to the primary circuit in two ways: there may be a significant time delay from diffusion across the graphite wall and an evaporation rate limited step at the cooled surface\(^{(30)}\). These processes are also taken into account in the codes such as FIPDIG\(^{(21, 23)}\).

To show the sensitivity of \(^{137}\)Cs release to the relevant physical parameters (diffusion coefficient and adsorption isotherm) the FIPDIG \(^{(23)}\) code has been run with a realistic set of input data to correspond with the 20% core height position of the peak channel of a notional UK design HTR\(^{(31)}\). The helium gap, time dependence of power and temperature, diffusion, partition and adsorption effects are all included but \(\bar{\phi}\) and \(\bar{\phi}'\) were assumed constant at the 10\(^{-4}\) level. The fuel tube diffusion and adsorption properties were then separately perturbed by a factor of ten, and the results, presented in Fig.5 show a linear dependence of release upon adsorption coefficient but no significant dependence on \(D\) after the first 50 days at power.

Again remembering our requirement for a factor five accuracy in overall release it appears that for \(^{137}\)Cs we need to know \(D\) within a factor 100, and evaporation coefficient (which is proportional to Cs vapour pressure at a particular loading) within a factor two. Note that a more stringent control of \(D\) might be required if \(\bar{\phi}\) were allowed to increase near the end of the dwell time.

This kind of analysis could be repeated for Sr, Ba and Ag for example, but the answers would differ because some releases are too low to warrant our factor five accuracy requirement. \(^{137}\)Cs has been chosen as the most critical case.

Turning to the present state of measurements of these data we find, that for gilsoncarbon fuel tube graphites, diffusion coefficients are available for rare gases, \(^{12}\)Cs, Sr and Ag, which, apart from small quantities of Sb and Ba, are the only fission products detected in the Dragon primary circuit\(^{(32, 33, 19)}\). The delay imposed by the fuel tube upon rare gases and iodine is negligible for safety assessment purposes\(^{(30)}\) and the accuracy of available diffusion and adsorption data is therefore adequate. In Fig.8 the diffusion coefficients for Cs, Sr and Ag in gilsoncarbon graphite are shown as a function of temperature. Measurements available at the \(\mu g/g\) level include laboratory tracer work\(^{(34)}\) and post-irradiation examination of Dragon fuel tubes\(^{(14, 35, 36, 37, 38, 39, 40, 41)}\). At this level there is no significant variation of \(D\) with concentration.
The Sr data are a consistent and reproducible set, with an uncertainty of a factor three. The Cs data show very clearly the two levels of diffusion rate which can be inferred from profiles, depending upon the time and temperature of the experiment. In the examples plotted, post-irradiation fuel tube profiles yield much lower D values than the short term laboratory experiments. It is believed that this anomaly results from a gradual diffusion into graphite grains occurring simultaneously with a relatively rapid diffusion between grains. Probably the more rapid process is relevant to our purpose, this being the assumption made in current safety analyses, and we then have a factor 10 uncertainty on the 'best estimate'. In the unlikely circumstance of the slow process proving to be the important one the 'best estimate' data must be reduced by factor 100 with the same uncertainty.

In the case of Ag we have at present only data from post-irradiation examination of fuel tubes, the radial concentration profiles being interpreted on the assumption of a steady input rate from the fuelled matrix. In view of the Cs experience it would be unwise to conclude that we know DAg with any precision until laboratory measurements are complete.

The evaporation coefficient in FIPDIG is controlled by the metal adsorption isotherms\(^{(30)}\). The programmes of work at AERE Harwell and at GGA have to date yielded the data presented in Fig.9. Much of the early work on this topic, as for diffusion constant measurements, was done at loadings far above the realistic \(\mu g/g\) range, where Freundlich adsorption occurs. The results shown are those at a representative 0.1\(\mu g/g\), at which level we find only Langmuir adsorption. Some available results for adsorption of Cs on matrix graphites are included, not only because they are relevant to directly-cooled designs, but also because in conjunction with the estimated partition factor they provide an indirect measurement of Cs pressure over gilsoncarbon graphite which is a more difficult measurement. The same structural details which complicate diffusion of Cs in fuel tube graphite also lead to vapour pressures which appear to fall with increasing time as grains take up some of the metal. As a result we have for the Cs adsorption isotherm three sets of results arising respectively from GGA\(^{(42)}\), Harwell work using long equilibration times, and an estimate via the matrix data. The GGA results have been used to give the present best estimate as in reference\(^{(30)}\), but the more recent Harwell work indicates that, for the fast process, this is an uncertainty of factor 100 towards higher pressures at 1000°C. Consideration of the slow process could produce the same degree of uncertainty towards lower pressures.

For Ag there are no adsorption measurements yet available on fuel tube or matrix graphite.

For Sr reproducible adsorption isotherms have been obtained at Harwell using H327 graphite, and Fig.9 shows these very low vapour pressures at the 0.1\(\mu g/g\) level. In view of the experimentally determined partition factor between matrix and fuel tube graphites of around unity, no effort has been made separately to measure a Sr adsorption isotherm for matrix graphite.

