ANALYSIS OF ACCIDENTAL SEQUENCE TESTS AND INTERPRETATION OF FISSION PRODUCT RELEASE: INTERDEPENCE OF Cs, Mo, AND Ba RELEASE.

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ABSTRACT

The behaviour of fission products during fuel life in a nuclear reactor is of particular importance as it plays a significant role in the release kinetics during hypothetical reactor severe accident transients.

A description of (Cs, Mo, Ba) chemical states after some UO_2 fuel irradiation stages is drawn. Molybdenum is supposed to be localised in a MoO_2 precipitate (volatile over 1273K) and chemical reactions with condensed (Cs, Ba) oxides and/or uranates are possible.

According to this description, a coherent interpretation is presented for some accidental sequence tests providing new ideas for release mechanisms of (Cs, Mo, Ba) and an attempt is done to calculate their release with MFPR code, which is being developed by IBRAE in collaboration with IRSN.

Keywords: Cs, Mo, Ba, fission product release, Vercors tests, MFPR code.

1. INTRODUCTION

The physics of Fission Products (F.P.) is a field which is still in progress and much effort has been expended in the past forty years, namely in the French "Institut de Radioprotection Nucléaire" (IRSN) to understand F.P. release under severe accidental conditions, in the field of nuclear safety, by the way of small or large scale experiments, development of physical models and computation codes. Special attention will be paid through this work to separate experimental results from simulation calculations, which can always be viewed with caution. The temptation is high, when computation results are close to experimental ones, to accept the models implemented, although they might not be so physically grounded.

This paper is dedicated to the analysis and interpretation of some analytical experiments called Vercors tests, performed at Commissariat à l'Energie Atomique (CEA), France. These tests are performed on a fuel pin element consisted by three pellets of irradiated UO_2 [1]. A temperature transient[1], without any irradiation, is applied to these specimens under various atmospheres (pure steam, pure hydrogen, or steam/hydrogen mixture). For all the tests, a temperature step is done (at around 1573K), under an oxidising gas flow, in order to get a full oxidation of the clad (Zr). The release kinetics are measured using online γ-spectrometry, which provides very accurate curves and high quality results. Therefore, Vercors test are very interesting to interpret in order to understand the release mechanism of fission products for a further interpretation of integral tests (Phebus tests [2] for instance) and for applications to simulations of a reactor accident scenario (with the ASTEC code [3] for example). The

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[1] generally from 293K to the fuel degradation and relocation which most of the time occurs over 2573K.

attention is focused here on (Cs, Mo, Ba) whose behaviour is not well known during temperature transients.

An interpretation of (Cs, Mo, Ba) release, during Vercors 4, 5 tests, is proposed here. It is based on the concept that F.P. release is ruled by the evolution of their chemical forms and localisations inside a fuel under a temperature transient and defined gas conditions.
This interpretation lays on observations of F.P. during the fuel life in reactor and a modelling (linked with an analysis of experimental results) of F.P. evolution during the test sequence.
Then, the model of F.P. chemical behaviour was implemented into MFPR (Mechanism of Fission Product Release) release computation code used for release calculations of Vercors 4 and 5 tests.
In this sense, the scope of the present work differs from classical approaches concerning Cs for instance with the classical models of "Booth" [4] where intragranular diffusion is the only limiting phenomena for release and where no chemical reaction is taken into account.

2. COMPUTATIONAL AND PHYSICAL BASES OF MFPR

F.P. release calculations were performed with MFPR code [5]. This mechanistic code is written in Fortran 77 language, and includes a driver block (11 subroutines) interacting with nearly 130 subroutines useful for chemical equilibriums, F.P. release, fuel oxidation, physical properties of [gas, bubbles, point defects (vacancies, interstials) or linear defects (dislocations)], and numerical and mathematical utilities.

The code is started with an input file written by the user. This file is divided into three parts:

- physical models for gas and fuel behaviour (bi/multi modal distribution of bubbles), bubbles nucleation, bubbles diffusivity and coalescence, use of vacancy calculations, capture of bubbles or not by dislocations, bubble thermal or irradiation resolution, fuel oxidation, grain growth...
- physical data of the fuel, grain size, dislocation density...
- [temperature/irradiation/(gas flow composition and rate)] history during irradiation and temperature transient, and the global time step (further used internally to establish a local time step consistent with the numerical scheme for equations resolution) chosen by the user at each temperature for calculation.

At each global time step, two major transport equations must be resolved in a grain, represented in the code as a tetrakaidecahedron[2] (TDK) [5]. The first one concerns the noble gas (Xe, Kr) behaviour:

$$\frac{\partial X_i}{\partial t} = F_i(X,t), \qquad (1)$$

for vector variable $X = \{C_g, C_b, \rho_d, C_d, C_v, C_i, N_b, V_b\}$ and $X_i \in [C_g, C_b, \rho_d, C_d, C_v, C_i, N_b, V_b]$

The basic space-time dependent variables involved here are the concentrations of gas atoms C_g and intragranular bubbles C_b, the average number of gas atoms within a bubble N_b, and the average bubble volume V_b. A sub-model for the gas bubble interactions with dislocations, vacancies and interstitials is described by three additional variables: vacancy concentration C_v

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[2] with, for mean values, when the grain is surrounded by its neighbours, 7 faces, 12 edges, and 6 corners.

interstitial concentration C_{int}, dislocation density (length per unit volume) ρ_d, and concentration of intragranular bubbles captured by dislocations C_d.

Additionally, at each time step, the code provides a gas distribution representation, not only for the grain bulk, but also at the grain faces, edges and corners. The intergranular bubble behaviour is of percolation type and is simulated as follows: grain faces are first filled and when the face bubbles reach a surface coverage over 50% of the face area, the edges and corners are filled. When edge and corner bubbles percolate, the content of atoms is released until the criterion of complete coverage of the edge length is unsatisfied again.

