the behaviour and chemical state of irradiated ceramic fuels

A panel to discuss the Behaviour and Chemical State of Fission Products in Irradiated Fuels was held at the IAEA headquarters in August. The meeting, chaired by Dr. K.E. Crouthamel of Argonne National Laboratory, was attended by 19 experts from seven countries, including three associated with EURATOM. Twenty-three written communications were presented and discussed. Publication of the proceedings is being prepared, but in the interim, this report of the conference summarises its work.

PURPOSE

The primary aim of the panel was to review our present understanding of the chemical behaviour of reactor fuels subject to irradiation under typical operational conditions, and to obtain a consensus regarding outstanding problem areas in the field. This behaviour relates to the many chemical interactions occurring between fuel, fission products, cladding and coolant, and how these interactions affect the design requirements for operational fuel. As such, this topic, with its important implications regarding plant safety and economy, ties in with one of the most critical problems in reactor technology – the devising of safe, reliable, high-performance fuel.

The Agency has not directly dealt with this specific subject before, and the convening of this panel at this time was considered timely in view of the broad effort being made in this field, especially in countries engaged in fast breeder reactor development. The emphasis on fast reactor fuels was reflected in the Panel's discussions which centered on oxide-type fuels, currently of greatest interest for LMFBR* systems. Nevertheless, much work has been going on with advanced carbide and other type fuels, and the Panel did not fail to include some discussions of recent results in this area.

The present Panel bears closely on the Agency's continuing program in the field of fuel technology, with other related meetings recently sponsored by the Agency, including: (a) the symposium on analytical methods in the nuclear fuel cycle (December 1971); (b) the panel on burnup physics (July 1971); (c) the panel on plutonium recycling in thermal power reactors (June 1971); and (d) the symposium on plutonium as a reactor fuel (March 1967).

* Liquid Metal Fast Breeder Reactor

The irradiation of reactor fuel results in a chemistry of unique complexity. The transformation of a certain fraction of the original actinide elements into new species, the generation of both soluble and insoluble fission products in the fuel matrix, the re-distribution of the mobile species, as well as their chemical reactions with fuel, cladding and coolant are just some of the changes which must be studied and understood to provide for safe and reliable operation of nuclear power plants.

Of the various reactor fuel types discussed, the oxide-fuel, stainless steel-clad design which is the preferred one at present for LMFBR's, received the most attention by the Panel because of the impending operation in the near future of various large LMFBR plants.

Fission product control, which represents a considerable part of the cost of a nuclear power station, includes many measures external to the fuel element itself. The most vital aspect of control, however, concerns the integrity of the fuel cladding, so that physico-chemical interreactions occurring between the fuel/fission products and the cladding are of interest as potential failure mechanisms. Internal fission gas pressure buildup, clad corrosion, fuel swelling or densification are some of the various clad failure mechanisms that must be considered. For mixed-oxide, stainless steel clad fuel elements, for example, internal corrosion is an important chemical interaction phenomenon, for which the oxygen potential at the fuel-clad interface is a key variable. The dependence of this quantity on burnup, temperature gradient, operating conditions and other parameters, is not well understood and remains an important problem because of experimental and analytical difficulties in its determination.

The Panel's review of the field bore on three main areas:

(a) Transport phenomena, including the time-varying distribution of oxygen-to-metal ratio in the fuel matrix; the redistribution of U/Pu; problems of fission gas transport, retention and release, as well as problems of the transport of the volatile and solid fission products.

(b) Fuel/fission product-cladding interactions, including, in particular, the important corrosion problem occasioned by the change in oxygen potential in oxide and mixed oxide fuels.

