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High burnup effects on fuel behaviour under accident conditions: the tests CABRI REP-Na

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Abstract

A large, performance based, knowledge and experience in the field of nuclear fuel behaviour is available for nominal operation conditions. The database is continuously completed and precursor assembly irradiations are performed for testing of new materials and innovative designs. This procedure produces data and arguments to extend licencing limits in the permanent research for economic competitiveness. A similar effort must be devoted to the establishment of a database for fuel behaviour under off-normal and accident conditions. In particular, special attention must be given to the so-called design-basis-accident (DBA) conditions. Safety criteria are formulated for these situations and must be respected without consideration of the occurrence probability and the risk associated to the accident situation. The introduction of MOX fuel into the cores of light water reactors and the steadily increasing goal burnup of the fuel call for research work, both experimental and analytical, in the field of fuel response to DBA conditions. In 1992, a significant programme step, CABRI REP-Na, has been launched by the French Nuclear Safety and Protection Institute (IPSN) in the field of the reactivity initiated accident (RIA). After performing the nine experiments of the initial test matrix it can be concluded that important new findings have been evidenced. High burnup clad corrosion and the associated degradation of the mechanical properties of the ZIRCALOY4 clad is one of the key phenomena of the fuel behaviour under accident conditions. Equally important is the evidence that transient, dynamic fission gas effects resulting from the close to adiabatic heating introduces a new explosive loading mechanism which may lead to clad rupture under RIA conditions, especially in the case of heterogeneous MOX fuel. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

The optimized use of nuclear fuel in pressurised water reactors (PWRs), and particularly the economic aspects of the reactor core management, entice the nuclear industry to change significant parameters of the nuclear reactor operating mode. Relying on very encouraging experience feedback concerning fuel behaviour under normal operating conditions, Electricité de France (EDF), the French electrical energy utility, has intro-

duced: the increase of the UO_2 fuel discharge burnup (from 33 000 to 47 000 MWd/t by mean assembly), the load follow operation (power variations according to the electrical grid requirements), as well as a new fuel, the MOX (mixture of uranium and plutonium oxides).

However, a study of the fuel behaviour under design basis accident conditions was not conducted for the increased discharge burnups. This particularly relates to the reactivity initiated accident (RIA) for which the postulated initiator is the ejection of a control rod bundle. For this accident, the main safety criteria currently in effect and intended to prevent accidental fuel dispersion, limit the energy injected during the accidental transient condition to 230 cal/g for fresh fuel and 200 cal/g for irradiated fuel.

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The EDF plan to request a new authorisation for burnup increase from 47 000 to 52 000 MWd/t (megawatt-day per ton of fuel) has led the safety authority to ask EDF to perform research on the behaviour of PWR fuel at high burnup in order to reassess the criteria and to evaluate the impact of the new reactor core managements. The IPSN (French Nuclear Safety and Protection Institute) was interested in participating to this programme.

The IPSN Department for Safety Research (DRS) was entrusted with this research programme through co-operative IPSN/EDF action, considering its competence as well as its unique experimental facilities.

2. Purpose of the tests

The postulated initiator of the PWR design basis reactivity accident is the ejection of a control rod bundle under the effect of the system pressure following a control rod housing rupture. The reactor's hot standby (280°C, 155 bar) was defined as an aggravating situation for this accident. The ejection of the control rods would lead to a temporary supercriticality and to a transient increase of the nuclear power in a group of fuel assemblies in the vicinity of the ejected bundle.

The danger associated to the reactivity accident power excursion resides in the rupture of the fuel rod cladding, followed by fuel dispersion that could finally lead to a steam explosion, the scattering of radioactive material and/or the loss of part of the reactor's core cooling possibility.

The CABRI REP-Na programme intends to study the early phase of the physical phenomena and the key mechanisms of the RIA transient. It mainly concerns the changes of the fuel (fissile material and cladding) induced by irradiation up to high burnup. Abrupt fuel overheating produces a mechanical interaction (Pellet clad mechanical interaction, PCMI) which reaches its maximum level in the near adiabatic phase, before the cladding temperature increases by thermal conduction. In a second phase, the cladding rapidly overheats and approaches the conditions to reach the critical heat flux (departure from nucleate boiling, DNB).

Three complementary parts characterize the IPSN research RIA programme for high burnup fuel:

- Global experiments in the sodium test loop of the CABRI reactor,
- Development of the transient thermo-mechanical fuel behaviour code SCANAIR,
- Measurement of specific high burnup properties for use in SCANAIR.

