

# Reactor Irradiation Techniques

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The problems encountered in carrying out precise investigations of the effects of neutron irradiation on the properties of materials are discussed. Various experimental techniques which have been developed to overcome these problems are described with particular reference to techniques which could be used in the Australian Atomic Energy Commission's research reactor, HIFAR.

TABLE 1—IRRADIATION FACILITIES IN HIFAR

Designation	Number	Position	Peak thermal flux	Peak fast flux > 1 MEV
6V	4	In heavy water	5-6 x 10 <sup>13</sup>	2.5 x 10 <sup>11</sup>
4V	5	" " "	3 x 10 <sup>13</sup>	1 x 10 <sup>11</sup>
2V	9	" " "	7-9 x 10 <sup>13</sup>	1 x 10 <sup>12</sup>
6VGR	6	In graphite reflector	3 x 10 <sup>12</sup>	2 x 10 <sup>9</sup>
4VGR	2	" " "	3 x 10 <sup>12</sup>	2 x 10 <sup>9</sup>
10VGR	2	" " "	3 x 10 <sup>12</sup>	2 x 10 <sup>9</sup>
10H	1	In heavy water	7 x 10 <sup>13</sup>	2 x 10 <sup>12</sup>
6H	1	" " "	8 x 10 <sup>13</sup>	6 x 10 <sup>12</sup>
4H	6	" " "	6 x 10 <sup>13</sup>	4 x 10 <sup>12</sup>
2 tan	1	" " "	9 x 10 <sup>13</sup>	4 x 10 <sup>12</sup>
6HGR	10	In graphite reflector	1 x 10 <sup>13</sup>	2 x 10 <sup>10</sup>
12HGR	2	" " "	7 x 10 <sup>12</sup>	2 x 10 <sup>10</sup>
4HTC	9	In graphite thermal column	7 x 10 <sup>10</sup>	4 x 10 <sup>8</sup>

## INTRODUCTION

The ever-increasing demands of reactor design engineers for materials which will withstand high doses of irradiation under extreme conditions means that irradiation damage investigations must now play an extremely important part in any atomic energy program. Of equal importance in this field is basic research, not only to obtain data on the mechanisms of irradiation damage, but to assist in a better understanding of the physics of the solid state.

With the commissioning of the Australian Atomic Energy Commission's research reactor, HIFAR, facilities will become available, both to the Commission's Research Establishment and to other research organisations, for investigations of the effects of neutron irradiation on the properties of materials. The purpose of this paper is to outline the problems encountered in carrying out these investigations in a reactor of the HIFAR type and to describe techniques which have been developed for similar reactors overseas.

Irradiation damage investigations can be divided broadly into two types on the basis of techniques used and difficulties encountered:—

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## Investigations on fissile materials

This includes small-scale investigations on fuel element materials and the testing of full-scale prototype fuel elements in complex "loops." The latter type of investigation will be mentioned only briefly, although the problems encountered are generally the same as for the small-scale work. Fuel element testing loops are usually a major engineering project and their operation and control is outside the scope of this paper.

## Investigations on non-fissile materials

Because of the lower heat generation and generally lower hazards associated with this type of experiment, the techniques used are often simpler than in the fissile material experiments.

## FACILITIES AVAILABLE

The types of facility available for the insertion of experiments vary considerably with the type of reactor. In HIFAR the facilities consist of horizontal and vertical thimbles which are situated either in the heavy water moderator adjacent to the fuel elements, or in the graphite reflector. These facilities, together with the neutron fluxes available are shown in Table I; the fast fluxes given in the last column are very approximate.

In the designation, the number refers to the

diameter of the hole, the V to vertical holes and the H to horizontal holes.

The fast flux in these facilities is not sufficiently high for some work on non-fissile materials where high neutron doses with energy greater than 1 MeV are required. In these cases a neutron "converter" must be used; this generally consists of a hollow fuel element. Fission in the fuel enhances the fast flux in the centre at the expense of the thermal flux.

In HIFAR it is hoped that some hollow fuel elements with a 2in. diameter central thimble will replace some of the standard fuel boxes to provide a fast neutron irradiation facility. Flux above 1 MeV in this facility will be approximately  $3 \times 10^{13}$  n/cm<sup>2</sup>/sec.

### DESIGN PROBLEMS

#### Space limitation

Limitations are often placed on the design of experiments by the small diameter of experimental holes, by the distance of the high flux region in a hole from the face of the reactor and by the limited amount of space usually available outside the pile shield in the immediate vicinity of the experimental hole. This limitation becomes particularly serious when in pile measurements such as the rate of strain in a creep test must be made.

#### Heat removal and temperature control

##### HEAT PRODUCTION

A major problem encountered in the design of experiments is that of removing the heat generated by various processes in the sample and associated apparatus while allowing control of the temperature in the specimen.

Heat production in fissile materials by fission is given approximately by:—

$$H = \frac{\phi \Sigma_f V}{3.1 \times 10^{10}} \text{ watts}$$

where  $\phi$  = mean flux in sample,  
 $\Sigma_f$  = macroscopic cross-section for fission,  
 $V$  = volume of sample

This heat production can reach very high levels, e.g., 4½ KW/gram of U235 in a flux of  $10^{14}$ .

Accurate determination of the mean flux in a specimen for this calculation is difficult. The undisturbed flux in the experimental thimbles is reduced by absorption in the sample and apparatus and by streaming along the thimble. Several empirical methods (Lewis 1954) have been used for estimating the flux depression, but the results are not accurate, particularly for strong absorbers, e.g., highly enriched fuel samples.

