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A proposal for in-pile vapor pressure measurements on cesium compounds

W. Breitung, G. Schumacher
Institut für Neutronenphysik und Reaktortechnik
Projekt Schneller Brüter

Kernforschungszentrum Karlsruhe

KERNFORSCHUNGSZENTRUM KARLSRUHE
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THE SIGNIFICANCE OF CESIUM IN LMFBR AND LWR SAFETY ANALYSIS

A proposal for in-pile vapor pressure measurements on cesium compounds

W. Breitung
G. Schumacher

Kernforschungszentrum Karlsruhe GmbH, Karlsruhe

Kernforschungszentrum Karlsruhe GmbH
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Abstract

The significance of cesium, which is the most abundant volatile fission product in reactor fuels, is shown for LMFBR and LWR safety analysis.

The conclusion for the LMFBR case is that apparently a high potential for fuel motion induced by cesium pressures exists. The magnitude of this effect should be clarified by in-pile vapor pressure measurements on Cs_2UO_4 and $Cs_2UO_{3.56}$.

In the LWR case, cesium isotopes play an important role in the overall health risk from released fission products. Cesium vapor pressure measurements on Cs_2MoO_4 and Cs_2UO_4 would provide fundamental input data for mechanistic LWR release codes which are presently under development at different laboratories.

A test matrix of five experiments is proposed. The tests are based on already existing in-pile techniques.

DIE BEDEUTUNG VON CÄSIUM IN LMFBR UND LWR SICHERHEITSANALYSEN.

Ein Vorschlag für in-pile Dampfdruckmessungen an Cäsiumverbindungen.

Zusammenfassung

Cäsium ist das häufigste der leicht verdampfbaren Spaltprodukte in Reaktorbrennstoffen. Seine Bedeutung für LMFBR und LWR Sicherheitsanalysen wird gezeigt.

Die Schlußfolgerung im Falle des LMFBR ist, daß offensichtlich ein bedeutendes Potential für eine Brennstoffbewegung existiert, die durch Cesiumdampfdruck hervorgerufen wird. Die Größe dieses Effektes sollte durch in-pile Dampfdruckmessungen an Cs_2UO_4 und $Cs_2UO_{3.56}$ bestimmt werden.

Im Falle des LWR wurde gezeigt, daß Cäsiumisotope eine wichtige Rolle im Gesundheitsrisiko von freigesetzten Spaltprodukten spielen. Cäsium Dampfdruckmessungen an Cs_2MoO_4 und Cs_2UO_4 würden grundlegende Eingangsdaten für mechanistische LWR Freisetzungscodes liefern die derzeit bei verschiedenen Laboratorien entwickelt werden.

Ein Versuchsprogramm aus fünf Experimenten wird vorgeschlagen. Die Tests basieren auf bereits entwickelten in-pile Versuchstechniken.

CONTENT

1. Introduction

Irradiated reactor fuel contains a whole class of fission products which during normal reactor operation exist in solid or liquid form within the fuel pin. However, during off-normal temperature excursions these substances can be transformed into the gaseous state, which may have consequences on the accident progression.

The most important members of the class of easily volatized fission products are Ba, Ce, Pd, Te, Sb, Cs, Rb and Cd. As will be discussed later in more detail, the effect from the vaporization of a given fission product should generally increase with increasing vapor pressure and increasing abundancy in the fuel of that fission product. So in order to separate out the important contributors, one can look at the product of vapor pressure times fission yield. If this "figure of merit" is calculated for the fission elements given above, (using the vapor pressure at 2000 K), the score for cesium exceeds that of the other fission products by one order of magnitude or more. Hence it appears reasonable to concentrate the investigation on the role of cesium in accident calculations. Compared to cesium, the other easily volatized elements appear unimportant.

As will be shown in this paper, the high vapor pressure and the large fission yield of cesium can have important implications for LMFBR and LWR accident scenarios.

The data will be identified which are required in order to quantify and include the cesium effects in reactor safety calculations.

2. Significance of Cs in LMFBR safety analysis

Much attention has been given in LMFBR safety research to detailed mechanistic calculations of Core Disruptive Accidents (CDAs). In such CDAs the fuel is subjected to a temperature ramp of several 100 K/s upto several 1000 K/s, depending on details of the accidents considered.

In order to describe the fuel pin behaviour under these transient conditions a number of codes were developed in different laboratories, e.g. SAS and FRAS at ANL, SANDPIN at SNL, LAKU at KfK and NEFIG in the UK. The main goal of these codes is the calculation of pin failure, as caused by the different sources of internal pin pressure, and the subsequent clad and fuel motion in the cooling channel. However, quite surprisingly the pressure effect from fission products is restricted solely to the fission gases Xe and Kr. Other highly volatile fission products are not included in the modelling, although, these volatiles can be much more abundant than the fission gases. Among the volatile fission products, cesium has by far the highest potential for a large pressure contribution in irradiated fuel.

The following paragraphs will first describe the production and migration of Cs in LMFBR fuel pins under nominal operating conditions. Then the behavior of the cesium in the various parts of an LMFBR fuel pin under a temperature transient is discussed.

Finally, theoretical estimates are given for the Cs pressures which can be expected from the cesium during a CDA temperature excursion.

2.1 Cs concentrations in LMFBR fuel pins

2.1.1 Cs production

A number of cesium isotopes appear in the fuel from the beta decay chains of fission products. Table I summarizes the information of interest. Most important in the present context are Cs-133, Cs-135 and Cs-137. These isotopes are accumulated to relatively high concentrations in the fuel during normal operation, because the masses 133, 135 and 137 are all situated in the second maximum of the fission product mass distribution. In the case of fast fission of Pu-239 the cumulative yields for Cs-133, Cs-135 and Cs-137 are 6.97%, 7.46% and 6.68%, respectively /17/.

Table I: Production of cesium isotopes during irradiation. Each field gives the fission yield in % and half-life $T_{1/2}$ of the nuclide, e.g. Sb-133. The yields are for fast fission of Pu-239 /17/. meta denotes the meta-stable state.

Element	Mass number of fission product decay chain					
	133	134	135	136	137	138
Sb	yield = 1.06 $T_{1/2}$ = 2.3m	.3 .85s 10.5s	.04 1.7s			
Te	4.54 12.5m meta 55.4m	3.51 41.8m	1.76 18s	.75 21s	.16 3.5s	
J	1.29 20.8h meta 9s	3.38 52m	4.5 6.6h	3.3 46s 83s	1.95 24.2s	.8 6.3s
Xe	.043 5.3d meta 2.2d	.19 stable	1.15 9.2h m.st. 15.3m	2.54 stable	3.79 3.8m	4.05 14.1m
Cs	$.16\cdot10^{-4}$ stable →	→ (n,γ) $.23\cdot10^{-4}$ 2.1a	$.8\cdot10^{-2}$ $2\cdot10^{6}$a →	→ (n,γ) .105 13d	.78 30.1a	.82 32.2m m.st. 2.9m
Ba		stable	stable	stable	stable	stable

The formation of Cs-134 and Cs-136 from beta decay is blocked by a stable Xe-isotope in the decay chain, however these isotopes can appear through neutron capture in Cs-133 and Cs-134, respectively. In a fast spectrum this production path is unimportant because of the small fast capture cross section, but in a thermal spectrum noticeable amounts of Cs-134 and Cs-136 are generated.

2.1.2 Cs migration

Numerous analyses of irradiated fuel pins have shown that the cesium which is produced under steady-state conditions according to the flux distribution in the fuel pin, does redistribute during the irradiation.

The dominant mechanism for the observed migration is successive evaporation and condensation of cesium in the open interconnected volume within the fuel pin. The cesium is transported along the existing radial and axial temperature gradients, that is, in the fuel from the fuel center into the fuel periphery and in the fuel cladding gap from the axial peak power node towards the colder ends of the fissile column. The Cs remains in the colder parts of the fuel pin because the higher oxygen potential and the lower temperature allow the formation of stable Cs-compounds which have low Cs vapor pressures.

A second transport mechanism may come into play for the isotope Cs-133, which has a Xe precursor of several days half-life. The Xe-133, which enters the free volume within the pin should pressure equilibrate in the existing temperature gradient, thereby accumulating in the cold regions. The beta decay of Xe-133 then deposits Cs-133 predominantly in the cold parts of the free volume. However, for the overall Cs-transport this gaseous migration is less important than the evaporative Cs transport.

Figure 1 gives an overall picture of the cesium redistribution which is typically observed for mixed oxide fuel pins having a peak burn-up of about 10% /20/.

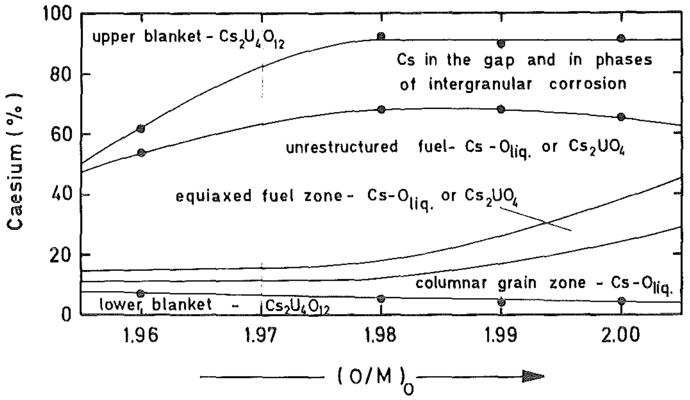


Figure 1 Typical cesium distribution in the various regions of an LMFBR fuel pin irradiated to 10% peak burn-up as function of initial fuel stoichiometry /20/. The fuel contains about 60% of the total cesium in typical LMFBR pins (O/M=1.97...1.99).

The Cs distribution among the different pin regions depends on the average O/M-ratio because this controlles the oxygen potential in the pin and the stability of Cs-U-O compounds. The general tendency is that with decreasing O/M less stable Cs-compounds with higher Cs partial pressures are formed in the fuel, leading to increased Cs transport. Most commonly used average O/M-values in LMFBR fuel pins range from 1.97 to 1.99. For these two limiting cases, the Cs distribution from Figure 1 is given numerically in Table II.

