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Simulation of the Chemical State of Irradiated Oxide Fuel; Influence of the Internal Corrosion on the Mechanical Properties of Zry-4 Tubing

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Simulation of the Chemical State of Irradiated Oxide Fuel; Influence
of the Internal Corrosion on the Mechanical Properties of Zry-4 Tubing⁺

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Abstract

Zircaloy is not compatible with oxide fuel nor with some fission product elements. Therefore, chemical interaction between the irradiated oxide fuel and the Zry cladding material take place, especially at temperatures that can be reached during reactor incidents (ATWS, LOCA). In order to find out which influence the chemical interaction between the fission products and the Zry cladding material have on the mechanical properties of Zry-4 tubing out-of-pile burst experiments and creep rupture tests have been performed at temperatures $\geq 600^{\circ}\text{C}$ with short tube specimens containing simulated fission products. First of all, assessments of the chemical state of irradiated oxide fuel were performed and a method is described for introducing simulated fission product species into fresh oxide fuel for irradiation tests. As the test results of the out-of-pile studies show, only iodine can lead to a low ductility failure of the Zry-tubing at temperatures $\geq 600^{\circ}\text{C}$. However, the influence of iodine on the deformation behavior of Zry-tubing can be neglected above 850°C .

Simulation des chemischen Zustandes von bestrahltem Oxidbrennstoff; Einfluss der Innenkorrosion auf die mechanischen Eigenschaften von Zry-4-Hüllrohren

Zusammenfassung

Zircaloy ist gegenüber Oxidbrennstoff und einem Teil der Spaltprodukte thermodynamisch nicht stabil. Es finden deshalb chemische Wechselwirkungen zwischen dem abgebrannten Oxidbrennstoff und dem Zry-Hüllmaterial statt, besonders bei Temperaturen wie sie bei Reaktorstörfällen (ATWS, LOCA) erreicht werden können. Um herauszufinden, welchen Einfluss die chemischen Wechselwirkungen zwischen den Spaltprodukten und der Zry-Hülle auf deren mechanische Eigenschaften haben, wurden out-of-pile Berst- und Zeitstandexperimente mit kurzen, spaltprodukt haltigen Zry-4-Rohrproben bei Temperaturen $\geq 600^{\circ}\text{C}$ durchgeführt. Zunächst wurden Abschätzungen über den chemischen Zustand von bestrahltem Oxidbrennstoff durchgeführt. Eine Methode zur Einbringung von simulierten Spaltprodukten in Oxidbrennstoff zur Durchführung von Bestrahlungsexperimenten mit spaltprodukt dotiertem UO_2 wird vorgeschlagen. Wie die out-of-pile Versuchsergebnisse mit simulierten Spaltprodukten zeigen, führt nur Jod zu einem verformungsarmen Versagen der Zry-Hüllrohre bei Temperaturen $\geq 600^{\circ}\text{C}$. Der Einfluss von Jod auf das Verformungs- und Bruchverhalten der Zry-Hüllrohre kann jedoch oberhalb 850°C vernachlässigt werden.

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1. Introduction

Knowledge of the fuel fission product chemistry in a LWR-fuel rod is important, because it can exert an unfavorable influence on the fuel rod behavior during normal reactor operation as well as in reactor incidents. In normal reactor operation it cannot be avoided that fuel element damage occurs although the number of failures is minimal. As shown by reactor operating experience, fuel element damage may take place preferably during startup of a reactor and in the course of power rises. Since fuel elements damage is both a problem of safety and economics, efforts are made to reduce it by continuous advancement of the fuel element technology and by optimization of the reactor operating conditions.

To obtain more information about the fuel rod behavior very comprehensive theoretical and experimental investigations are being made worldwide. Special attention is being paid to the fuel rod behavior in case of power ramps. During power increases, especially after medium and high burnups of fuel rods, quite considerable mechanical interactions might take place between the fuel and the cladding material. The hoop stress of the cladding tube may then attain very high values locally so that the fuel element cladding is subject to mechanical failure. However, contrary to earlier concepts, it is felt today that a mechanical stress of the cladding tube alone is not the cause of fuel rod failure but that chemical environmental influences have to occur in addition. Meanwhile, even the conviction prevails that the fuel rods mainly fail as a result of stress corrosion cracking /1 - 7/. Stress corrosion cracking (SCC) has received wide spread attention as a mean of explaining failure of Zry clad fuel rods. Brittle cracks have been found on rods subjected to power increases after having operated for sustained periods at low power levels.

However, heavy mechanical stresses of the cladding tube might not occur only during power ramps but also in reactor accident transients (ATWS, LOCA). Generally, the question therefore arises how the

mechanical properties of the Zry-4 cladding material might be influenced by chemical interactions of the fuel/fission product system. This question can be studied by in-pile experiments and partly also by out-of-pile experiments. In-pile experiments are close to reality, however very time consuming, and expensive and the results are often difficult to interpret. By contrast, out-of-pile experiments are comparatively cheap; they can be quickly performed and therefore allow extensive parameter studies to be made on the fuel rod behavior. Moreover, laboratory scale experiments offer the advantage that single effects can be investigated specifically. However, the out-of-pile test results must be confirmed always by in-pile experiments since not all the nuclear parameters can be simulated. A stage between the in-pile experiments and laboratory investigations are the short term irradiation experiments on fuel with simulated fission products. A method for introducing simulated fission products into test fuel rods is described in the appendix.

2. Chemical State of Irradiated Oxide Fuel

It is important to know the chemical state of the irradiated LWR fuel to perform out-of-pile experiments or in-pile experiments with fission product doped UO_2 . In other words, it is necessary to know the chemical state of the different fission product elements contained in UO_2 as a function of the O/U ratio and temperature. Besides, it is important to know which fission product elements and compounds must be considered as aggressive with respect to the Zry-4 cladding material. To obtain this information, both thermodynamic estimates in the complicated multi-component system of UO_2 -fission products - cladding material and careful post-irradiation examinations of LWR fuel rods can be performed. The drawback of the thermodynamic estimates lies in the exclusive application of equilibrium thermodynamics and in the lack of the respective thermodynamic data for many possible reaction products. Nevertheless, satisfactory statements can be made for a great number of fission products concerning their possible chemical states in UO_2 . Post-irradiation examinations supplement the theoretical estimates

and confirm partly the possible reaction products although they do not provide a comprehensive picture of the fuel - fission product - cladding material chemistry. The reasons are that already in sample preparation the easily volatile and hygroscopic reaction products get lost.