In summary, it would appear that we require more accurate Cs adsorption data, since our evaporation coefficient precision falls short of the required factor two. The broad question of which set of diffusion constants is the relevant one also requires an answer. Both of these areas are under examination at Harwell and in the Dragon Project. The Ag data also must be obtained to provide diffusion and evaporation constants. The position for Sr is satisfactory.

It has long been realised that a direct check of the calculated predictions of metal release from fuel tubes is essential to give confidence that the model
is correct and that the estimates of data uncertainty are realistic. At the present time at least three such integral checks of the whole metal release chain, from defective particle to evaporation into the coolant, are under way but none is yet ready to be reported. It is surprisingly difficult to interpret the encouragingly low Dragon circuit activities\(^{(32, 33)}\) in terms of a 'FIPDIG' release because:-

(a) the average fuel element residence times fall far short of a 1000 day dwell,

(b) the $\phi$ and $\phi'$ values of the elements are generally not known as a function of dwell time,

(c) there is a very inhomogeneous mixture of element types in the core at any one time,

(d) plate-out measurements are very restricted in frequency and position,

and so single channel loop experiments have been initiated for the purpose\(^{(30, 43)}\). Other attempts at this experimental check are being made by means of $^{85}$Sr tracer in Dragon.

Finally, it is relevant to the directly-cooled element of the future to note that metal vapour pressures over matrix type graphite have been measured for Cs and Sr (Fig.9), without the ambiguity which arises in the more porous fuel-tube graphites. No measurements are yet available for other metals.
7. THE FATE OF FISSION PRODUCTS IN THE PRIMARY CIRCUIT

The quantities of fission products leaving the fuel elements during their irradiation in the HTR are the most important numbers in a safety assessment and we have tried to show the present state of the calculation route used in their derivation. The description of the behaviour of those fission products, during both normal and accident conditions, is however an important area, the study of which can in principle eliminate some of the pessimistic assumptions which have otherwise to be made.

7.1 PLATE OUT OF γ EMITTERS

The difficulty of maintaining a primary circuit in which long-lived γ-emitting fission products have accumulated over 20 years or more can result in a situation where $^{134}$Cs, $^{137}$Cs, $^{137}$I (and perhaps $^{110}$Ag) release is a limiting condition in the specification of the life of the fuel. The kinetics of the plate-out process are very relevant here because the distribution pattern around the primary circuit can be considered alongside the specification of key maintenance points, local shielding or remote handling equipment. In particular, it is of benefit to know that a fraction of the released metals is re-adsorbed within the core itself.

The basic data required for a calculation of this type are the helium flow and circuit surface conditions together with adsorption isotherms and diffusion constants for the fission products on steel surfaces and within graphite blocks. Computer codes are available for the calculation itself. Results of this type of approach have included estimates that 50% of the $^{137}$Cs will re-adsorb within the core and that boiler surface saturation may cause $^{137}$Cs deposition profiles to advance through a boiler with increasing time. The state of diffusion and adsorption data for the Cs-graphite system has been surveyed earlier and is subject to order of magnitude uncertainty. There are no data on Ag or I$_2$ adsorption to cover the range of core graphite temperatures, although order-of-magnitude estimates have been made. Work is in hand in all these areas. The situation regarding adsorption on steel surfaces is a very difficult one because not only are the adsorption isotherms for Cs, Sr, I$_2$ and Ag uncertain to the extent of a factor of 5, but the surface roughness which also controls the adsorption per unit superficial area is capable of a factor ten variation. The adsorption of fission products on for instance graphite dust, or the removal of graphite dust containing fission products from the fuel tube by corrosion, can further complicate the plate-out profile by reducing the rates at which species diffuse to the tube surfaces. Work is in progress in Dragon Project and at CEGB (BNL) to improve the accuracy of adsorption isotherms for steel surfaces and to determine the size of fission product carrying species in the Dragon reactor.

At the present time, correlation of plate-out profile prediction with experiment is limited to a discussion of the $^{137}$I profile in a Dragon heat exchanger in terms of the recirculation fraction. A coolant sampling probe has in this case provided evidence that the I$_2$ was in molecular form.

7.2 THE LEVEL OF HE-BORNE RADIOACTIVITY

In predicting the consequences of helium leaks and of depressurisation accidents at a HTR station it is necessary to know the fraction of those fission products leaving the fuel elements, actually present in the helium phase.

The same plate-out calculations, based upon adsorption at the various temperature zones around the primary circuit and the mass transfer coefficients appropriate to the helium flow conditions, can in principle determine the partial pressures of significant species (I$_2$, Cs, Sr) at any point. Rare gas concentrations may be directly derived from the fuel emission rates.
The data uncertainties referred to above have resulted in the use of a rather less quantitative approach. Based upon experience in other gas-cooled reactors and upon observations of I_2 levels in the Dragon probe an overall 'plate-out factor' of 10^5 or 10^4 is assumed for all species. Certainly the available evidence indicates 10^5 to be a pessimistic factor in normal operation.

The resuspension of fission product deposits following water ingress or very rapid depressurisation is another relevant area where pessimistic assumptions must be made until data become available.