Table II: Cs distribution in the various regions of 10% burnup LMFBR pins for two limiting O/M-ratios. (From Fig.1)

pin region	percent of all cesium in pin			
	O/M=1.97		O/M=1.99	
lower blanket	6.5		5.0	
columnar grains	5.0	fuel	11.5	fuel
equiaxed grains	4.0		9.5	
unrestructured fuel	47.5		42.0	
fuel cladding gap	19.0		23.0	
upper blanket	18.0		9.0	
total in fuel	56.5		63.0	

Another important variable for the degree of Cs migration in an LMFBR pin is burn-up. Note that Fig.1 which shows the Cs distribution for a peak burn-up of about 10%, is typical for the end-of-life situation. At lower burn-up a larger fraction of the produced cesium is found within the fuel pellet. Main reasons are the solubility of cesium, which is e.g. .8±.2% at 1300 K in UO_2 /2/, and the limited kinetics of the evaporative transport.

Another example for the observed cesium transport in LMFBR fuel pins is shown in Figure 2a. Since the average Cs concentration in the 5.4% burn-up fuel should be .48%, the fuel periphery contains significantly more Cs than was produced there locally. The excess cesium migrated from the inner towards the outer fuel regions, along the open interconnected porosity in the fuel pellet.

It is useful to compare the concentrations measured for Cs (Figure 2a) with those found for Xe (Figure 2b). The cesium concentration is practically everywhere one order of magnitude higher than that of Xe, although the cumulative fission yield for both elements is nearly the same. The reason for this lies in the different release behaviour of the two elements during steady-state irradiation: most of the gaseous Xe is liberated from the fuel, whereas around 60% of the produced Cs remains in the fuel (Table II). In order to quantify the importance of the relatively large amounts of cesium for fuel motion, the possible pressures from Cs will be estimated in paragraph 2.3.

2.2 Cs behaviour during a temperature excursion

The purpose of this paragraph is to discuss the relative importance the cesium in the different regions of an LMFBR pin has for the transient pin behaviour during a CDA. For each pin region the following points must be considered:

- relative amount of cesium present in this region,
- chemical stability of the dominant Cs-compound,
- temperature level reached during the excursion,
- possible action of Cs pressure on clad or fuel.

The lower blanket contains only a few percent of the total cesium inventory (Table II) and it is the coldest part of the pin during a CDA temperature excursion. Therefore the cesium in this region will not be important for the transient fuel pin behaviour.

The upper blanket contains more Cs than the lower blanket, typically 10 to 20%. The cesium compound in the blanket region often has a complex composition of the type: $(Cs,Ba,Sr)_2 \cdot (U,Pu,Te)_4 O_{\approx 12}$ where the Cs-, Ba-, and Sr-concentrations follow the relation $Cs \gg Ba > Sr$ /18/.

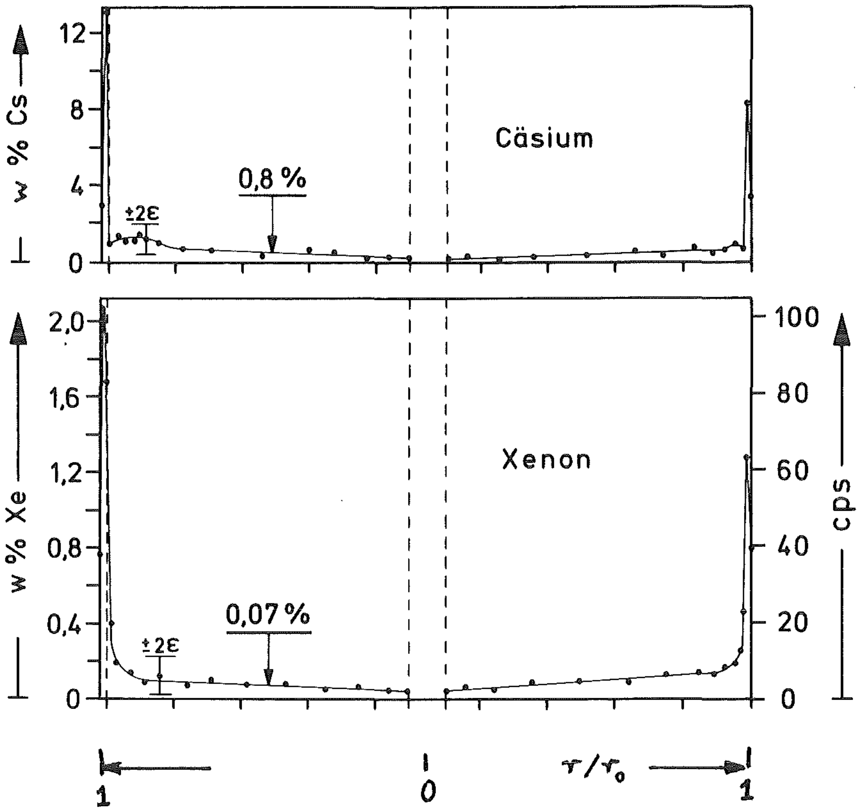


Figure 2a,b Measured radial Cs and Xe concentration in irradiated LMFBR fuel /1/. (Pin PNL-10-12, 5.4% burn-up in EBR II, 29 kW/m linear heating rate). The cesium concentration in the fuel is about one order of magnitude higher than that of xenon.

The formation of such phases containing a large number of elements and showing no definite stoichiometry, can be understood on the basis of vapor transport, which is a practically independent process for each of the elements present. The particle flux in the vapor phase and hence the relative concentration of a given element in the Cs-phase will mainly be determined by the abundance and the volatility of that element or its compounds. Apparently the chemical stability of the vapor deposited Cs-phase is quite low. This is demonstrated by the fact that the Cs-phase is not found in the hot fuel regions of an irradiated pin. Consequently the moderate temperature increase in the upper blanket region during a CDA scenario may result in appreciable pressures. On the other hand the cladding in this part of the pin remains strong for long times into the accident. Furthermore, there is generally significant mechanical bonding between fuel and cladding in the fissile region of an LMFBR pin. Therefore it can be expected that most of the pressure from the dissociating Cs-phase relaxes along the radial unobstructed gap of the blanket region into the upper fission gas plenum. So the cesium in the upper blanket has probably no significant influence on the behaviour of the fuel or cladding in core disruptive accidents.

The radial gap betwen fissile pellets and cladding contains up to about 20% of the total cesium inventory. As in the upper blanket, the Cs is contained in a vapor-deposited multi-element phase of low chemical stability. The temperature rise of this phase during a CDA temperature excursion will be significant because it is very close to or even in direct contact with the fuel. The action of the dissociation pressure on the clad will depend on the heating rate. In a fast TOP with several thousand K/s temperature increase, the dissociation pressure from the Cs-phase will increase the internal load on the cold cladding and support early clad failure.

This was observed e.g. in the experiments of the FD2/4 serie (Paragraph 2.4.2). Also from the absorption spectroscopy used in these tests, it was learned that the cesium containing phase in the gap very easily dissociates, releasing metallic cesium vapor on clad failure.

In a slow LOF with several hundred K/d heating rate, a considerable part of the cesium phase can destill towards the cold lower blanket region, (see e.g. CABRI experiment BI1, Fig.6). The cesium in the gap can also remove the weak clad from the fuel pellet column, as was seen in FD 2/4 experiments (paragraph 2.4.2). Generally, the main effect of the gap cesium will be to support early clad failure. The gap cesium will probably have little influence on fuel motion during the accident, which is an important aspect in CDA scenarios because of the strong usually negative reactivity feed-back.

The fuel region in an LMFBR pin contains typically 60% of the total cesium for 10% burn-up pins (Table II). For burn-up values below 1% the cesium fraction in the fuel can rise close to 100%. So the majority of the total cesium inventory is retained in the fuel region of the pin. Of this cesium in the fuel region a certain fraction is in solution within the fuel grains and the remainder is found in grain boundary pores and the interconnecting porosity network between the grains. Due to the evaporative transport, most of the migrated cesium is found in the porosity of the outer unrestructured fuel region. The high cesium concentration in this part of the fuel is apparent in Fig.1 as a wide band. The inner hot regions of the fuel contain mainly cesium which is in solution within the grains.

During a CDA temperature excursion both parts of the fuel cesium can influence fuel motion in certain ways. The cesium on the grain boundaries will exert pressures between the grains and support a sudden break-up of the fuel into grain-sized particles, as soon as the restricting force from the clad is relieved. Of the cesium within the fuel grains a certain part - which depends on details of the temperature history - will accumulate in small bubbles (may be together with Xe) because the Cs solubility decreases with increasing temperature. When the fuel becomes plastic or liquid the expansion of the bubbles can cause massive fuel swelling. During subsequent slumping of liquid fuel the remaining Cs which is still in solution may leave the liquid and lead to fuel foaming. So both fractions of the fuel cesium have important potential for driving fuel motion after pin failure.

The above discussion of the relative importance of the different cesium parts in the various regions of an LMFBR pin indicates that the cesium remaining in the fuel region has the highest potential for driving fuel motion. The main reasons are:

- most of the cesium in LMFBR pin remains in the fuel,
- the cesium in the fuel reaches the highest temperatures during a CDA, and
- there exist microscopic mechanisms which can drive solid and liquid fuel quite effectively, provided the heating rate and the pressures from Cs are large enough.

The next important question that must be addressed is therefore the possible magnitude of Cs-pressures in the fuel.

2.3 Theoretical estimates for Cs pressures

The Cs partial pressure over a cesium containing condensed phase depends on the binding forces within that phase and hence on the chemical constitution of the Cs phase. To obtain a first order estimate for the range of possible Cs pressures in reactor fuels, the cesium pressure over metallic Cs and over the most likely Cs compounds was calculated.

2.3.1 Cs pressure from metallic cesium

The results for metallic cesium are an upper limit for possible Cs pressures in reactor fuels. For calculating the Cs pressure within fuel pores the following information is needed:

1. Cs concentration, e.g. Cs atoms per pore.
2. Cs vapor pressure up to the critical temperature.
3. Cs Van-der-Waals constants of the gaseous state.

The data for point 1 were estimated using Fig.2a. The vapor pressure curve was taken from /3/. At the critical temperature of 2050 K a pressure of 11.7 MPa is reached. Van-der-Waals constants for description of the gaseous state were calculated from the measured critical data, using corresponding states relations.