The second major equation to solve is a coupled transport /mass action one concerning all the others F.P., which are chemically active. It consists in a system of n_e=17 atoms (Ba, Ce, Cs, Eu, H, I, La, Mo, Nb, Nd, O, Ru, Sb, Sr, Te, U, Zr) and n_s=134 species, which are built on these 17 atoms, and whose form can be the condensed or the gaseous one. This chemical solver (consisting in a minimisation of the global Gibbs's energy of the whole system) involves 117 chemical reactions.

The reaction of formation of each species S_i from the 'basic' components (n_e) can be written as:

$$S_i = \sum_{j=1}^{n_e} b_{ij} S_j \qquad i = 1, \dots n_s \quad (2)$$

where b_{ij} are the stoichiometric coefficients. The stoichiometric matrix, $\boldsymbol{B}$, with the elements b_{ij}, is a rectangular one of n_e columns and n_s rows.

For equilibrium in the reactions of formation (2), the law of mass action yields:

$$A_i = K_i \prod_{j=1}^{n_e} (A_j)^{b_{ij}} \quad (3)$$

where A_i is the activity of the i-th specie.

A transport equation type is coupled to the solution of (2) + (3), that gives the concentration Y_i, when the specie S_i is supposed to diffuse, i.e. when it is located in solid solution in the fuel. This equation takes the form:

$$\frac{\partial Y_i}{\partial t} = B_i + \frac{1}{r^2} \frac{\partial}{\partial r} (D_i r^2 \frac{\partial Y_i}{\partial r}) \quad (4)$$

where B_i expresses the created concentration of the S_i specie by fission (B_i=0 in annealing regime) and where D_i is its diffusion coefficient in solid solution. When the specie S_i has reached a grain boundary (a face of the TDK), and if its solubility limit is also reached, it precipitates following thermodynamics laws. Five phases are considered in MFPR calculations: the solid solution and the separate phase, which includes three sub-phases, that are ternary compounds (Cs_2MoO_4 for instance), metallic phase (metallic Mo for instance), CsI phase. These three sub-phases are in equilibrium with each other, and each one is in equilibrium with the final modelised phase, i.e. the gas phase, whose volume is set equal to the volume of face bubbles at each time step. Therefore, equation (4) is only applied to solid solution and resolution of equations (3)+(4) gives at each time step de total distribution of single elements in different S_i species and the release amount.

3. INTERPRETATION OF VERCORS 4 AND 5 TESTS

Vercors 4 and 5 (see table 1 for experimental conditions and total amounts released) were deeply investigated on the basis of experimental release kinetics (figure 1), with a focus on Cs, Mo, and Ba.

If many thermodynamical studies have been performed on the systems (Cs, Ba)-(Mo, U)-O, only few are devoted to describe the real localisation of (Cs, Mo, Ba) inside an irradiated UO_2 fuel. Kleykamp [7, 8] indicates that Cs, Mo, Ba, and their respective oxides have a very low solubility. Moreover, due to oxygen liberation during fission, these three fission products are oxidised. Cs is supposed to precipitate under uranates forms of type Cs_xUO_y, [7, 9-13] the simplest one being Cs_2UO_4. Ba has been observed under BaO precipitate [7, 8, 14, 15], and Mo under MoO_2 precipitate [7, 8, 14-19] coming from the oxidation of Mo existing inside a metallic precipitate which contains also Pd, Rh, Ru, and Tc [7, 8, 14-19].

It is therefore possible to submit the following interpretation (figure 2): at the initial time (t_0) of the test, (Cs, Mo, Ba) are over their solubility limit (reasonable regarding the fuel specimen burnups) and Cs is supposed to be localised into Cs_2UO_4 precipitate, Mo into MoO_2 precipitate and Ba into BaO precipitate (or Ba_xO_y).

It was demonstrated [15, 17, 19] that MoO_2 has special vaporization properties. If it is considered alone, under 1 atm, thermodynamic data predict a vaporization of solid MoO_2 only towards 2800K whereas this vaporization occurs in nuclear fuels over 1273K. As shown in figure 3 for Vercors 5 test, Mo seems to be released under two chemical forms: there is first specie which condenses quickly (it is supposed to come from a reaction between the atmosphere and MoO_2). The second specie whose amount is about 50% of the Mo release, seems to be fully collected into the impactor (a multistage filter for aerosol trapping). Therefore, during Vercors 4 and 5 tests, all the MoO_2 is supposed to vaporise over 1273K and about 50% of this amount enters in reaction with Cs, or Ba separate phases. Gaseous MoO_2 is supposed to fill the intergranular porosities and to be transported through the intergranular bubble network already established by noble gases. A part of MoO_2 can react with some precipitates located at the grain boundaries. A similar mechanism of vaporization/reaction was observed in Phénix reactor bundle pins [19], where no Mo was found in the centre of the pellet whereas molybdates of Ba or Cs were found at the periphery. In this case MoO_2 is supposed to vaporize from hot parts of the fuel then condense and react in colder parts.

During the clad oxidation phase, as hydrogen is produced between the clad and the fuel, there is a lowering of the oxygen potential ($\Delta GO_2=RT\ln(PO_2)$) leading to a decrease of the stability field of Cs_2UO_4 (a 2-d field where the first dimension is T and the other one is ΔGO_2). This can provide some gaseous Cs release. Cs_2MoO_4 seems also to be affected as Mo release is observed at the end of oxidation temperature step.