2.1 Fission Product Yield for U-235 and Pu-239 Fission

To be able to make statements about the maximum possible oxidation of the cladding inner surface by the oxygen released from UO_2 or $(U,Pu)O_2$ by nuclear fission, estimates on thermodynamics were made for the oxide fuel-fission product-cladding material quaternary-component system. In order to make thermodynamic considerations relative to the complex system and to be able to perform experiments with simulated fission products, it is important to know which fission products are formed at which concentration by the fission of uranium and plutonium. The fission product yield depends on various parameters such as the mass number of the fissioned isotope, the neutron spectrum, the duration of the neutron irradiation, the cross sections for neutron capture reactions, and the decay time. The fission product yield for the fission of the U-235 and Pu-239 nuclides as a function of the parameters indicated was calculated and summarized in a report /10/.

For the fission of U-235 and Pu-239 by thermal neutrons, the fission product yields contained in Table 1 are obtained for a decay time of one year. In compatibility studies it is reasonable to combine the fission products into chemical groups. The fission product concentration of these groups (e.g. alkaline metals, earth alkaline metals, lanthanides) is then almost independent of the irradiation and decay times /10/.

Since in the future also Pu-239 is to be used as a fissionable isotope in LWR fuel rods, it was taken into account in the thermodynamic estimates in addition to U-235.

Fission Products	Fission Product Yield	
	U-235	Pu-239
Cs, Rb	22.95	21.63
Ba, Sr	16.04	9.35
Se, Te	2.98	3.83
I, Br	1.31	2.37
Kr, Xe	25.41	25.12
Ru, Rh, Pd	15.38	39.89
lanthanides	48.79	43.64
Zr	31.13	19.39
Nb	0.19	0.16
Mo	24.73	23.83
Tc	6.16	5.90
Y	5.00	1.80
Cd	0.08	0.51

Table 1: Comparison of fission product yields of stable and long-lived elements for the fission of U-235 and Pu-239 by thermal neutrons; data about fission product yields in at.% related to a total value of 200%. Decay time: 1 year

2.2 Thermodynamic Estimate of the Chemical State of Fission Product Elements in the Oxide Fuel

During the nuclear fission of oxide fuels oxygen is released which is bound by the fission products formed, the fuel, and the cladding material. It depends on the thermodynamic stability of the oxides, the concentration of the oxygen binding fission products, and initial O/M-ratio of the fuel (M = U,Pu) to which extent the oxygen will react with the different substances.

The free enthalpies of formation of the different fission product oxides are represented in Fig.1 as a function of the temperature /1, 11/. By a comparison of these values with the partial free enthalpy of the oxygen in the fuel we find that all those fission products will be present as oxides whose amounts of free enthalpy of formation are more negative than the free enthalpy of the oxide fuel.

The fission products can be classified according to the stability of the oxides as

- the very stable lanthanide (rare earth metal) oxides and Y, Zr, Ba, Sr, which in the oxide form are present either in solid solutions with the fuel or as pure fission product oxides;
- fission products whose partial free enthalpy of oxide formation is almost identical to the partial free enthalpy of the stoichiometric fuel (O/M \sim 2.00), such as Cs, Rb and Mo;
- Ru, Rh and Pd which do not form oxides and are therefore present in the fuel as metals or intermetallic phases /1,11/.

On the basis of the thermodynamic data from Fig.1 and the fission product yields from Table 1 the average valency of the fission products can be calculated and compared with that of the oxide fuel (valency $\geq 4^+$). Both for the fission of U-235 and of Pu-239 the average valency of the fission products is smaller than that of the fuel, which means that less oxygen is bound by the fission products than is released by the fission of UO_2 or $(U,Pu)O_2$. Therefore, the burnup exerts a decisive influence on the chemical behavior of the irradiated system, since, besides the fission products, oxygen will be available for reactions with the Zry cladding material.

2.3 Shift of Fuel O/M-Ratio during Burnup

Careful post-irradiation examinations on fuel rods of power reactors yielded that the UO_2 in the fuel rod has a stoichiometric O/M ratio /12/. The oxygen potential of UO_2 is determined by the oxygen gettering of the Zry-cladding material. In power reactors the cladding temperature

is sufficiently high so that part of the oxygen released during nuclear fission of UO_2 gets bound by the cladding material. However, if the fuel/fission product system is considered separate from the cladding, the O/U ratio of UO_2 will always increase during irradiation because the fission products cannot bind the entire oxygen released by nuclear fission. Therefore, an average stoichiometric shift can be determined which, besides on the fissile isotope and the neutron energy, depends on the state of oxidation of the fission products. However, the estimate of the stoichiometric shift is complicated by the inadequate knowledge of the oxidation state of some fission products and by the plutonium fission increasing with burnup. Figure 2 represents the change of the O/U ratio as a function of the burnup for different oxidation states of the fission products Cs, Mo and the lanthanides. The most probable case is that Cs and Mo largely occur in a nonoxidized state in UO_2 ; the formation of Cs_2MoO_4 and/or Cs_2UO_4 or other oxygen containing complex compounds was not considered in these calculations.

Depending on the valency of the rare earth metals (lanthanides) the stoichiometric shift of MO_2 ($M = U, Pu$) as a function of the burnup for U-235 and Pu-239 fission is presented in Figure 3. The rare earth metals were assumed to be trivalent or tetravalent. In this way, a scattering band is obtained in which the O/M-increase of the oxide fuel takes place. A higher amount of oxygen release can be expected from Pu-239 fission because less oxygen binding fission products are formed. However, the oxygen in excess estimated in this way is completely gettered at sufficiently high cladding material temperatures as encountered in power reactors so that the O/M-ratio of the fuel takes the value 2.000 /12/. Therefore, using these estimates, the maximum oxygen uptake of the Zry-cladding can be determined and, consequently, the maximum thickness of the oxide layers to be expected on the cladding inner surface can be estimated which is significant for the performance of simulation experiments ($1 \mu m$ $ZrO_2 \hat{=} -\Delta O/M \approx 0.001$).

