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Specific Features of Cesium Chemistry and Physics Affecting Reactor Accident Source Term Predictions

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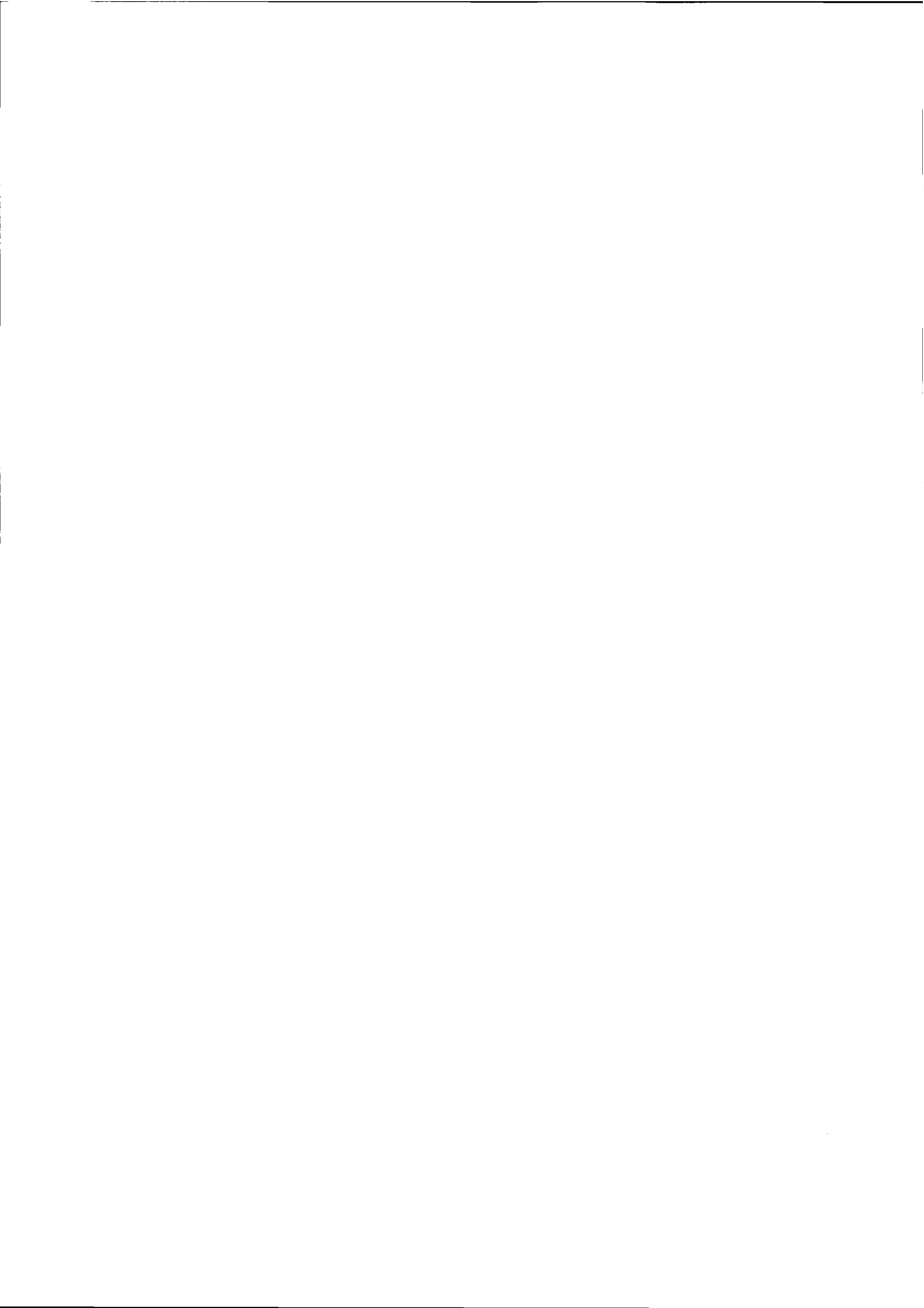
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Abstract

In the process of assessing remaining uncertainties in predicting the source term of severe reactor accidents a special investigation is devoted in this report to the case of cesium. The cesium isotopes, especially Cs137 and Cs134, are among those nuclides which could have a major impact on the environment in the event of a release.

The processes for release from fuel and retention in the reactor coolant system and the containment are treated. The intention is to identify and discuss those specific features of cesium chemistry and physics that strongly affect source term predictions.

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1 Purpose and background

For many years after the TMI-2 accident one of the major issues in reactor safety research was the analysis of the behaviour of iodine during a severe accident. Many studies, workshops and seminars have been devoted to this topic and much progress has been achieved in this complex area. At the workshop on iodine chemistry in reactor safety 1991 [1] it was noted that the studies should be broadened to examine fission products other than iodines such as cesium and tellurium.

In the process of assessing other remaining uncertainties to predict the source term, a proposal was given to carry out a special assessment for cesium, even if the behaviour of cesium may be less complex than that of iodine. On the other hand, cesium is at least as important as iodine, when the environmental impact is concerned, especially long term land contamination.

The general aim of the present report is to contribute to the assessment of what gaps in our present state of knowledge are detrimental to our ability to accurately predict the cesium source term for specified accident sequences.

The original purpose was to summarize exploratory work on cesium source term issues with particular emphasis on specific information needed for consequence modelling. Some initial work and the preparation of an outline of a report were carried out in 1992.

After the issue of the draft (now printed) of the state of the art report on Primary System Fission Product Release and Transport [2], which is a comprehensive treatment of the source term subject and includes a variety of processes for cesium as well as other fission products, the scope of the present report was somewhat reduced. Thus references are made to the many documents available instead of repeating and describing facts and phenomena well known to the expert reader.

The present investigation, however, implied that quite a number of documents were studied and therefore some comments are given to put the more cesium specific material included into perspective.

That concerns cesium behaviour before and during the release from the fuel as well as chemical and physical interaction with core and structural materials. Recent results of re-vaporization of cesium are also included. Some observations of cesium behaviour in the Chernobyl fallout are given as an Appendix.

For the processes in the containment a report on the physical and chemical characteristics of aerosols has been issued by FPC [3]. That report addresses several topics of special relevance to cesium, e.g. reactions with boric acid and the stability of CsI during hydrogen burning.

2 Relevance of cesium source term to environmental impact assessment

The cesium isotopes, in particular Cs137 and Cs134, are important potential contributors to ground contamination and radiation doses in the environment in the event of a major reactor accident which has resulted in core degradation and containment failure. This is due to the following special features of the cesium isotopes

- substantial inventory in the fuel
- long half-life
- high mobility and volatility in the fuel
- high solubility which influences transport in the environment
- extensive uptake into food products
- high radiation dose factors

The Windscale accident in 1957 focused the attention on the importance of the radiation doses from the iodine isotopes, especially iodine-131. Much effort was spent in the 60's and 70's in various countries and still is being spent, to study the behaviour of iodine and various measures to retain iodine by the use of scrubbers, sprays and filter systems. Much progress has also been achieved in the understanding of the variety of processes and use of systems for reducing the release of iodine in the event of accidents.

Significant amounts of cesium (20 TBq) were in fact also released during the Windscale accident. [4]. The experience also gave interesting results concerning the rate of decrease of the contamination level on the ground (weathering effect) [5].

At an IAEA symposium in 1973 on environmental surveillance, Beattie [6] argued for a shift of attention from iodine to cesium. This also became apparent from the WASH-1400 [7] results.

During the TMI-2 accident in 1979 the fission products released from the damaged core were retained in the primary circuit and the containment except for small quantities of noble gases and minor amounts of iodine, which were released to the atmosphere. The high retention of iodine was a surprise to quite a number of people and was not considered compatible with the US source term prevailing at that time, predicting iodine to be in elemental form [8]. However, several European experimental results [e.g. 9-11] showed quite extensive retention even for elemental iodine. A special task force was initiated by the NRC to evaluate the technical basis for estimating fission product behaviour during LWR accidents. A final report was issued by the NRC [12] in 1981. The objective of the report was also to identify where gaps exist in our knowledge. A major issue was the discussion of the chemical form of iodine after an accident in a LWR. This discussion also had relevance to cesium. Important conclusions were that the RCS certainly would retain a lot of the fission products and that experiments were needed to obtain a better database for predictions. To provide that database was the aim of the large scale Marviken ATT (Aerosol Transport Test).

In March 1986 the CSNI published a report on Nuclear Reactor Accident Source Terms [13], which discussed uncertainties regarding source term assessment. Examples of reasons for uncertainties were shortcomings in models and data, omissions of important phenomena, specification of accident sequences and plant geometry. A list of areas of major importance to source term uncertainty was also presented.

At the Chernobyl accident, which occurred in April 1986, the release of cesium to the environment was reported [14] to be 10–13 % of the core inventory. The most recent estimates [15] suggest 40–60 % (see Appendix 1). The relevance of the Chernobyl accident to source terms for severe accidents in water-cooled and water-moderated reactors of Western design has been discussed [16]. Even if there are substantial differences with respect to the design, safety features and operational practice compared to Western reactors, there were a few observations of the fission product behaviour, which it could be of some interest to examine further. One such item was the chemical form of iodine in the environment, which apparently was not in the form of cesium iodide but appeared to a large extent as gaseous iodine. Some aspects of the cesium behaviour is included in an appendix to this report.

In April 1987, the GREST group of CSNI reported on selected source term topics [17], among them studies of the reactions between cesium hydroxide and stainless steel surfaces also on the behaviour and properties of various cesium compounds (CsI , Cs_2O , Cs_2UO_4 and Cs_2MoO_4).

In 1992 the expert group on Fission Product Phenomena in the Containment issued a report on Source Term Uncertainties – Recent developments in understanding fission product behaviour [18]. This latter report emphasized, as one topic among many others, the need for further experiments on revaporization of deposited fission products. A note on this issue has been prepared for the FPC [19]. Some recent information and results on this topic concerning cesium are also included in the present report.

3 Features of cesium chemistry and physics

Long before the TMI-2 accident it was evident that the major release of iodine and cesium from overheated fuel occurs as atomic or ionic species and not as chemical compounds. The release from solid fuel grains is a process of atomic or ionic diffusion. Another reason is that e.g. CsI and CsOH molecules are dissociated at 1800–2200°C, which is the relevant temperature range for extensive cesium release during a severe accident.

On the other hand, as soon as iodine and cesium meet a cooler environment they can combine and form compounds. Minor amounts e.g. from gap activity, gas bubbles and grain boundaries may appear in molecular form.