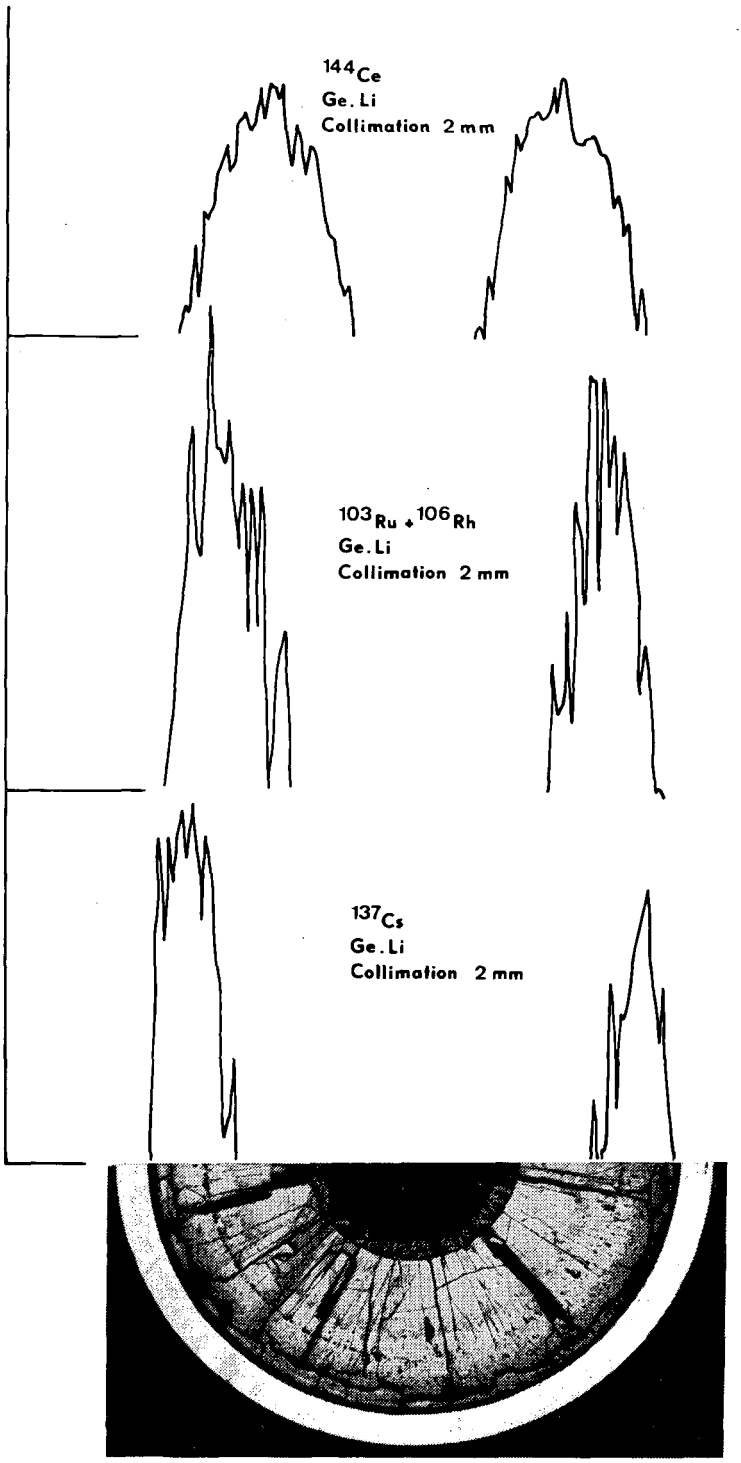
(c) Thermodynamics and phase equilibria, including application of the available thermodynamic and phase equilibria data to obtain information on the chemical state of irradiated fuel.

A number of problems areas were identified where further work is required. Among the more important ones were:

1. The need to measure oxygen redistribution directly and to evaluate it in terms of the relevant oxygen-carrying species;

2. Mechanistic concepts and redistribution models are needed to predict the changes which take place in the fuel under a variety of conditions. Similarly an understanding of the kinetic factors which influence oxygen and volatile fission product transport are needed in order to predict swelling of fuel pins and cladding attack by certain reactive constituents;

3. To fully understand the conditions which initiate cladding attack, more information is required of the thermodynamics and phase relationships of the various reactants and products, of the fission product attack on the protective properties of the oxide layers of the cladding, the role of caesium and tellurium in fuel-clad interaction, and the effect of getters to prevent oxidation of the cladding.



Radial analysis of fission products by Gamma ray spectrometry.

4. Measurements of the oxygen potential in various systems is essential to a fuller understanding of the processes taking place. These measurements include the oxidation potential of the mixed U-Pu-O system at temperatures above 3000°C and its dependence on the rare earth ions present, and the oxygen potential at which Mo-containing inclusions oxidize in irradiated fuels.

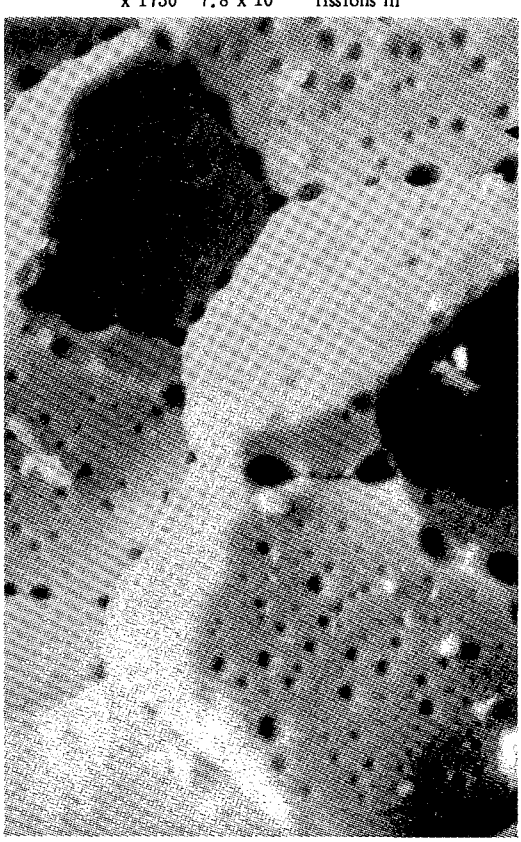
RECOMMENDATIONS

The Panel recommended that the Agency consider sponsoring an international inter-comparison of analytical predictive techniques of the chemical composition of irradiated fuels.

The decision to hold a future panel meeting in this field should be based on developments taking place during the next few years. The present Panel's area of discussion was felt to be a small part of an active field which is rapidly developing, and that consideration should be given to a future meeting of wider scope.

Scanning electron micrographs of irradiated UO_2 grain faces.

x 1750 7.8×10^{24} fissions m^{-3}



x 3500 1.2×10^{25} fissions m^{-3}