The results are listed in Table III for 2500 and 3000 K. The calculated pressures are indeed enormous. Even if one would decrease the measured Cs-concentrations of Fig.2a by a factor of 2 to 3 and increase the fuel porosity, pressures of 100 MPa and above seem possible from metallic cesium.

Table III Calculated Cs pressures in fuel pores, assuming metallic Cs state.

relative fuel radius r/r_o		.5	.8	.9
P_{Cs} (MPa)	2500 K	40	45	500
	3000 K	70	75	650

The calculated pressures are roughly a factor of 10 larger than corresponding Xe pressures. The basic reasons for this high pressure potential of Cs are the following:

Below the critical temperature of 2050 K Cs can be accumulated in the fuel pores in a condensed state. Since the density of condensed phases is orders of magnitude larger than that of gases, a given pore can contain e.g. 100 times as much Cs as Xe. When the critical temperature of Cs is exceeded, all Cs becomes gaseous, or in other words, the specific volume (m^3/kg) of Cs increases drastically and a tremendous local pressure develops within the small fuel pore.

2.3.2 Cs pressure from Cs-uranates

The cesium generated in reactor fuel under irradiation may undergo chemical reactions with other fuel constituents. The only important reaction partner for cesium is the fuel oxide UO_2 and $(U,Pu)O_2$, respectively. Fission products other then Cs generally have too low abundancies to bind a significant fraction of the cesium.

The only exception is molybdenum. It has about the same fission yield as Cs and can in an operating fuel pin bind Cs according to the following reaction:

$$2\ Cs + MoO_2 + O_2 = Cs_2MoO_4 \qquad (1)$$

However in typical LMFBR pins, the oxygen potential is too low to allow the formation of molybdenum dioxide, the necessary reaction partner for cesium. At low oxygen potentials the molybdenum remains metallic and is nearly completely accumulated in metallic inclusions of irradiated fuel /19/. In LWR fuel, which has a higher oxygen potential than LMFBR fuel, cesium molybdate formation is important. For the LMFBR fuel reaction (1) can be neglected and it is sufficient to evaluate the compounds which are possible in the Cs-U-O-system.

The chemistry of the Cs-U-O system was controversial until about three years ago a complete picture developed, at least in the temperature range from 800 to 1400 K /4,5/. The data of interest are summarized in Fig.3.

The stability of Cs-U-O compounds depends on the temperature and the oxygen potential $\Delta\bar{G}_{O2}$. Figure 3 therefore represents a stability map of such compounds. The phase boundaries give the compounds which exist in equilibrium at the respective $\Delta\bar{G}_{O2}$-T-lines. Between two lines those compounds exist which are written along both phase boundaries. Also included is the oxygen potential line for $(U,Pu)O_{2.00}$. The gray area is the region of interest for the fuel periphery in LMFBR fuel pins, where the lower bound of the grey area is somewhat arbitrary. Also shown are Cs-isobares which denote the Cs partial pressure over the respective Cs-compound field.

The following can be said about the chemical state of Cs and cesium pressure in the fuel pin periphery ($T<1500$ K):

- The Cs pressures range between about 10^{-5} Pa (in the fuel-cladding gap) and 10^{5} Pa (at the 1500 K - isotherm of the fuel). These pressures and pressure gradients are large enough to sustain an effective vapor transport of Cs towards colder regions under normal operating temperatures.
- Under normal operating temperatures the dominant Cs compound in the outer region of the fuel pin, is Cs_2UO_4. The so-called z-phase ($Cs_2UO_{3.56}$) may exist in smaller amounts.
- The existance of metallic Cs appears unlikely because of the low oxygen potential required.

In a severe CDA scenario the uranates in the fuel will be heated up to or even beyond 3000 K. In order to obtain some information for typical CDA temperatures, the data of Fig.3 were extrapolated linearly up to 3000 K. The result is shown in Fig.4. Besides the two phase boundaries for uranates (denoted 1 and 2) the oxygen potential lines of $(U_{.75}Pu_{.25})O_{2-x}$ are given for the O/M range of interest. (O/M=1.97...2.00).

The results of the thermodynamic extrapolation are:

- For LMFBR fuel of O/M=1.98,1.99 and 2.00 the uranate Cs_2UO_4 dissociates to $Cs_2UO_{3.56}$ at 700,1220 and 1880 K, respectively. The corresponding Cs pressures are roughly 10^3,10^4 and 10^5 Pa.
- For the same O/M ratios as given above, the $Cs_2UO_{3.56}$ dissociates to metallic Cs at 1430, 2350 and 2750 K, respectively. The corresponding Cs pressures range between 1 and 100 MPa.
- Compared to the Cs pressures over metallic Cs, the pressures over the uranates can be smaller by orders of magnitude, e.g. 2 orders of magnitude of 1600 K, and one at 2300 K.

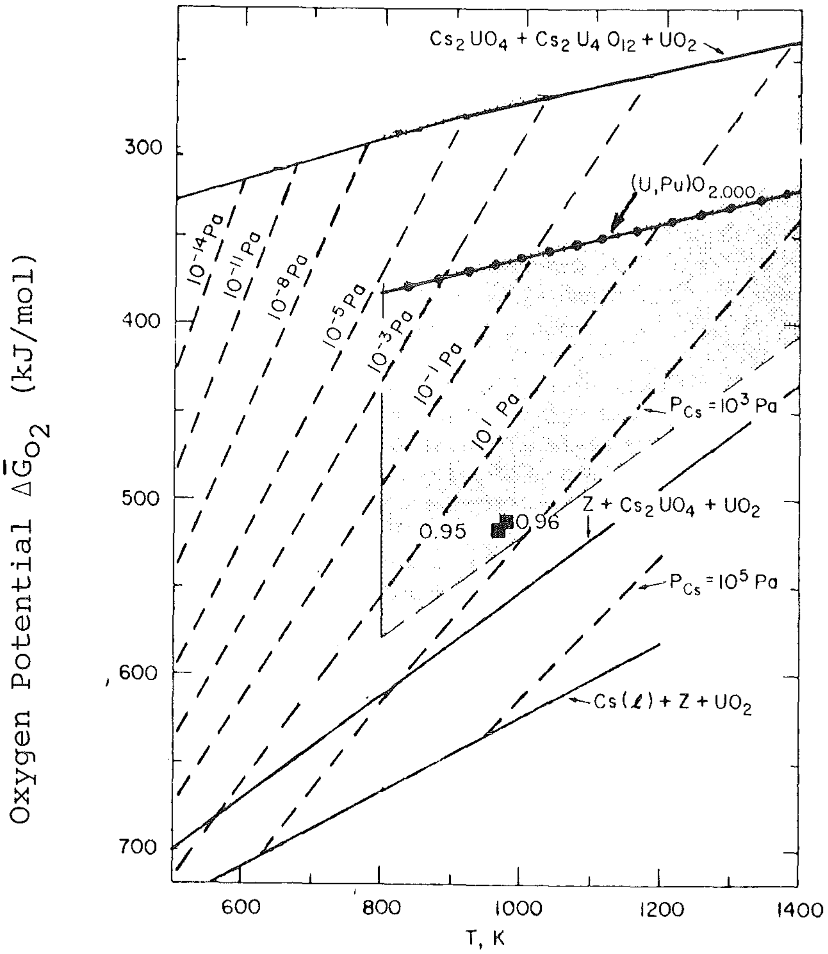


Figure 3 Selected portion of the Cs-U-O system /4/. The dashed lines are cesium isobars in Pa. The solid lines are phase boundaries for cesium compounds. The dotted line shows the oxygen potential of (U,Pu)$O_{2.00}$. The grey region is the area of interest for the fuel periphery in typical LMFBR pins.

The dominant cesium compound in the fuel periphery is Cs_2UO_4. The z-phase ($Cs_2UO_{3.56}$) may exist in smaller amounts.