This leads to a first step of Cs release at the end of the oxidation phase (~20%), all the non-released Cs is located into the Cs_2MoO_4 phase coming from the reaction of Cs_2UO_4 and gaseous MoO_2. It seems that (following the experimental results) during this oxidation stage no Ba-molybdates is destroyed, because no Ba release is observed.
Therefore, at the end of the clad oxidation phase, it is supposed that there is still some gaseous MoO_2 in the intergranular bubbles network, all the Cs is located in the Cs_2MoO_4 phase, and Ba is dispatched between oxides, and molybdates.
Then, when temperature is raised again to 2573K, the evolution changes from Vercors 5 to Vercors 4 because of a different injected atmosphere:

- for Vercors 5, under pure steam, Cs_2MoO_4 is vaporized (~2200K) leading to some Cs and Mo release, BaO is vaporized (2500K) leading to Ba release, and some Ba and Mo stay trapped into BaMoO4 phase which is stable at 2573K under oxidising condition,
- for Vercors 4, under pure H_2, Cs_2MoO_4 and $BaMoO_4$ can be affected by the low oxygen potential coming for fuel reduction; this would lead to a decomposition of these two precipitates giving Cs and Ba release and a return of Mo into the metallic phase. Moreover it seems following experimental Mo release in Vercors 4 that a small part of Cs_2MoO_4 can be vaporized. There is a kind a competition between the molybdates destruction and its vaporization. In fact, for Vercors 4, 10% of Mo was collected into the impactor (instead of 50% for Vercors 5) coming from Cs_2MoO_4 vaporization.

For Vercors 4 and 5, 93% of Cs is released. The non-released part may come from a remaining part in solid solution (at the solubility limit) and other phases as Cs_2ZrO_3 whose thermal stability is high even under the reducing conditions of Vercors 4. 92% Mo is released for Vercors 5. If the 8% remaining is considered to be located in $BaMoO_4$ precipitate, this leads to a non-released part of Ba equal to 8x(0.248/0.084)=24%, 0.248 and 0.084 being the respective fission yield of Mo and Ba. For Vercors 4, 80% of Ba is released instead of only 55% for Vercors 5. As seen on figure 2, no Ba is expected to be trapped into $BaMoO_4$ phase. Therefore, following this scheme, the theoretical value for Ba release in Vercors 4 should be 55%+24%=79% which is closed to the experimental value (80%). The non-release part of Ba for Vercors 4 (20%) test can come from a remaining part in solid solution (at the solubility limit) and other phases as $BaZrO_3$ whose thermal stability is high even under reducing conditions. The 45% remaining part of Mo, in Vercors 4, is supposed here to be trapped into the metallic Mo condensed phase.

Additional comment: it is supposed here that, under reducing conditions, Mo coming from destruction of (Cs, Ba) molybdates returns into the metallic phase. This should have sense only if the fuel becomes sub-stoichiometric. As there is no measurement of the oxygen potential in Vercors tests, it is not evident that, under the reducing conditions of Vercors 4 test, the fuel is sub-stoichiometric at the end of the test. If it is still over-stoichiometric, a return of Mo into MoO_2 precipitate could be a better representation.

4. MFPR CALCULATIONS

Among the output files given by MFPR, several are devoted the follow the evolution step by step of the behaviour of chemically active fission products.

Fission flux during irradiation at 1100K under 25 atm of He and injected gas phase during the transient are taken into account to start the calculation. The gas phase was estimated to about 1% of the real injected flow. As there is no clad modelisation in MFPR, this estimation comes from the fact that cladding plays the role of a geometric barrier between gas and fuel. Moreover it is also a chemical barrier during the oxidation phase so that, no oxidising atmosphere was introduced, for the calculation, before the clad is fully oxidised, i.e. before the end of the oxidation step.

The main problem here is that neither MoO_2 nor BaO exist as solid precipitates in the code database. They can be found only in solid solution. Therefore, an additional input file where Gibbs energy, activity, diffusion coefficient (...) can be changed for each specie S_i, is useful. To start from the required localisation of Cs, Mo, and Ba, solubilities of MoO_2 and BaO were increased by decreasing their activities in solid solution. The same additional input file was used for Vercors 4 and 5 and the calculation provided an initial repartition presented in figure 4. These initial results are satisfying: 100% of Ba is located in BaO (as dissolved into solid solution), 94% of Cs is in the Cs_2UO_4 precipitate, and MoO_2 (as dissolved into the solid solution) contains 84% of total amount of Mo.

Figure 5 and 6 display calculation results of ternary compound evolution during Vercors 4 and 5 tests. It can be seen that the formation of Cs_2MoO_4 from Cs_2UO_4 and MoO_2 is represented and starts at the expected vaporization temperature of MoO_2 precipitate. This was done by forcing the code to make a reaction between MoO_2 (in solid solution) and Cs_2UO_4 (in ternary compound sub-phase). This operation was not done for the formation of $BaMoO_4$ from MoO_2. That is why no formation of $BaMoO_4$ is predicted until the end of the clad oxidation phase. Nevertheless, the final state of $BaMoO_4$ is satisfying for Vercors 5, with a large remaining part, and for Vercors 4, with a low remaining part.

The calculated release kinetics are presented in figure 7 and 8 for Vercors 4 and 5. Figure 7 is associated to figure 5, and figure 8 to figure 6. The first step of Cs release corresponds to the destruction of Cs_2UO_4. The last stage of Cs release is due to Cs_2MoO_4 decomposition in Vercors 4 and vaporization in Vercors 5. It can be seen that the Cs calculated kinetic fits quite well the experimental one for both tests. It is the same for Mo, supposed to be released as MoO_2 that was already present in grain porosities for temperatures over 1273K. Nevertheless, the release kinetic is delayed comparing the Vercors 5 for experimental results as well for calculated ones. For barium, calculated kinetics correspond to the expected phenomena but they do not reproduce very well experimental ones.