Both Dragon Project and Harwell aim to gain further direct measurements of fission product burden in the helium phase under realistic operating conditions, and in particular the Pluto Loop B will allow direct investigation of resuspension effects.

8. CONCLUSIONS

1. A survey of the mechanisms of fission product release has shown four areas in which data improvements are required to improve current safety assessments:

   (a) the time-dependent fractional release of Sr, Cs, Ag and rare-earth metals from bare kernels, particularly at the lower core temperatures.

   (b) The time dependent fractional release of Cs and Ag from pyrocarbon-coated particles.

   (c) The mechanism of adsorption and diffusion of Cs in fuel tube graphites.

   (d) The adsorption of fission products on primary circuit metals.

2. Because failure has been shown to be critically dependent on free space, the distribution of total free space (from the kernel and the buffer layer) within a batch needs to be evaluated to determine the 1 in 10^4 failure criterion.

3. The potential improvement in burn-up associated with the higher strength SiC will, if demonstrated, result in somewhat less reliance being placed on the PyC layers. Further data are still required on shrinkage and creep of pyrocarbons as a function of density, isotropy and coating conditions in order to optimize the specification. Once these data have been obtained it seems unlikely that within batch variation will need to be measured routinely. However, as a consequence there will be a need to measure the within batch variations of the fracture stress of the SiC.

4. A long term integral check of the entire mechanism of fission product release from a fuel element should be carried out, and this should include measurements of plate-out and resuspension in a circuit of realistic materials and temperatures.

ACKNOWLEDGMENTS

The authors would like to acknowledge the considerable help given by Messrs. J.P. Audebeau, R.L. Faircloth, J. Holliday, R.L.R. Lefevre, Dr. D. Martin and Mr. R.E. Siddall in providing data for this report.
REFERENCES

5. AERE-R 4215 (1962) - Some measurements of the diffusion of $^{235}$U fission products and $^{233}$Pa from 'coated' fuels. R.H. Flowers, J. Pattison, F.C.W. Pummery, R.E. Watts.
17. DPR-687 (1969) - The significance of fission product release to reactor operations. F.P.O. Ashworth, R. H. Flowers.

18. J. Shepherd, Dragon Project, Private communication.


DPR-628 (1969) - Description of the computer code FIPDIG. G. Preinreich.


23. R.L. Faircloth, AERE, Harwell - to be published.


27. Paper 11 of this Symposium.


30. AERE-R 6575 (1970) - The relation of fission product release limitations to the design and operation of a large HTR station. R.H. Flowers.

31. DPTN-70 - Notional core parameters for the safety analysis of 1500 MW(T) HTR Power Station. G. Mancini.


34. R.L. Faircloth and F.J. Sandalls, AERE - private communication.


37. AERE-R 4434 (1964) - Post-irradiation radiochemical analysis of the Loop Pluto IIIA. E. Groos, H.J. de Nordwall.
38. AERE-R 5040 (1965) - Post-irradiation radiochemical analysis of Charge 8 for Pluto Loop 'A'. P.E. Brown, H.J. de Nordwall.
41. DW(71)4 - Fission products in the Dragon Charge II centre rod elements Nos.414, 417, 430 and 431. H.T. Baker et al.
43. DPTN-199 - A note on the data used in FIPDIG calculation for $^{137}$Cs and $^{80}$Sr in Pegase Experiment IDYLLE 01. B. Barré and P. Rowland.
44. L.F. Jones, CEGB (BNL) - private communication.
45. D.V. Freck, R.S. Radliffe (CEGB internal document).
46. R.E. Siddall - private communication.
47. DPTN-194 - Quality control of production for a particle coating process - achieved and achievable properties. J.P. Audebeau.
48. DFR-744 (1971) - The presence of Ag.111 in the Dragon primary coolant circuit. P.R. Rowland et al.
49. W V Goeddel, Private communication.
50. F.P.O. Ashworth - Paper 1 of this Symposium.
51. DFID/400 Pt.5 - Post-irradiation examination of Studsvik 17. J.H. Evans, K.S.B. Rose.
54. J.P. Audebeau - private communication.
57. G. Horsley, G. Weldrick - to be published as DPTN.
59. N. Hibbert - private communication.

- 477 -
60. DPTN-113 (1971) - The effect of the deposition conditions on coating efficiency and density in a 3-inch diameter fluidised bed. J.P. Thomas.


64. H.B. Grubmeier and G.P. Scheidler (ORNL Trans.2127).

65. P. Koss (OSGAÆ, Seibersdorf) - private communication.


68. K. Mayr - private communication.


70. D. Martin - private communication.

71. G K Williamson, P Horner - private communication.


74. J. Shepherd, G. Mancini - Paper 3 of this conference.

FIG. 1. OUTLINE OF THE VARIATION OF $^{133}\textit{Xe}$ R/B AS A FUNCTION OF BURN-UP.
STUDSVIK 17
FIG. 2. VARIATION IN BURN UP AT FAILURE RATE DUE TO STATISTICAL VARIATIONS IN VARIOUS PARTICLE PARAMETERS
Release Particle $40 \times 10^{-20} = 13\%$ B.U.  Fiss gas release fraction = 0.9
Solid f.p. swelling = 0.8 per $\%$ B.U.