As regards the chemical interactions with the Zry-cladding material, the fission products iodine and tellurium as well as cadmium, in addi-

tion to oxygen, have found to be aggressive with respect to Zry from thermodynamic estimates and laboratory scale experiments /9,13/. It should be further noted that even the UO_2/Zry initial system, which is free from fission products, is not stable thermodynamically /16, 17/. However, since the normal cladding material temperatures are sufficiently low, the chemical interactions with the cladding material can be tolerated. Nevertheless, the question arises to which extent the mechanical properties of Zry are influenced accordingly. The impact of iodine on Zry has appeared as a special problem since iodine caused stress corrosion cracking. The necessary iodine partial pressures leading to stress corrosion cracking of Zry in the out-of-pile experiments are considerably higher than the partial pressure expected from equilibrium thermodynamics in the fuel rod. It is absolutely necessary to clarify this discrepancy by appropriate experiments.

3. Influence of Internal Corrosion on the Mechanical Properties of the Zry-4 Tubing

Under the Nuclear Safety Project investigations have been performed in Karlsruhe to show whether also in reactor transients (ATWS, LOCA) volatile fission products, especially iodine, exert an influence on the mechanical properties of Zry. There was particular interest to answer the question whether iodine might cause stress corrosion cracking also at cladding material temperatures above $400^{\circ}C$. The following fission product elements and compounds have been examined to find out whether the presence of these substances will exert an influence on the strain and rupture behavior of the Zry-cladding tube at high temperatures ($\geq 600^{\circ}C$):

- I, Cs, Te, Se, Cd, Sn, Sb
- ZrI_4 , TeI_4 , Cs_2Te , CsI , Cs_2O , TeO_2 , I_2O_5
- Cs_2ZrO_3 , Cs_2MoO_4 .

Moreover, the influence of oxide layers on the cladding tube inner surface as well as of defects in the cladding tube has been investigated /8,14,15/.

As appears from the results of the burst experiments at 800⁰C, all substances caused a decrease of burst strain of the Zry-cladding tubes as compared with the specimens filled only with argon. However, only the test specimens containing elemental iodine or iodine compounds, excepts for CsI, indicated a conspicuous reduction in cladding tube rupture strain at temperatures below 850⁰C as a result of stress corrosion cracking. Therefore, the following statements are restricted solely to the influence exerted by iodine on the mechanical properties of Zry-4 cladding tubes

These are the most significant test results of the creep rupture and burst tests with the short iodine containing Zry-4 tubular specimens under inert gas conditions /8,14,15/:

- Also, at cladding material temperatures above 350⁰C, which can be expected in LWR accidents, elemental iodine exerts an influence on the mechanical properties of Zry, especially on the maximum circumferential burst strain of the cladding tubes (Figure 4). This results in a low ductility failure of the Zry-cladding tubes due to stress corrosion cracking, caused by iodine. The burst strain of iodine containing Zry-4 cladding tube specimens is clearly lower than that of iodine free reference specimens. However, the influence of iodine on the deformation behavior of Zry tubes decreases with increasing temperatures and can be neglected above 850⁰C (Fig. 4).
- Thin oxide layers (<30 μm) on the cladding tube inner surface combined with iodine lead to a further reduction in ductility of the Zry cladding tubes below 850⁰C. In the temperature and pressure range typical for a hypothetical LOCA, the burst strains of pre-oxidized Zry-tubing amount to 5 - 15% (Figure 5).
- Preflawned (machine grooves) Zry-4 cladding tubes containing iodine suffer from SCC at much lower nominal stresses than tubes without incipient cracks. The time-to-failure at 800⁰C is much shorter than for specimens not containing iodine. The burst strains amount to values <5% (Figure 6).
- If the true hoop stress at rupture of the Zry cladding tubes is determined, taking into account the burst strain, it can be

recognized that this rupture stress is smaller below 850°C for the specimens containing iodine than for the argon reference specimens (Figure 7). The hoop stress so determined might be the necessary critical stress in the cladding material which, in the presence of corrosive media, such as iodine, results in SCC and hence in the failure of the cladding tube. At temperatures above 850°C the presence of iodine exerts practically no influence on the deformation and rupture behavior of the Zry cladding tubes. Most probably the critical stresses in the cladding tube required for failure are no longer attained at the elevated temperatures, since these stresses get reduced before as a result of plastic deformation. The extent to which the two-phase character of Zry above 850°C is codeterminant of this behavior has not yet been clarified.

- The critical iodine concentration above which low ductility failure results of the Zry-cladding tube due to stress corrosion cracking mainly depends on the Zry temperature and on the presence of oxide layers on the cladding tube inner surface. Thin oxide layers (7 - 15 µm) cause a slight shift of the critical iodine concentration towards lower values. According to preliminary test results the critical initial iodine concentration amounts to about 1 mg/cm³ for the as-received Zry cladding tubes at about 800°C and for the specimens with thin oxide layers on the cladding tube inside surface to about 0.1 mg/cm³ (Figure 8). By contrast, at about 950°C no influence of iodine on the mechanical properties of Zry was found up to the presently investigated maximum iodine concentration of 100 mg/cm³ (Figure 9).
- SEM-investigations of the cladding tube inner surface of burst specimens show clearly that quite a number of intergranular incipient cracks are formed in the cladding tube surface in the presence of iodine at cladding material temperatures <850°C. The specimens damaged by iodine SCC at temperatures <850°C always exhibit great intergranular crack zones and small transgranular cleavage zones followed by ductile fracture zones (Figure 10). In the absence of iodine (reference specimens) or temperatures >850°C, the fractography of the tubes shows an entirely ductile mode of failure (dimpled crack surface).

4. Conclusions

In-pile and out-of-pile experiments have clearly shown that the impact of fission product in the fuel rod may lead to a low ductility failure of the Zry cladding tubes, even at high temperatures. It was proved by extensive out-of-pile experiments that of the fission product elements present in the fuel rod, mainly iodine is responsible for brittle failure of Zry. At a sufficiently high tensile stress in the cladding material, the impact of iodine results in cladding tube failure due to stress corrosion cracking. The iodine partial pressures required are determined from out-of-pile studies. These partial pressures are considerably higher than those expected in the fuel element on account of theoretical estimates. It is assumed in these estimates that the iodine is bound by Cs at low oxygen potentials of the UO_2 and occurs as CsI.