The TMI-2 accident in 1979 triggered a new period of extensive studies of the behaviour of iodine in severe accidents. Now also cesium properties and behaviour were included and NUREG-0772 [12] presented important results: Equilibrium calculations for the system Cs-H-O show that CsOH is the dominant cesium containing species in oxidizing and inert environments (H/O ratio between 1.5 and 2.0.) In reducing environments CsOH is the major species at lower temperatures. However, as the temperature is increased above 1000°C there

is an increase in the concentration of Cs vapor. The concentration of Cs vapor is markedly enhanced when there is a large excess of hydrogen present (H/O greater than 3). In the extreme case of very high temperature (greater than 1700°C) and a very large excess of hydrogen (H/O = 30), the calculations indicate that more than 80% of the cesium is present as Cs vapor.

At the symposium on chemical phenomena associated with radioactive releases during severe nuclear plant accidents 1986 [20], several papers discussed the behaviour of cesium, e.g. cesium gas phase species, behaviour in the TMI-2 accident and revaporization from the RCS.

Kinetic calculations of the system Cs-H-I-O were presented by Torgerson et al [21] which showed a rapid formation of the equilibrium compounds discussed above (see Figures 1 and 2). Also Cronenberg and Osetek [22] have presented these types of calculations (see Figure 3). The real system is, however, much more complex than that which can be modelled. Therefore integral experiments are still of interest.

A note on low temperature/low pressure chemistry inside the containment has been prepared by Borioli for the FPC [23]. That note contains information on the chemical and physical behaviour of cesium and properties of some common compounds of cesium.

4 Cesium in the fuel

4.1 Inventory of Cs137 and Cs134

Cs137 is produced by the fission of U235 and Pu239 with the fission yields of 6.2% and 5.2%, respectively. Cs134 is produced by neutron capture in Cs133, which is the daughter of the fission product Xe133. The inventories are dependent on the burnup and can be calculated by the use of e.g. the ORIGEN 2 code. Figure 4 gives the ratio of the two cesium isotopes as well as that for relevant cerium and ruthenium isotopes for various burnups. The increasing importance of Cs134 compared to Cs137 should be taken into account for high burnup fuel. As an illustration some data from the Chernobyl accident are given below.

The average Chernobyl burnup was 10 MWd/kg [14] and the Cs 134/Cs 137 ratio 0.59 according to measurements of fallout [24]. Of the first year total open air cesium dose equivalent due to ground deposition from Chernobyl, about 55 % was caused by Cs 134 and 2 % by Cs 136. The remaining slightly more than 40 %, was due to Cs 137 [25]. The dose from ingestion is a small fraction of the total Cs dose. Of the 50 year total Cs dose the Cs 134 contribution is about 20 % [24].

4.2 Migration and distribution in the fuel

A small fraction (typically a few percent) of the cesium content in the fuel migrates from hotter parts of the fuel during reactor operation and concentrates at cooler regions of either the fuel pellet or fuel rod or at the cladding. This is also the behaviour of iodine and noble

gases and to a lesser extent also of tellurium. There are several studies of this radial and axial redistribution [e.g. 26, 27].

Power ramps and high burnup favour the fission product release from the pellet structure [e.g. 28]. Due to the cracking and formation of gas tunnels a certain fraction may reach the gap and the cladding. Normal 96% TD (Theoretical density) LWR fuel is more migration resistant than e.g. 92% TD FBR fuel.

There are also some differences in migration behaviour between the cesium isotopes due to different properties of the precursor [29]. Some typical observations for mild transient conditions are given in the next few paragraphs.

At a linear heat rate around 25 kW/m, relocation of the volatile elements, xenon and iodine, starts from the central part of the pellet. The semivolatile tellurium and cesium start to migrate towards the colder cladding at around 30 kW/m in rough figures. At 43 kW/m for some hours the redistribution from the centre may be around 10% for cesium. Tellurium is known to condense on its way to the cladding earlier than cesium and iodine due to the higher boiling point, 900°C, compared to 679°C for cesium and 184°C for iodine [27].

A fraction of the redistributed cesium and iodine will reach the cladding wall and will preferably deposit at the pellet interface positions on the cladding. A transient at 43 kW/m for some hours revealed that an iodine/cesium ratio on the cladding was almost five times greater than the calculated total inventory ratio. An additional measurement twelve days later revealed a loss of iodine 131 of more than 30%, confirming previous Studsvik observations which show that iodine is not efficiently bound in the deposits on the cladding and was probably present in even higher amounts prior to opening the rod [27].

The chemical state of the fission products in intact fuel has been discussed by many investigators [26, 30, 31, 32, 33].

Unlike iodine, cesium reacts with the fuel and forms various cesium uranates, which affects the behaviour in the fuel. It also appears as gaseous molecules of cesium oxides or in elemental form. A low O/M (oxygen potential) favours a higher cesium partial pressure, which enhances the radial and axial transport [26].

In the modelling of fission product behaviour [34] in the FASTGRASS code the following cesium and iodine species were considered: I, Cs; CsI, Cs₂UO₄, Cs₂MoO₄. When comparing calculations of xenon, iodine and cesium migration with SFD experiments, iodine showed the same characteristics as xenon and no CsI formation was predicted [22, 35]. See also Figure 5. Cesium either reacted with the fuel to form uranate or migrated in atomic form. No CsMoO₄ was predicted for the SFD fuel.

Thermodynamic equilibrium calculations of the chemical state of fission products in irradiated UO₂ fuel have been performed by using the SOLGASMIX-PV code [30]. The results showed that cesium at low oxygen potentials existed mainly as zirconate with minor fractions of telluride, cesium gas and CsI. At higher oxygen potential, the cesium form was molybdate and 10% CsI. It is to be noted that cesium uranates were not included in the calculations.

In a review from 1985 [31] the system Cs-Zr-I-O for 400-900°C is discussed in detail. In summary, Cs_2ZrO_3 (as well as higher zirconates) is a good cesium sink and is stable at least up to 915°C. CsI in the fuel was suggested to react with ZrO_2 to give Cs_2ZrO_3 and ZrI_4 . CsI may react also with ZrI_4 to form Cs_2ZrI_6 (melting point 788°C). The authors of Ref 27 are convinced that CsI is not the inert fission product sink so commonly assumed.

On the other hand, moisture will cause hydrolysis of cesium zirconates and other metalates and CsOH will form. Zirconium iodides may also hydrolyze or oxidize to HI or I_2 and CsI can form [31]. In the survey of Kleykamp [26] he states that the chemical state of iodine within oxide fuel is not completely clarified. The high iodine mobility in oxide fuels is associated with the chemical state of this fission product. The formation of CsI within the fuel is therefore not compatible with the well documented high iodine mobility.

In conclusion, a few percent of the cesium inventory in the fuel is collected in the gap and more easily available for release after cladding rupture.

The chemical state inside the fuel before cladding rupture is not expected to have a substantial effect on the release of cesium, which is almost as mobile as the noble gases. The release from fuel at relevant high temperatures is expected to be as cesium atoms. Cesium hydroxide will form in contact with water or water vapour .

5 Reactor accident scenarios

The present study is focused on those severe accident scenarios which start with core degradation and may progress to early or late containment failure, containment by-pass or diffuse leakage from an isolated containment.

The case of a successfully terminated LOCA is analyzed in Chapter 9.1 from the cesium behaviour point of view by using a recent intercomparison exercise within CEC [36].

6 Release from fuel

The release of fission products from the fuel and the core during a core melt accident will occur as a result of a variety of processes and at several phases of core degradation. The heatup rate during a severe accident is dependent on accident scenarios.

A fraction of the gap activity will be released when cladding starts to fail above 1050–1400 K, depending on pressure. The chemical form may be elemental or in the form of a compound. If a LOCA is successfully terminated at this point, no more activity will be released. Such a case is discussed in Chapter 9.

In the early phase thereafter, the fission product release is by diffusion through solid fuel grains but is influenced by cracks and grain-boundary tunnels. The diffusion out of fuel grains will occur as atomic species [35]. Materials collected in bubbles may form different chemical compounds and may be released as molecules [35]. Cracking will enhance the fission product release.

When the fuel liquifies, which can occur at lower temperature (2800 K) than the melting point (about 3100 K) of the uranium dioxide, due to formation of eutecticum between fuel and cladding, the rate of release may increase. A typical rate is 0.1%/s during the in-vessel melt progression [32].

At the symposium on source term evaluation for accident conditions in 1988 [37] cesium releases from fuel during accident conditions were compared to those of krypton and iodine and found to be similar.

Recent studies of fission product release from irradiated zircaloy-clad UO_2 fuel of LWR relevant burnup have been performed at ORNL, SNL, KfK, Grenoble, Idaho and Chalk River. A review of experimental results on LWR core melt progression [38] contains comparisons of these studies. The ORNL studies have been summarized and compared to other relevant results [39]. Figure 6 shows the similarities in release rate for Kr 85 and Cs 134 in a release test in steam but the same behaviour was also found in hydrogen. The most volatile fission product elements, krypton, iodine and cesium are released almost totally at temperatures up to 2700 K, with little effect of atmosphere [39]. Deposition profile on the outlet thermal gradient tube showed maximum iodine deposition at 875 to 775 K as would be expected for CsI, but several deposition peaks for cesium indicated the presence of several different cesium compounds [39].

In steam atmospheres, about 10 % of the cesium has behaved like CsI and the remaining 90 % as the highly reactive CsOH [39]. The aerosol formation was higher with steam compared to hydrogen. According to [39] this behaviour suggests that, in an accident with abundant steam, a smaller fraction of the released cesium would condense on cool surfaces than would occur under reducing conditions. Hence, for cesium, oxidizing conditions appear to represent a greater hazard than reducing conditions [39]. On the other hand as remarked by Sich [15] more Cs was retained in the melt at the (oxidizing condition) Chernobyl accident than in the TMI-2 accident (reducing condition). Release data from melt

experiments have been used for code (CORSOR, VICTORIA, FASTGRASS) improvements and comparisons.

The TMI-2 post accident core mass has been divided [40] into four groups (with fraction of core material in parantheses): intact fuel rods (15 %), upper debris bed (20 %), consolidated mass (45 %) and lower plenum (20 %). The distribution of the total cesium core inventory long time after the accident was as follows [40]: reactor building basement and tanks (42 %), RCS (1 %), auxiliary building systems (5 %), letdown demineralizer resins (5 %). This amounts to 53 %. Thus 47 % remained inside the RPV.

Analyses of the fission product retention in TMI-2 fuel debris showed for Cs 137 a retention of 9-22 % (average 16 %) in the lower plenum debris bed and 6-32 % (average 21 %) in the upper plenum bed [40].

According to a cesium transport model for TMI-2 post accident conditions [41], about 60 % of the cesium released from the fuel deposited on surfaces of the RCS and was slowly leached by the RCS water. This agrees with the STCP calculations [41].