5. CONCLUSIONS

The present interpretation concerning (Cs, Mo, Ba) release in Vercors 4 and 5 tests highlights that their behaviour is fully interdependent. Cs should not be considered as a "volatile" specie treated in the same way and with the same models as noble gases. Cs, Mo and Ba have interdependent chemistry in nuclear fuels. MoO_2 precipitates can enter into reaction with Cs_2UO_4 and BaO precipitates to give some (Ba or Cs) molybdates during the reactor operation and during a temperature transient. For Vercors test, a first step of clad oxidation tends to decrease oxygen potential of the fuel, leading to a destabilisation of oxides and ternary compounds. A first Cs release step is observed and attributed to the destruction of Cs-uranate and some Mo can be released form the gaseous form MoO_2. Then when temperature is raised nearly to fuel degradation, in oxidising conditions, Cs and Mo are release due to Cs_2MoO_4 vaporization and Ba from BaO; in reducing conditions, molybdates tend to decompose with a return a Mo in condensed phase, which leads to additional Cs and Ba release without any Mo release, trapped into the fuel.

The work done with MFPR gives some good results on the release kinetics of Cs, Mo, and Ba for Vercors 4 and 5. Some thermodynamics laws were "disabled" because it seems that the true understanding of fission product release phenomenon must mix thermodynamic and mechanic/mechanistic sciences. This was specially observed for MoO_2 precipitates.

Nevertheless, such an interpretation needs some microscopic experimental evidences. Scanning and Transmission Electron Microscopy should be useful to identify phase distribution before and after tests on irradiated UO_2. X-ray diffraction with an increasing temperature on irradiated pellet should be also a precious tool to follow the phase evolution.

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FIGURE CAPTION

Table 1: Vercors 4, 5 experimental conditions and corresponding Cs, Mo, and Ba released amounts

Figure 1: Vercors 4 and 5 release kinetics (% of the initial inventory after irradiation) for Cs, Mo, and Ba.

*Figure 2: (Cs, Mo, Ba) release interpretation for Vercors 4 and 5 tests. *: P1 describes the destruction of* Cs_2MoO_4 *giving Cs release and a return of Mo into the metallic phase, P2 describes the destruction of* $BaMoO_4$ *giving Ba release and a return of Mo into the metallic phase.*

Figure 3: Difference between Mo released and Mo collected in the impactor for Vercors 5 test

Figure 4: initial repartition given by MFPR calculation on Vercors 4 and 5 tests

Figure 5: MFPR calculation of ternary compound evolution during Vercors 4 test

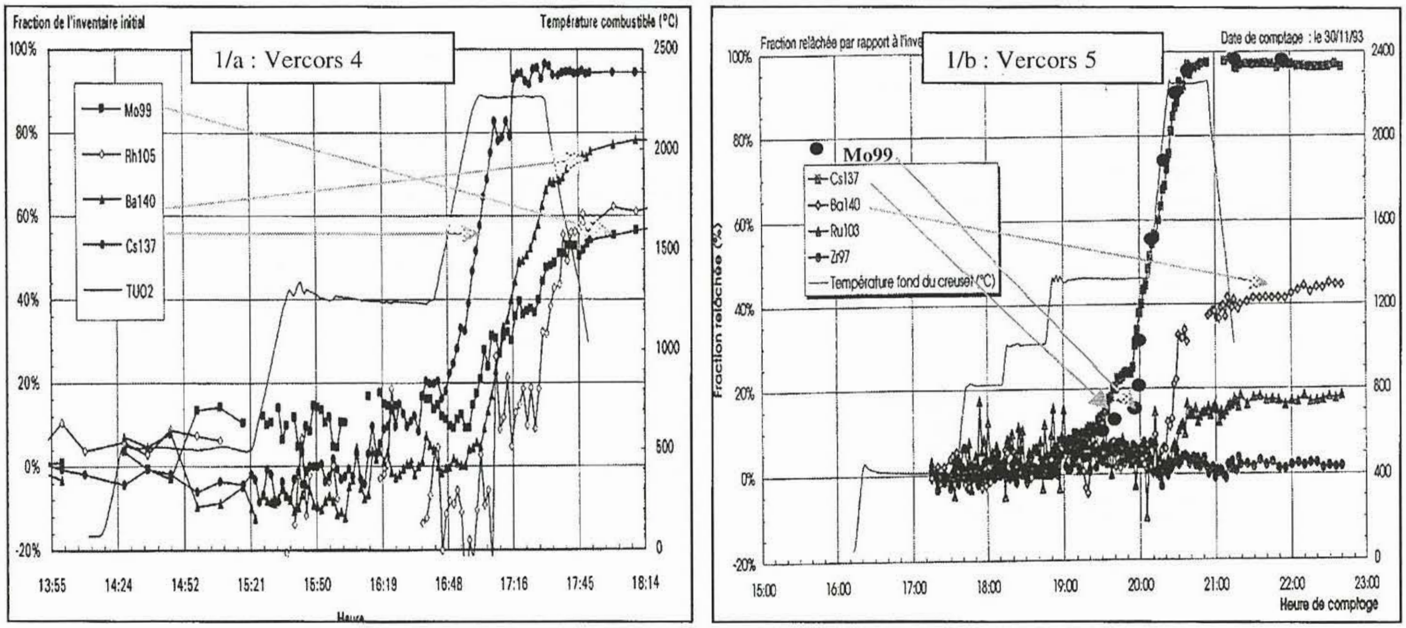
Figure 6: MFPR calculation of ternary compound evolution during Vercors 5 test