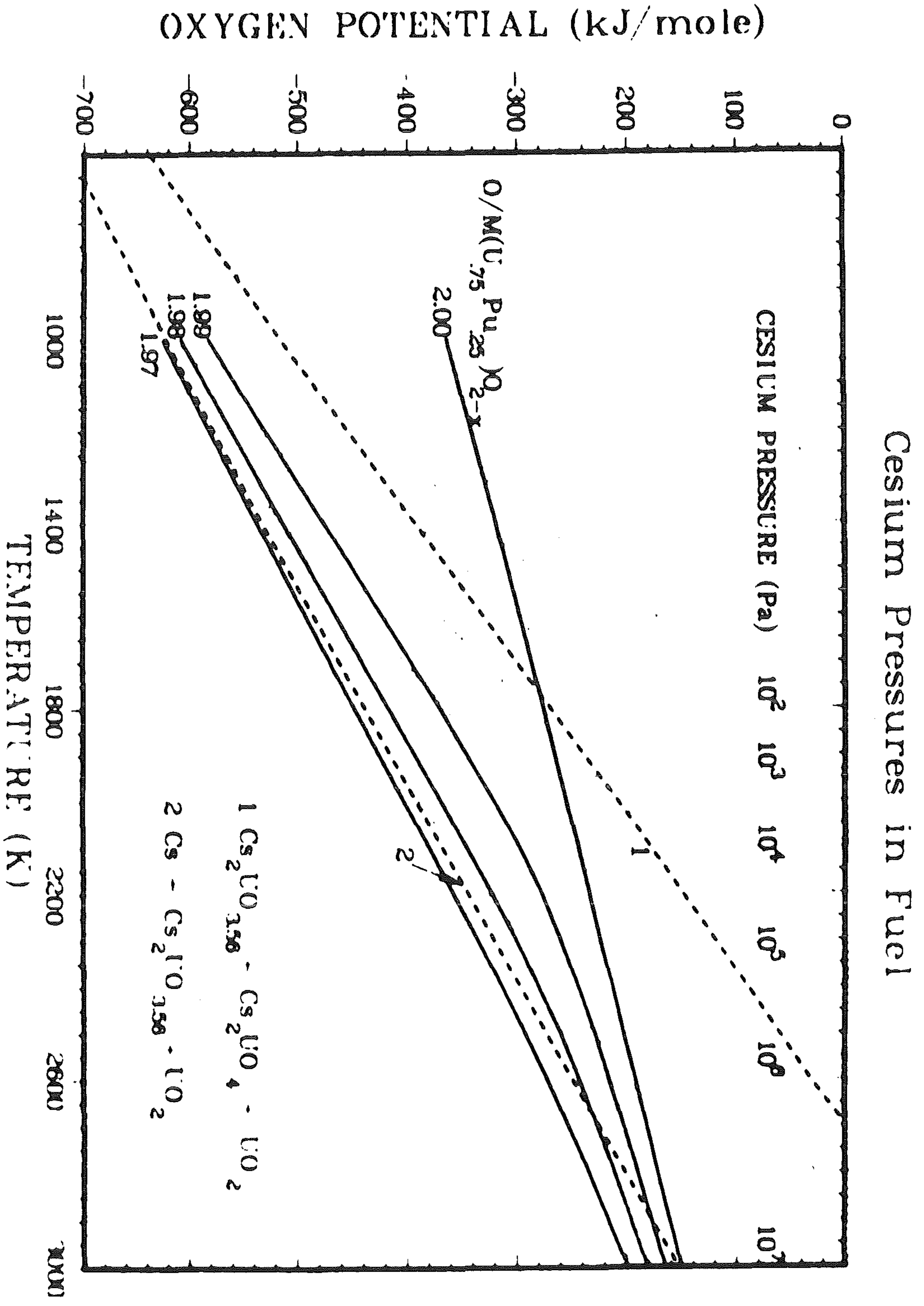


Figure 4 Extrapolation of the Cs-U-O data from Figure 3 to higher temperatures. The dashed lines are phase boundaries. The solid lines represent the oxygen potential range of interest (O/M=1.97...2.00). For $(U_{.75}Pu_{.25})O_{2.00}$ the extrapolation predicts a phase change from Cs_2UO_4 to $Cs_2UO_{3.56}$ at 1720 K, and a phase change from $Cs_2UO_{3.56}$ to metallic cesium at 3000 K. At lower O/M these phase changes occur at lower temperatures.

The magnitude of Cs pressures during a temperature excursion is therefore governed by the question of uranate stability at higher temperatures. However, due to uncertainties in the extrapolation of Fig.4, the predictions of uranate dissociation temperatures and Cs pressures are not reliable enough to be incorporated into existing codes.

Conclusive results for the temperatures encountered in CDA scenarios, can only be obtained experimentally. Also, since time could be required for an uranate dissociation process the pressure build-up must be measured on typical CDA time scales.

2.4 Experimental evidence for Cs pressure effects

To the authors' knowledge the role of Cs in fuel disruption was not addressed experimentally in the past. It was only in some recent experiments that cesium was monitored. In these instances quite obvious indications for the existance of Cs pressure effects could be gathered.

2.4.1 Sandia test FD1.7

In the Sandia experiment FD1.7 a fuel sample from pin PNL-10-12, having 5.4% burn-up, received a total energy deposition of about 1500 J/g from room temperature. The heating was performed by two 5 ms wide neutronic pulses. Maximum fuel temperatures varied between 2600 and 3000 K, depending on radial location. Fuel behavior was recorded on high speed movies. After the experiment the specimen was examined by micrographs, SEM and electron probe analysis /21/.

During the power transient massive swelling occurred in the columnar grain and the adjacent equiaxed grain region. Large voids appeared in this inner fuel region. The outer unrestructured region expanded due to inter- and intragranular cracking. The combined fuel swelling amounted to 55 volume %.

When the radial distributions of Xe and Cs after the test (Fig.5) were compared to those of the sibling sample (Fig.2a,b) it was found that about equal amounts of Xe and Cs were released during the transient. Therefore Cs appears to have similar importance as Xe for the fuel motion observed in test FD 1.7.

The data from test FD 1.7 allow one more important conclusion: Due to the relatively mild temperature excursion of FD 1.7, the fuel lost only a fraction of its Cs and Xe inventory. The release was only 13% for cesium but 50% for Xenon, so that after the test, five times more Cs than Xe is found in the FD 1.7 sample /21/. Therefore in more severe tests where the fuel is brought to higher temperatures for longer times, cesium should dominate the pressures from fission products. Especially the late fuel motion in the cooling channel could be mainly driven by cesium since its inventory in the fuel is not depleted as rapidly as that of the fission gases.

2.4.2 Sandia test serie FD 2/4

Due to the obvious involvement of cesium in the fuel motion observed in FD1 experiments, subsequent FD tests employed absorption spectroscopy to specifically analyse the escaping vapor clouds for cesium. Absorption spectroscopy was used in experiments 2.6, 4.2 and 4.3. In all three tests the following general trends were observed.

The first cesium is detected directly after clad failure in the ejected cloud of aerosols and vapors. This cesium stems very likely from the cesium containing phase in the fuel-cladding gap.

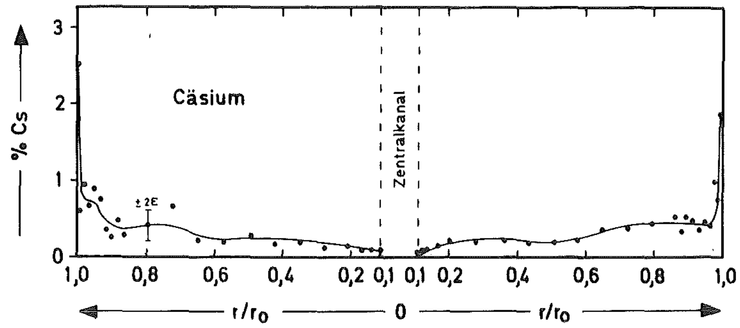


Figure 5 Measured radial cesium concentration in the fuel sample of Sandia experiment FD 1.7. (Xe was detected by dissolution fission gas analysis)/1/.

Comparison with the original Cs and Xe concentrations (Fig.2a,b) showed that about equal amounts of cesium and xenon were released in test FD 1.7. More importantly, the remaining cesium inventory in the test sample is five times that of xenon. So in more severe tests with higher fuel temperatures and of longer times, cesium should clearly dominate pressures from fission products.

The next sudden event after clad failure seems to be a fuel dispersal in the solid or plastic state. This fuel break-up is also accompanied by cesium release, indicating that this cesium probably came from the pores and grain boundaries in the outer fuel region.

The third observed event in the tests is fuel swelling, which is again accompanied by a distinct release of cesium vapor. This cesium may be intragranular cesium or cesium from grain boundary pores.

After the experiment the deposit on the bottom of the experiment capsule was examined by gamma spectroscopy. The by far dominating element in the deposit was cesium, all other volatile fission products showed only minor concentrations.

In summary, the events of clad failure, fuel dispersion and fuel swelling in the above tests was always accompanied by a corresponding discontinuous release of cesium. The amounts of cesium released are significant when compared to the possible Xe release. The authors consider these experimental results to be quite convincing evidence for the existance of important cesium effects leading to fuel disruption under CDA typical heating sequences.

However the now existing experimental data do not allow a quantitative estimate for the magnitude of the Cs pressures. The separation of the Cs effect from other not well known pressure sources like fuel melting or cracking, seems very difficult if not impossible.

The magnitude of Cs pressures needs to be determined in separate cesium vapor pressure measurements which have the advantage of being free from competing pressure effects.

2.4.3 CABRI experiments

The recent CABRI experiments have provided valuable information about the Cs behaviour in transient heating experiments.

For several test pins, the axial distribution of Cs was measured before and after irradiation in the CABRI reactor using the characteristic γ-line of Cs^{137}. Since there is probably not much cesium accumulated in the gap of the 1% burn-up pins, the signal measured at a given axial location should be closely proportional to the radial average of the cesium concentration.

Experiment BI1 was an LOF without additional TOP /7/, having a characteristic time constant of 7 seconds. The clad melted nearly completely along the full pin length. The fissile column remained essentially intact, only from a small part in the pin center about 6g of liquid fuel were ejected. The axial Cs distributions before and after test are shown in Fig.6. Cs migrated from the hot upper pin section down into the lower cooler part. The depletion in the hot zone reaches up to 50% of the original inventory.

Experiment AI2 was a pure TOP test with 20 ms pulse FWHM and a total peak energy deposition of 880 J/g /8/. The fissile column showed central melting over a large fraction of its length. A moderate amount of fuel was released into the cooling channel. Fig.7 shows that the Cs release in this test is more extensive than in test BI1. Except in the lower cooler 20% of the pin the Cs concentration decreased by up to 80% from the original level.

Experiment AI3 was again a TOP, (30 ms FWHM), with an increased peak energy deposition of 1280 J/g /9/. The fuel melted nearly completely and a large amount was ejected into the cooling channel. As can be seen from Figure 8, Cs was practically driven out completely, only minor fractions of the initial Cs remained at both pin ends.

It is interesting to note, that in AI3 the hodoscope detected extended fuel motion long after voiding of the cooling channel. The experimenters suspect either fuel vapor (if there is any) or fission gas as the driving force. Recalling from experiment

Figure 6 Axial cesium distribution before and after CABRI test BI1 /7/. In this mild test with little fuel melting, cesium migrated from the hot upper pin section down into the cooler part.