The characteristics of the sodium coolant allow to study the early PCMI phase of the transient sequence of events, i.e., the PCMI loading phase. As already mentioned, the evaluation of the failure risk during this early

phase represented the major objective at the time when the programme was launched. From the beginning it was clear that this approach would not solve all the aspects of the high burnup issue, in particular, the failure risk related to DNB and post failure phenomena in the pressurized water environment.

The development of the SCANAIR code aimed at both, preparation and interpretation of REP-Na experiments and transposition to the reactor case.

Finally, three major separate effect programs have been adopted in order to understand the integral test results from the CABRI REP test program:

PROMETRA: an out-of-pile test program to measure mechanical properties of high burnup cladding under transient temperature and loading conditions.

PATRICIA: the determination of the cladding to water heat transfer correlation during rapid power transients.

SILENE: quantification of the kinetics of fission gas behaviour in the fuel during rapid power transients.

The data from these separate effect test programs are used to improve the modelling of the physical phenomena in the SCANAIR code. SCANAIR will then be validated against the REP-Na integral test data before being used for evaluating rapid reactivity transients in power reactors.

3. Test matrix

At the beginning of the programme, the fuel rod burnup and the transient energy deposition were the only parameters of the test matrix. Soon, through experiment feedback, other important parameters were identified such as the amplitude and the fine structure of corrosion as well as the energy injection kinetics (width of the power-pulse). Finally, nine tests, six UO₂ tests and three MOX tests (Table 1) were programmed.

4. Fuel evolution under reactor operation

The power operation of the fuel inside the reactor leads to important cladding and fissile pellet modifications.

Firstly, *the cladding* is submitted to a creep induced plastic strain under the effect of the PWR primary system pressure, 155 bar, and is plated against the fuel. This process of fuel/cladding 'gap closure' is actually ended around the middle of the second cycle (~1.5 years, ~20 000 MWd/t).

Henceforth, the fuel is in direct contact with its cladding and any rapid fuel expansion, with kinetics

Table 1
CABRI REP-Na test matrix and main results

Test (carried out)	Tested rod	Pulse (ms)	Energy at pulse end (cal/g)	Corrosion (μ)	RIM (μ)	Results and remarks
Na-1 (11/93)	EDF Grav5c, span 5, 4.5% U5, 64 GWd/t	9.5	110 (at 0.4 s) (460 J/g)	80, important initial spalling	200	Brittle failure at $H = 30$ cal/g, $H_{\max} = 115$ cal/g; fuel dispersion: 6 g including particles other than RIM, sodium pressure peaks
Na-2 (6-94)	BR3, 6.85% U5, 33 GWd/t	9.5	211 (at 0.4 s) (882 J/g)	4	–	No rupture, $\Delta\phi/\phi$ (max): 3.5% average value, FGR/5.54%
Na-3 ^a (10/94)	EDF, 4.5% U5, 53 GWd/t	9.5	120 (at 0.4 s) (502 J/g)	40	100	No rupture, $\Delta\phi/\phi$ (max): 2% max, FGR/13.7%
Na-4 (7/95)	EDF Grav5c, span 5, 4.5% U5, 62 GWd/t	75	97 (at 1.2 s) (404 J/g)	80, no initial spalling	200	No rupture, transient spalling, $\Delta\phi/\phi$ (max): 0.4% average value, FGR/8.3%
Na-5 (5/95)	EDF Grav5c, span 2, 4.5% U5, 64 GWd/t	9.5	105 (at 0.4 s) (439 J/g)	20	200	No rupture, $\Delta\phi/\phi$ (max): 1% max, FGR/15.1%
Na-6 (3/96)	EDF MOX, 3c, span 5, 47 GWd/t	~35	165 (at 1.2 s) (690 J/g)	40	–	No rupture, $\Delta\phi/\phi$ (max): 3.2% max, FGR/21.6%
Na-7 (2/97)	EDF MOX, 4c, span 5, 55 GWd/t	~40	175 (at 1.2 s) (732 J/g)	50	–	Rupture at 120 cal/g, pressure peaks, examination currently carried out
Na-9 ^a (4/97)	EDF MOX, 2c, span 5, 28 GWj/t	~40	228 (at 1.2 s) (953 J/g)	<20	–	No rupture, examination currently carried out
Na-8 (7/97)	Grav 5c, span 5, 4.5% U5, 60 GWd/t	75	106 (at 1.2 s) (443 J/g)	130, cladding presenting spalling	200	Rupture at 83 cal/g (or lower b) gas blow-out, no fuel dispersion, examination currently carried out

^a Improved cladding i.e. low tin.