Figure 7: Vercors 4 release calculation and comparison with experimental values

Figure 8: Vercors 5 release calculation and comparison with experimental values

		Vercors 4		Vercors 5	
Fuel (Burnup)		UO_2 (40 GWd/t_{UO2})		UO_2 (40 GWd/t_{UO2})	
Clad oxidation temperature (K)		1523 (70 minutes)		1573 (80 minutes)	
Gas conditions					
Clad oxidation	**After oxidation**	H_2O/H_2	**H_2**	H_2O/H_2	**H_2O**
Cs released fraction (%)		93		93	
Mo released fraction (%)		**55**		**92**	
Ba released fraction (%)		**80**		**55**	

Table 1: Vercors 4, 5 experimental conditions and corresponding Cs, Mo, and Ba released amounts



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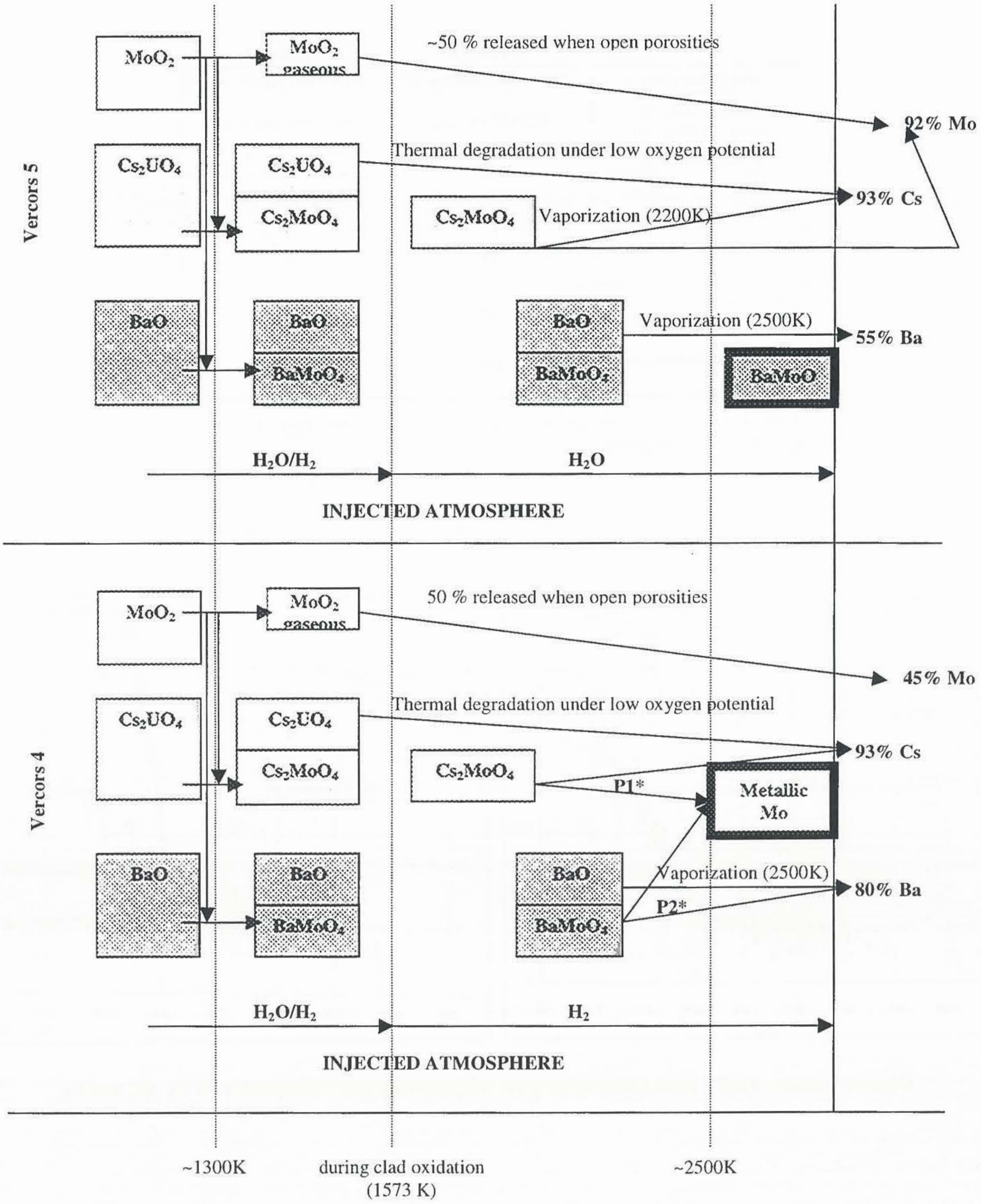