Hobbins et al [38] report that the noble gas (as well as iodine and cesium) release rate in in-pile integral-effects tests increases rapidly when measured fuel temperatures rise above 1570 K and levels off at a steady rate of about $5 \cdot 10^{-4}$ /s from 2200 K (see also Figure 5 from Ref 22 and Figure 6 from Ref 39)

The FASTGRASS code [42] has been used to calculate the cesium release (see Figure 7) after preirradiation and cooldown to simulate normal operation followed by simulated LOCA with a heatup rate of 2 K/s to temperatures in the 1800-2800 K ranges with the fuel then held at constant temperature for up to several hundred hours.

In the Source Term Code Package (STCP) analyses [58], the iodine and cesium are assumed to be in the form of CsI and CsOH and are treated as vapors as they are transported from the core. However, in calculating the transport and retention in the reactor coolant system they can condense on walls and aerosol particles, evaporate from where they have condensed, or become chemically adsorbed by the surfaces. The STCP does consider Cs irreversible deposited as an insoluble specie. Generally, the treatment of fission product chemistry in the STCP is simplistic when it exists, and there remains a very high degree of uncertainty in the chemical forms of released fission products [58].

In general the experimental data base clearly reveals that Cs, I, NG show the highest release rates and that the rates are similar for these elements.

7 Behaviour in the primary system

A state of the art report on fission product release and transport within the primary system was recently prepared for CSNI [2]. That report identified several weaknesses in the current understanding of fission product release. These were not concerned with cesium behaviour but rather low-volatile fission products from molten fuel and releases at high temperatures in air or other high oxygen potential conditions.

A major part of the fission products can be retained in the reactor coolant system. A re-release of the primary retained material can occur by a variety of processes: resuspension, revaporization, reentrainment and revolatilization. In a note by Sugimoto et al [19] these processes are discussed together with information on recent results. Several of the experiments discussed concern the behaviour of cesium and cesium compounds in the RCS or containment. It was concluded that revaporization had a significant effect on source term evaluation.

In a lecture for the seminar on fission product transport processes in reactor accidents in Dubrovnik 1989 Powers [43] discussed the uncertainty distributions for release during core degradation and revaporization (see Figures 8 and 9). For cesium the calculations using STCP showed a more than 80% release at a 50% probability, see Fig. 8.

Cesium deposited on RCS surfaces can partially be removed by water and revaporization. A fraction forms cesium silicate and is not leachable [44]. To illustrate what effect the chemical form might have on predictions of revaporization, results of calculations [44] are shown in Figure 10.

The retention of cesium and some other fission product was analysed and calculated for some accident scenarios assuming the released cesium exists as predominantly CsOH [45]. Effects of chemisorption on aerosols and deposition velocities were discussed [45].

The reactions of CsOH and CsI with 304 SS at high temperatures in steam atmosphere have been studied [46] and it was found that cesium silicates were formed.

Studies of revaporization of CsOH from stainless steel have recently been reported [47]. Previously it was shown that prolonged heating of samples after exposure to CsOH vapour, particularly at increased temperature, results in transformation of soluble into insoluble product. When samples were gradually reheated to 1500°C, three types of release were observed, one at 500–800°C, the second at 800–1300°C (tentative as Cs₂CrO₄) and the last one at 1300–1500°C (probably atomic cesium vapour after the break up of cesium silicate formed within the steel oxide layer).

In separate-effects experiments it has been observed that boric acid used in the coolant of PWRs can react with the otherwise thermodynamically favoured fission product species CsI and CsOH to produce CsBO₂ and HI or polyborates. In the only integral-effects in-pile test that included H₃BO₃ in the coolant (LOFT FP-2), there was no evidence of the formation of HI; rather AgI was proposed as the predominant chemical form of iodine. Contrary to earlier thermodynamic calculations, experimental studies have demonstrated that boric acid will react with cesium iodide to form cesium metaborate and hydrogen iodide [48]. New calculations confirms experimental results on this system [48].

In the Falcon facility it was demonstrated that the formation of volatile iodide species from reaction between cesium iodide and boric acid drastically affected the transport behaviour of the released iodine [49, 50].

Also for cesium the transport from fuel to the containment was increased (in the order of a factor of two) when experiments were conducted with boric acid [49,50].

Several integral tests and experiments on the fission product behaviour in the primary system and in the containment have been performed during the past several years:

Marviken-V, LACE, LOFT-FP2, FALCON, ACE and PHEBUS. Observations of specific relevance to cesium were the strong hygroscopicity, especially of CsOH, which can cause relocations.

Hygroscopicity influences aerosol transport in pipes because of increased stickiness of aerosols [51]. It will also increase the deposition due to rapid growth in size due to water attraction to hygroscopic aerosols [51]

Predictions of the Cs release from fuel for the LOFT-LP-FP-2 run ranged from 3 to about 45 % by use of CORSOR (45 %), FASTGRASS (3 %) and MAAP (40 %) compared to the 2.9 % measured release [52]. However, in addition to the transient release of Cs an additional release occurred during reflood and amounted to 16 % [53]. It was concluded that the use of mechanistic models can reduce uncertainties from an order of magnitude to a factor of two to four. They are also useful in demonstrating key sensitivities [52]. CsI was not present in significant concentrations. Instead, AgI and CsOH appear to have been the primary fission product species for I and Cs [53]. The deposition in the upper plenum was calculated to 8-36 % compared to the 17 % measured [52].

8 Behaviour in the containment

The influence from chemistry inside the containment on the behaviour of fission products has been discussed by Borioli [23] in a note to FPC. The physical and chemical characteristics of aerosols in the containment have recently been summarized [3].

8.1 Effect of hydrogen combustion

Several studies have addressed the effects of hydrogen combustion on fission products and aerosols [54]. They are mostly related to iodine behaviour but may also have some effect on cesium. If iodine occurs as CsI, hydrogen combustion can oxidize CsI under dry conditions to form molecular iodine. In wet conditions, a conversion of CsI to CsOH and HI will take place in addition to the gas phase thermal decomposition.

Surakka and Raunemaa [55] have performed experiments on the behaviour of CsI aerosols in the presence of PVC-cable smoke. They found higher sedimentation rates in the mixture compared to CsI only and there were indications of some decomposition of CsI.

8.2 Aerosol behaviour

At the workshop on water-cooled reactor aerosol code evaluation and uncertainty assessment in 1987 comparisons were made between various codes and with experiments per-

formed [56]. The behaviour of CsOH aerosols as well as other relevant aerosols were treated.

8.3 Containment source term

In the more recent severe accident source term analysis [57, 58], the release into the containment $ST_{CON}(i)$ is represented by

$$ST_{CON}(i) = ST_{INV}(i) + ST_{VB}(i) + ST_{EXV}(i) + ST_{REV}(i)$$

where (i) represents the radionuclide group, $ST_{CON}(i)$ represents the total source term for species (i), $ST_{INV}(i)$ represents release from the reactor coolant system into the containment prior to vessel failure, $ST_{VB}(i)$ represents the releases during vessel breach, $ST_{EXV}(i)$ represents the ex-vessel releases into the containment, primarily during core-concrete interactions, and $ST_{REV}(i)$ represents the releases due to late revolatalization from the reactor coolant system.

For cesium typically 10–100% (average about 60%) is released into the RCS before vessel breach according to STCP calculations for some specified scenarios (Figure 11). This means that a significant fraction could remain in the melt and escape retention within the RCS and thus remain accessible for release into the containment atmosphere after corium discharge from the RPV [58]. Of that fraction released before breach typically 3–40% (mean values for some accident types in BWR and PWR) is transmitted to the containment. For illustration the total release to the containment under various accident conditions for PWR and BWR are given in Figures 12 and 13. The supplementary cumulative distribution function of the release to the environment for cesium is given in Figure 14.

9 Releases to the atmosphere

9.1 Successfully terminated LOCA, releases to the atmosphere

Within the CEC two benchmark studies were completed in 1988 to predict the consequences of a successfully terminated large LOCA at a PWR [36]. In a recent report the calculation methods were further developed. For cesium-137, step-by-step results from calculations starting with the core inventory and ending with the release to the environment were presented (reproduced here as Figure 15). The spread of initial benchmark calculations between the three participating countries is shown in the figure as well as the results of Case 1 – 3, which reflect different design features.

In the benchmark calculations 10 – 100% of fuel cladding was assumed to fail. Even if there is some uncertainty the recommended values were in the range 3 – 33%.

The release of activity from the gap and also as a result of fuel fragmentation during a LOCA was assumed to be 2% for Cs137. For I131 it was assumed to be 0.65%.

Of this gap activity, 10% was assumed to be released during the dry phase and 90% after the rupture was recovered. 90% of the iodine and cesium released from fuel were assumed to be transported directly to the containment sump without being airborne in the containment atmosphere.

The initial spread of the Cs137 release to the atmosphere (about 0.1 GBq to 10 TBq) in the exercise among the three contributors was five orders of ten, a range which, however, also included differences in designs. After the detailed analysis of assumptions and differences followed by an evaluation of best estimates the final release results were within one order of ten (0.1 to 1 GBq) for two of the designs. For the third design (case 3) the release was just a small fraction of that, because of strong retention in the secondary containment.

9.2 Severe accidents releases of cesium to the atmosphere

The supplementary cumulative distribution function for the release of cesium to the atmosphere in severe accidents is given in figure 14 (from NUREG-1150).

This atmospheric release of cesium is strongly affected by the release of cesium during vessel breach (ST_{VB}) when containment failure occurs soon thereafter.

When containment failure is late, the atmospheric release of cesium is strongly affected by the late revolatilization of cesium trapped in the reactor coolant system (ST_{REV}) since it occurs over a long period of time.

In containment bypass situations the strong hygroscopicity of especially CsOH aerosols influences their transport through pipes because of increased stickiness.

Thus gaps in our present knowledge of physical and chemical features of cesium most detrimental to our ability to accurately predict atmospheric releases relate to:

- cesium aerosolization at vessel breach (ST_{VB}) in early containment failure situations
- late revolatilization of cesium from the reactor coolant system in late containment failure situations and
- transport of sticky cesium aerosols through pipes in containment bypass situations.

This perception of the situation is recognized by those organising source term research.

