Reactivity and Neutron Emission Measurements of Burnt PWR Fuel Rod Samples in LWR-PROTEUS Phase II

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Measurements have been made of the reactivity effects and the neutron emission rates of uranium oxide and mixed oxide burnt fuel samples having a wide range of burnup values and coming from a Pressurised Water Reactor (PWR). The reactivity measurements have been made in a PWR lattice moderated in turn with: water, a water and heavy water mixture, and water containing boron. An interesting relationship has been found between the neutron emission rate and the measured reactivity.

KEYWORDS: burnt fuel, zero-energy reactor, reactivity measurement, neutron emission measurement

1. Introduction

Optimised fuel cycle costs, improved fuel utilisation and reduced spent fuel inventories are important goals in the present day design and operation of nuclear power plants. One facet of this optimisation process is to lengthen the time the fuel remains in the reactor, i.e. to increase the burnup. This is possible with modern fuel because of the continual improvement in design, construction, and material compositions. In order to obtain higher burnup, the initial ²³⁵U-enrichment of the fuel is increased. In fact, the current tendency of fuel management and core design is to increase the initial ²³⁵U enrichment from about 4% to more than 4.5%, and the batch discharge burnup level is now typically about 50 GWd/t (instead of 30-40 GWd/t).

Fuel rods with average burnup values beyond 50 GWd/t are characterised by relatively large amounts of fission products and a high abundance of major and minor actinides in a degraded uranium oxide matrix. Moreover, the composition of actinides and fission products becomes ever more complex at increasing burnup levels. Of particular interest is the change in the reactivity of the fuel as a function of burnup and the capability of modern codes to predict this change. In addition, the neutron emission from burnt fuel has important implications for the design of transport and storage facilities.

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2. LWR-PROTEUS Phase II

PROTEUS [1] is a zero-power research reactor that has been operated at the Paul Scherrer Institute (PSI) since 1968. In broad terms, the reactor consists of an annular block of graphite with a 1.23 m diameter central cavity to contain the system under investigation. All the instrumentation sensors and control systems are contained within the outer graphite region. The ongoing LWR-PROTEUS Phase II programme [2] is part of a continuing collaboration between the Swiss nuclear power plants and PSI and is the framework for the study of five uranium oxide (UO₂) and three mixed oxide (MOX) burnt fuel rod samples. These were all fabricated by the same fuel vendor (*Framatome ANP*) and discharged from the same Swiss Pressurised Water Reactor (PWR) (*Kernkraftwerk Gösgen-Däniken*).

Fuel with a wide range of burnup levels (up to well over 50 GWd/t) is under investigation, and this will be extended to even higher values. Furthermore, two Boiling Water Reactor (BWR) samples will be added to the list. The experimental investigations cover the following three independent but complementary approaches: post-irradiation examinations, chemical assays, and reactivity and neutron emission measurements (in PROTEUS). The present paper reports principally on the reactivity effect investigations and neutron emission measurements carried out with the five UO_2 and three MOX fuel rod samples available to date.

3. Investigations of Reactivity Effects

3.1 Fuel Configuration

The LWR-PROTEUS facility has a 160 mm x 160 mm central PWR test zone of fresh, fulllength UO₂ fuel rods of 4.3w%²³⁵U-enrichment. The Gösgen-Däniken nuclear power station supplied this fuel, with support from their current fuel supplier Framatome ANP. Analogously, the Leibstadt nuclear power station and Westinghouse Atom have provided the eight SVEA96 OPTIMA2 UO₂ BWR fuel assemblies placed in the outer 450 mm x 450 mm region of the test tank. The fuel configuration is shown in Figure 1. The two regions of fuel of different type reflect the collaborative nature of the LWR-PROTEUS programme, the first phase of which was devoted to the detailed investigation of fresh, highly heterogeneous BWR fuel assemblies [3]. A guide tube in the centre of the PWR region facilitates the insertion of test samples into the core centre. At the sample position, the neutron spectrum is representative of a full PWR core. A range of neutron-spectrum conditions has been investigated by changing the moderator in the PWR region: full-density H₂O, H₂O/D₂O mixture, and borated H₂O.

3.2 Transport and Measurement

Each of the 400 mm long burnt fuel samples has been overcanned in the PSI Hot Laboratory with a special Zircaloy cladding, which was welded using a certified procedure to guarantee leak tightness and freedom from contamination. A combined transport flask and sample changer (Figure 2) is used for the simultaneous transfer of up to four burnt fuel samples from the Hot Laboratory for their subsequent measurement in PROTEUS. The sample changer has a control unit that also acts as a data logger during the measurements. The transport flask remains in place above the reactor throughout the reactivity measurements, and serves to move the samples in and out of the reactor during the experiments.

The reactivity difference between burnt fuel and fresh 3.5w% enriched UO₂ fuel, and its variation with burnup, is deduced from the effect on the neutron balance of the PROTEUS

reactor of introducing 400 mm long test samples, one at a time, into the core centre. Two methods of reactivity measurement are used, compensation and inverse kinetics. Each derives a reactivity value by analysing the change in the reactor state when a rod sample is inserted, but each in a different way.



Figure 1 A View of the Fuel Configuration for LWR-PROTEUS Phase II.