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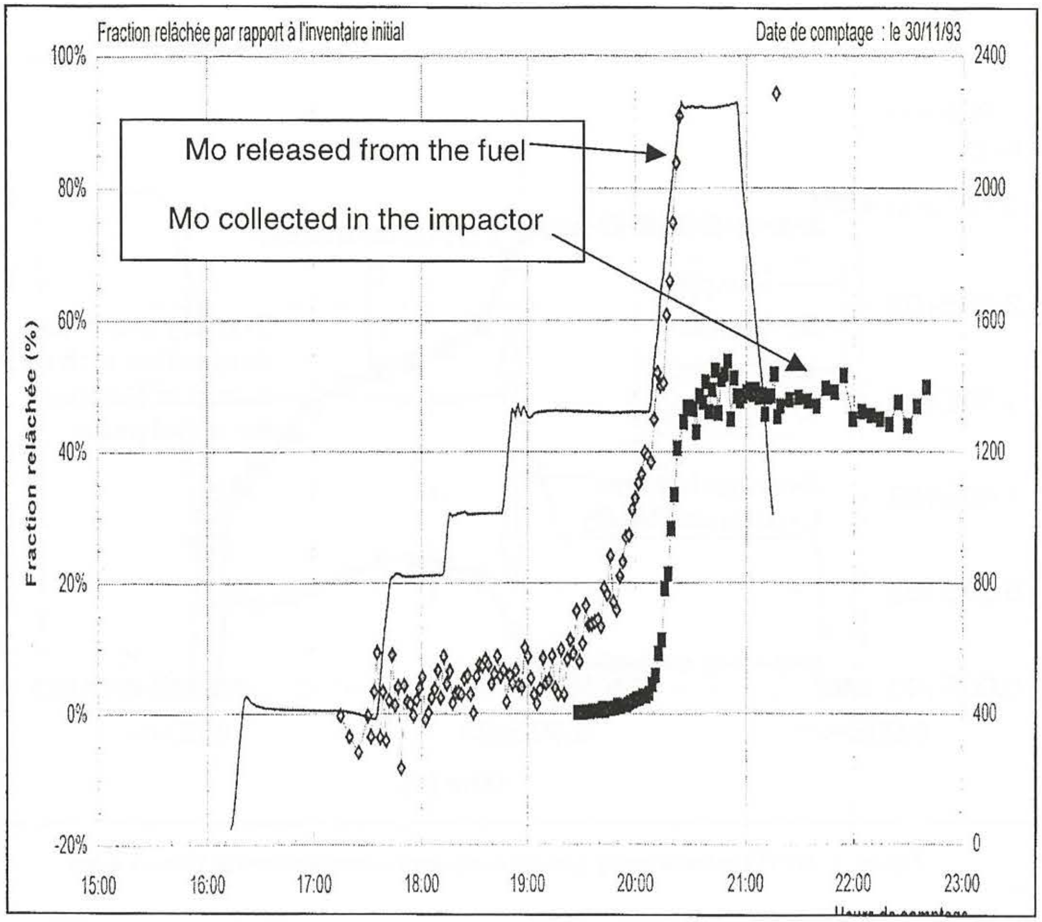

Figure 3: Difference between Mo released and Mo collected in the impactor for Vercors 5 test