10 Conclusions and recommendations

Review of information from various sources relevant to the objective of the present report which is to identify areas of remaining uncertainty regarding the cesium source term for severe reactor accidents has shown:

- The inventories in the fuel of the most important cesium isotopes Cs 137 and Cs 134 can be calculated by established computer codes with sufficient degree of accuracy.
- Cesium release from overheated fuel is well known and occurs at a rate and to an extent similar to that of the noble gases and iodine. Minor amounts can be trapped in the ceramic part after a core melt.
- Cesium will react with water or water vapour after the release from the fuel and form cesium hydroxide and other compounds e.g. mixed oxides. Cesium hydroxide is very reactive and will be transformed to more stable compounds. At high temperatures cesium compounds partly dissociate.
- Cesium and its compounds will deposit on and react with stainless steel surfaces in the reactor coolant system. The deposited cesium can partly be removed by water leaching or vaporization. Studies are in progress to study the processes involved and the revaporization at various temperatures. It is of interest to obtain a further understanding and quantification of these phenomena.
- Different observations are at hand regarding the importance of the interaction between cesium and boric acid in a PWR. Further evaluation is recommended.
- The transport of cesium compounds as aerosols through pipes may be affected by hygroscopicity which can cause clogging and translocation.
- Hydrogen combustion can decompose CsI under certain circumstances, but this is more of relevance to iodine than to cesium.
- The behaviour of relevant cesium compounds as aerosols in the containment has been studied extensively from laboratory to full-scale experiments and several computer codes are available. Special effects, e.g. hygroscopicity, which cause particle size growth, are taken into account in the further development of codes.

11 Acknowledgement

The authors would like to thank Prof. J-O. Liljenzin for providing information and initial discussions and Prof. R. Segal and Dr.D. Powers for valuable comments and suggestions.

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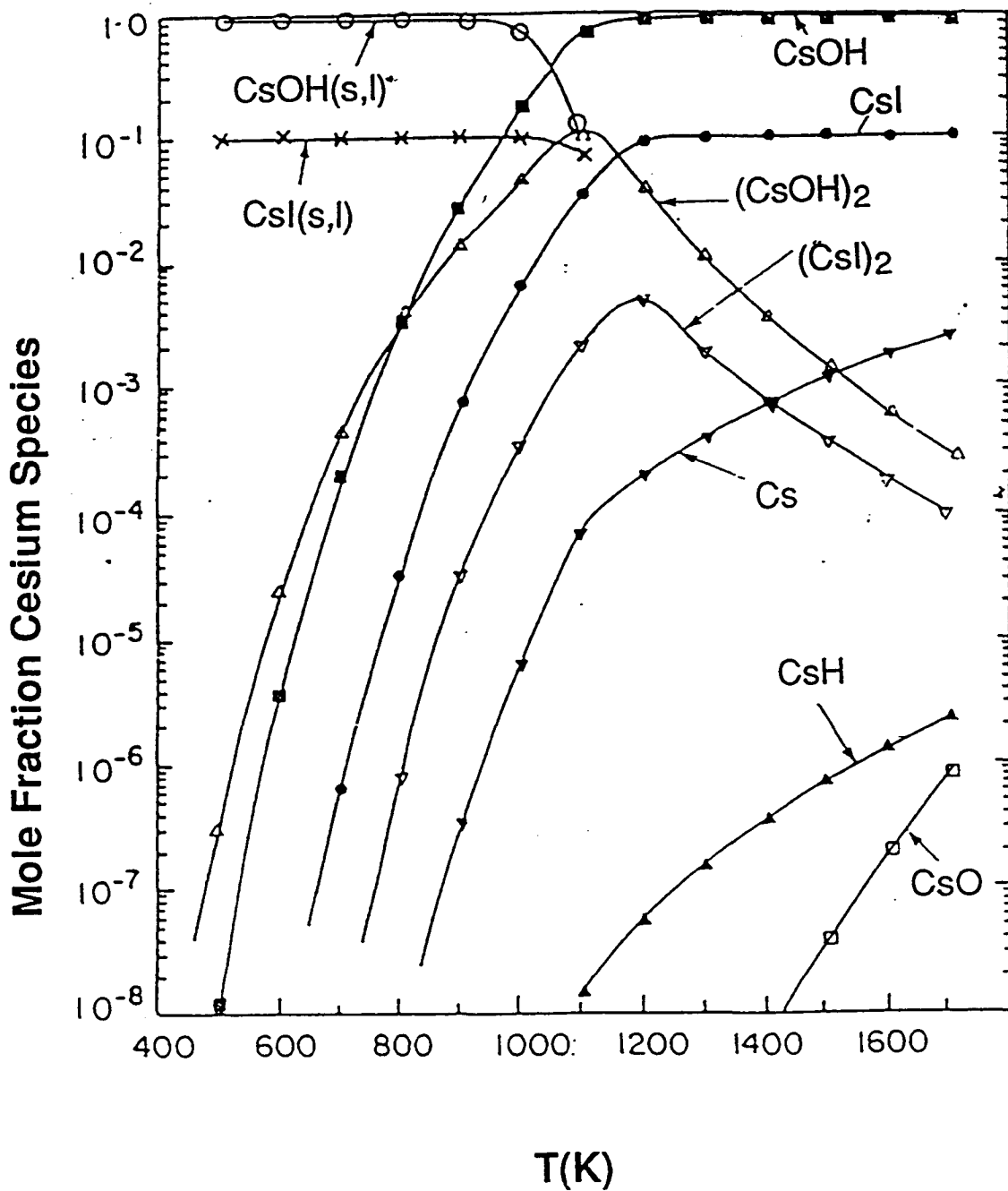


Figure 1: Cesium species distribution diagram for a system containing steam (4 moles), iodine (10^{-2} moles) and cesium ($Cs/I = 10$) at a pressure of 1.2 MPa. (D Torgerson, Report to CSNI)

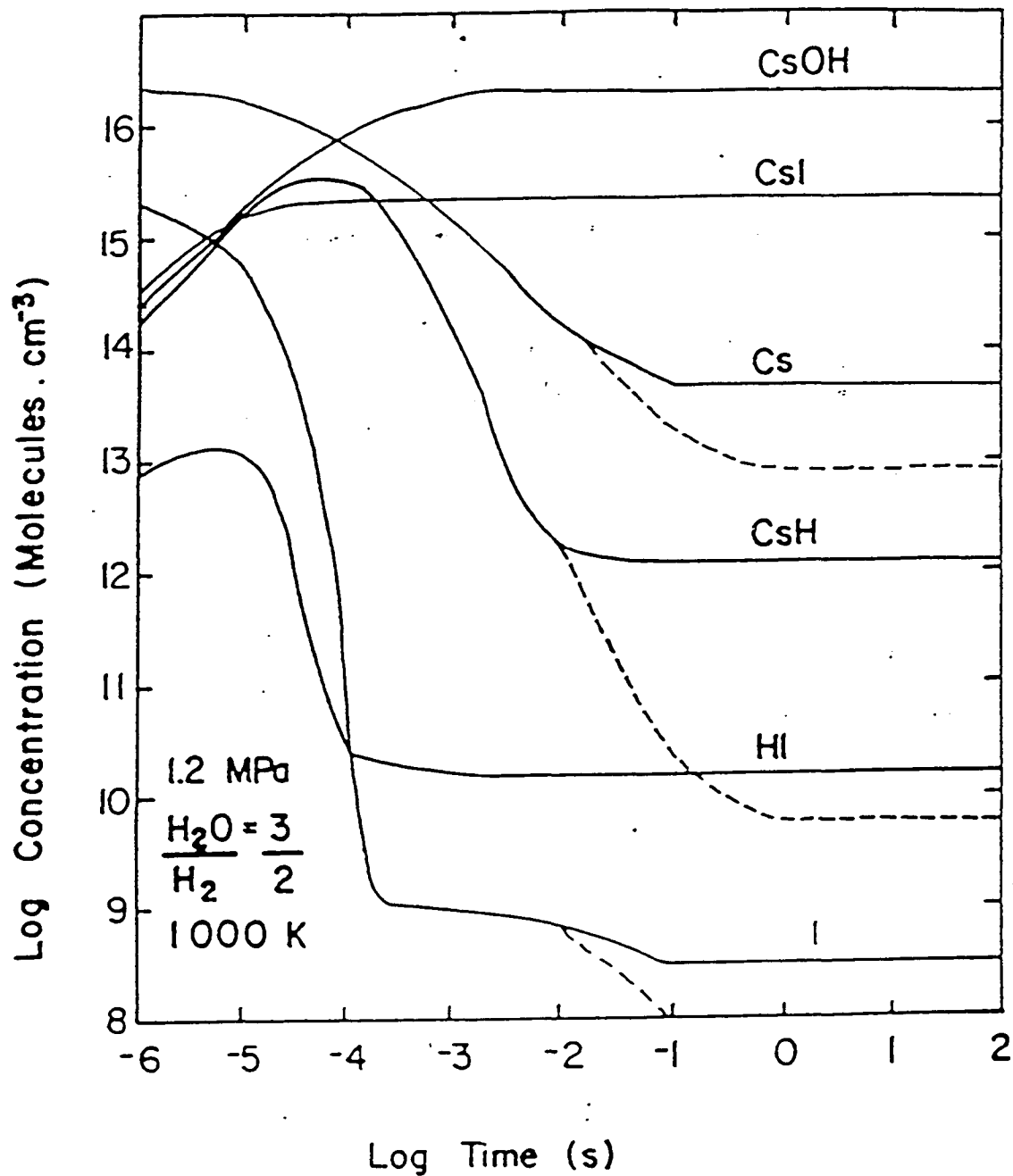


Figure 2: Chemical kinetics of cesium and iodine in a steam/hydrogen atmosphere at 1000K. The dashed lines include thermal reaction contributions only, while the solid lines include both thermal and radiation effects for a gamma radiation dose of $3 \times 10^{-1} W \cdot g^{-1}$. The initial Cs and I atom concentrations are 5×10^{16} and $5 \times 10^{15} \text{ atoms} \cdot \text{cm}^{-3}$, respectively. (D Torgerson et al)

