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Nuclear Fuel

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ALTHOUGH SCK•CEN already could irradiate fuel in its high-flux BR2 Materials Testing Reactor (MTR) and could perform high-quality postirradiation examinations, it had not emphasized modelling. Presently, it is making a great effort to enhance its ability to model the fuel behaviour up to high burnup, under stationary and transient irradiation conditions, in view of the following considerations:

- the utilities pledge for improving the general availability of nuclear power plants in terms of load following and extended operation at reduced power, with subsequent restoration of nominal power;
- fuel designers and cladding manufacturers continuously explore new products (nuclear fuel, cladding materials, new concepts, etc.);
- economics and prudent use of natural resources provide strong incentives for extending the average burnup levels of fuel in commercial reactors.

These tendencies might impact fuel integrity and, accordingly, safety. The answer to these questions largely relies upon models containing many parameters, tuned to yield the experimental results, but with little understanding of the physical mechanisms involved. A strong correlation between thermal, mechanical, and chemical performances of the nuclear fuel makes the analysis even more complex. While integral experiments provide useful information for validating the models, there is a need for extending the knowledge of the underlying basic phenomena and for improving the models. This theoretical approach in turn requires a more sophisticated scientific analysis and more experimental data on separate effects, especially at high burnup (in excess of 60 GWd/Mt). These effects include thermal and nonthermal fission-gas releases, fuel swelling, fuel thermal conductivity, fuel specific heat, size (relocation) and thermal conductance of fuel-pellet gaps, fuel rim effects (thermal conductance and gas release), and chemical and mechanical fuel–cladding interactions.

Objective The present R&D programme aims at better predicting quantitatively the operational limits of nuclear fuel and at assessing the behaviour of fuel under incidental and accidental conditions. The ability to predict fuel behaviour at high burnup under stationary and transient conditions will increase both the safety and the efficiency of LWR fuel in general, and of MOX fuel in particular.

SCK•CEN wants to improve existing fuel modelling tools by looking more closely at improved modelling of fission-gas release, at improved modelling and more accurate parameters for thermal conductivity, at basic physical phenomena involved in fission-gas release, and at relevant experimental tools for studying fuel in incidental situations and even in accidental ones.

Modelling of fission-gas release in LWR fuel We began developing an improved fission-gas-release model in 1994. In a first step, we recapitulated the different mechanisms for fission-gas release in nuclear fuel. In a second, we made an overview of the relevant models, from which we proposed a new one.

The model we proposed for fission-gas release consists of two modules. The first, termed intragranular, tackles the transport of the fission gases in the grains of the UO_2 matrix. It incorporates three mechanisms: gas atom diffusion in the grain, resolution and trapping associated with intragranular bubbles, and resolution of fission products from bubbles situated at the grain faces into a resolution layer adjacent to the grain boundary. The second module, termed intergranular or macroscopic, handles the transport of the fission gases at the grain boundaries.

We implemented the microscopic and macroscopic module by means of a finite-element code, and compared the global model with experimental results. Without surprise, this exercise suggested further improvements, in accordance with the initial proposal. A discussion on the role of grain-boundary diffusion led us to introducing secondary phases at the grain boundary and to formulating additional improvements mathematically, in addition to developing an alternative macroscopic module.

The first improvement, namely the trapping and resolution of fission products at intergranular phases or traps (such as bubbles or metallic precipitates), is currently being implemented. We tested the precision of several routines by comparing their results with an analytical solution under particular conditions. We chose and implemented the routine D03PDF from the NAG library, and started to optimize the numerical parameters in the model, in terms of precision and calculation time.

From the experimental point of view, we defined a test matrix for validating the assumptions regarding the behaviour of fission products at the grain boundaries and for calibrating the final model. The first series of measurements will be performed at the Laboratory for High and Medium-level Activity (LHMA), whereas the second was already performed in the Halden BWR programme.

Perspectives for 1998 We will further optimize the numerical parameters in the fission-gas-release model and complete a parametric study to point out the dominant factors in the release of fission products. We will then implement the other improvements and compare the model with experimental results. We will also start implementing the alternative macroscopic module.

The validation of the underlying assumptions in the fission-gas-release model is planned to start in early 1998, while the calibration of the final model is planned for the end of 1998.