^b Pertinence of signals at 45 cal/g to be investigated by post-test examinations.

exceeding the creep velocity of the clad material produces a strong mechanical interaction.

During the whole irradiation cycle, a corrosion process in the reactor forms a layer of zirconium oxide (ZrO_2) on the cladding external surface and introduces into the metal an important amount of hydrogen, proportional to the zirconia thickness. At high burnup (>50 000 MWd/t), it is possible to reach or even pass, for Zircaloy4 cladding, a zirconia thickness of 100 μm and ~ 800 ppm of hydrogen (Fig. 1).

An aggravating aspect of corrosion is produced when the oxide layer 'spalls' locally. The absence of oxide then produces a cold point towards which the hydrogen migrates and an accumulation of hydrides is formed at the cladding's surface (*blister*). The presence of a blister can lead locally to the total loss of the cladding's ductility (Fig. 2).

At very high burnup (~ 60 000 MWd/t) a very high degree of spalling was observed on certain assemblies fitted with standard, unimproved cladding. The new cladding materials, now introduced in the EDF plants, should not spall at this level of burnup; however, the precise mechanism of this phenomenon is not yet understood.

The *fuel pellets* are subject to a deep transformation under irradiation: cracking, accumulation of fission products and swelling. Among the fission products, the gaseous elements (Xe and Kr), retained under the form of nanometric bubbles on intra-, or inter-granular sites

in the fuel, play a predominant role during fuel rapid overheating. At 60 000 MWd/t their STP volume is equivalent to about $1.6 \text{ cm}^3/\text{g}$, 16 times the volume of the fuel. Increased under rapid overheating, this gaseous volume presents considerable swelling, fragmentation and dispersion potentials.

Beyond about 45 000 MWd/t a peripheral zone is created at the fuel surface through a neutronic effect. The characteristics of this zone are a high plutonium content generating a very high local burnup rate, a submicronic grain size as well as very important porosity ($\sim 20\%$). This width of the rim-zone is in the range of 200 μm . This structure formation is called 'rim effect' and represents a phenomenon characterizing highly irradiated fuel. Fundamental studies are currently being performed and aim at the clarification of the rim effect, in particular, the subdivision of the fuel grains into submicronic fragments.

The *MOX fuel* shows specific differences compared to the classical UO_2 fuel. The MOX fissile material is plutonium. During the preparation of the MOX following the MIMAS procedure, a mother blend of uranium/plutonium mixed oxide is added to natural or depleted uranium oxide. Pelletizing and sintering of this powder mixture create an heterogeneous final product, with mixed oxide $(UPu)O_2$ agglomerates or clusters imbedded in the matrix of natural UO_2 . During reactor irradiation, the fission occurs in the clusters which reach very high burnup rates compared to the nominal mean