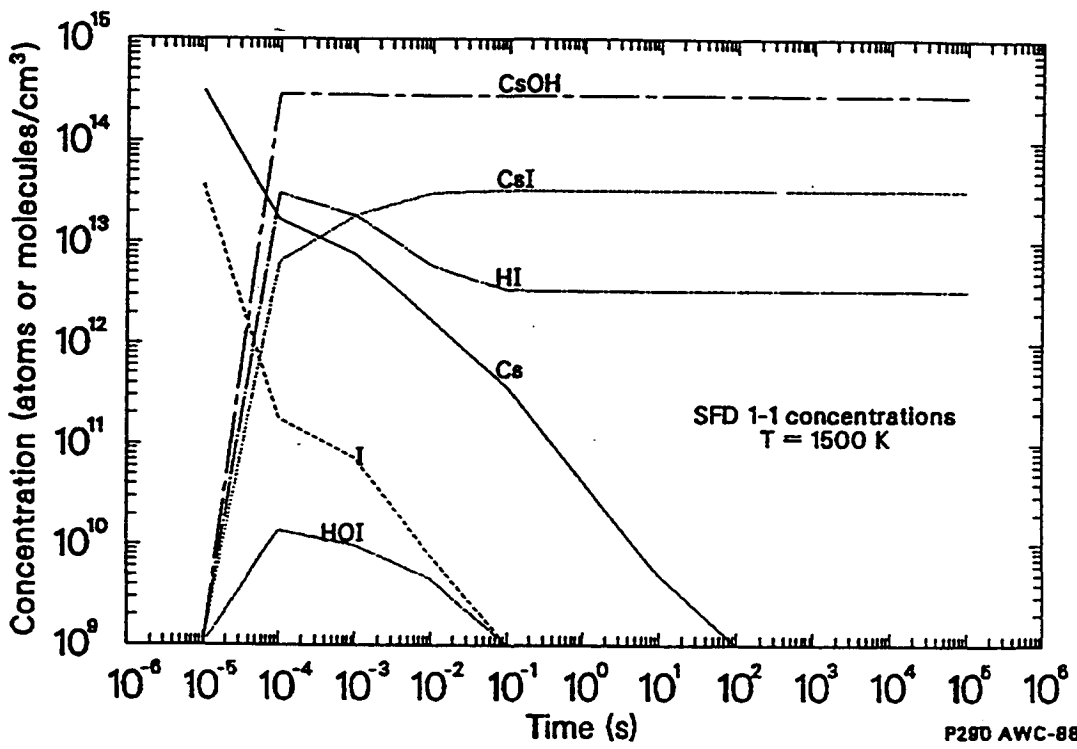


Figure 3: Results of chemical kinetics calculations for the SFD 1-1 concentration conditions. (Cronenberg and Osetek 1986)

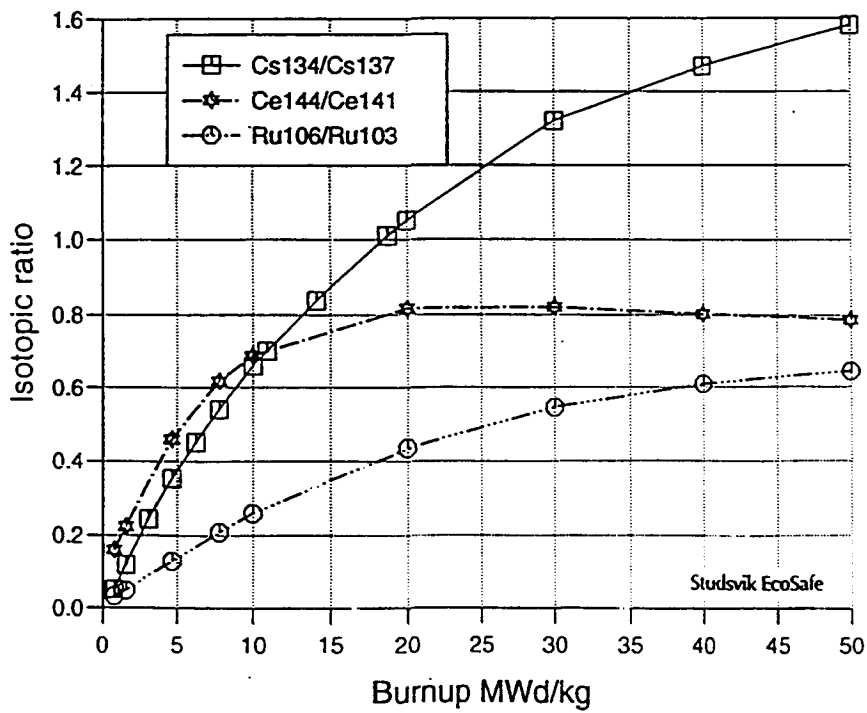


Figure 4: Isotopic ratios versus burnup

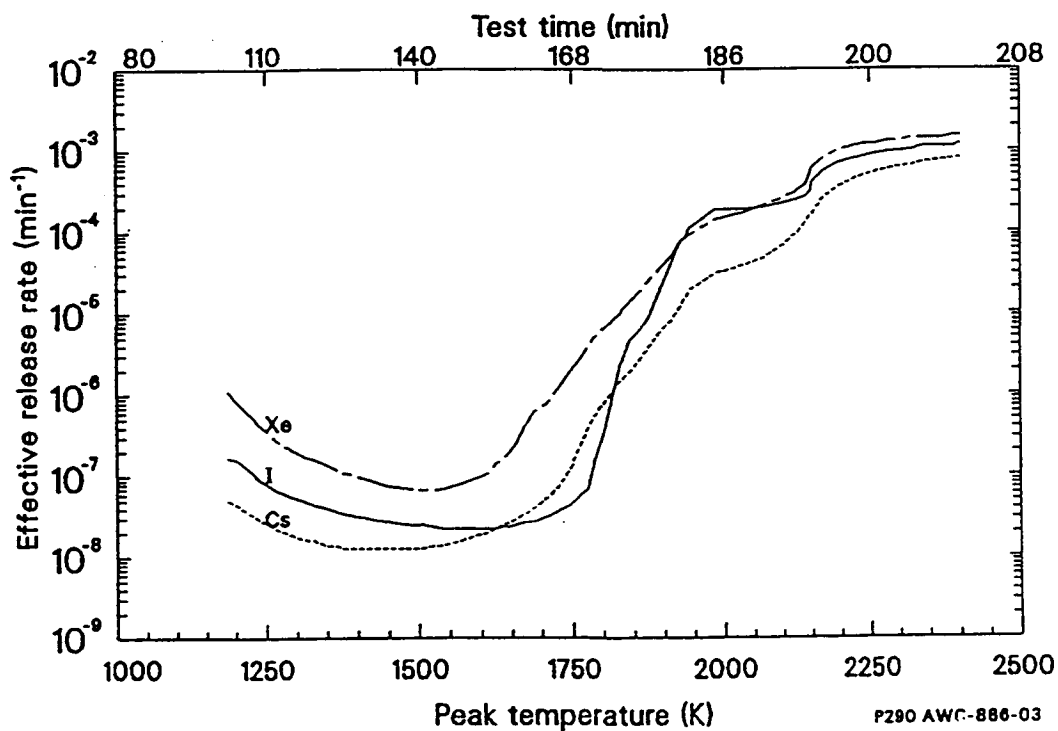


Figure 5: Comparison of the measured I, Cs, and Xe release rates for the PBF-SFD-Scoping test. (Cronenberg and Osetek 1986)

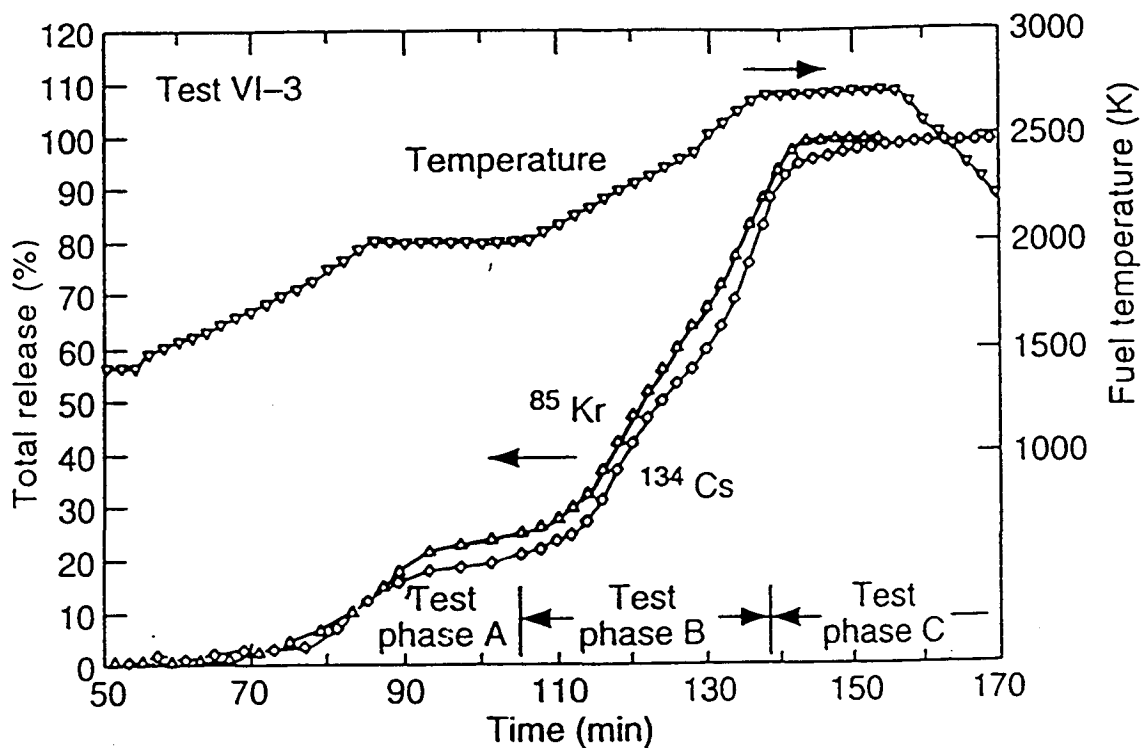


Figure 6: Fission product release from fuel specimen with cladding in steam. (Osborne and Lorenz 1992)