$200 \rightarrow$ Type III
inner fracture stress

$100 \rightarrow$ Type I & II
inner fracture stress

$20 \rightarrow$ Type I
outer fracture stress

FIG. 3. STRESS AT INNER AND OUTER SURFACES OF SiC vs BURN-UP AND DOSE

Critical values:

Dose of B.U. Fracture is $\times 10^{20}$

Type I 27.1 8.8 Outer surface
Type II 35.5 11.5 Inner surface
Type III 47.8 15.5 Outer surface
DECREASE IN PARTICLE LIFE AS A FUNCTION OF TEMPERATURE TRANSIENT AT A CONSTANT TEMPERATURE OF 1200°C. TYPE II SIC BREAKS AT 11.54% B.U.
REFERENCE CASE USING BEST ESTIMATE DATA COINCIDES WITH D IN MATRIX ÷ 10 CASE

---

PARTITION FACTOR REDUCED FROM 25 TO 1

--- D IN FUEL TUBE ÷ 10

--- H ÷ 10

FIG. 5. EFFECT OF MATRIX AND FUEL TUBE GRAPHITE CHARACTERISTICS ON $^{137}$Cs RELEASE FROM A PEAK TEMPERATURE POSITION OF AN HTR CORE. (Φ = 10^{-4})
FIG. 6. DIFFUSION COEFFICIENTS FOR FISSION PRODUCT METALS IN MATRIX GRAPHITE.
FIG. 7. Cs PARTITION BETWEEN NATURAL GRAPHITE AND GILSOCARBON AT THE 1µg/g LEVEL.

\[ \log_{10} K = \frac{4.55 \times 10^3}{T} - 1.72 \]

\[ T = 0^K \]
FIG. 8. DIFFUSION COEFFICIENTS OF FISSION PRODUCT METALS IN FUEL TUBE GRAPHITE.
FIG. 9. VAPOUR PRESSURE OF Cs AND Sr OVER MATRIX AND FUEL TUBE GRAPHITES AS A FUNCTION OF TEMPERATURE AT A CHARACTERISTIC LOADING OF 0.1 μg/g.
DISCUSSION AND CONCLUSION

Paper No. 15

Paper No. 16
Although this Colloquium is concerned with steam systems, it is perhaps appropriate, as it draws to its end, to remind ourselves of the safety implications of systems which may, in the future, more fully exploit the capabilities of the HTR to produce very high gas temperatures and to accommodate fast transients. Two applications are prominently in mind:

Firstly, plant to provide chemical process heating, such as steam reforming, has been proposed by the Dragon Project and others. Reactor gas exit temperatures of about 950°C are needed in this application, which would extend the market for the HTR beyond the more familiar electricity generation.

Secondly, there is an economic incentive to use direct cycle gas turbines as an alternative to steam systems, due partly to the expectations of reduced capital cost, and partly to the expectation of enhanced net station efficiency as higher gas temperatures are achieved. The break even point with steam, from which net station efficiency of about 42% is achievable, is with a maximum cycle temperature of about 900°C, and one can expect 4-5% points improvement for each 100°C increase. Figures from studies of gas turbine systems with fairly low power densities, compared with a typical steam system design are given in the following Table.

With the low power densities of the gas turbine system in this table, peak fuel temperatures are no higher than for the steam system for comparable net station efficiency, and in addition the temperature gradients in the fuel will be smaller, so that in-service fuel particle life can be expected to be as good, if not better. Even in the case quoted with higher gas temperature and a higher net station efficiency, the increase of about 60°C in fuel temperature above the steam system value will tend to be offset by the lower temperature gradient, so that little improvement, if any, is needed beyond present fuel quality to give a satisfactory life in these conditions.

In all high temperature plant, chemical or gas turbine, diffusion and evaporation rates of fission products will be greater because of the higher graphite surface temperatures, so that the contamination and broken particle fraction of fresh fuel may need to be controlled more carefully. But this problem differs in degree rather than principle, and is amenable to the same sort of analysis.

Regarding hazards arising from accidents, maloperation or partial plant failure, in the case of chemical processing plant the issues and arguments are not appreciably different in principle from those of electricity generating steam plant. The pressure vessel and steam generator arrangements will be similar, although design will be more critical because of the higher gas temperatures. The possibility of water ingress and its results are similar, but possibly different in degree.

*"High Temperature Reactor for Chemical Purposes" - Commission of the European Communities - EUR 468e.
<table>
<thead>
<tr>
<th>Power Density MW/m³</th>
<th>Nett Station Efficiency %</th>
<th>Maximum Gas Temperature °C</th>
<th>Peak Random Temperature °C (2 standard deviations)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas turbine</td>
<td>6.2</td>
<td>950</td>
<td>Fuel 1350</td>
<td>D.P. Report 711</td>
</tr>
<tr>
<td>Steam</td>
<td>8.5</td>
<td>850</td>
<td>Surface 1250</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Fuel 1100</td>
<td>Paper 3 of this colloquium</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Fuel 1052</td>
<td>-</td>
</tr>
</tbody>
</table>
For gas-turbine systems, the engineering arrangements affecting hazards will be different in several respects. An important one is that there is no high pressure water circuit adjacent to the primary coolant circuit. The only water adjacent to the primary circuit is the small quantity in the intermediate circuits leading to the precoolers and intercoolers.