Thermal conductivity The temperature is in itself an important parameter for fuel-rod safety. Moreover, because it strongly influences many other fuel parameters (such as fission-gas release and pellet-cladding mechanical interaction), its accurate description is also indispensable for the description of these parameters. Our research programme, started in 1995, aims to improve the description of the thermal conductivity and hence the description of the temperature distribution in the fuel rod.

A theoretical analysis of various descriptions of the thermal conductivity reported in the literature revealed differences of 50 to 100°C in the prediction of the fuel central temperature, even at very low burnup. This analysis concluded that, though the theoretical description of the thermal conductivity need not be re-evaluated, the fitting parameters should be determined more accurately. In view of this, our research programme limits itself to the development of an innovative, more accurate measurement technique of the fuel thermal conductivity. This technique is applicable in both in-pile and out-of-pile conditions, so the programme considers both.

Our approach to determine the thermal properties of fuel, called power-variation method, applies periodic temperature variations to the fuel rod and measures the response on, for example, the fuel central temperature. In this way, it distinguishes between the thermal properties of the fuel and those of the gap and cladding. It requires such basic techniques as the instrumentation of the fuel rod and the ability to produce temperature variations in it.

For developing the power-variation method, we validated the heat-transport code HEATING for several cases of temperature variation in a fuel rod, by comparing its results with those of an analytical expression, for both in-pile and out-of-pile cases. The modelling of the fuel rod was limited to the fuel column only, while the cladding and gas gap were simulated by means of the boundary conditions. This approach was required by the lack of analytical expression describing the fuel rod in more than one material zone with a variable fuel power.

The irradiation programme BACCHANAL (Burnup ACCumulation for High-performing fuel ANALysis) aims at obtaining a set of fuel rods with several burnup values for subsequent use in the fuel research programme. It uses the CALLISTO loop. In this framework, we irradiated several fuel rods from BR3 in BR2 during three cycles. After the first and second cycles, we unloaded the fuel rods and performed gamma-spectrometric measurements to determine the linear power of each rod. A comparison with neutronic calculations showed that the calculation of the relative power distribution was correct within 3%.

Perspectives for 1998 With the help of the code HEATING, we will investigate a realistic model of fuel rod under temperature variation and determine basic design parameters for an irradiation rig. As regards BACCHANAL, we will also irradiate nine fuel rods for five cycles.

Basic physical phenomena A better modelling of the fuel behaviour and, particularly, of the fission-gas release requires a better understanding of the basic physical phenomena involved. SCK•CEN currently studies two topics: gas bubbles and fuel grain boundaries.

The local analysis of retained noble gas in nuclear fuel is inherently difficult because the physical form under which it is stored varies from atomic dispersion to bubbles with a diameter of several hundred nanometres. The Electron Probe MicroAnalysis (EPMA) technique, applied since more than twenty years, yielded many important results, but its application to highly inhomogeneous materials is limited. We therefore developed a method to analyse a system of gas bubbles distributed in a solid matrix, based on multiple-voltage EPMA measurements combined with a Scanning Electron Microscopic (SEM) analysis of the bubble-size distribution. A more accurate analysis of gas retained in bubbles leads to a better insight in the gas release of power-bumped fuel rods, especially in relation to the role of grain boundaries.

One particularly interesting item in the safe exploitation of nuclear fuel at higher burnup is the behaviour of volatile fission products. The theoretical study presently conducted at SCK•CEN showed that some transport mechanisms are little established experimentally. The present study aims at a more thorough investigation of grain boundaries and the role they can play in the transport of fission products. In collaboration with the Université de Limoges and the EG Transuranium Institute, we initiated a surface analytical study of grain boundaries in both UO_2 and MOX fuel.

Postirradiation examination for fuel performance assessment Nuclear fuel vendors and operating nuclear power plants continuously endeavour towards higher performance, reliability, and safety. To that end, both current and new improved primary core components (such as fuel rods, control rods, and structural internals) have to be qualified. The qualification of these highly radioactive objects requires specialized infrastructure, qualified techniques, and appropriate expertise—all available at SCK•CEN's LHMA. The project aims to provide independent expert services based on our extensive experience in PostIrradiation Examinations (PIEs).