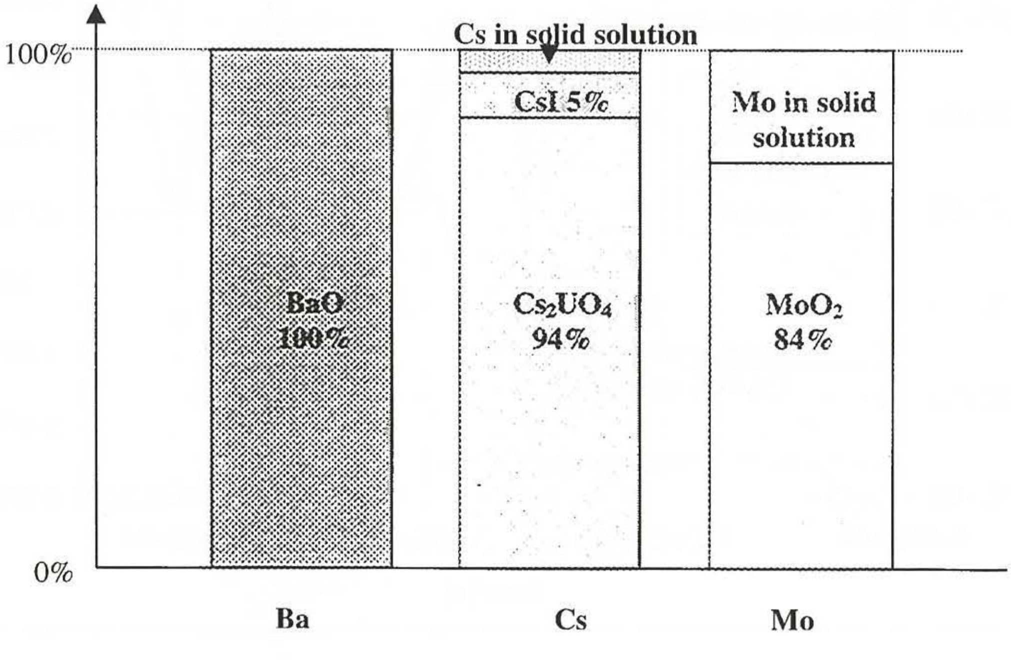

Figure 4: initial repartition given by MFPR calculation on Vercors 4 and 5 tests

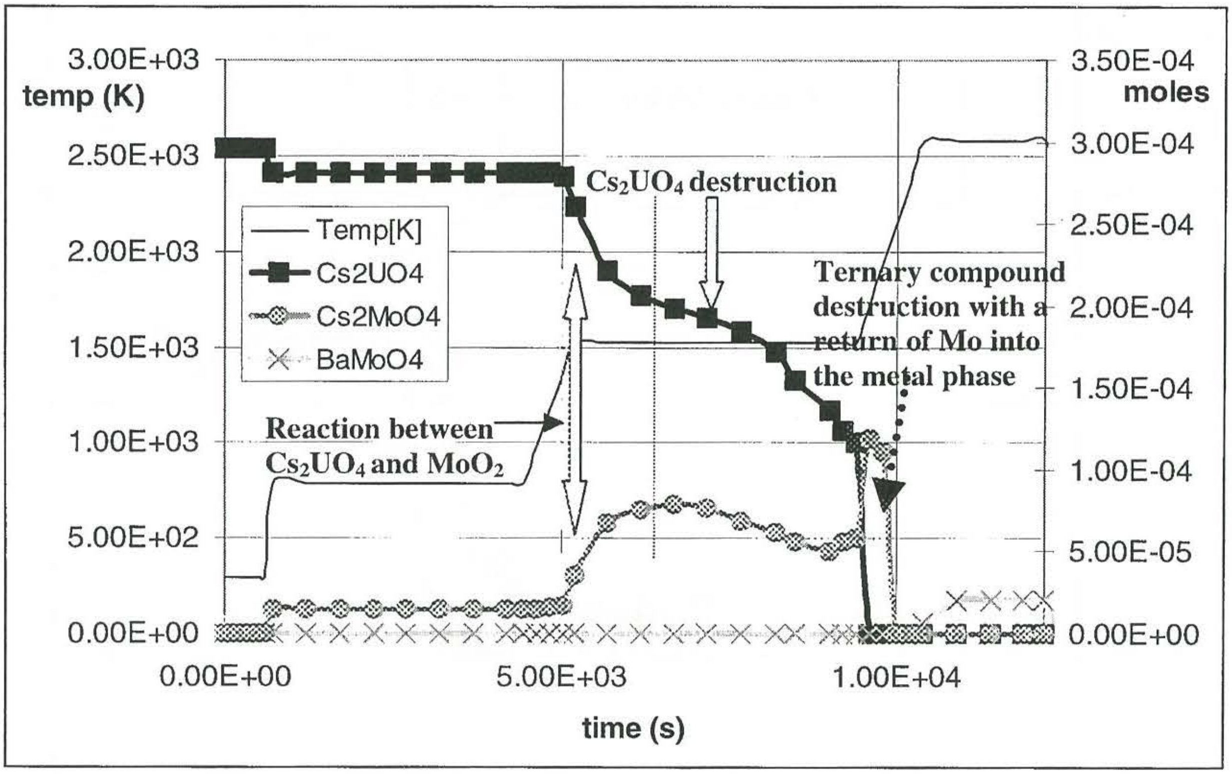

Figure 5: MFPR calculation of ternary compound evolution during Vercors 4 test

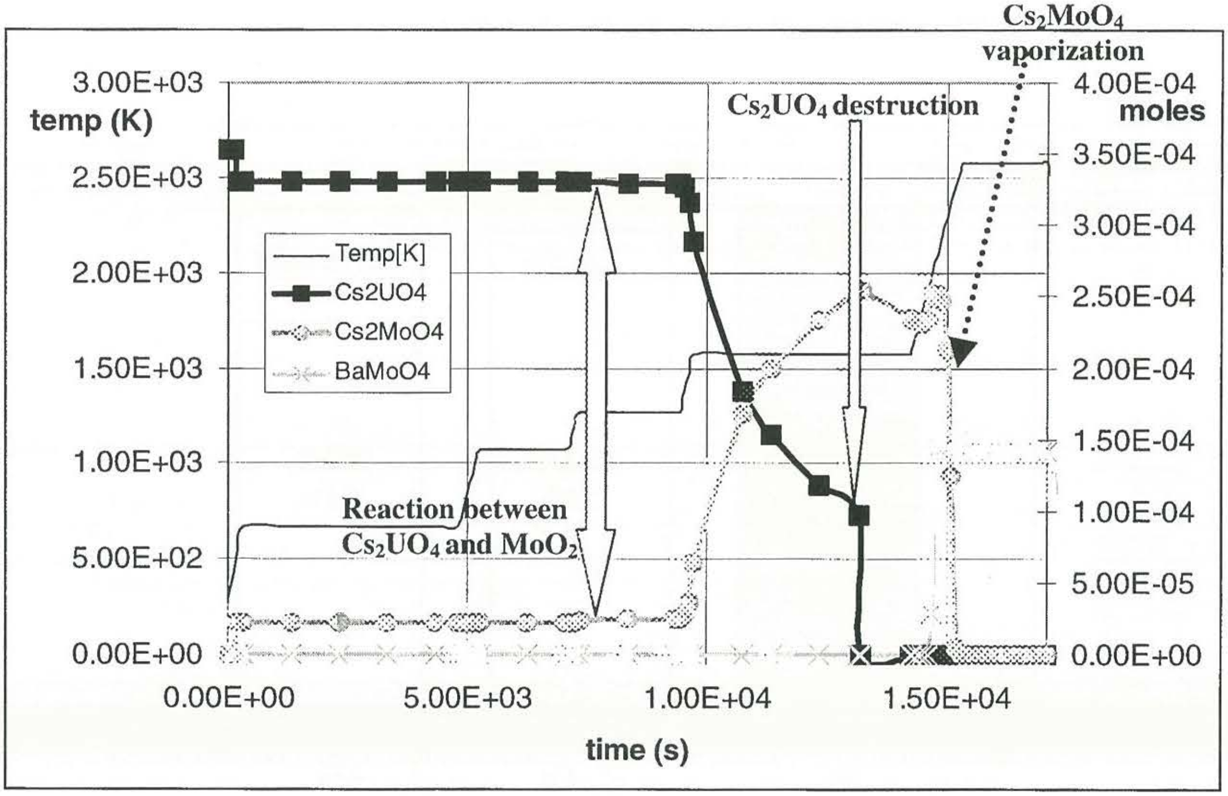