To clarify this discrepancy between the necessary iodine partial pressures determined from out-of-pile studies and the estimated partial pressures expected in the fuel element, specific in-pile experiments are required, particularly because the reactor radiation field takes some importance (reference: reduced stability of cesium iodine in the radiation field which results in the increase in iodine activity in the system). However, to cut the long irradiation periods and to be able to study better the effects of individual species the in-pile experiments should be performed with fission product doped fuel. Moreover, it is important to take care of the proper setting of the cladding material temperature in these experiments since the cladding material temperature has a decisive influence on the fission product/fuel chemistry.

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APPENDIX

Method for Introducing Simulated Fission Products into Test Fuel Rods

A method is described for introducing fission product species into fresh oxide fuel for irradiation tests.

Cesium, iodine and tellurium are regarded as aggressive fission products with respect to the Zry cladding material. For simulating irradiated oxide fuel, the fission product elements Cs, I, Te and Molybdenum should be added to the UO_2 by experimentally developed means. Mo is also an abundant fission product and it plays a role in the transport mechanism of Cs. These four elements can be introduced into the fuel as free elements or as compounds. Since cesium and iodine have a strong affinity for one another at low oxygen potentials it is convenient and preferable to introduce them in the form of the stable compound CsI (in addition, elemental cesium can be introduced only in an oxygen free and moisture-free atmosphere). More cesium is generated during irradiation than iodine, however, the additional amount of cesium needed for a given burnup can be added as well defined complex oxides such as Cs_2UO_4 or Cs_2MoO_4 . The additional oxygen included will also simulate the increase in the oxygen/metal ratio which occurs during irradiation. Mo may be added either in elemental form or as MoO_2 if additional oxygen is needed to reproduce the irradiation effect. Tellurium is added in the elemental form.

The amounts of fission products which have to be added to the fuel per 1 g of UO_2 for different burnups are indicated in Table 2. The use of Cs_2MoO_4 or Cs_2UO_4 , will increase the O/U ratio of the fuel to a value approximately equal to theoretical estimates, which are based on a fission product-oxygen balance. Thus, fuel oxidation is unnecessary.

The change of the O/U ratio in the fuel as a function of burnup is calculated on the assumption that the lanthanides (rare earth) are present in the trivalent state, while Cs and Mo occur as metals in UO_2 . This means that the O/U ratios represent the maximum possible oxygen potential of UO_{2+x} , provided that the Zry-cladding does not undergo

oxidation on its inner side. However, such values are not applicable since at sufficiently high cladding material temperatures the excess oxygen reacts with Zry (UO_{2+x} will be reduced to $UO_{2.000}$).

During a short in-pile conditioning period of the oxide fuel rods, part or all of the excess oxygen will react with the Zry-cladding, on the premisses that the cladding material temperature is sufficiently high. In this way conditions are attained which are relevant to practical application. "Relevant to practical applications" means that the oxygen released in nuclear fission will react with the Zry-cladding, thereby stabilizing the oxide fuel at an O/U ratio of about 2.000 in conformity with the thermodynamic data. This means for the simulated irradiated fuel rods that during the short pre-irradiation in the reactor the oxygen of the simulated fission product oxides (Cs_2MoO_4 , Cs_2UO_4) should react with the Zry-cladding (problem of kinetics).

The fission product mixtures are to be introduced as small pellets into holes provided in the UO_2 -pellets. For a simulated burnup of 5 at.%, about 0.1 g of the fission product mixture must be added per pellet. However, it seems reasonable to add fission product mixtures to only each second or third fuel pellet.

To accommodate the fission product mixtures, the UO_2 pellets introduced into the fuel rod can be provided with central holes. In the case that the fuel pellets have already been provided with central holes for the installation of thermocouples additional non-axial holes must be drilled to accommodate the fission products. The size of the holes (diameter, depth) is dependent, among other factors, on the amount of fission products and hence on the simulated burnup, the density of fission product mixtures, and on the number of sample locations.

During fuel rod assembly, the fission product mixtures should be placed into the dried UO_2 pellets immediately before loading into the cladding tubes. Exposure of the fission product mixtures to air should be minimized because CsI is hygroscopic. This means that with

respect to fuel rod assembly humidity in the atmosphere is more critical than the presence of oxygen. However, the humidity can be largely eliminated by vacuum drying.

When a given burnup is to be simulated by addition of fission products under the method described, it must be considered that in this method a 100% release of the Cs, I and Te fission products is assumed. However, this will probably not be the case in practice (with LWR's). The percentage actually released depends primarily on the fuel temperature. For accurate simulation of irradiated fuel, the expected fission product release must be estimated. A higher gas pressure in the fuel rod must be simulated too.

First irradiation experiments with simulated fission product doped oxide fuel, according to the proposed method, were already successfully performed in the Power Burst Facility in Idaho, USA.⁺)

⁺) R.R.Hobbins, T.F.Cook, S.A. Ploger;
Postirradiation Examination Results for the Irradiation
Effects Test IE-5
TREE - NUREG - 1201 (1978)

Table 2: Concentration of Some Fission Products and Fission Product Compounds. Respectively, in 1 Gram Irradiated UO_2 for Different Burnups

U-235 fission by thermal neutrons ($E_M = 0.025$ eV); decay time 1 year:

1 at.% \approx 9130 MWd/t

Weights are indicated in gram per 1.0 g $UO_{2.000}$

Simulated fission products and fission product compounds, resp.	Simulated Burnup (At.%)			
	1	3	5	10
Cs (Rb)	0,001173	0,003591	0,00611	0,012903
J (Br)	0,000063	0,000194	0,00033	0,000698
Te (Se)	0,0001463	0,0004479	0,000762	0,001609
CsJ	0,000129	0,000397	0,000676	0,001429
Cs_2MoO_4	0,001771	0,00542	0,009222	0,019475
Cs_2UO_4	0,002365	0,007327	0,012312	0,025999
⁺ Theoretical increase of O/U ratio during fission of $UO_{2.000}$	2,004	2,013	2,022	2,045
⁺⁺ Increase in O/U ratio by the use of Cs_2MoO_4 and Cs_2UO_4 , resp.	2,004	2,014	2,03	2,05

⁺When changes in stoichiometry are calculated the lanthanides are assumed to be trivalent while Cs and Mo are assumed to occur as metals; therefore the value indicated represent the maximum variation of the O/U ratio in the fuel.