The CALLISTO programme, studying the overall behaviour of advanced PWR fuel, was successfully completed, with the remaining mechanical tensile tests on the fuel-rod claddings. It resulted in the following spin-offs:

- the STEAM programme analyses how irradiation influences the precipitate size distribution of a new type of Zircaloy cladding;
- the INCA programme investigates at the microstructural level the material characteristics leading to unusual corrosion or hydriding of a particular fuel-rod cladding;
- for the international FIGARO programme, we performed microstructural fuel examinations to elucidate the basic phenomenological mechanisms that govern the fission-gas behaviour in high-burnup PWR MOX fuel;
- microstructural examinations on the BR2 driver fuel addressed the presence of localized corrosion on some driver-fuel plates;
- we characterized and qualified former BR3 rods, to be irradiated in the CALLISTO loop of BR2 for future R&D programmes on the thermal characteristics of nuclear fuel and the fission-gas behaviour;
- we provided various hot-cell services on request of both internal and external clients.

Achievements The SEM examination of the size distribution and number density of intermetallic $Zr(Fe,Cr)_2$ precipitates in Zircaloy, a main parameter governing the alloy's corrosion and related hydrogen uptake, turned out to be a challenge: there was no straightforward sample-preparation procedure, giving rise to appropriate SEM images adequately reflecting the precipitates population. Although we successfully established an appropriate procedure on unirradiated samples, the transfer of this procedure to hot-cell conditions for application on irradiated specimens proved very difficult. Specifically, it was difficult to control the exact timing of the successive etching and rinsing steps. As a consequence, only part of the irradiated specimens could be examined successfully in 1997.

Optical microscopy, SEM, and EPMA of a particular CALLISTO fuel rod revealed its detailed corrosion and hydriding characteristics (extent, morphology, and chemistry).

In the framework of the FIGARO programme, four instrumented MOX fuel segments were transported from Halden to our laboratory. Out of each segment, we manufactured a specimen, conditioned it for microstructural examination, and performed a general optical microscopic survey before submitting it, next year, to elaborate EPMA and SEM research on the fuel.

Extended microstructural examinations of the grain structure and secondary-phase particles in the aluminium cladding of several BR2 driver-fuel plates allowed us to exclude metallurgical effects and intergranular attack as the possible corrosion mechanisms. Galvanic or deposition corrosion were assessed by EPMA. Despite a clear tin contamination on one particular sample, subsequent investigations did not confirm this mechanism as responsible for the localized corrosion phenomena of some BR2 driver-fuel plates. In view of the high costs involved in a systematic (destructive) microprobe investigation of a large number of samples, we are now running a nondestructive analysis campaign.

Thirty-seven former BR3 MOX fuel rods, with different plutonium contents and with a burnup ranging from 12 to 20 GWd/tM, have been characterized in view of their anticipated application in future fuel R&D programmes; they were found to be qualified for their further irradiation in the PWR-simulating CALLISTO loop in BR2. Out of them, fifteen rods selected for irradiation in BR2 cycles 1/97 and 2/97 were analysed nondestructively for the power experienced during their irradiation.

Several miscellaneous hot-cell services have been provided on behalf of both internal and external clients, among which SEM MOX powders characterization on behalf of Belgonucléaire, and SEM energy-dispersive spectrometry analysis of a vitrified waste sample corroded in situ (HADES) and of the laser traces on a Zircaloy tube on behalf of FBFC International.

Perspectives for 1998 We will complete the SEM evaluation of size distribution and number density of secondary precipitates in irradiated Zircalloy cladding, perform comprehensive EPMA and SEM research on the FIGARO fuel to elucidate the basic phenomenological mechanisms governing the fission-gas behaviour, carry out additional research to address further the localized corrosion of some BR2 driver-fuel plates, and evaluate, on behalf of MOX fuel vendors, fresh MOX fuel in terms of plutonium homogeneity and porosity characteristics. Several international programmes are also being negotiated with external customers.

Conceptual studies of incidental and accidental fuel experiments Increasing the allowed burnup of nuclear fuel can provide considerable savings on the fuel-cycle costs of nuclear power plants. However, one must investigate the behaviour of high-burnup fuel, particularly under severe transient conditions, to ascertain that the operation margins are safe. Such an investigation requires representative in-pile experiments. The project therefore aims to investigate the industrial needs for, assess the feasibility of, and develop in-pile rigs allowing severe tests on LWR fuels, with a high probability of fuel failure or even melting and in representative thermohydraulic conditions. It envisages two irradiation devices: COSAC for single-rod and DESTIN for multirod testing, respectively.