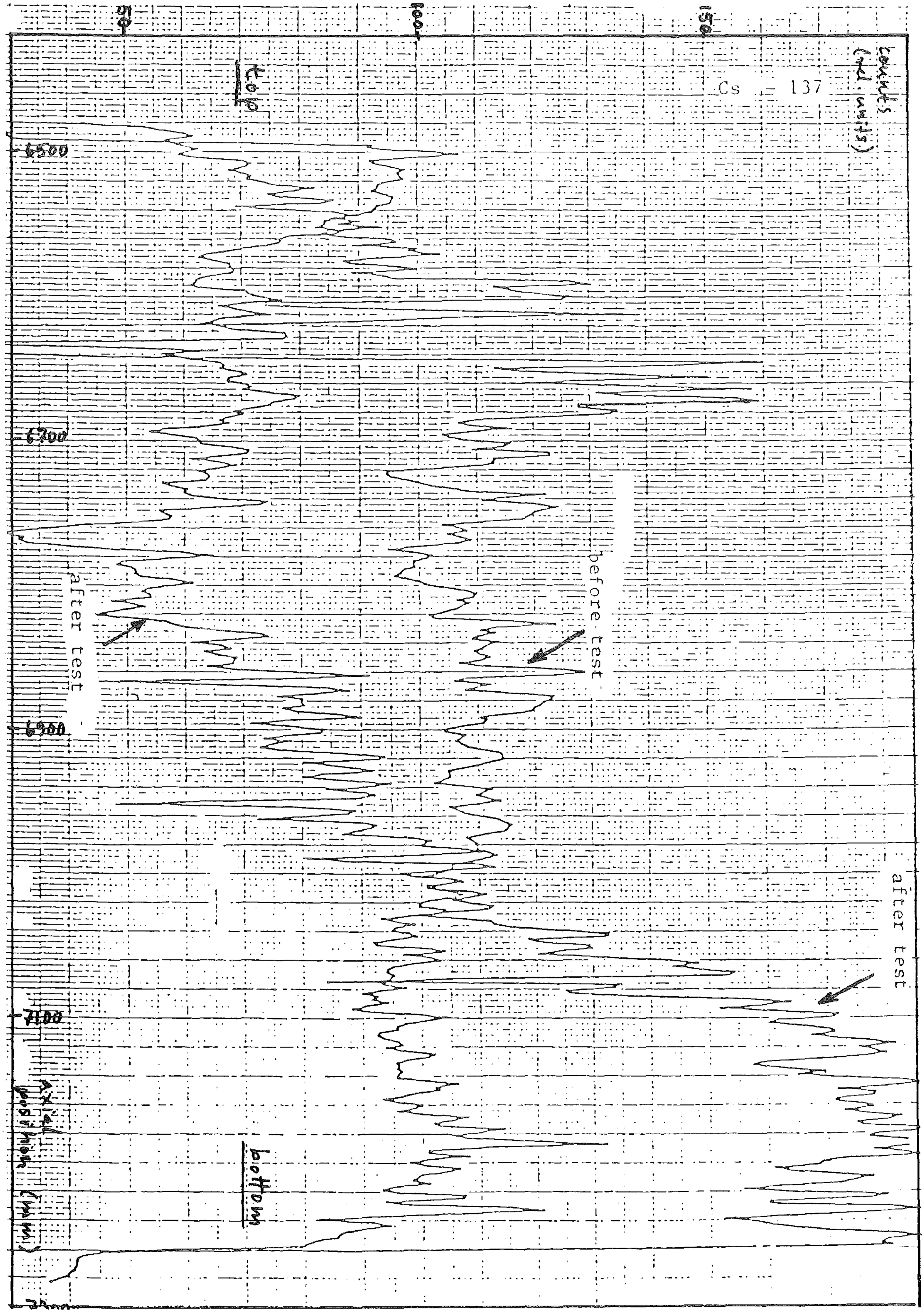


Figure 7 Axial cesium distribution before and after CABRI test AI2 /8/. The peak energy deposition was 880 J/g, central fuel melting occurred over a large fraction of the pin. Cesium release in this test is more extensive than in BI1.

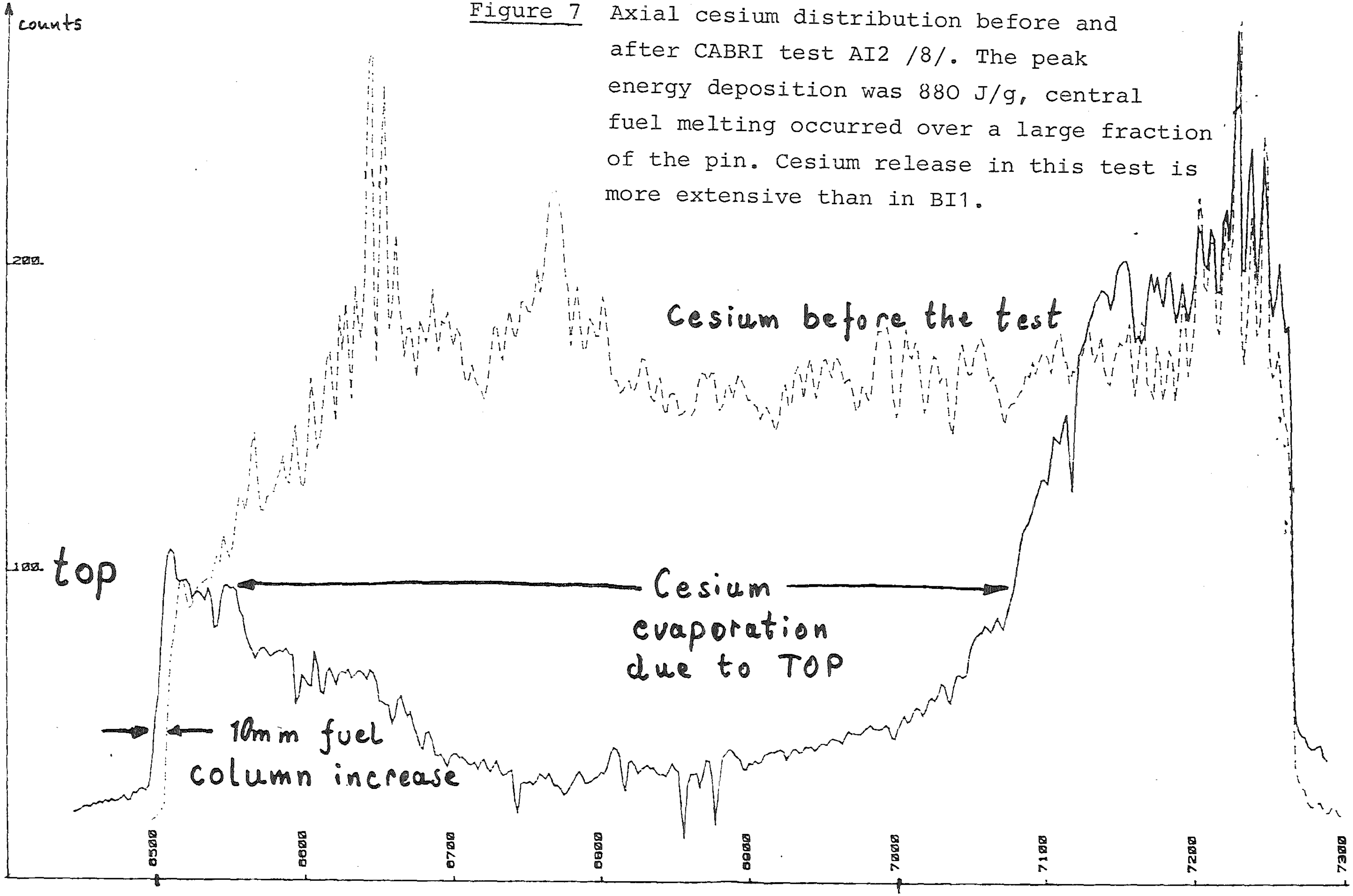
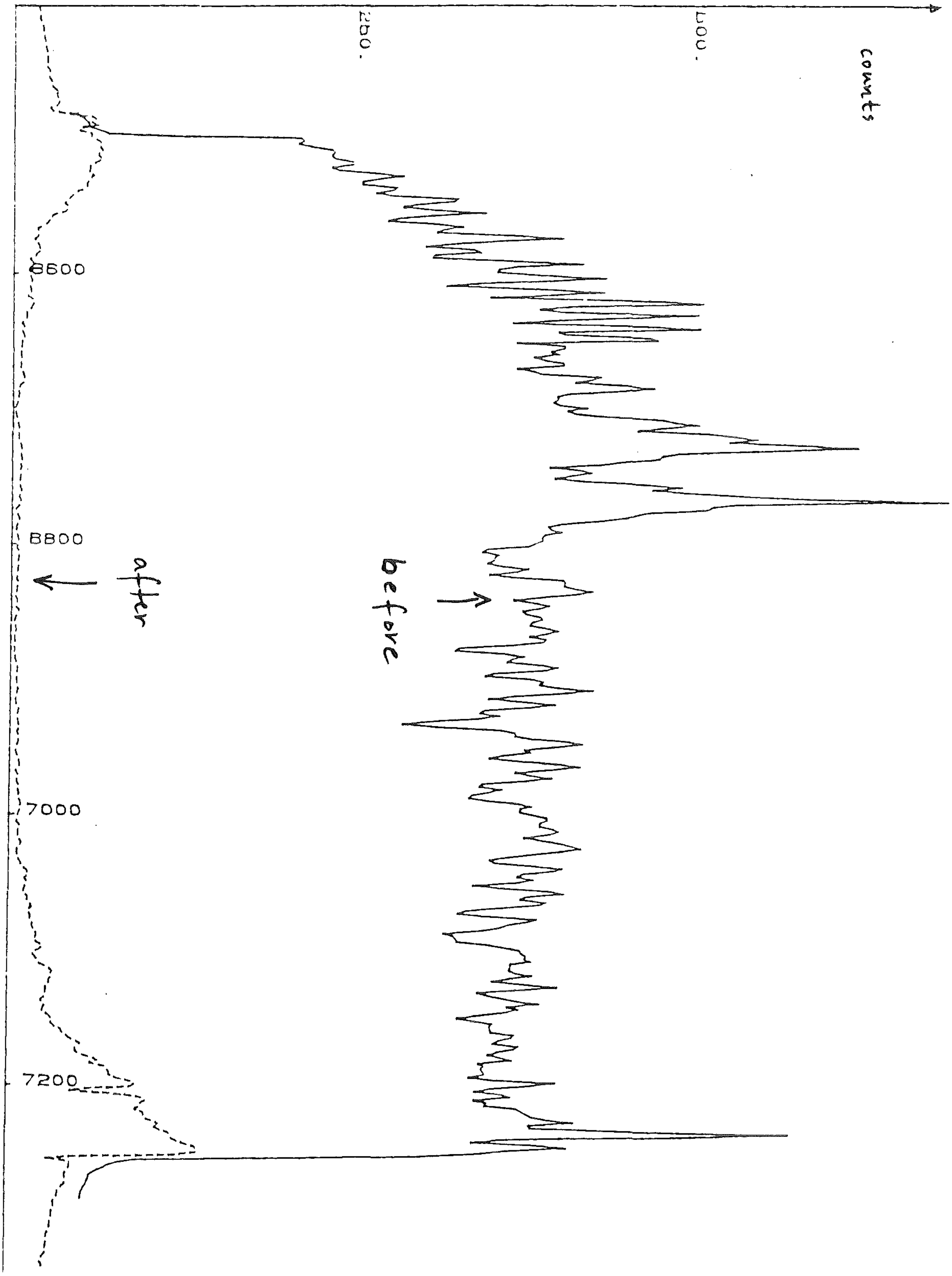


Figure 8 Axial cesium distribution before and after CABRI test AI3 /9/. The peak energy deposition was 1280 J/g. Large amounts of liquid fuel were ejected into the cooling channel and significant fuel motion occurred. Practically all cesium has disappeared from the fuel column.