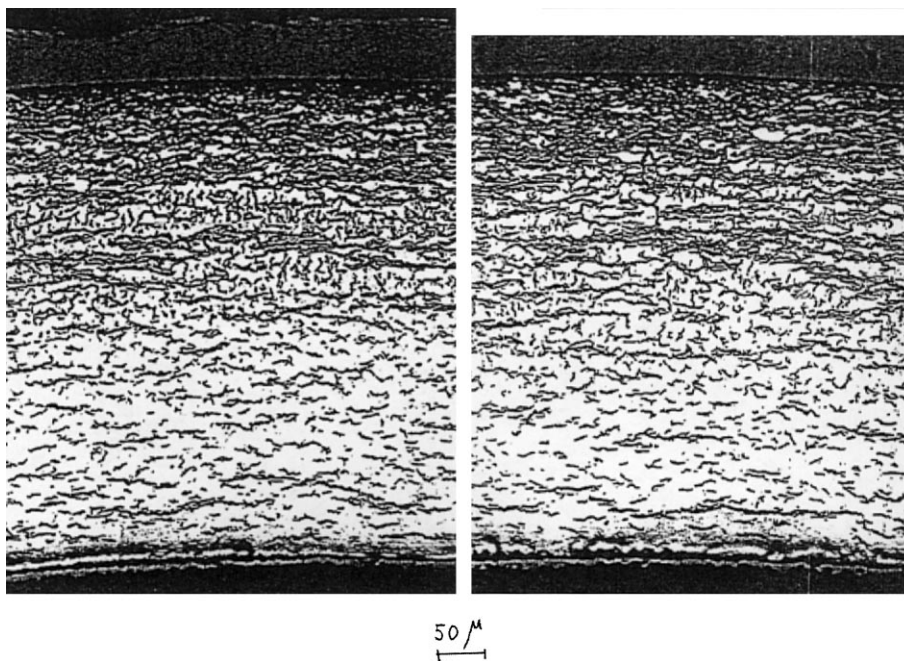


Fig. 1. Metallographic cut of the REP-Na4 rod cladding after CABRI test. The hydride plates are revealed by etching. The upper dark layer represents the ZrO_2 oxide layer with a thickness of about 80 μm (left). Large transient spalling occurred under this test (right).

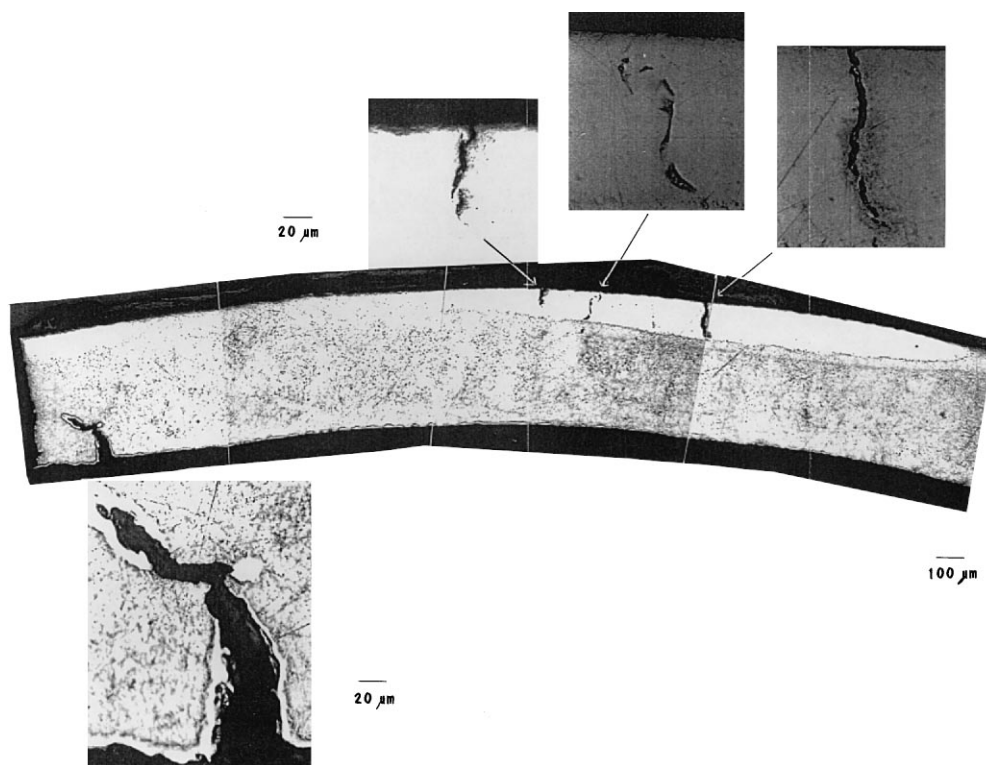


Fig. 2. The hydrogen migration towards a cold point of the cladding aggravates its embrittlement under irradiation in the reactor. The pre-existing cracking of the hydride phase can represent an incipient rod failure location under reactivity transient conditions. The photograph shows one of the REP-Na1 cladding blisters which are most probably the cause of the multiple ruptures during the test.

burnup (matrix average plus clusters). The structure and composition of the irradiated MOX clusters can be compared to those of the UO_2 fuel RIM, however, with a four to five times higher volume fraction.

5. Test results and phenomenological understanding

The main results currently available are presented in Table 1. The cladding rupture observed in the REP-Na1, Na7 and Na8 tests are remarkable and spectacular and contribute to the understanding of the failure mode and to the formulation of a failure criterion. The non-failure tests have produced valuable quantitative and qualitative results, for the understanding of physical mechanisms, and therefore for the development and validation of the SCANAIR code.