COSAC (Contaminant Operation SAfety Capsule) is inspired by our existing pressurized-water capsules. The modifications aim at reaching the prototype stage of the thermohydraulic irradiation conditions in the capsule, by adding a so-called jet-pump system that improves the circulation along the fuel rod, not unlike a BWR's primary water circulation. In addition, we will adapt the waste-water treatment of the device to allow destructive transients and the operation of leaking rods over periods of several days. In 1997, we started the concept definition and feasibility study.

DESTIN (DEStructive Testing IN-pile loop) is a compact integrated PWR loop installed in the central channel of BR2, which can accommodate 3×3 or 5×5 test fuel bundles. This modular installation can be used in a wide spectrum of experiments, with only adaptation of (relatively) inexpensive internal parts. Thanks to the unique geometry of the BR2 core, it can accept actual-size PWR fuel rods, beside refabricated and instrumented ones. It is currently targeted at three main fields of application: RIA-type tests, fuel-bundle degradation processes (for example, during LOCA events), and behaviour of severely degraded fuel. In-pile fuel quenching is an application where an integral experiment would help address still open questions.

In 1997, work on DESTIN focused on establishing contacts with potential partners. An international programme called FATE, aiming at the verification of the criteria for the high-burnup fuel in case of a LOCA, was proposed in cooperation with Belgonucléaire. The Forschungzentrum Karlsruhe / Institute für Reaktorsicherheit (Germany) and the Institut de Protection et Sûreté Nucléaire (France) showed an initial interest, to be further concretized.

Perspectives for 1998 The COSAC project will be proposed for first-phase approval to SCK-CEN's Committee for Examination of Experiments (CEE) in early 1998. This should lead to a decision to proceed with the detailed study.

The DESTIN team will continue its efforts to assess the needs for international safety experiments and to stimulate the interest of possible partners and sponsors, both in Belgium and abroad. The fifth framework programme of the EC should be the occasion to concretize project proposals.

Partners, sponsors, and customers

Scientific partner Belgonucléaire (BN)

Customers Belgonucléaire (BN) — British Nuclear Fuels Ltd (BNFL) — Franco-belge de fabrication de combustibles International (FBFC Int.) — Mitsubishi Heavy Industries Ltd (MHI) — Nuclear Fuel Industries Ltd (NFI)

Scientific output

Presentations delivered in 1997

Ph. BENOIT, A. DELBRASSINE, B. ARIEN, Ch. DE RAEDT, "Destin—An Integrated PWR Loop for the Destructive Testing of Fuel Bundles," TopFuel'97: Manchester, United Kingdom, June 9-11, 1997.

L. BORMS, P. DE REGGE, A. GYS, V. WILLEKENS, "Validation of a New Method for Burn-Up Determination by Means of Gamma-Spectroscopy," Annual meeting on Nuclear Technology, German Nuclear Society: Aachen, Germany, May 13-15, 1997.

J. C. GARCIA, P. VAN UFFELEN, "Analysis of Halden Fuel Experimental Data with the ENIGMA Code," Meeting of the Sociedad nuclear española 1997: La Corona, Spain, November 5-7, 1997.

V. SOBOLEV, P. VAN UFFELEN, K. VAN DER MEER, "Problems of In-Pile Measurement of the Nuclear Fuel Thermal Properties," TopFuel'97: Manchester, United Kingdom, June 9-11, 1997.

Y. VANDERBORCK, L. MERTENS, J. DEKEYSER, L. SANNEN, "Fuel R&D International Programmes, a Way to Demonstrate Future Fuel Performances," TopFuel'97: Manchester, United Kingdom, June 9-11, 1997.

M. VERWERFT, C. MARTENS, L. SANNEN, A. DELBRASSINE, "Quantifying Xe in Nuclear Fuel: (Im)Possible by Microprobe Techniques?," Topical day on Fuel Behaviour: SCK•CEN, Mol, Belgium, November 5, 1997.

Thesis published in 1997

D. SCHROYEN, "Validation du code HEATING pour une pastille de combustible soumise à des flux sinusoïdaux," final-year project, Industrial Engineering (Institut Supérieur Industriel de Bruxelles), September 1997.

Reports published in 1997

P. VAN UFFELEN, "An overview of fission product release mechanisms," internal SCK•CEN report (1997). R-3144.

Ten reports were also issued to our customers. They are commercial and confidential, and may not be released to third parties.