Water ingress to the reactor from these becomes a consideration only if a mechanism is postulated whereby the pressure in the primary circuit adjacent to a broken water-pipe is forced below atmospheric. Water ingress to the reactor could only take place if there is a vessel rupture at the same time as a pipe rupture, and there is no reason for supposing that one may cause the other, as there might be with high pressure water circuits. Although water interactions with graphite would be faster at higher temperatures, they would be limited in total extent by the small amount of available water.

The accident philosophies for steam systems always assume guaranteed integrity of the tendon-stressed concrete pressure vessels, containing the whole of the primary circuit. These are envisaged as cylindrical (possibly with steps) with axial penetrations. Large gas-turbines cannot operate with vertical shafts, and anyway they are too large to be accommodated parallel to the reactor vessel axis, so their penetrations into the vessel of an integrated design, (i.e., wholly inside one vessel) would of necessity be radial, posing special problems of design and stress analysis. Designs proposed so far have ranged over the whole spectrum between fully integrated and fully separated, with the Dragon proposal of B.P. Report 711 probably most closely approaching full integration. In this design two separate vessels with horizontal axes, housing power turbines, stand below the main vertical vessel, each connected to it by two very short ducts carrying medium pressure, hot gas (37 bar, 780°C) and low pressure intermediate temperature gas (19 bar, 560°C). All other ducting is in the form of lined passages in the vessels. Turbo compressor units are housed vertically in the main vessel wall. This is a split shaft system: a single shaft unit (all compressors and a single turbine on one shaft), as favoured on the European continent for economic reasons, is much less compatible with integration.

Non-integrated designs, including the three vessel Dragon proposal, are particularly vulnerable to earthquakes. Pseudo integrated designs have been proposed in which the reactor is contained in one pressurised chamber and the machinery, with heat exchangers and ducting in separate chambers of the same concrete structure. This is tantamount to amalgamating the reactor vessel and a containment vessel. The interesting prospect of combining concrete and steel vessels in nuclear gas-turbine plant is offered by the increasing confidence in steel vessels, discussed in this colloquium, based mainly on the principle that a detectable crack will always develop to give ample warning of incipient failure.

A further factor in vessel analysis for gas-turbine systems is that power changes inevitably mean pressure changes. Although by-passes are proposed for rapid changes, and the reactor gas exit temperature can be reduced for slower changes, these both substantially reduce the cycle efficiency. The only way, at present recognised to be feasible, to reduce power and maintain good efficiency is to partly depressurise the system. With the degree of load following predicted for the end of this century this could mean daily pressure reduction to 40%, say.

In depressurisation accidents, the same criteria of acceptability of rates of pressure change apply to gas-turbine systems as to steam systems; viz: machines must not be damaged to an extent that destroys their ability to remove decay heat from the system, and the insulation must continue to protect the vessel.
Design of insulation arrangements needs special care with the higher temperatures, to ensure that it remains simple and not specially susceptible to damage, and preferably repairable.

Air must be excluded after depressurisation, as with steam systems, probably by Nitrogen injection.

There is considerable incentive to use high pressures in gas-turbine plant since the sizes of all parts of the plant, including machines, are reduced as pressure increases. The currently proposed reactor pressure of 60 bar means that decay heat must be removed after a depressurisation accident at 1/60 of the initial pressure, which would require all of the compressors running at normal speed in conjunction with Nitrogen injection.

The contribution which the turbines could make to the gas circulating power needed to remove decay heat, whether pressurised or depressurised, is uncertain at present because they would be working so far from normal conditions. Some external power could be made available through the starting devices used to spin the turbo-compressors up to self-sustaining speed. These could be motors, or perhaps turbines operated by injected gas, the same gas as is needed to prevent air ingress after depressurisation.

If the load of a gas-turbine system is lost suddenly, for example by the loss of electrical load, it is necessary to guard against two consequences. Firstly, overspeeding of the turbo-generator must be restricted to about 10% to avoid damage; that is about the increase which would occur in 1 s if full power were absorbed by the acceleration of the power turbine and generator of a split-shaft system. Secondly, gas which has not given up heat in doing work in the turbines is too hot to be admitted directly into the heat exchangers. Because of the high thermal inertia of the moderator and reflector graphite, which during normal operation has acquired a temperature close to that of the gas, the reactor exit temperature will be sustained for relatively long periods after fission power is removed. (A change of 100°C in core average temperature is equivalent to about 30 full power s). Proposals to deal with these conditions include the rapid diversion of gas round the power turbine, probably through a heat sink to cool it while the mass flow is great, but also mixing it with sufficient gas from a cooler part of the cycle to cool it when the mass flow falls. The diversion valve would need to operate in a fraction of a s. Another proposal is to apply an artificial braking load to the power turbine, for example by a Pelton wheel.