5.1. Mechanism and mode of rupture

In the first test of the matrix, the REP-Na1 test, a very early cladding rupture was recorded. This unexpected result was followed by a detailed metallographic examination programme and a series of calculations to

identify the rupture conditions as well as its characteristics in order to conclude on the failure's cause and mechanism. The rupture aspect (Fig. 3) shows a purely brittle fracture and the CABRI reactor measurements locate it at an instant which is described by the SCANAIR code calculation as a state where the RIM zone alone exceeds the nominal operating conditions. Details of the metallographic cuts show the presence of hydride accumulations (blisters) in the cladding. It is, therefore, possible to conclude that the rupture originated from a mechanical interaction due to the RIM effect, assisted by cladding embrittlement due to the presence of hydride (hydride assisted PCMI failure). It was demonstrated through the satisfactory rod behaviour during other UO_2 REP-Na matrix tests, that in case of moderate clad corrosion, the rod sustains PCMI charging even at a burnup greater than 60 000 MWd/t (REP-Na4, REP-Na5).

A second cladding rupture was observed in the REP-Na7 test, MOX test at 55 000 MWd/t. Examination of the tested rod is still to be carried out. However, a rupture mechanism such as during REP-Na1 appears unlikely, given the absence of spalling of the oxide layer. The sound cladding condition leads to the conclusion

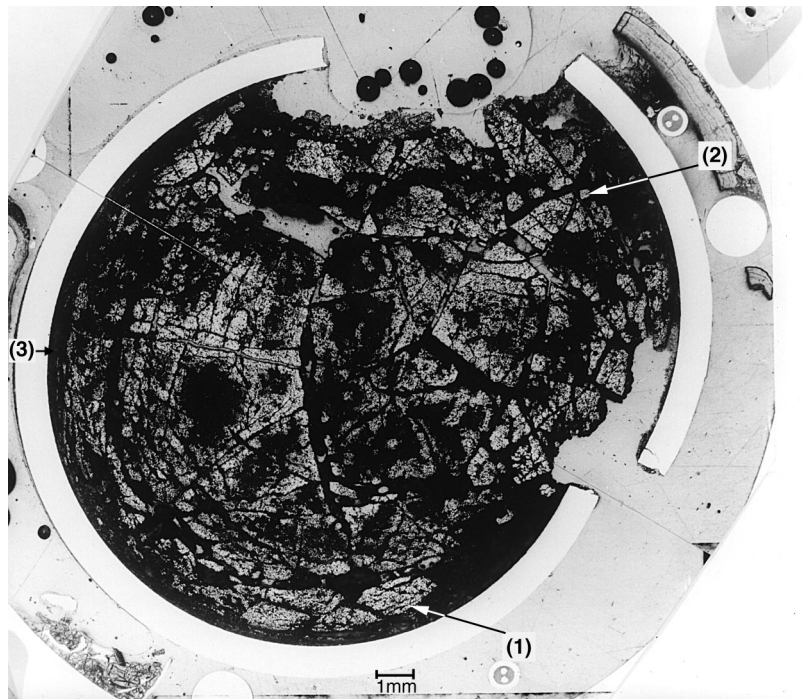


Fig. 3. Metallographic section (X4) of the REP-Na1 rod after test. The brittle aspect of the ruptures (perpendicular cracking) and the fuel fragmentation constitute outstanding facts of this observation. The numbers indicate locations of detailed examination: (1) RIM structure, (2) fragmented fuel, (3) intact cladding.

that the rupture mechanism is dominated by the contribution of fission gas to transient fuel swelling that could, in the case of MOX, be more important than for the UO_2 (also in discussion in Section 7). An examination programme of REP-Na 7 has been formulated with the aim to identify the rupture mechanism.

The cause and conditions of the rupture observed during the REP-Na8 test are currently the subject of investigations.

5.2. Cladding plastic strain

The fuel thermal expansion and the transient swelling are the two main factors contributing to cladding loading and cladding rupture occurs if the ultimate yield strength and the cladding's plastic strain capability are exceeded. In the CABRI tests without rupture, cladding strain is measured by profilometry. These examination results constitute valuable data for validation of the thermo-mechanical model of the SCANAIR code. Fig. 4 shows the REP-Na2 profilometry.