Figure 6: MFPR calculation of ternary compound evolution during Vercors 5 test

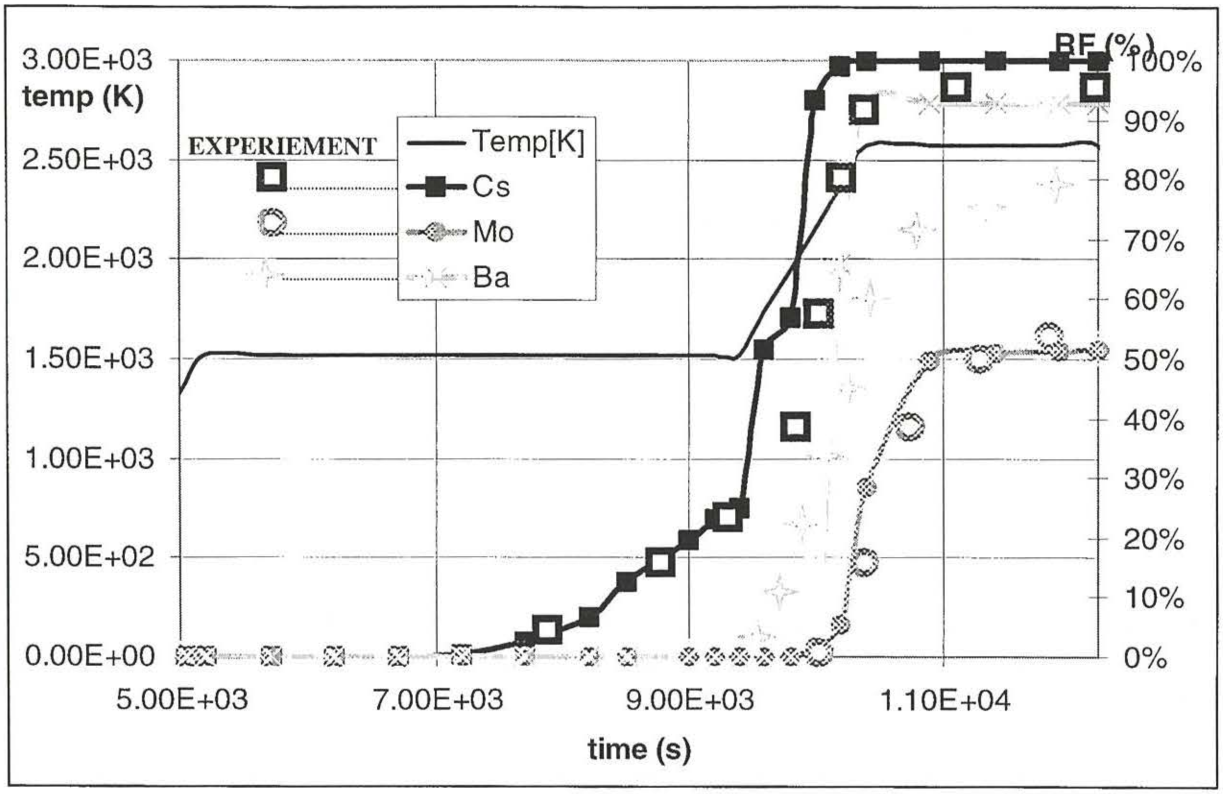

Figure 7: Vercors 4 release calculation and comparison with experimental values

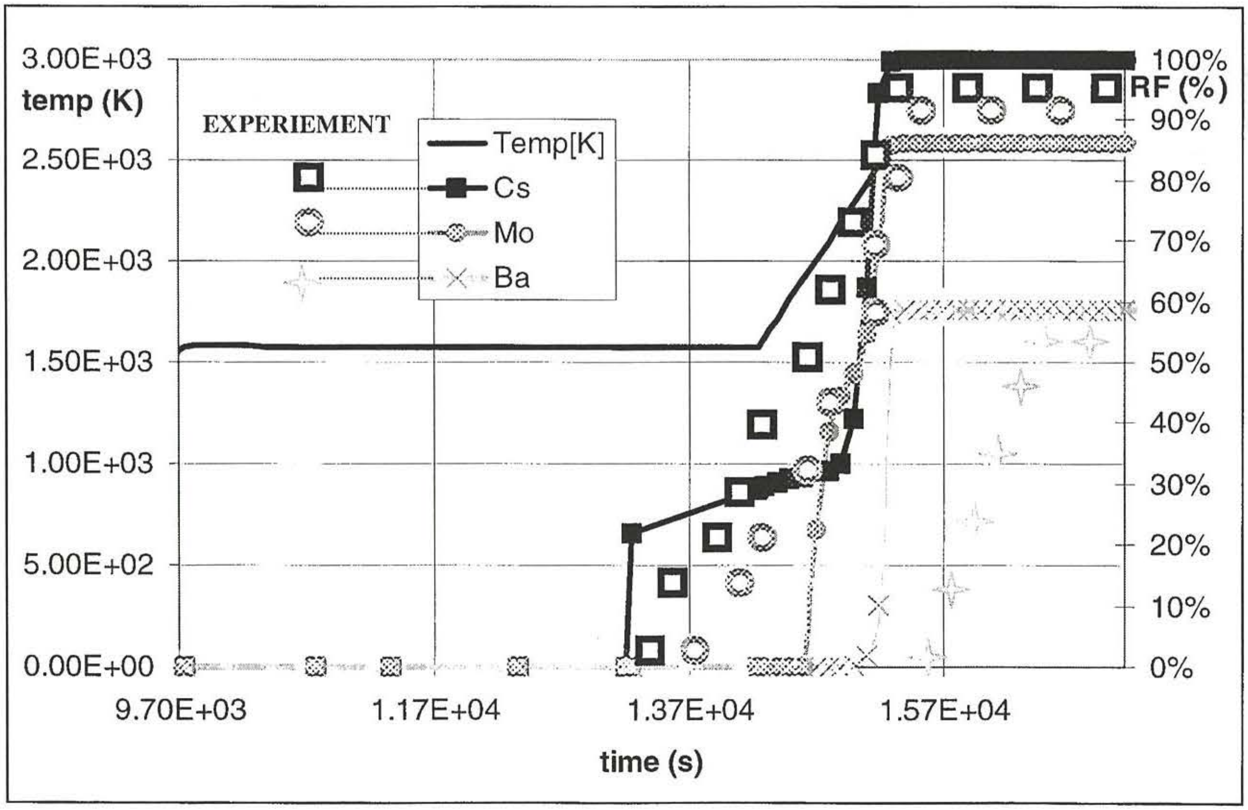

Figure 8: Vercors 5 release calculation and comparison with experimental values