FD 1.7 that cesium should have its highest driving potential late in time, and considering the complete cesium release in AI3 (Fig.8), the observed fuel motion may well have been due to cesium pressures.
A special feature of the CABRI experiments should be mentioned here. Due to the short heating time and the flux inversion, only small radial temperature gradients exist in the test fuel, meaning that during the transient nearly all of the fuel at a given axial location reaches the liquidus at the same time. The action from released cesium under these circumstances may be different from that in reactor accidents. For more realistic experiments, slower transients would be preferable which allow the formation of typical radial temperature gradients due to thermal relaxation of the fuel.

In summary the above described CABRI tests show a strong correlation between the degree of fuel movement and the amount of Cs released. This correlation is not accidentally because

- similar effects were observed for iodine /8/, which is another volatile fission product (of low concentration however),
- no such correlation was found for fission products of low volatility, like Zr or La. These fission products simply stay within the liquid fuel.

The observed redistribution of relatively large amounts of Cs in confined space and in short times, appears as a strong indication for Cs driving pressures which are comparable to or even larger than fission gas pressures. Cesium appears to have an especially high potential for fuel motion when the fuel reaches liquidus temperatures for long times because under such conditions the full cesium inventory within the fuel can become active, at a time when the fission gases have already been released.

2.5 Conclusions

At present, all codes describing transient fuel behavior under CDA conditions neglect volatile fission products, they only account for pressure effects from the fission gases Xe and Kr.

The conclusions for the significance of Cs in the analysis of Core Disruptive Accidents are:

- The fuel of irradiated LMFBR pins contains significantly more Cs than fission gases, e.g. 10 times more in the example of Fig.2.
- Calculations assuming a metallic state for the cesium, result in equilibrium pressures which are more than 10 times higher than corresponding fission gas pressures.
- The possible but unresolved formation of Cs-uranates (Cs_2UO_4, $Cs_2UO_{3.56}$) can reduce the Cs pressures by orders of magnitude, compared to those of metallic cesium.
- Theoretical estimates for the stability of Cs-uranates and their corresponding Cs pressures are not conclusive at the high temperatures of interest (1000-3500K).
- There is no experimental information about the stability of Cs-uranates and their Cs pressures beyond 1400 K. The vapor pressure of metallic Cs is only known up to 2050 K.
- Strong indications exist for an important pressure source from Cs vapor in in-pile tests conducted at SNL and at CABRI.
- The magnitude of cesium pressures effects cannot be quantified from the complex in-pile tests.

In view of this situation, the vapor pressure over Cs_2UO_4 and $Cs_2UO_{3.56}$ should be measured up to about 3500 K on typical CDA time scales. The measurement of a well defined pressure-temperature relation would reveal:

1. the stability range of the Cs-uranates,
2. the required Cs vapor pressure data, and
3. the importance of rate effects on Cs pressures, by comparing results from two largely different heating rates, representing LMFBR and LWR situations, respectively.

If the measured Cs pressures should be comparable to or larger than fission gas pressures, their incorporation into new transient fuel behaviour codes which are presently under development - e.g. CALIPSO at KfK for interpretation of the CABRI experiments and SANDPIN at SNL for modelling of the STAR experiments - would significantly extend the capabilities of these codes.

3. Significance of Cs in LWR safety analysis

The main risk to the public from Light Water Reactors derives from the potential fission product release into the environment during severe accidents. Therefore the main goal of LWR safety studies is estimation of the amount of fission products released from the reactor containment, the so-called source term, and calculation of the resulting health hazard.

3.1 Health hazard from Cs release

Both the American Reactor Safety Study (WASH-1400) and the German Risk Study identified Cs as an important contributor to the public risk from released radioactivity. The main reasons are large fission yield, high volatility, and high decay energies of Cs-134 and Cs-137. The combined concentration of both cesium isotopes in LWR fuel is typically around 1.3 kg/tU at a burn-up of 30.000 MWd/t /17/.

The importance of cesium is demonstrated by Fig.9, which shows calculated total body doses from surface deposited radionuclides as function of time /11/. During the first week after the release, the dose is determined by iodine species, which however decay fairly rapidly. At later times the body dose is governed by Cs isotopes, reaching 90% at 50 years. Note that the dose received from the external surface, which is shown in Figure 9, is considered to be the most important path for radioactive exposure of the public /12/.

Cesium appears as the dominant long-term contributor to the health hazard from released fission products. Consequently a detailed understanding of the Cs release in degraded core accidents is important for reliable risk estimates.

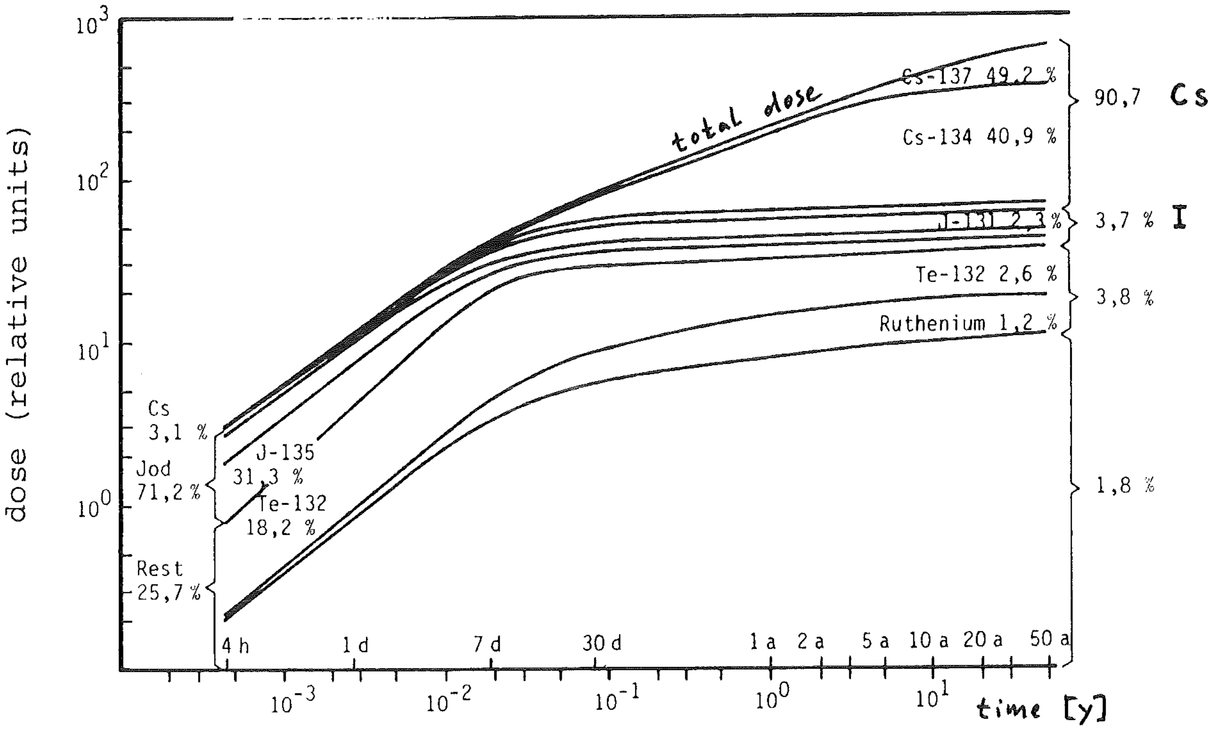


Figure 9 Development of the total body dose from external ground radiation /11/ (Release category 2, neutral weather, distance 10 km). During the first week after the fission product release the body dose is determined by iodine isotopes, at later times the cesium isotopes become dominant, reaching 90% at 50 years.

3.2 Analysis of Cs release

The in-vessel fission product release from the fuel during a melt-down accident is generally separated into three phases.

1. Gap release: When the heated clad bursts under the internal fill gas pressure gaseous and volatile fission products in the fuel-cladding gap are vented into the pressure vessel. The Cs gap release is largely determined by the Cs vapor pressure of the relevant Cs compound in the gap.

2. Solid fuel release: During the heat-up phase, which raises the average fuel temperature from typically 1200 to 2200 K within minutes to hours, depending on the accident considered, Cs release is supplied from two main sources. Firstly, the Cs which was transported during the steady state operation by evaporation-condensation process into the interconnected porosity of the outer fuel region, now vaporizes and can be released fairly rapidly. Secondly, the Cs which is still in solution within the fuel grains, migrates to the grain boundaries. Because also the stable fission gases Xe and Kr diffuse to the grain boundary, a rather porous interlinked structure can develop, which provides newly created free surfaces for the release of Cs vapor. Since vapor transport is involved in the release of both grain boundary and intragranular cesium, the total Cs release during this accident phase will depend on the Cs vapor pressure.

3. Melt-down release: After fuel melting it is expected that a melt pool will be formed in the lower part of the pressure vessel, consisting of molten fuel, Zircaloy and structural material. Because of the high temperatures involved, the remaining Cs will rapidly leave the molten pool. For detailed release calculations the cesium partial pressure over the pool is required for temperatures up to 3000 K.

The above outline of the Cs release shows that the Cs vapor pressure of the relevant Cs compound plays an important role in each of the three release steps. The basic reason is that the transition of cesium into an airborn form which can be carried away through the atmosphere, requires an initial evaporation process. An important quantity needed for the calculation of Cs release rates from the fuel is therefore the Cs pressure of the dominant Cs compound in the fuel.