⁺⁺ Cs_2MoO_4 and Cs_2UO_4 cause approximately the same increase in the O/U ratio of the fuel for the quantities indicated.

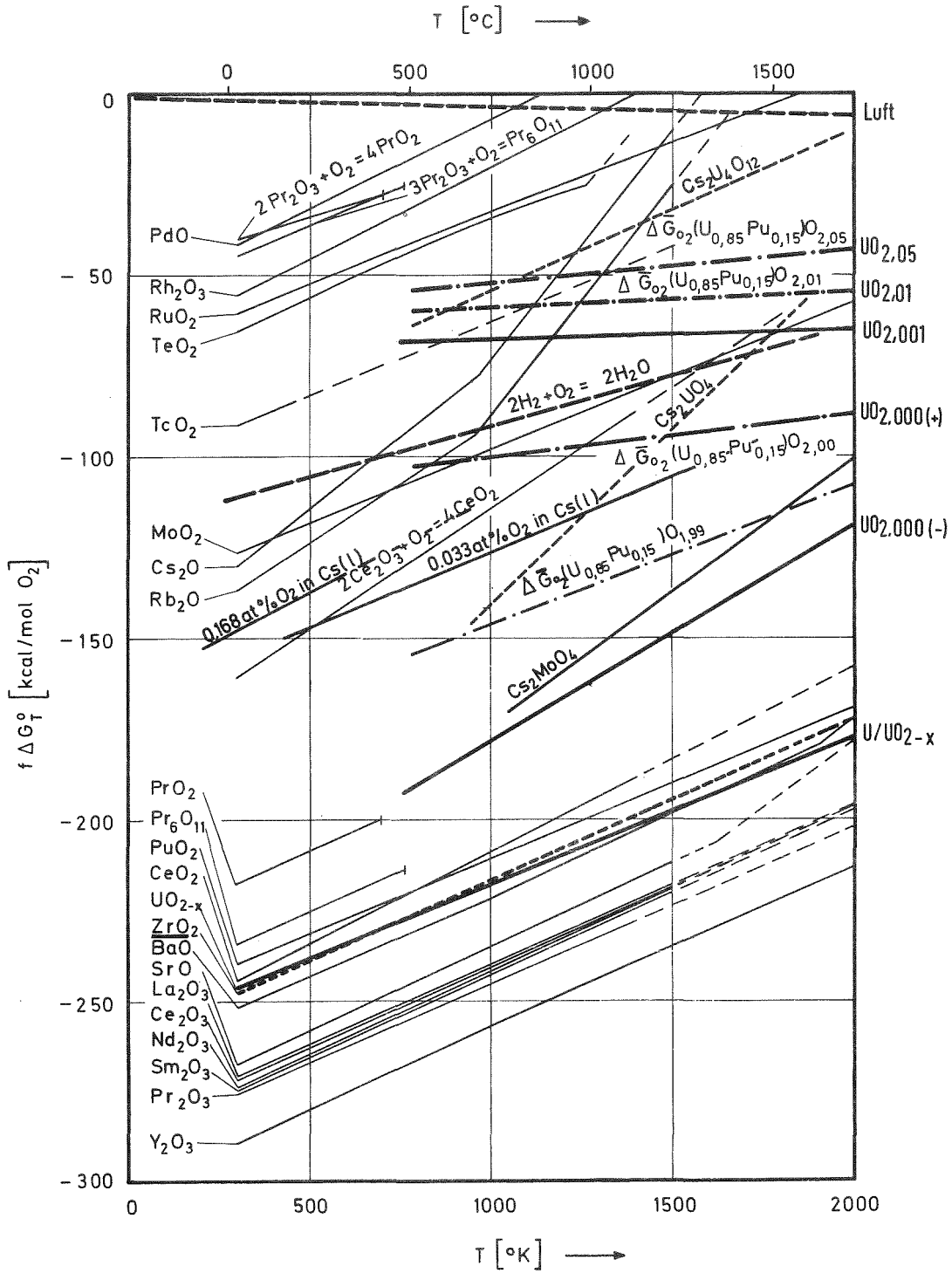


Fig. 1: Oxygen potential of UO_2 , $(U,Pu)O_2$ and fission product oxides as a function of the temperature [11].

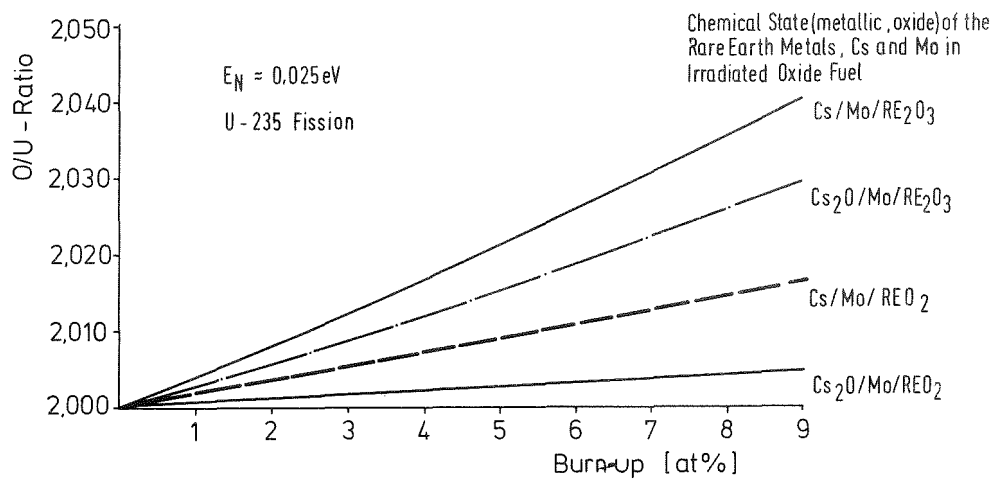


Fig. 2: O/U-shift of UO_2 as a function of the burnup for different states of oxidation of Cs, Mo and the lanthanides (rare earth metals).