As mentioned in Mr. Soubelet's paper, 6B, we now have, in the Dragon Project, a mathematical model of gas-turbine systems which can be used on the PACE analogue computer at Winfrith for transient performance studies under normal, controlled or accident conditions. It has been used in its earlier form, with a simple representation of particular machines, to study control methods and some depressurisation and loss of load accidents. It has now been considerably refined, in collaboration with leading gas-turbine authorities, so that realistic machine designs can be represented by definitive parameters in the plant data. It is intended to continue to use this model for further control and accident studies, and to interpret the transient effects of plant performance on the core by means of fine structured digital computer models, particularly the model CIPAY, as explained in Paper 6B. Then can follow analyses of possible particle failure, fission product migration, and fuel pin stresses, using the techniques discussed in this colloquium.

It should be borne in mind that demonstrably feasible, reliable, and safe designs embodying the high temperature and fast response principles, which the

- 492 -
HTR offers as its development potential, still depend on development work, particularly on engineering materials. Among the problems are fatigue behaviour of high temperature turbine blades in the HTR primary circuit gas, the proving of control rod materials in terms of interactions of their constituent materials, and the procurement and testing of high temperature thermal insulation materials in suitable mechanical forms.

However, all the problems of gas-turbine systems are straightforward engineering design and materials problems, soluble with the appropriate effort. There are no new, fundamental safety problems, and those associated with an active circuit are natural extrapolations from the steam systems. With the danger of high pressure water ingress removed, the gas-turbine system should on balance be intrinsically safer than the steam systems.

DR. L. R. SHEPHERD (QUESTION PAPER 16)

There seems not to be very much emphasis on the effects of fast neutron dose. Whilst we are currently exceeding specified burn-up by quite a large margin, there does not appear to be sufficient evidence that we are succeeding in the same manner with respect to fast neutron dose.

The results so far with methane pyrocarbon coated particles seem to indicate that failure at the end-of-life is more dependent on fast neutron dose than on burn-up.

MR. J. B. SAYERS (ANSWER)

Yes this is true we have concentrated on the pressure failure although we do also comment in the paper on failure initiated by fast neutron failure of the pyrocarbon. We do appear to have a problem of a strain limit for the pyrocarbon at dose levels of around $3 \times 10^{21}$ n/cm$^2$ (DNE) for methane-derived pyrocarbon. Unfortunately at this time we do not have enough data on the dependance of pyrocarbon parameters on this strain limit under fast neutron dose. The present specification for the pyrocarbon coats arise not only from their fission gas retention capabilities but also on the requirement to maintain the SiC in tension. It has been rather loosely assumed that there was no practical strain limit. What we are now claiming is that the duties of the pyrocarbon can be relaxed with the advance of the stronger silicon carbide and that the pyrocarbon specification should be primarily based on the strain limit. Further than that, this development does allow a re-examination of the present particle design to be made with the objective of re-optimising it with respect to burn-up.

MR. M. S. T. PRICE (COMMENT)

One of the problems with the assessment of pyrocarbon properties is that the methods to make the assessment are still under development. The existence of adequate test methods is vital to the development of any material. With the new methods such as the KFA "cold oxidation" technique we can now proceed at a faster rate to improve the qualities of the pyrocarbon.

MR. F. P. O. ASHWORTH (QUESTION PAPER 16)

Would the authors comment on the difficulty of measuring SiC strength and porosity as a production quality control?
MR. M. S. T. PRICE (ANSWER)

We have said if porosity were to be measured on say 1,000 particles, this is a method which has a fairly large margin of error. Micro-methods are now being actively developed and in fact Dr. Bildstein has already developed one such method for use in post-irradiation examination.

My opinion is that now the problem has been clearly stated we will soon have suitable production control methods.

I am less certain about suitable methods for routine measurement of SiC layer strength. Davidge, et al., (AERE, Harwell) have recently published a paper and work has also been under way, at KFA Jülich. But with the possible exception of Drittler's technique the research methods available are somewhat tedious.

DR. L. R. SHEPHERD (QUESTION PAPER 16)

If reliance were placed on SiC to provide the required strength, what then would be the function of the outer pyrocarbon?

MR. J. B. SAYERS (ANSWER)

To put this question into perspective I think one has to remember that one of the reasons for putting an outer pyrocarbon coat over the SiC is to prevent evaporation of the SiC in service. If it can be shown that this is a redundant requirement, then it is quite likely that the outer pyrocarbon is not worth too much in particle life, then clearly this is a development that needs to be demonstrated by experiment.

MR. H. F. ENZMANN (COMMENT)

This Colloquium has shown that with the possibilities given to build a safe HTR Plant, the safety problems as such are not yet resolved. The safety requirements will have to be further examined and will have to be more clearly formulated. I would like to propose the formation of a Safety Working Party, which:

(a) .could continue the examination of the safety case for the HTR

(b) which would maintain the contacts between the different bodies represented here in this meeting

(c) and which would provide also means of clearing detailed problems which do not receive sufficient attention in a Colloquium.