The in-vessel cesium release as described above, is of course only the first step in a sequence of transport processes. Other transport mechanisms, especially the aerosol behaviour, determine the fraction of cesium which is carried out of the primary circuit or the containment. It appears, however, that for these processes sophisticated codes exist (CONTAIN,NAUA), which allow a quite advanced modelling of the involved transport phenomena. The theoretical modelling of the in-vessel release seems to be in a less satisfactory state. One important reason for this is the widely scattered experimental information on the cesium release from LWR fuel.

3.3 Measured Cs release values

Experimental data on fission product release from LWR fuel were obtained by heating single pellets or small pins in out-of-pile tests. The time dependent release is commonly fitted to a fractional release curve of the form

$$f(t) = 1-\exp(-\alpha t)$$

where α is the experimentally determined "release rate constant". The constant α is assumed to depend only on the temperature at which the experiment was run. The influence of other test parameters like e.g. surface areas or carrier gas flow rates is omitted.

The resulting data base for measured Cs release rate constants is shown in Fig.10 /13/. The data points in the most important temperature range (1200-2000°C) vary by three orders of magnitude.

It is obvious that on this data base a reliable pre-calculation of the fuel release in a degraded core accident is difficult. The problem with the "release-rate-approach" is that the effects from a number of interrelated physical transport phenomena are buried into one single quantity α, which in addition is only depending on one single variable, namely the fuel temperature.

In order to overcome these difficulties, release rate calculations done in the past are generally based on the highest measured values of α, leading to essentially complete in-vessel cesium release in any accident. This approach leads however to rather pessimistic estimates of the cesium release. The authors of /13/ feel that the base case shown in Fig.10 is more realistic.

A more promising approach to description of release fractions appears to be the mechanistic modelling of the microscopic release steps, using the relevant thermophysical data. The authors believe that in this way cesium release values will turn out to be significantly lower than the total release which was used as an upper bound in the past. Preliminary calculations using cesium vapor pressure estimates and gas-phase mass transfer correlations did indeed indicate the potential for order of magnitude reductions in release-rate estimates (A.R. Taig /13/ p.A-9). One of the most important input data for mechanistic release calculations is the Cs vapor pressure of the relevant cesium compound in the fuel.

3.4 Cs compounds in LWR fuel

The Cs release depends on the chemical state of the Cs in the fuel pin because the Cs partial pressure over different Cs compounds can vary by orders of magnitude (see e.g. Fig.3). It is therefore necessary to assess the dominant chemical form of Cs in LWR fuel.

Initially the newly created cesium atoms reside in interstitial sites within the urania lattice. By random walk processes the Cs atoms can meet other fission products or reach the grain boundary where gas phase reactions with oxygen and/or gaseous fission products can occur. An estimate for the dominant chemical form in which the Cs will finally end up, can be made by comparing the free energy change associated with the consumption of Cs in the various chemical reactions.

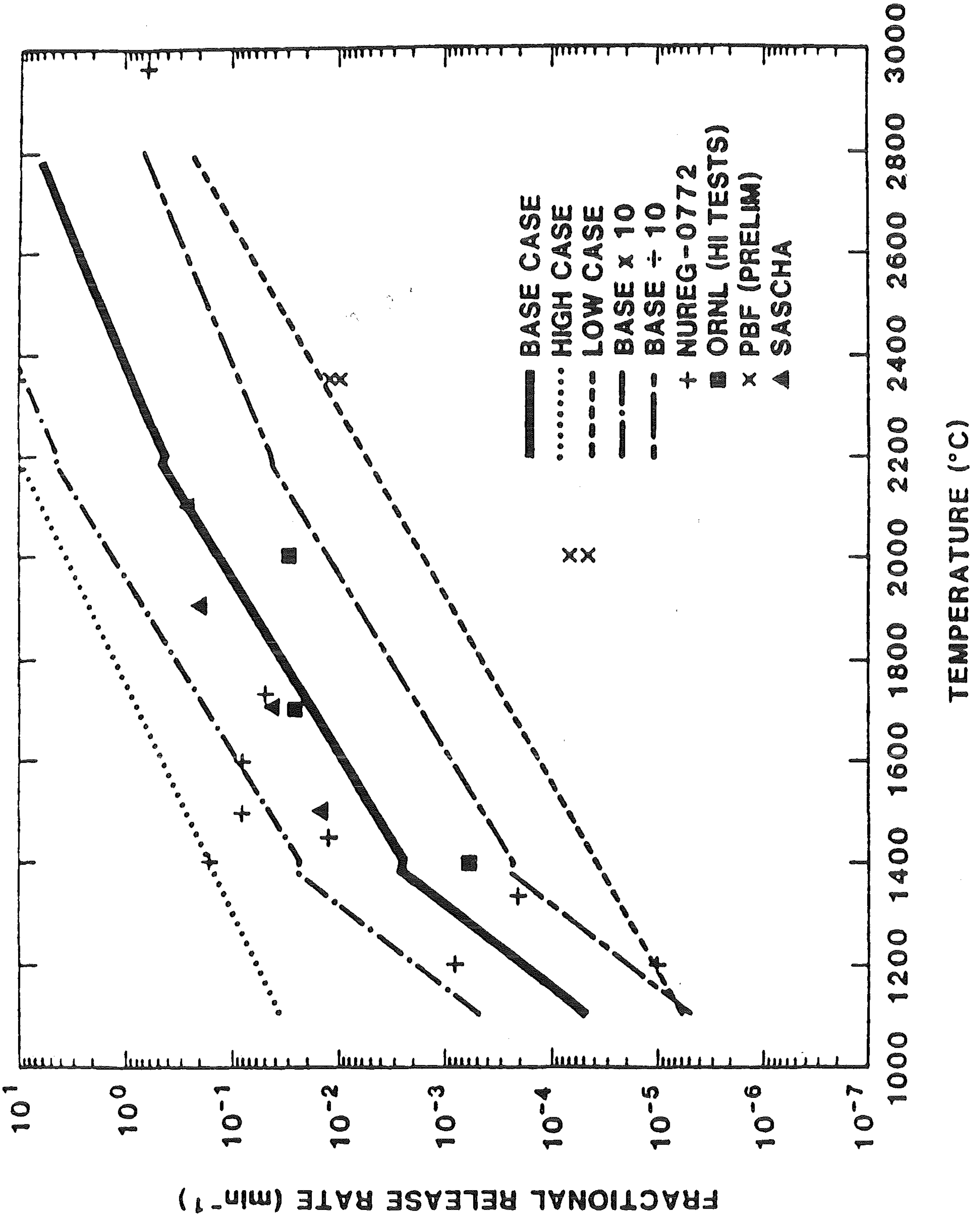


Figure 10 Comparison of release rate constants for cesium as measured in LWR fission product release experiments /13/. The data points in the most important temperature range (1200-2000^{o}C) vary by four orders of magnitude.

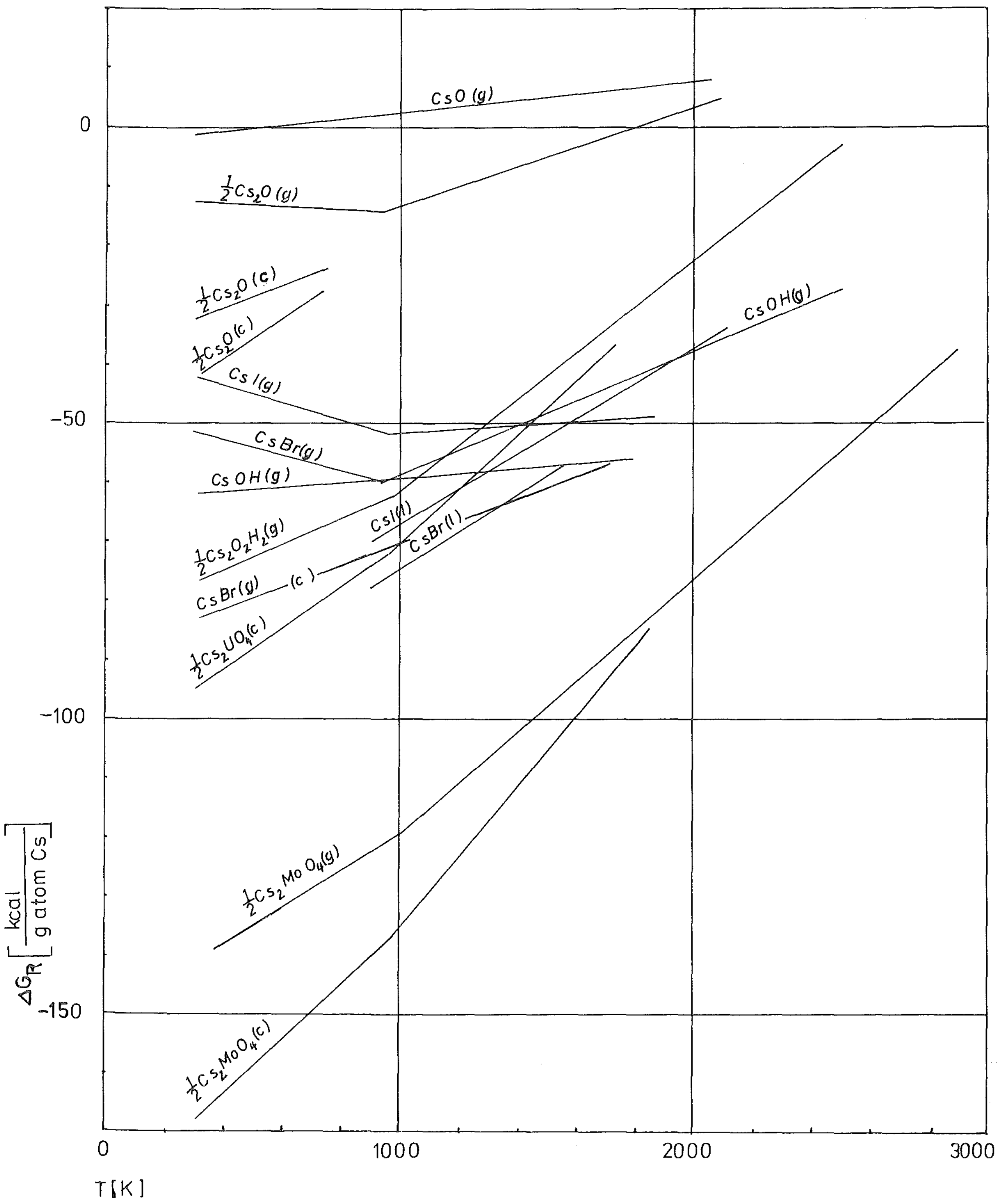


Figure 11 Free energy change in the reaction of cesium with irradiated LWR fuel constituents.