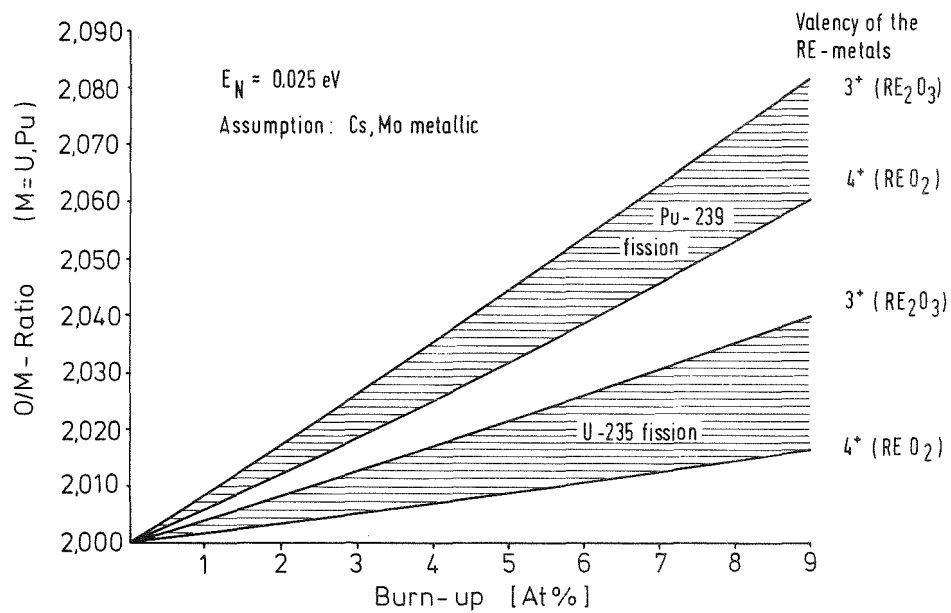


Fig. 3: O/M-shift of the oxide fuel as a function of the burnup for the fission of U-235 and Pu-239.

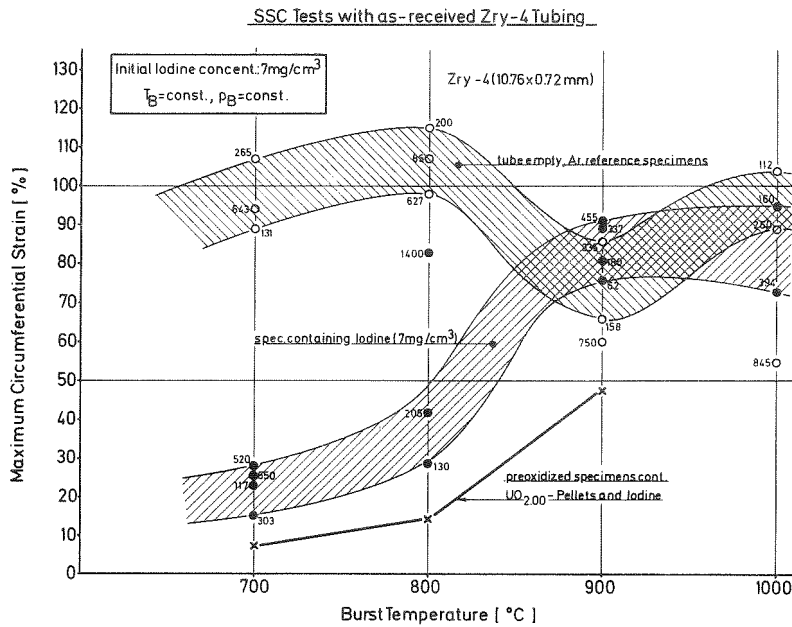


Fig. 4: Circumferential burst strain vs burst temperature of as-received Zry-4 tubing internally pressurized with argon or argon and iodine-gas mixtures (with the indication of the time-to-failure).

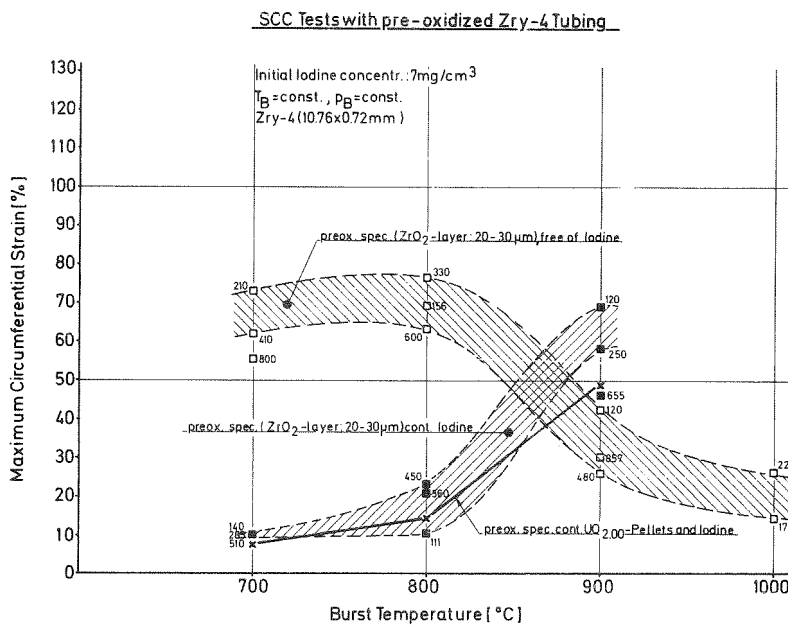


Fig. 5: Circumferential burst strain vs burst temperature of pre-oxidized Zry-4 tubing internally pressurized with argon or argon and iodine-gas mixtures (with the indication of the time-to-failure).