3.3 The Compensation Method

Figure 2 The 700 mm diameter Transport Flask/Sample Changer

For the compensation method, the reactor control rods are used to keep the reactor power constant by compensating for the reactivity change caused by the introduction of a sample. In particular, an automatic fine control rod is used (known as the Autorod). This rod is controlled by a servo feedback system, which compensates automatically for small reactivity changes. The positions of all of the control rods are recorded every second by a data logger that is part of the sample changer control system.





Figure 3: Autorod Movement during a Compensation Measurement.

Figure 3 shows the movements of the automatic control rod during a typical measurement sequence. A reference sample (usually 3.5w% UO₂) is inserted first, followed by the first burnt sample to be measured and then the reference sample again, and so on, usually repeated three times. After corrections for slight non-linearities, the movements of the control rods are indicative of the reactivities of the test samples. The difference in reactivity between the burnt sample and the reference is found by time-wise interpolation. This method of analysis has the advantage of cancelling any long-term drift in the state of the reactor. Because the samples emit significant numbers of neutrons, the measurements are made at several different reactor powers. Then an extrapolation is made to infinite power where the influence of the intrinsic neutron source is zero. To put the reactivities measured by the compensation method on an absolute basis, the Autorod is calibrated via reactor period measurements analysed by the inverse kinetics method.

3.4 The Inverse Kinetics Method

In the inverse kinetics method, the control rods remain fixed whilst a sample is inserted and the resulting change in reactivity causes the reactor power to diverge from its steady value. After a short time, the divergence has the simple exponential form of a stable reactor period. Using the data logger mentioned previously, count-rates are recorded from six neutron detectors located at different positions in the reactor. Figure 4 shows the change in power recorded during a typical set of inverse kinetics measurements with a similar measurement sequence as for the compensation method. The initial reactor balance is arranged such that the reference sample gives a positive period and the burnt sample gives a negative period. By analysing the recorded flux evolution, the reactivity of a sample in the PROTEUS reactor can be determined on an absolute basis. The results of such an analysis are shown in Figure 5. As with the compensation method, time-wise interpolation removes long-term reactor drifts. Inverse kinetics measurements are usually less precise than the compensation method, but they represent a completely independent approach and have the advantage of directly providing absolute values of reactivity.



Figure 4: Flux Variation During an Inverse Kinetics Measurement.



Figure 5: Reactivity Values Derived from an Inverse Kinetics Measurement.

3.5 Special Samples Reactivity Measurements

Special samples were manufactured by Westinghouse Atom in order to provide reference reactivity worths and thus added value to the series of measurements with burnt samples. Additives corresponding to certain important fission products (neodymium, rhodium, samarium, and gadolinium) were blended with uranium oxide and then sintered to produce pellets. The measurements indicate that there is a linear relationship between reactivity effect and the amount of additive but that the accuracy of the additive concentration data for the special rod samples needs to be improved. Accurate chemical analysis of representative pellets will be made in the Hot Laboratory as part of the ongoing analysis of the burnt samples. It should be mentioned that checks on the homogeneity of the additives have already been made by neutron radiography in the SINQ facility [4] at PSI.

4. Neutron Emissions

The intrinsic neutron source of burnt fuel, after many years cooling, is mainly due to the spontaneous fission of ²⁴⁴Cm. However, other spontaneous fission and (α , n) sources are also present, to a small extent, in the samples. Determination of the neutron emissions from each of the burnt fuel rod samples has been primarily achieved by inserting them into the reactor, one by one, with the reactor in a slightly sub-critical state. In this condition, the neutrons emitted from the sample into the multiplying reactor environment result in a suitably significant count rate on the reactor instrumentation neutron channels. A secondary method used was a by-product of the compensation reactivity measurements at different powers. As part of the extrapolation process of reactivity against the inverse of the power, the neutron source appears as the slope of the resulting straight line. By inserting a calibrated ²⁵²Cf neutron source, the neutron emission rates were put on an absolute basis. At the time of measurement, the cooling time was about seven years for the UO₂ samples and between two and four years for the MOX samples. The neutron output results were decay corrected using the ²⁴⁴Cm half-life of 18.1 years.

5. Experimental Results

The experimental results obtained to date (Figure 6) indicate that, for each of the three different moderator conditions, there appears to be a simple exponential relationship between the reactivity effect and the neutron emission rate for each type of fuel (i.e. UO_2 or MOX).



Figure 6. Relationship between measured reactivity effects and neutron emission rates of burnt PWR fuel rod samples for three different moderation conditions. For each combination of fuel type and moderation condition, the corresponding curve is in terms of increasing burnup when viewed from bottom right to top left, i.e. the higher the burnup, the greater the neutron emission and the absolute measured reactivity effect.

The suggested implication of this observed relationship is that, with only two ranging measurements of burnt fuel reactivity effects, results for other samples in the same fuel family and in the same moderating condition can be deduced from the neutron emission rates alone. This applies, of course, for the range of burnups currently investigated.

6. Conclusions

In the LWR-PROTEUS Phase II programme, the reactivity worths and neutron emissions of eight PWR burnt fuel rod samples have been measured in three different neutron spectra. These experimental results, in conjunction with the chemical assays currently underway, will be used to validate a number of production reactor codes.

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