5.3. Fission gas driven fuel fragmentation

In all the CABRI REP-Na tests with significant plastic strain, a large fuel fragmentation zone is ob-

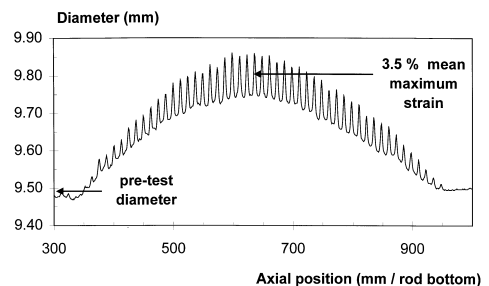


Fig. 4. REP-Na2 diametral straining over the length of the test rod. The shape of the curve traces the axial power distribution in CABRI. The fine structure demonstrates each fuel pellets strain (hour glass type). This type of result provides precious elements for the SCANAIR code validation.

served (Fig. 5). This fragmentation results from fuel grain decohesion under the effect of the fission gas fraction accumulated in micro bubbles in the intergranular zones. The bursting of the gas bubbles under the effect of fast transient heating leads to instantaneous increase of the fuel/clad contact pressure (PCMI) at high burnup when the fuel/clad gap is closed and also represents the driving force for grain separation. During the cooling process, when the cladding's permanent strain

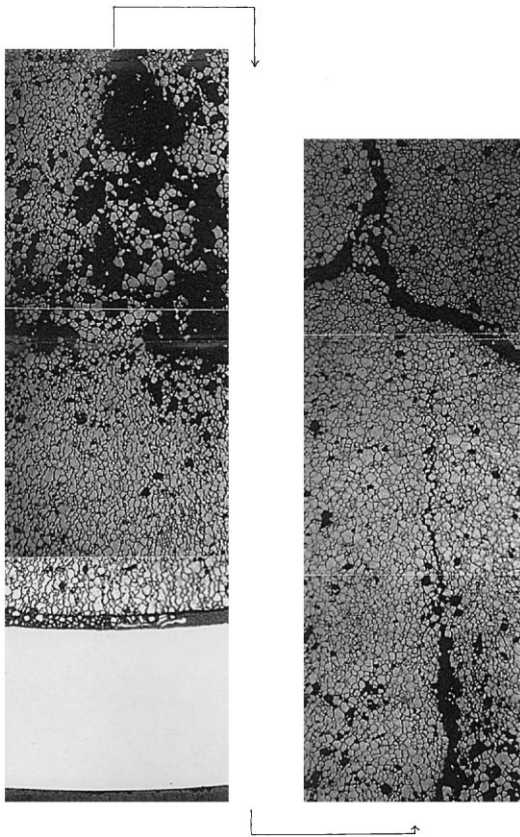


Fig. 5. Metallographic section of the REP-Na2 rod after CABRI testing. The grain decohesion and the loss of a large number of grains during preparation of the metallography demonstrate the fuel fragmentation. The fission gases accumulated in the fuel grain boundaries are the driving force of the transient fragmentation.

offers to the fuel an additional free volume, the grain-boundaries can open, producing the structure which is observed by ceramographic examination. This phenomenon of gas driven fuel expansion is a characteristic of the high burnup and can contribute, during cladding rupture, to the associated dispersion of fuel through the dynamics of the pressure relieve. The dispersion potential and its consequences are amplified in the RIM and in the MOX clusters due to the high local gas concentration and by the potential of emission of plutonium rich and submicronic particles.

5.4. Fission gas release

The fraction of fission gas, released during the test, is given in the results column of Table 1. Gases in intergranular sites alone are released in the very short RIA transient time. The activation of diffusion mechanisms releasing intragranular gases (majority fraction) only

takes place for very high energy deposition (~ 200 cal/g). The amounts of released gas are significant, they increase the internal pressure of the rods not ruptured during the accident's first phase. The risk of creep-induced late rupture increases if the internal pressure exceeds the system's pressure. The release measurement results allow for validation of the gas behaviour models and to quantify the thermal and mechanical effects of the fission gas during the RIA transient [1].

5.5. Transient spalling

In several, highly corroded, REP-Na tests, a transient spalling of the oxide layer has been observed. In the very short RIA transient time, an important part of the oxide layer is detached from the cladding's metallic surface. In the case of an accident, this phenomenon introduces, even in the absence of cladding rupture, an important amount of debris into the reactor's coolant channels in a very short time and creates a risk of flow reduction and clogging. In addition, the cladding/water heat transfer could be reduced in the crucial phase of the accidental scenario when the fuel approaches critical thermal flux conditions and spalling oxide tiles influence the cooling conditions.