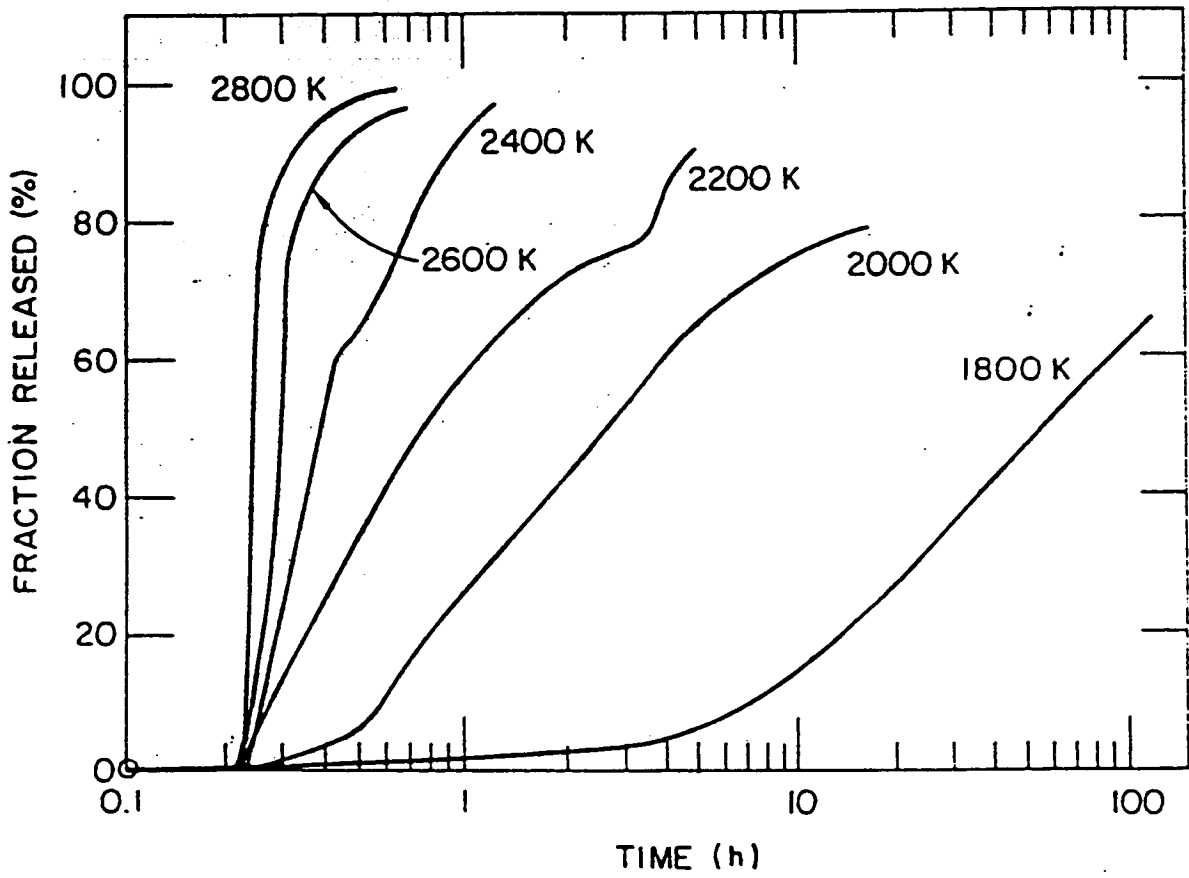


Figure 7: FASTGRASS-calculated Cs release during fuel heatups to 1800–2800 K and during subsequent hold periods. (Rest and Zawadski)

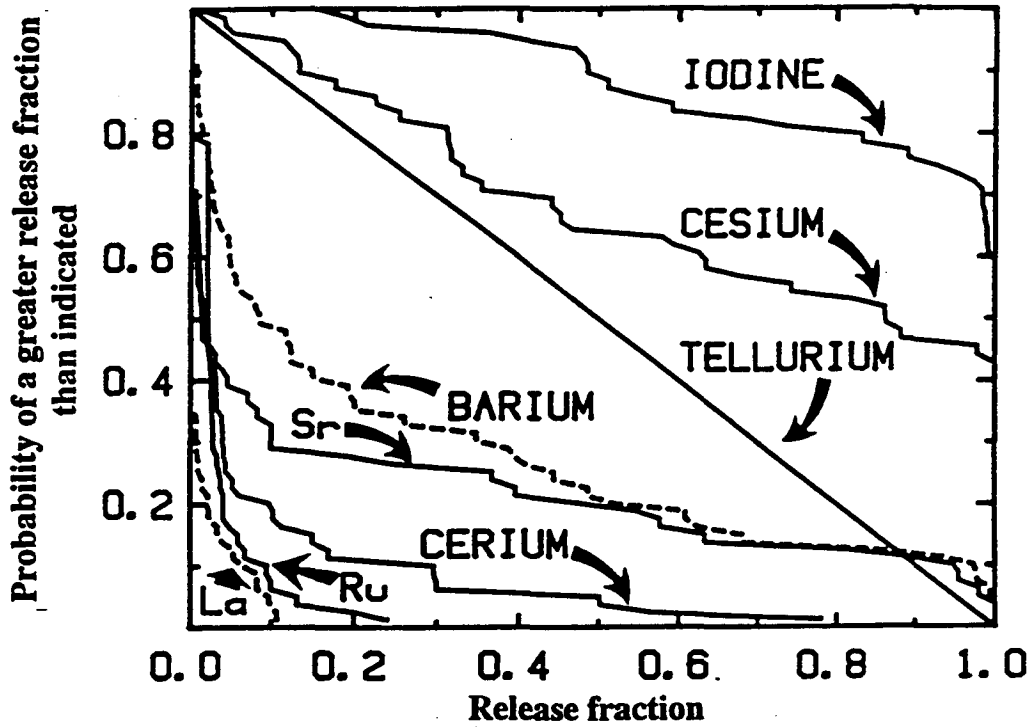


Figure 8: Uncertainty distributions for the releases of radionuclides during core degradation. (Powers 1989)

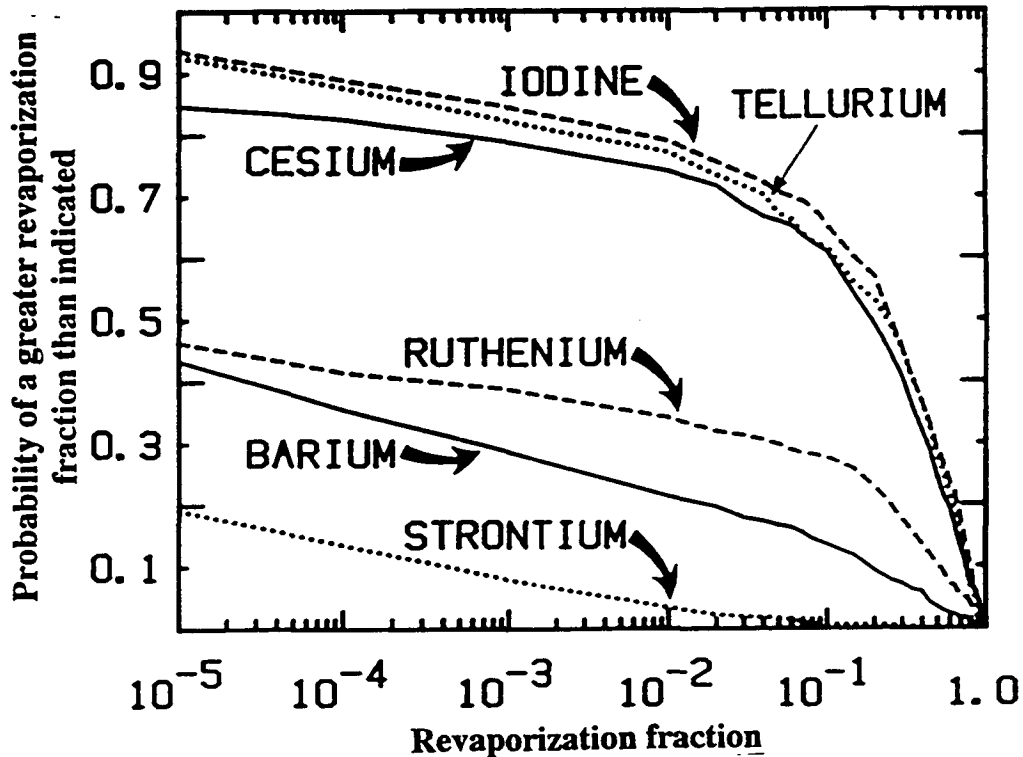


Figure 9: Uncertainty distributions for the revaporization fractions of deposited radionuclides. Revaporization fractions for La and Ce were always less than 10^{-7} . (Powers 1989)