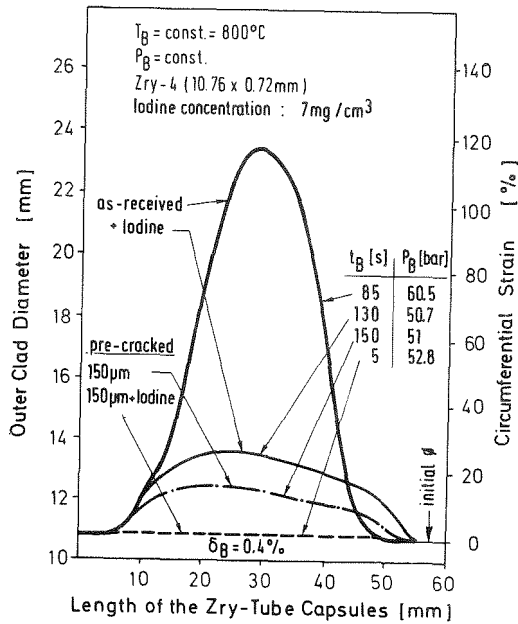


Fig. 6: Deformation of as-received and pre-cracked (150 µm) Zry-4 tubing after stress-rupture testing in Ar at 800°C. The tube specimens were internally pressurized with Ar or Ar/iodine-gas mixtures.

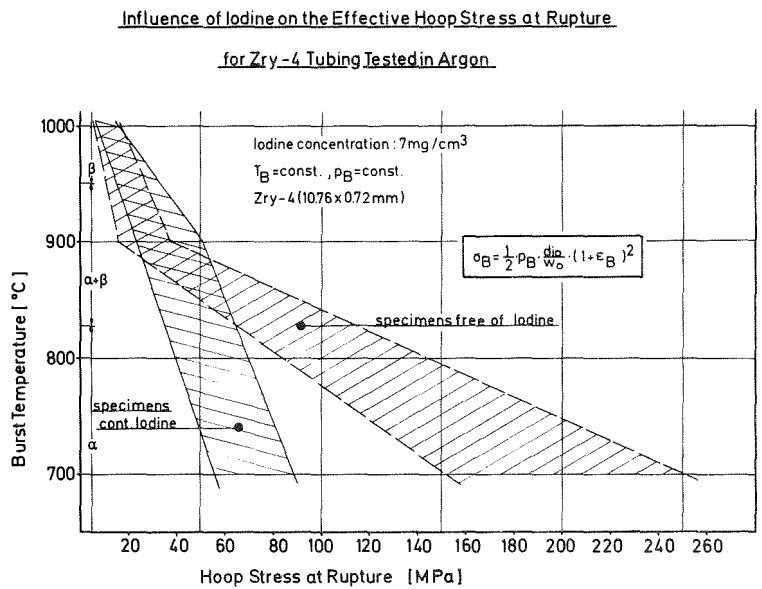


Fig. 7: Effective hoop stress at rupture for as-received and pre-oxidized Zry-4 tubing vs burst temperature; influence of iodine on the stress at rupture (isothermal, isobaric creep rupture experiments).

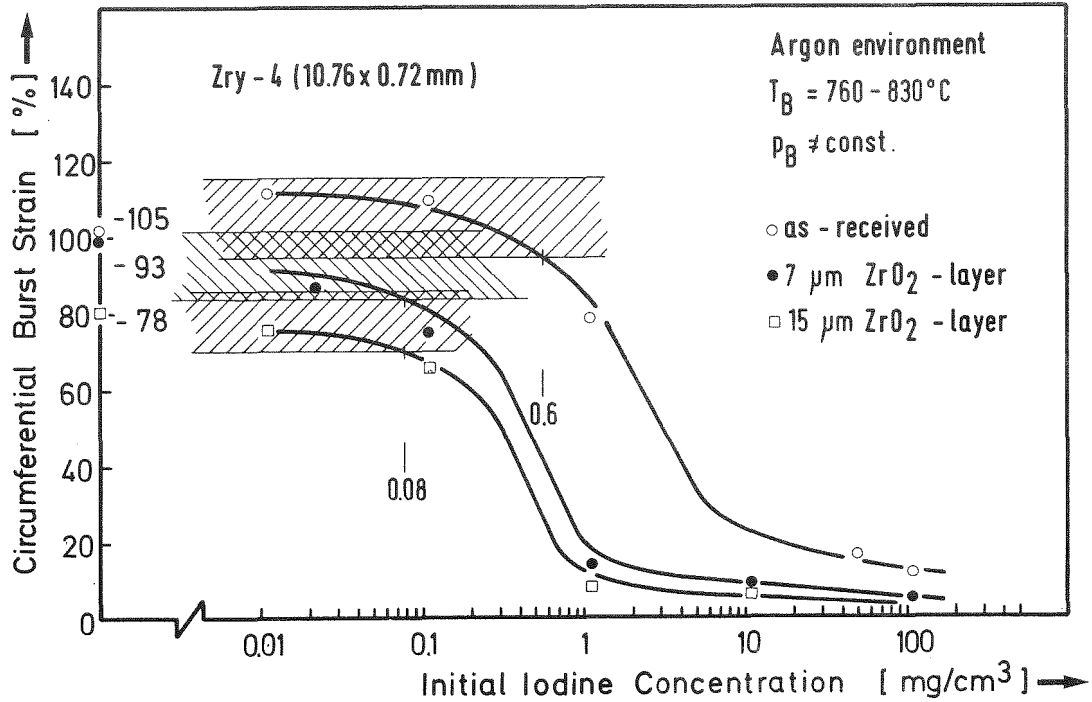


Fig. 8: Influence of the initial iodine concentration on the circumferential burst strain of as-received and pre-oxidized Zry-4 tubing at burst temperatures between 760 and 830°C.