6. National and international co-operation

In this programme [2], the IPSN co-operates with several partners. EDF and FRAMATOME's active participation provides a stimulating complementarity [3]. The services and assessments issued from numerous laboratories of the CEA/DRN (neutronics, fuel codes, support tests, radiometallurgy) are essential to the programme's progress [4]. JAERI (Japan) is the senior international partner. In its NSRR test reactor, JAERI has been conducting RIA tests for many years and has also been observing high burnup cladding ruptures strongly associated to the cladding's corrosion level. Other observations (FGR, $\Delta\phi/\phi$) confirm and complete the REP-Na programme results [5]. NSI-KI (Kurchatov Institute, Russia) is a contractual partner and transmits, in the scope of the contract, its theoretical as well as experimental know-how (RIA programme in the IGR reactor in Kazakhstan) [6]. US-NRC has signed a co-operation agreement in June 1995 enabling it to access the CABRI REP-Na programme results as well as the support tests. Frequent and fruitful discussions, within the scope of this agreement, include American specialists from research and industry (ANL, INEL, BNL, PNL and EPRI) [7,8]. OECD-NEA has finally become the meeting ground for contacts with numerous other countries. The 'CSNI Specialist Meeting' in Cadarache, in September 1995 assembled more than 125 experts from 15 countries and this conference's proceedings [9]

provide a very complete view of the problematic of the light water reactor reactivity accident.

7. Discussion, conclusion and perspectives

Fig. 6 shows the RIA test database in terms of either maximum or failure enthalpy as a function of burnup of the test rods and underlines the contribution of the CABRI tests in the high burnup range.

This compilation, established by US-NRC, presents a large number of tests that should be sufficient to understand and validate the calculation codes. The following list indicates briefly the major non-prototypical conditions of the tests compared to RIA conditions in a PWR:

- CABRI: sodium coolant, low pressure: sodium cooling properties keep the clad temperatures low and low internal pressure mitigates the transient dynamic gas effect.
- NSRR: capsule tests, low pressure, low temperature, narrow pulse (~ 5 ms): the cladding remains during a significant time period below the brittle to ductile transition-temperature, the radial fuel temperature profile is anomalously peaking and critical heat-flux conditions are inadequately simulated.
- IGR: capsule test without instrumentation, low pressure, very wide pulse (>500 ms), low temperature, imprecise energy deposition: the radial fuel temperature profile is too flat.

- CDC/PBF: fuel not representative of the PWRs, most tests carried out in capsule, low temperature, low pressure: low fission gas retention and fuel restructuring (central hole formation) due to high power level during pre-irradiation.

The main drawback of this representation (Fig. 6) is the fact that it does not allow to fully assess the influence of clad corrosion and/or pulse width which are clearly identified as the high burnup key parameters. Nevertheless, examination of the data in Table 1 and Fig. 6 suggest that the fuel failure enthalpy is reduced significantly with fuel burnup. In addition, the REP-NaI test underlines the unacceptable performance when oxide spalling and blisters are present in the cladding, i.e., a power excursion of low amplitude can result in fuel rod rupture. The original safety criteria of 230 (fresh fuel) and 200 (irradiated fuel) cal/g, presently being used, do not appear to be applicable to high burnup fuel.

It is suggested that the reduction in failure enthalpy with burnup, both for UO_2 and MOX, is due to the formation of very high burnup regions in each fuel type, i.e., the RIM structure in UO_2 and the clusters high in Pu (fissile material) in MOX. These very high burnup regions result in high concentrations of intergranular fission gas which produces fuel swelling during the transient and acts as an additional loading mechanism on the cladding. In the case of the MOX fuel, the high Pu clusters act similar to the RIM at high burnup with approximately five times the volume fraction of material