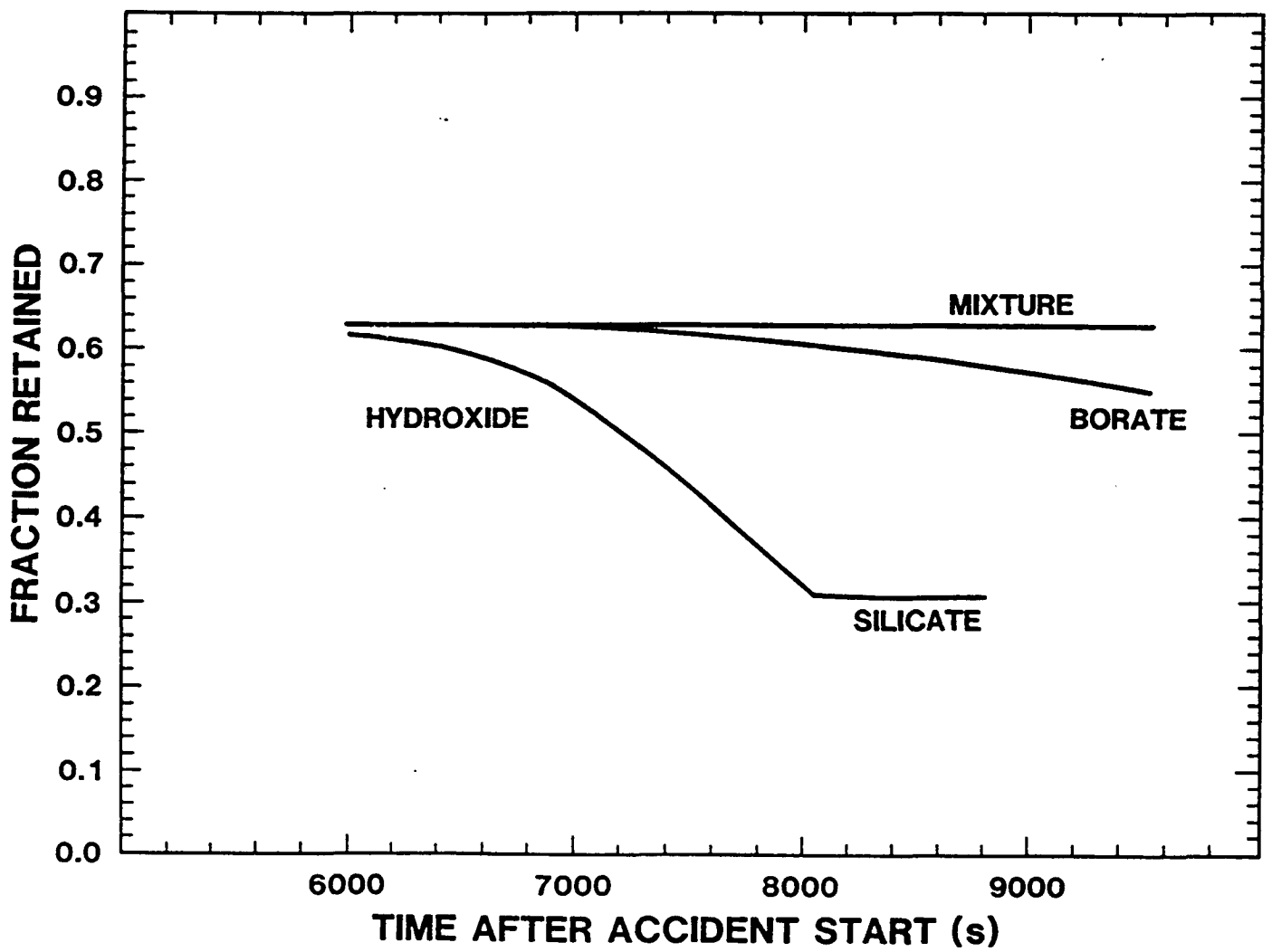
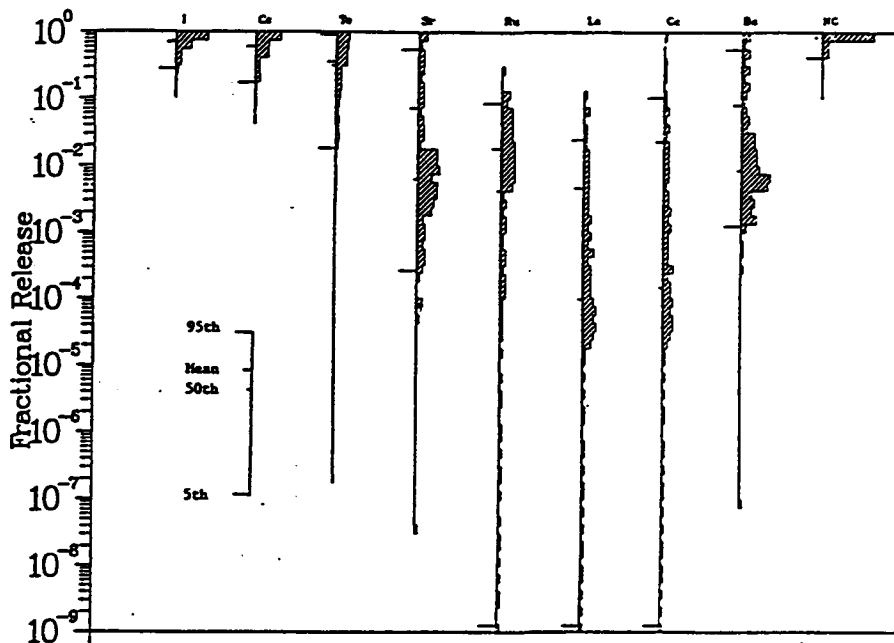
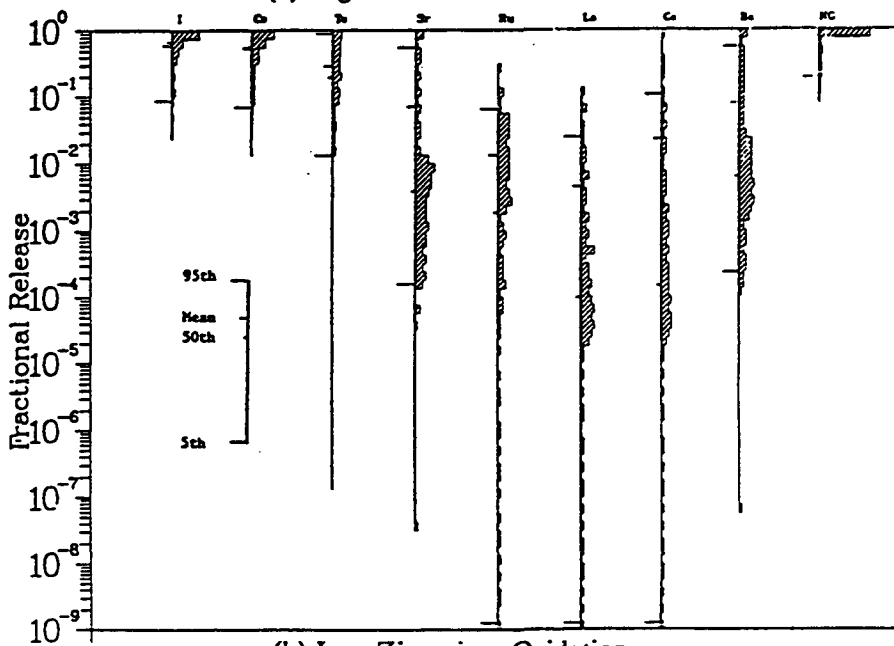


Figure 10: Revaporization of cesium in various chemical forms during a hypothetical reactor accident. (POWERS 1986)



(a) High Zirconium Oxidation



(b) Low Zirconium Oxidation

Figure 11: Uncertainty Distributions for Release of Radionuclides From Core Into the RCS Before Vessel Breach (FCOR) for PWRs. (NUREG/CR-5747)

Conditions		ST _{CON}							
		I	Cs	Te	Sr	Ba	Ru	La	Ce
Setpoint Pressure, High Zr Oxidation, Limestone Concrete, Dry Cavity	Median	0.45	0.50	0.25	0.05	0.05	0.006	0.006	0.004
	Mean	0.47	0.49	0.28	0.11	0.11	0.03	0.016	0.018
	95th Percentile	0.90	0.92	0.62	0.36	0.37	0.16	0.059	0.078
Low RCS Pressure, High Zr Oxidation, Limestone Concrete, Dry Cavity, Two Openings After VB	Median	0.77	0.81	0.49	0.08	0.06	0.002	0.002	0.001
	Mean	0.75	0.74	0.46	0.16	0.14	0.013	0.017	0.022
	95th Percentile	0.997	0.997	0.87	0.57	0.54	0.068	0.098	0.15
Setpoint Pressure, High Zr Oxidation, Basaltic Concrete, Dry Cavity	Median	0.45	0.50	0.25	0.04	0.03	0.009	0.007	0.006
	Mean	0.47	0.49	0.28	0.10	0.09	0.03	0.016	0.019
	95th Percentile	0.90	0.91	0.63	0.38	0.35	0.16	0.06	0.077
Low RCS Pressure, High Zr Oxidation, Basaltic Concrete, Dry Cavity, Two Openings After VB	Median	0.77	0.81	0.49	0.04	0.03	0.001	0.002	0.003
	Mean	0.75	0.74	0.46	0.14	0.12	0.01	0.016	0.025
	95th Percentile	0.997	0.997	0.865	0.65	0.57	0.068	0.099	0.139

Figure 12: Some Statistical Parameters for Total Release Into a PWR Containment Using NUREG-1150 Methodology. (NUREG/CR-5747)

Conditions		ST _{CON}							
		I	Cs	Te	Sr	Ba	Ru	La	Ce
High Pressure Fast Station Blackout, High Zr Oxidation, Limestone Concrete, Dry Pedestal, High Drywell Temperature	Median	0.56	0.58	0.35	0.07	0.07	0.009	0.0085	0.011
	Mean	0.56	0.55	0.34	0.19	0.17	0.031	0.017	0.04
	95th Percentile	0.94	0.96	0.71	0.58	0.54	0.166	0.06	0.24
Low Pressure Fast Station Blackout, High Zr Oxidation, Limestone Concrete, Dry Pedestal, Low Drywell Temperature	Median	0.75	0.73	0.55	0.09	0.09	0.001	0.003	0.007
	Mean	0.70	0.68	0.50	0.29	0.25	0.009	0.016	0.022
	95th Percentile	0.997	0.997	0.872	0.94	0.87	0.07	0.088	0.11
High Pressure ATWS Sequences, High Zr Oxidation, Limestone Concrete, Water Injection After VB	Median	0.68	0.66	0.35	0.06	0.07	0.009	0.008	0.011
	Mean	0.62	0.61	0.34	0.18	0.16	0.03	0.016	0.05
	95th Percentile	0.975	0.962	0.70	0.58	0.53	0.16	0.059	0.22

Figure 13: Some Statistical Parameters for Total Release Into a BWR Containment Using NUREG-1150 Methodology (NUREG/CR-5747)

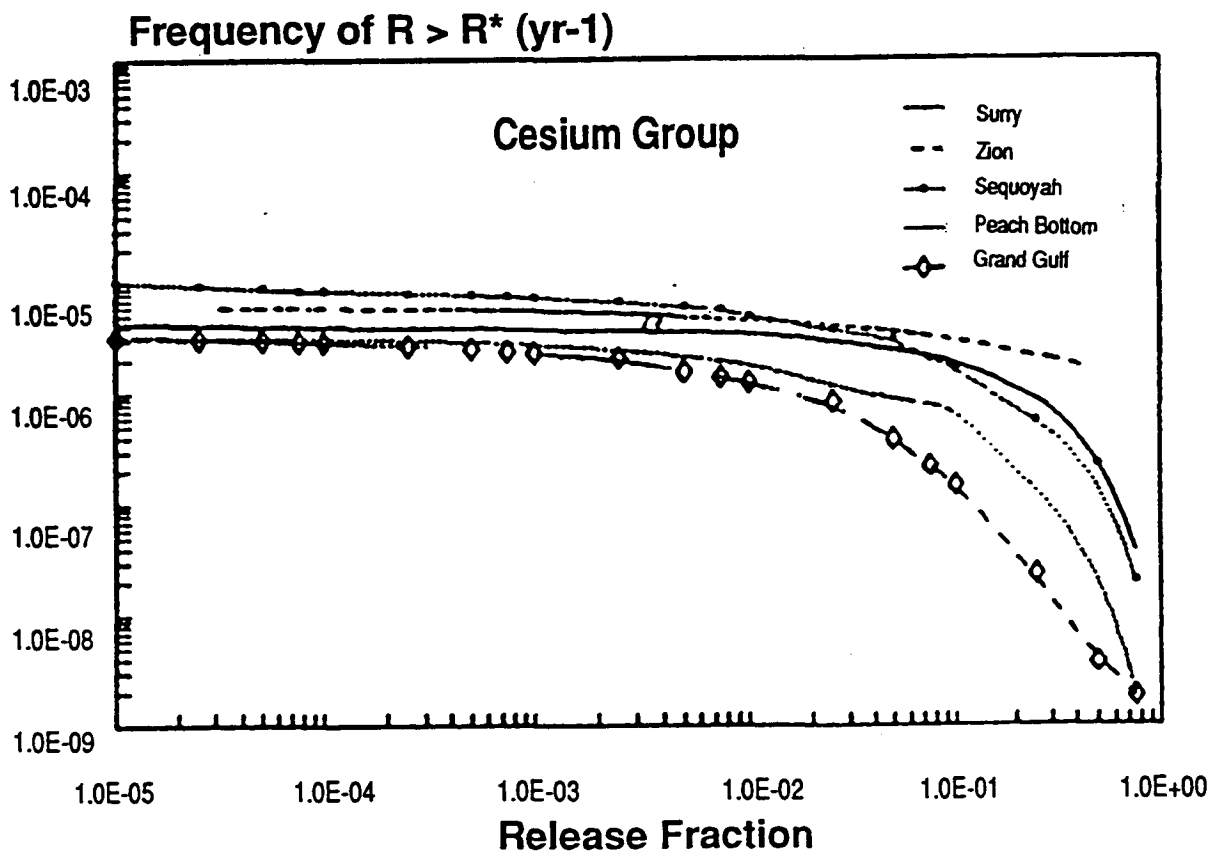


Figure 14: *The complementary cumulative distribution function of the release to the environment. (NUREG-1150)*