DR. L. R. SHEPHERD (COMMENT)

The suggestion made by Mr. Enzmann is interesting and there may well be many people present who would like to consider this. During the Colloquium it has become apparent that there is a need for further discussion. There have been many different opinions given and data is not always interpreted in the same manner.

The forum in which these discussions take place should be considered. Whatever may be the outcome of these considerations it would be beneficial I think, if we were to repeat the present colloquium at not too infrequent intervals so that there can be a mutual exchange of information and ideas.
If it is the wish to form a Working Party the Project will be happy to act as an intermediary. We shall therefore be interested to know the views of those present and if anyone has a suggestion to make on this matter they should in the first instance contact Mr. Ashworth.

**DR. L. R. SHEPHERD (CONCLUDING REMARKS)**

We have heard many opinions during the Colloquium on problems affecting the safety of the High Temperature Reactor ranging from the optimism of those operating or engaged in HTR development, to the more cautious approach of the licensing authorities.

With the safety of nuclear reactors being a mandatory matter it is correct that the licensing and inspection authorities should be concerned over the information given by the developers and constructors.

While the High Temperature Reactor does have many inherent safety features which can only inspire confidence, there are still a number of safety problems that need to be resolved. This does not detract from the fact that the economics of the system are attractive and the costs of construction of HTRs is lower than for other gas cooled systems. With this margin in hand, one could consider allocating some of this advantage into extra safety facilities. This applies particularly to the first generation of HTRs in order to ensure higher standards of engineering that will give an improved guarantee of safety.

What has been clearly defined at the Colloquium is the nature of the safety problems that exist. None of the problems raised so far have been unforseen and there is a general agreement concerning these issues, even though there are differences of opinion as to the severity of the problems.

The Colloquium has however bought out those points that have to be taken into account when assessing or guaranteeing the safety of the HTR.

I am particularly optimistic about the fuel and the core of the HTR. I believe we have built-up sufficient knowledge that we now have the possibility of envisaging a reactor core that can be guaranteed as being safer than that of other systems. I think that if we were to meet again in one years time we would be even more confident, but this still has to be demonstrated, as with many other aspects of the system.

In concluding I would like to thank all of those who have contributed to the success of the Colloquium and in particular to the authors for their co-operation in having their papers ready for circulation well in advance of the meeting so that they could be studied prior to the meeting.

I would also like to acknowledge the generous assistance of ENEA and to thank OECD for the excellence of the accommodation and facilities provided for this Colloquium. I hope that this meeting is the first of a series of such meetings where we gather to talk about this very important topic of the safety of High Temperature Reactor Systems.
A BRIEF DESCRIPTION OF "DAGGER III" AN HTR REACTOR

KINETIC PROGRAMME

by

D. A. WARD

BRITISH NUCLEAR DESIGN & CONSTRUCTION LIMITED

The programme "DAGGER III" is a digital code available either in ALGOL or FORTRAN. It is a multi plane representation of three fuel channels in each of which two coolant flows, a tubular fuel element and a surface and bulk moderator region are simulated. Nuclear kinetics are represented by the one-group neutron diffusion equation. The programme simulates one "driver channel" and two "driven channels". Solution of the neutron equations is obtained for the driver channel conditions and the resultant flux levels are applied to all three channels for the solution of the heat transfer equations. Starting from a steady state solution of the neutronic and the heat transfer equations the programme follows the behaviour of the channels during a transient fault specified by the user. Normal practice is for the driver channel to be a representation of an average channel in the reactor, one driven channel to be a representation of a peak rated channel and the second driven channel to represent a channel used to provide a trip output.

A detailed representation of the fuel elements is provided with the inner and outer fuel tubes each represented by a bulk and two surface regions and the fuel by four bulk and two surface regions, which allows conductive, convective and radiative heat transfer mechanisms to be accurately represented.

The driver channel is used to calculate temperature feed-back and the temperature dependence of fuel, fuel tube and moderator coefficients of reactivity are included. Six groups of delayed neutrons and six fission product groups are incorporated in the neutronic equations.

Two control rod lifts are represented, one of which can be used as an optional, automatic power control system; this operates from a thermocouple signal recording the mixed gas outlet temperature of the driver channel. Either or both control rod lifts can be tripped into the core when the thermocouple in any of the three channels exceeds a specified trip level or when the driver channel flux exceeds a specified trip level. Both temperature and flux trip levels can be automatically reset in either direction and will maintain a fixed trip margin unless the rate of change exceeds a specified limit. The coolant gas temperature change is calculated during its passage through the upper and lower reflectors. The thermocouples situated in the lower reflector respond to the gas temperature change with a mass flow dependent time constant.

Fault transients can be represented or induced by means of a number of time dependent inputs, namely control rod positions, reactivity perturbations, core mass flow, reactor gas inlet temperature, the overall power level and/or the driven channel power (relative to the driver channel power).

A time stepping method is adopted in the transient solution. The programme follows a series of user-specified time steps and calculates the conditions in all channels at the end of each step. To avoid iteration of the transient solution the equations are solved using values of some parameters calculated in the previous time step. These parameters are identified and some user judgement is involved in selecting time steps consistent with their rate of change.
TRITIUM IN HTR'S

This information is a summary of two reports, which are at present in draft, on the behaviour of tritium in the Dragon Reactor and predicted behaviour of tritium in a notional 1500 MW core \([1]\).