Most stable and abundant cesium compounds in LWR fuel are the cesium molybdate and the uranate Cs_2UO_4. Iodides and bromides are not important due to the low inventory of J and Br. (See Table IV).

The important Cs reactions and the corresponding free energy changes were taken from the most recent assessment of LWR fission product chemistry /14/. Figure 11 shows the free energies of reaction as function of temperature: Most stable Cs compounds in the LWR fuel are in decreasing order the molybdate, bromide, uranate and iodide. However, besides the thermodynamic stability also the abundancy of the Cs reaction partners in the fuel must be taken into account. In view of the low inventories of I and Br (Table IV) the iodides and bromides become unimportant, both elements together cannot bind more than 7% of the existing cesium.

The dominant chemical Cs compounds in LWR fuel are therefore the Cs-molybdate Cs_2MoO_4 and the uranate Cs_2UO_4. The Cs vapor pressure of these substances should therefore be important for the Cs release from LWR fuel.

Table IV: Inventory of Cs reaction partners in irradiated LWR fuel with 2.9% burn-up /15/.

Reaction partner	Cs	Mo	I	Br	U (as UO_2)
Inventory (atom %)	.53	.64	.03	.005	93.9

3.5 Conclusions for LWR safety analysis

The discussion given above leads to the following conclusions concerning the role of cesium in LWR safety analysis.

- Cesium isotopes play an important role in the health hazard from released fission products, they dominate long-term body doses (Fig.9).
- The cesium release calculations done for LWR accident studies lead to overly pessimistic upper bound estimates. More realistic release estimates can be obtained with mechanistic modelling of the release process.

- For mechanistic modelling of Cs release rates from LWR fuel, the Cs vapor pressure of the relevant Cs compounds belongs to the most important input data.

- The important Cs compounds in LWR fuel, bearing the highest Cs release potential, are Cs molybdate (Cs_2MoO_4) and Cs uranate (Cs_2UO_4).

Based on these findings, it is proposed to measure the unknown vapor pressure of Cs_2MoO_4 and Cs_2UO_4 as function of temperature. The temperature range should extend to about 3000 K to cover typical molten pool temperatures.

Compared to the more phenomenological integrated release experiments carried out in the past, vapor pressure measurements would investigate only one isolated release process, namely that of vaporization. While the results of the complex integral release experiments are generally very difficult to interprete, e.g./16/, the proposed vapor pressure measurements provide well defined thermophysical input data for mechanistic release calculations. These data are needed for release codes like PARA-GRASS (ANL) and VICTORIA (SNL), which are presently under development.

4. Comparison of possible experimental techniques

The proposed investigation of Cs pressures calls for an experimental technique which allows
- to heat cesium compounds up to around 3500 K,
- to measure pressures up to 100 MPa, and
- to vary the heating rate to investigate rate effects.

There are basically three techniques which can be considered, namely the existing in-pile EOS technique at SNL, a "laser boiling point" method, recently developed at KfK-INR, and autoclave measurements. The apparent trade-offs and capabilities of these methods are compared in Table V.

Autoclave measurements have the serious drawbacks of high costs, long development time and limited temperature range. On the other hand the effort to develop a suitable autoclave technique would lead to complete p-v-T information of high accuracy.

The laser method is a proven technique for metals, which allows fast and low cost investigations. However, at present it seems not possible to extend the measurements beyond pressures of 1 MPa. Due to this limitation it will not be possible to extend the vapor pressure data base for metallic Cs and probably only a little for the Cs uranates. Rate effects cannot be studied because the pressure measurement is made at a stationary temperature. Nevertheless, it appears that laser experiments on Cs compounds could result in valuable complementary information on Cs pressures below 1 MPa, mainly due to the superior temperature determination (optical pyrometer) and the large number of data points possible.

The in-pile method seems to be the only one which can cover the temperature and pressure ranges of interest for all the materials of interest. Due to its transient heating mode it is also the only method which allows the investigation of rate effects, which may be important for the LMFBR case. Compared to other methods, the temperature resolution is inferior, but the uncertainty of ± 4% is still better than typical temperature uncertainties in the code calculations where the p-T data would be used.

Table V: Comparison of methods for measuring vapor pressure data of Cs-compounds

	Advantages	Disadvantages
reactor	- proven method - short lead-time after start of program - continuous p-T-curve from one shot - high pressures possible (100 MPa) - required temperature range is fully covered - rate effects can be investigated (transient heating method) - heating method typical for LMFBR - simple interpretation - equilibrium very likely (closed system, large evaporation surface)	- limited number of experiments - highest temperature uncertainty (± 4%) of methods
laser	- proven method for metals - available method - high temperatures possible - lowest costs	- pressure limited to below 1 MPa - equilibrium not certain (open system, photon heating, temp. gradients) - point by point measurements - no investigation of rate effects possible (stationary temperature method) - empirical method (in depth interpretation difficult)
autoclave	- most precise method - many data points possible - measurement of complete p-v-T surface possible	- long lead-time - not proven for melting points above 700 K - most expensive method - limited to below 2500 K

5. Proposed in-pile tests

5.1 In-pile method

The experiments could be conducted in the ACRR at SNL, using the existing data aquisition system and the hardware design developed for the recently completed unirradiated fuels EEOS tests. Several minor modifications could tailor the method for the samples of interest here, e.g. lower energy deposition by increasing the spectrum hardness or lowering the sample enrichment.

The analytic tools for data reduction and analysis are those developed for the unirradiated EEOS series.

5.2 Test matrix

Table VI shows the recommended test matrix for five tests. The investigated substances are metallic Cs, Cs_2UO_4, $Cs_2UO_{3.56}$ and Cs_2MoO_4. The two uranate phases could be prepared from enriched uranium at LANL, at relatively modest costs /10/. The metallic Cs and the Cs-molybdate could be heated in-pile by adding enriched UO_{2-x} powder to the powder samples. In this case the UO_{2-x} would simply act as an inert heating medium via radiation and conduction. The vapor pressure of UO_{2-x} is negligible in the temperature range of interest.

5.3 Test objectives

The experiment on metallic Cs would be an initial proof test of the in-pile method for samples heated indirectly by admixed UO_2 powder. The results can be compared to the existing out-of-pile data, which extend up to the critical temperature of 2050 K. Since pressures up to 100 MPa can be measured in the

Table VI: Proposed test matrix for cesium vapor pressure measurements on Cs compounds

Number of Experiments	Design Goal	Sample Composition	Heating Time (ms)	Maximum Temperature (K)
1	Proof test	$Cs + UO_{2-x}$	10	3000
1	LMFBR	Cs_2UO_4	10	3500
1	LWR	Cs_2UO_4	100	3500
1	LMFBR	$Cs_2UO_{3.56}$	10	3500
1	LWR	Cs_2MoO_4 $+ UO_{2-x}$	100	3500

transient in-pile technique, it should be possible to extend the vapor pressure data base of Cs metal close to 3000 K, far into the supercritical region of cesium.

The two experiments on Cs_2UO_4 are designed to measure the vapor pressure data of this compound under fast and slow heating rates, representing LMFBR and LWR situations, respectively. Since the acting pressure depends strongly on the constitution of the Cs-U-O system, dissociation of the uranate into metallic Cs and UO_2 should be detectable as a sudden increase of the system pressure at the dissociation temperature. Measurement of the p-T relationship would reveal if and at what temperature such a phase transition occurs. Comparison of the slow and fast heating tests allow conclusions about rate effects in pressure development.

The experiments on $Cs_2UO_{3.56}$ and Cs_2MoO_4 have the objectives to measure vapor pressure data and to investigate dissociation phenomena for these Cs compounds. The choosen heating rates represent LMFBR excursions for $Cs_2UO_{3.56}$ and LWR situations for Cs_2MoO_4.

If sufficient funds should become available, an additional sixth test would be conducted with the cesium containing multielement compound which is often found in the fuel cladding gap of reactor pins.

6. Summary

The significance of cesium - which is the most abundant volatile fission product in reactor fuels - is shown for LMFBR and LWR safety analysis.

For the LMFBR case the conclusions are:

- The cesium pressures during a CDA temperature excursion are governed by the stability of the initially present Cs-uranates (Cs_2UO_4, $Cs_2UO_{3.56}$). Should the Cs-uranates dissociate, pressures much larger than fission gas pressures must be expected.

- Theoretical estimates of the stability of Cs-uranates and the corresponding Cs pressures for the high temperatures of interest (up to 3500 K) are not conclusive.
- Recent in-pile experiments at CABRI and SNL have shown strong indications for a considerable pressure source from cesium.
- The magnitude of cesium driving pressures cannot be deduced reliably from already existing or near-future in-pile tests like the STAR or the CABRI experiments.
- The apparently large potential for fuel motion from cesium pressures could be clarified by CDA-typical in-pile vapor pressure measurements on Cs_2UO_4 and $Cs_2UO_{3.56}$.
- Without such experiments the potentially advantageous role of cesium in driving fuel dispersion will remain unresolved.

The conclusions for the LWR case are:

- Cesium isotopes play an important role in the health hazard from released fission products, they clearly appear to dominate the long-term body dose from released fission products.
- The important Cs compounds in LWR fuel are the molybdate Cs_2MoO_4 and the uranate Cs_2UO_4.
- The unknown Cs vapor pressures of these compounds are among the most fundamental input data for mechanistic fission product release codes, presently under development at different laboratories.
- The Cs pressures of Cs_2MoO_4 and Cs_2UO_4 could be measured for the temperature range of interest using existing in-pile techniques.

A test matrix for five experiments is presented. Conduction of the proposed tests would largely resolve the presently existing data deficit for Cs pressures in LMFBR and LWR accident analyses.

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