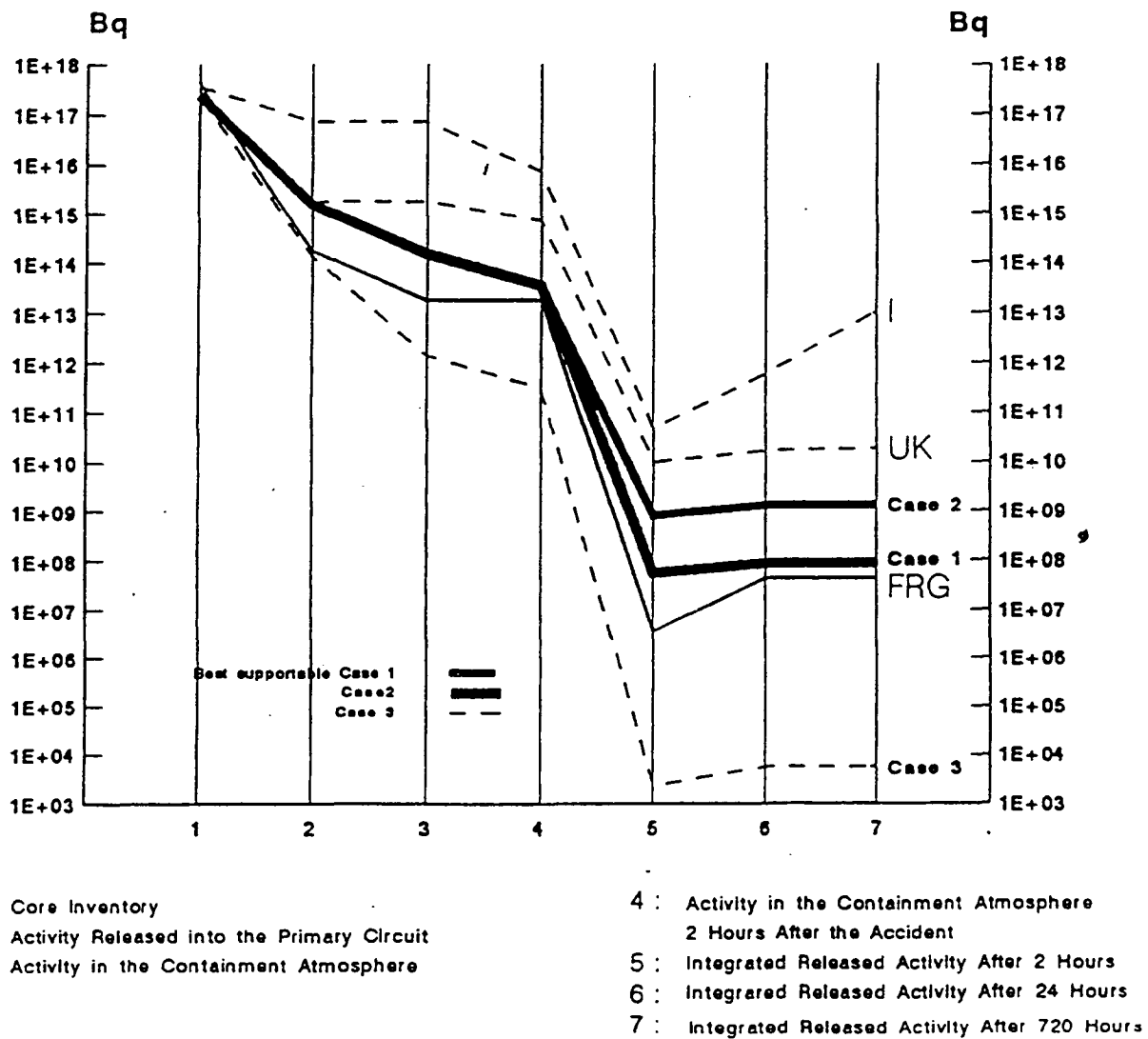


Figure 15: *Best Supportable Release Results for Cs-137. (EUR 14179/EN 1992)*

Appendix 1 Cesium behaviour in the Chernobyl accident fallout

The total release of Cs to the environment was reported [A1] to be 10–13 % of the core inventory. This estimate was based on the integration of the total Cs deposition within the Soviet territory. It was suggested [H. J. Dunster, UK] already at the IAEA conference in August 1986 that there should be an adjustment of the reported release figures by a factor of two due to the world wide dispersion. Various indications of substantial higher release were soon reported [A2]. The world wide total contamination was about twice that within the Soviet territory [A3].

A comprehensive study of the Chernobyl accident [A4] presents an in-depths discussion of the release fractions based on analyses of core material. The total release of cesium to the environment was estimated to be 40–60 % of the core inventory from these analyses. Unexpectedly 35 % of the cesium was retained in the solidified melt despite the oxidizing conditions, which is much higher than TMI figures (3 % retained in the corium, 19 % retained in the upper prenum debris) [A4]. Cesium was assumed to exist in low or mid-volatile chemical compounds at high temperatures. It was proposed that high-temperature chemical retention mechanisms for cesium ought to be investigated [A4]. However Akers (see chapter 3) reports 9–32 % Cs retention in TMI-2 core debris.

The initial observations [A5] of fallout from the Chernobyl reactor accident revealed extensive cesium and iodine concentrations. Cesium was in particulate form with a constant Cs134/Cs137 ratio of about 0.57. Iodine was found to be mainly in gaseous form.

Hot particles were associated with the fallout. Later analysis [e.g. A6] showed that the hot particles were of two types. One type consisted of almost pure ruthenium. The other type was fuel fragments including fission products. It was shown [A7] from observations of different Ru106/Ru103 and Ce144/Ce141 ratios that the particles originated from various parts of the core with different burnup. The fuel fragments were depleted in cesium and other volatile elements [A8].

In extensive studies of fuel fragments [A9–A12] it was shown that the depletion of cesium was 0–99% and 60% as an average. In larger particles, the depletion was lower than in smaller particles, which is compatible with a diffusion process at high temperature from fuel grains.

For the fraction of cesium remaining in the fuel fragments there was a range of the Cs134/Cs137 ratio from 0.2 to more than 1.0, thus showing various burnups and no significant cesium mixing in the core before release. This is an indication of the release of fuel fragments prior to any extensive fuel melting. These particles may belong to the first explosive excursion from the core and may not be representative of the later part of the release period. Analyses, concerning the variation of isotope ratios, in the core debris would give information on mixing completeness.

Cesium in the fallout behaved differently from iodine and low volatile elements and there was no indication of chemical interaction or compounds between cesium and iodine or tellurium [A8, A13, A14].

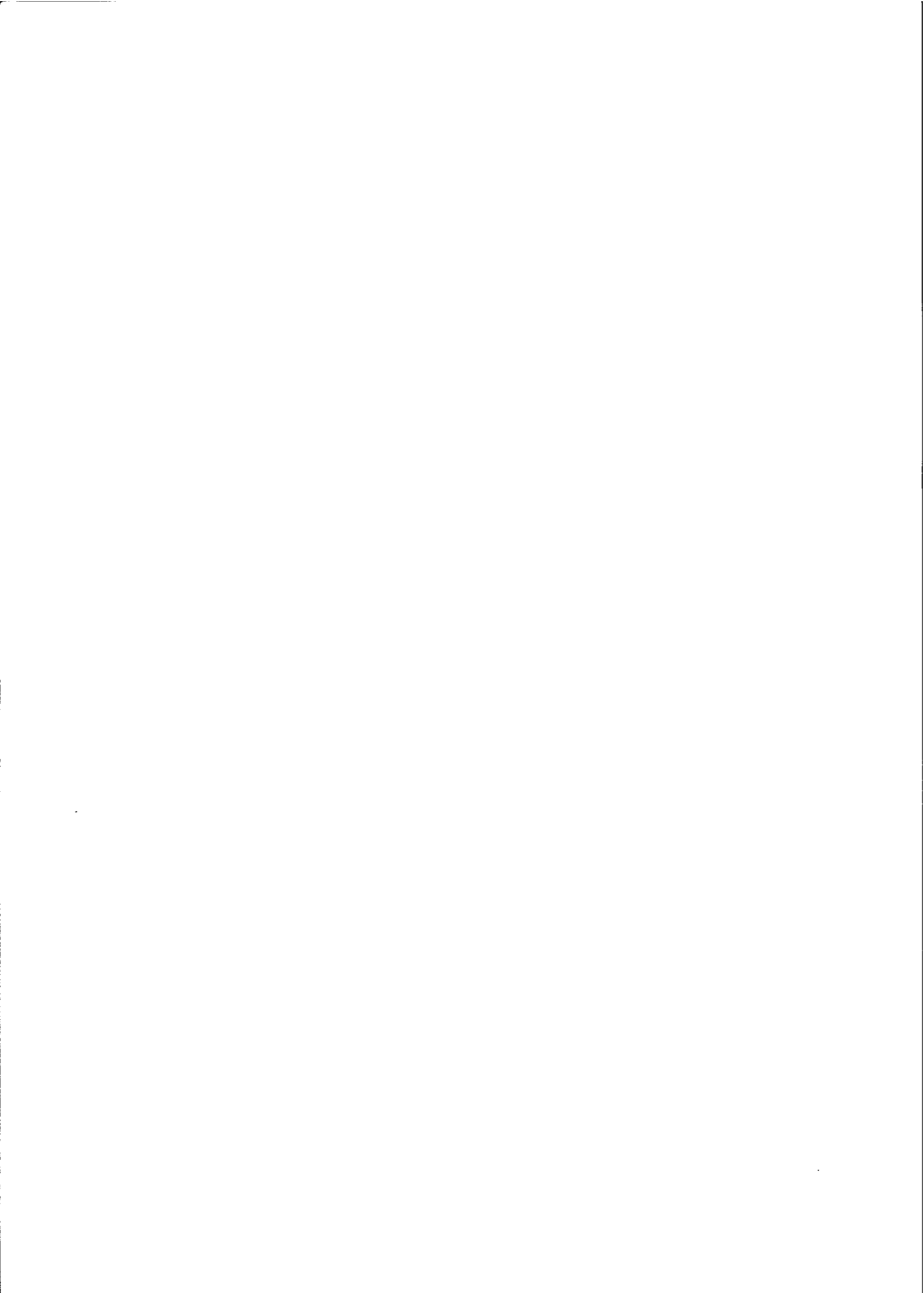
The physical and chemical forms of cesium released to the environment influence on the uptake into organisms and transfer e.g. to milk [A15-17].

The results of Salbu, who has studied the properties of Chernobyl fallout cesium, indicate that typically only 25% of cesium appeared as cation in rain water [A18]. The remaining part was in colloidal form.

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