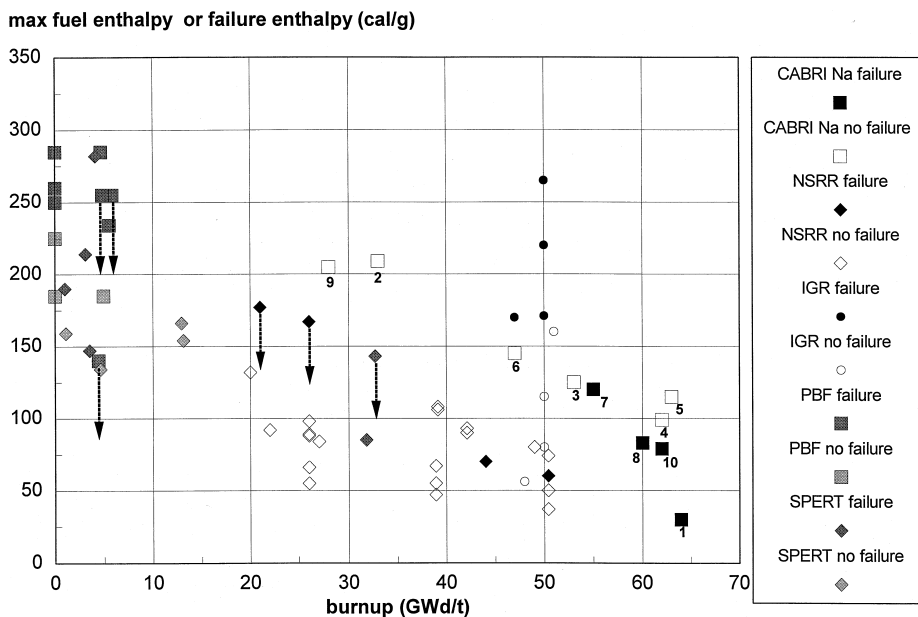


Fig. 6. A large number of experimental simulations of reactivity accidents has been carried out by several countries (US, Japan, Russia and France). The CABRI contribution includes all tests with burnup rates superior to 50 000 MWd/t as well as all of the irradiated MOX tests. The variety of results underlines the need to perform tests in realistic, representative conditions.

as the RIM in UO_2 . This results in significantly higher loading for MOX rods than the UO_2 rods and increased failure potential at similar burnups and energy deposition levels as evidenced by the REP-Na7 test result, i.e., low failure threshold without hydride blisters.

It is further suggested that the increase in FGR and resulting high pressures with burnup observed in Table 1 may result in rod ballooning and rupture for rods that reach critical heat flux (CHF) during an RIA in PWR. This is not evident from RIA tests to date because they have all been tested in either sodium coolant or under low temperature and low pressure conditions where CHF is not easily achieved.

The study of this phenomenology requires PWR representative conditions. The sodium channel conditions, in the current CABRI facility, do not allow to reach this representativeness of the reactor situation. The diagram presented in Fig. 7 shows the cladding temperature evolution calculated by SCANAIR under sodium and pressurized water conditions for compari-

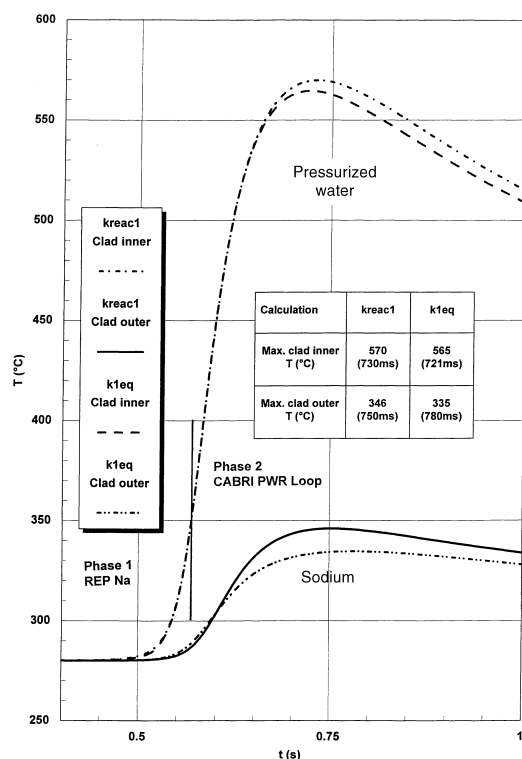


Fig. 7. The CABRI REP-Na tests were defined to study the PCMI phase of the accident phenomenology. Under sodium cooling conditions, the cladding temperatures remain comparatively low, as shown by the SCANAIR calculations. Under pressurized water cooling conditions, the departure from nucleate boiling leads to rapid clad overheating with risk of clad rupture by ballooning, as a consequence of the increase of the internal pressure by transient release of fission gas.

son. Only the phase 1 of the phenomenology could be studied by the REP-Na tests. The study of phase 2 requires experimental conditions which are representative of PWR conditions.

The installation into the CABRI facility of a pressurized water loop will enable the study of the whole spectrum of the accidental phenomenology (phases 1 and 2). The design and engineering work for this important transformation of the Cabri facility has been in progress for several years. The final decisions for this work should be made in 1999 and the first experiment of a programme with international co-operation is expected to be performed at the end of year 2003. This programme will provide, for the future fuel design, the experimental database for the assessment and updating of the burnup dependent safety criteria for the design basis reactivity accident.

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