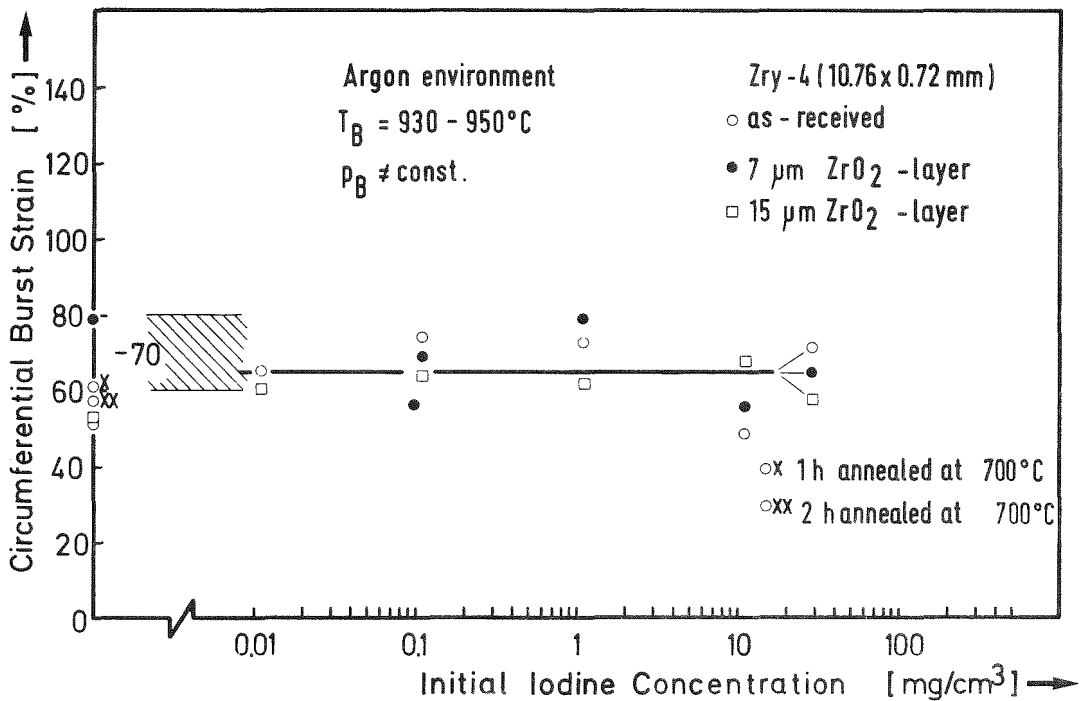


Fig. 9: Influence of the initial iodine concentration on the circumferential burst strain of as-received and pre-oxidized Zry-4 tubing at burst temperatures between 930 and 950°C.

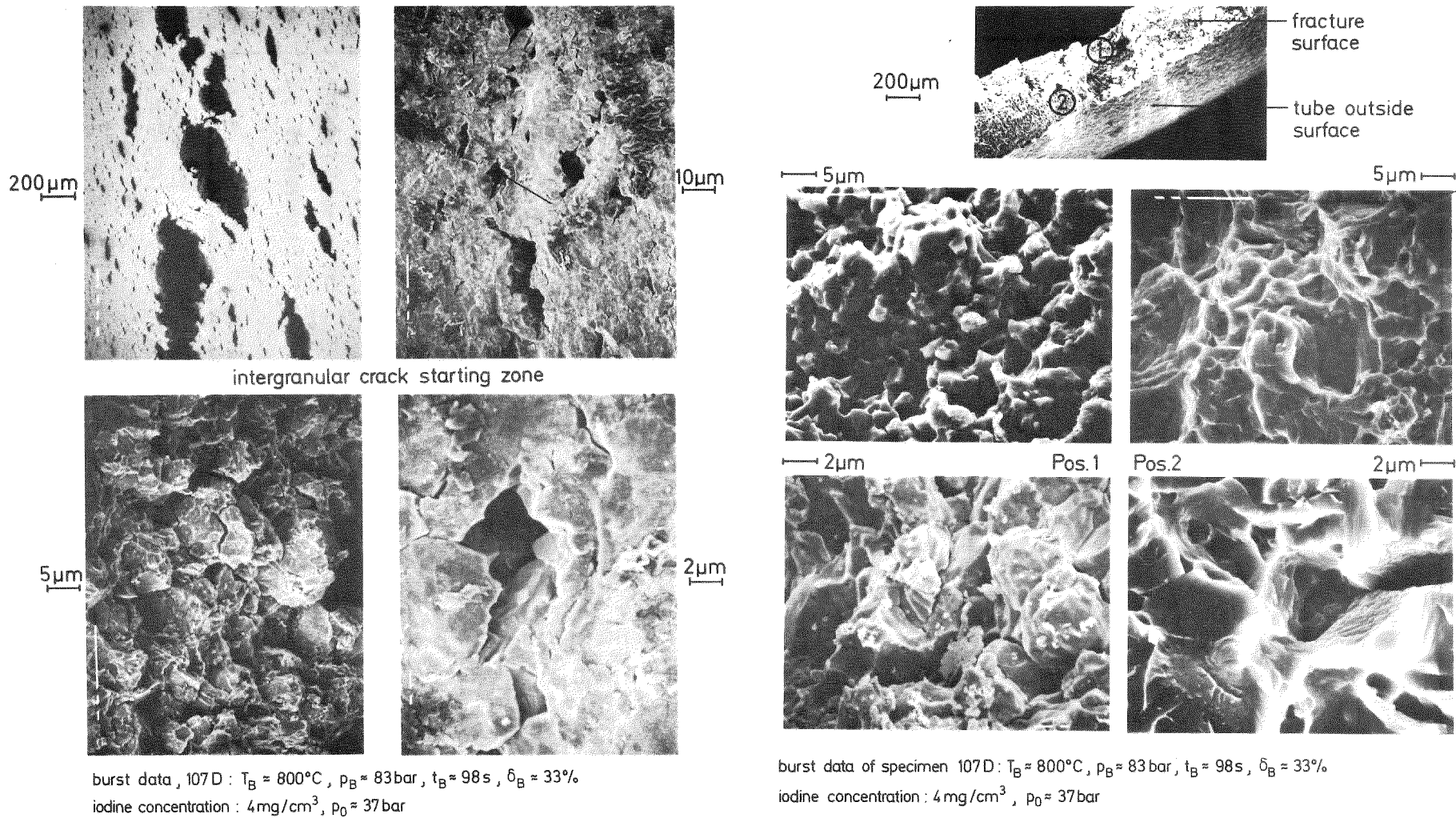


Fig.10: Scanning electron micrographs of the cladding tube inside surface and fracture surface of as-received Zry-4 tubing after failure under argon and iodine-gas pressurization.