Tritium is produced in HTR's in three ways:

- **In Graphite** by irradiation of \( \text{Li}^6 \) present as an impurity:
  \[
  \text{Li}^6 + n \rightarrow \text{H}^3
  \]

- **In Helium** by irradiation of \( \text{He}^3 \)
  \[
  \text{He}^3 + n \rightarrow \text{H}^3
  \]

- **In Fuel** by fission
  \[
  \text{U-235} \rightarrow \text{H}^3
  \]

The data and equations used in the calculation of the production rates are given below.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Dragon</th>
<th>1500 MW</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li concentration in Pile Grade A Graphite ((N_L))</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Neutron flux across core ( \phi_G ) ([2])</td>
<td>(1 \times 10^{13} ; \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1})</td>
<td>(2.8 \times 10^{13} ; \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1})</td>
</tr>
<tr>
<td>Thermal neutron capture cross section of ( \text{Li}^6 \sigma_L )</td>
<td>(95 \times 10^{-22} ; \text{cm}^2)</td>
<td></td>
</tr>
<tr>
<td>Mass of graphite in core</td>
<td>30 tonne</td>
<td>180 tonne*</td>
</tr>
<tr>
<td>Thermal power ((P))</td>
<td>20 MW</td>
<td>1500 MW</td>
</tr>
<tr>
<td>Number fission s MW (N_f)</td>
<td>(3.2 \times 10^{16})</td>
<td></td>
</tr>
<tr>
<td>Fission yield of tritium (Y)</td>
<td>0.005%</td>
<td></td>
</tr>
<tr>
<td>Concentration of ( \text{He}^3 ) (N_H)</td>
<td>(4.5 \times 10^{16} ; \text{atoms} \cdot \text{g}^{-1})</td>
<td></td>
</tr>
<tr>
<td>Neutron flux absorbed by circulating ( \text{He} \cdot \phi_H )</td>
<td>(1.35 \times 10^{12} ; \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1})</td>
<td>(6 \times 10^{12} ; \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1})</td>
</tr>
<tr>
<td>( \text{He}^3 ) neutron capture cross section ( \sigma_H )</td>
<td>(5.4 \times 10^{-21} ; \text{cm}^2)</td>
<td></td>
</tr>
<tr>
<td>Mass of circulating ( \text{He} )</td>
<td>70 kg</td>
<td>3 \times 10^5 ; \text{kg} ([3])</td>
</tr>
</tbody>
</table>

*calculated from data in \([1]\)
Use of these three equations gives the following tritium production rates:

<table>
<thead>
<tr>
<th>Source</th>
<th>Production Rate in Dragon Ci Day</th>
<th>Production Rate in 1500 MW HTR Ci Day</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li⁶</td>
<td>0.46</td>
<td>7*</td>
</tr>
<tr>
<td>He³</td>
<td>0.14</td>
<td>18</td>
</tr>
<tr>
<td>Fission</td>
<td>0.14</td>
<td>11*</td>
</tr>
<tr>
<td>TOTAL</td>
<td>0.74</td>
<td>36</td>
</tr>
</tbody>
</table>

*Half-life for Li removal: in Dragon 862 days in 1500 MW HTR 220 days

Tritium production rates are based on initial concentrations of Lithium.

*It is assumed from studies of other fission products that only broken particles release tritium, i.e., 0.01% is released by fuel to primary circuit. The rest remains in the fuel and will be released during reprocessing. One fuel pin irradiated for 800 days in a 1500 MW HTR would contain 0.2 Ci of tritium.

**Tritium Removal Rate and Primary Circuit Content**

The tritium purification rate in Dragon during periods when no injections are occurring = ca. 0.01% per day, i.e., 2% of the tritium produced by irradiation of Li⁶ and He³ is removed from the primary circuit coolant. Most of the rest remains on the core graphite where it acts as a sink for exchange with hydrogen. During impurity injections an increase in hydrogen content is followed by a corresponding increase in tritium content. The specific activity of tritium during Core 2, Charge IV in which the hydrogen content of the primary circuit coolant varied between 1 and 100 vpm, remained approximately constant at 0.01 CiH⁳:gH₂. The tritium clean up rate during periods of high hydrogen content is accordingly higher than during periods of low hydrogen content.
The clean up rate in the proposed 1500 MW HTR [3, 4, 5] is greater than in Dragon. This coupled with the higher rates of tritium production in the 1500 MW HTR, will give rise to large amounts of tritium oxide in the liquors and vapours removed from the purification plant on regeneration.

REFERENCES


REFERENCE TO COLLOQUIUM SUPPORT PAPERS

G. Mancini, "Notional Core Parameters for Safety Analysis of 1,500 MW(th) HTR Power Station", DPTN/70.


B. Barre and P. R. Rowland, "A Note on the Data Used in FIPDIG Calculation for Cs-137 and Sr-90 in the Pegase Experiment IDYL 01".