Heat is produced in materials by several processes other than fission:—

- (i) Capture  $\alpha$  heating due to n,  $\alpha$  reactions.
- (ii) Capture  $\gamma$  heating due to n,  $\gamma$  reactions.
- (iii)  $\gamma$ -heating due to exposure to the pile  $\gamma$  flux.
- (iv) Radioactive decay heating.

(v) Heating due to fast neutron scattering.

The relative importance of these effects varies between different materials, for instance, for light materials such as carbon or beryllium the heating due to fast neutron scattering is predominant. Knowing the pile neutron and  $\gamma$  flux, heating due to these effects can be calculated. Experimental measurements have been made in the U.K. reactor, DIDO (which is identical to HIFAR), and the expected heat production in various materials in several positions in HIFAR are given in Table II.

With very highly rated experimental reactors, such as the United States Atomic Energy Commission's Engineering Test Reactor in Idaho, this heat production can be as high as 50 watts/gm in the core.

TABLE 2.—HEAT PRODUCTION IN VARIOUS NON-FISSILE MATERIALS IN 2V, 4V AND 4VGR POSITIONS IN HIFAR.

Material	Heat production watts/gm		
	2V	4V	4VGR
Beryllium	0.65	0.10	0.015
Carbon	0.71	0.10	0.014
Aluminium	0.75	0.15	0.02
Iron	0.67	0.093	0.014
Zirconium	0.72	0.088	0.012
Lead	0.82	0.11	0.017

##### HEAT REMOVAL

Removal of heat generated by any of the above processes can be accomplished in several ways:—

- (i) Direct insertion of the experiment into the main reactor coolant stream (gas or water). This method is used for simple experiments in MTR, and has been used in BEPO. The disadvantage of the method is that any escape of activity from the experiment could contaminate the whole of the reactor circuit.
- (ii) For low heat outputs, the experiment can be suspended in an experimental thimble, conduction and convection to the main coolant being used to remove the heat.
- (iii) Forced cooling using a liquid or gas circuit distinct from the main reactor circuit. This system can vary in complexity up to a full-scale fuel element testing loop when the coolant would be the same as in the reactor system being investigated. Gas, high pressure water, liquid metal and organic liquid in-pile loops have all been operated in various reactors overseas (see, for instance, Martinec, 1957, and Francis, 1955). For simpler experiments gas or water cooling at low pressures can generally be used. If a guaranteed cooling circuit is required (i.e., if cooling of the experiment during reactor operation is essential to the safety of the reactor), adequate instrumentation must be provided

to trip the reactor if the supply fails, and a standby supply should be provided.

#### TEMPERATURE CONTROL

Materials must often be irradiated at elevated temperatures, and use can be made of the various forms of nuclear heating to attain these temperatures. A thermal barrier is placed between the sample and the coolant, the thickness and conductivity of the barrier being chosen so that the calculated heat production under irradiation will raise the temperature to the desired level. A gas gap, the most satisfactory barrier, has been used successfully in the U.K. (Plail, 1956), and will be used in HIFAR for investigations of the stability of fuel materials for use in a high temperature gas cooled reactor. For highly rated samples, it is usually necessary to provide a greater heat transfer area at the gas gap than the specimen surface area, or the width of the gas gap becomes prohibitively small. This greater transfer area may be accomplished by immersion of the samples in liquid sodium or sodium potassium alloy.

The above methods have two limitations:—

- (i) Uncertainties in calculation of the heat output mean that the desired temperature cannot be obtained with an accuracy of better than  $\pm 20$  per cent.
- (ii) Variations in neutron flux in the experiment due to reactor control arm movement and the insertion and removal of other absorbers in the vicinity, e.g., other experiments, will result in a variation in the specimen temperature over the irradiation period.

For high temperature irradiation of samples with low heat output, these disadvantages can be overcome to a certain extent by the use of radiation cooling across a wide gas gap of low conductivity, in which conduction and convection can be neglected. This method has the advantage that the temperature varies as the fourth root of the heat output, and hence is not very sensitive to changes in heat output or inaccuracies in calculation.

If more precise temperature control is required, some form of auxiliary heating must be used. In its simplest form, this consists of a resistance winding of small heat output compared with the nuclear heating. The thermal barrier is chosen so that the specimen temperature with nuclear heating alone would be somewhat lower than the desired temperature. The auxiliary heating is then used to raise the temperature to the required level, and to cater for variations in the nuclear heating during irradiation.

This method, however, results in temperature cycling at reactor shut-downs. In some cases, e.g., irradiation of uranium metal, this may be undesirable because the thermal cycling will itself produce damage which could override any irradiation effects. In this case, greater auxiliary heating must be applied, not only to control the temperature during irradiation but

also to maintain the temperature during shut-downs.

An apparatus of this type was developed at the Atomic Energy Research Establishment at Harwell, by the author, for irradiation of small samples of fuels (up to 1cm. in diameter by 2cm. long) with a heat output of 500 watts. The main problem in design of this rig was to provide a path to conduct the fission heat away during operation, but at the same time to incorporate sufficient auxiliary heating in the very small space available to maintain the specimen temperature at reactor shut-downs.

A sketch of the apparatus is shown in Figure I. The specimen was immersed in liquid sodium in a double-walled stainless steel can, which was a push fit in a finned zirconium furnace former. This was surrounded by a zirconium tube, which was separated from a water-cooled

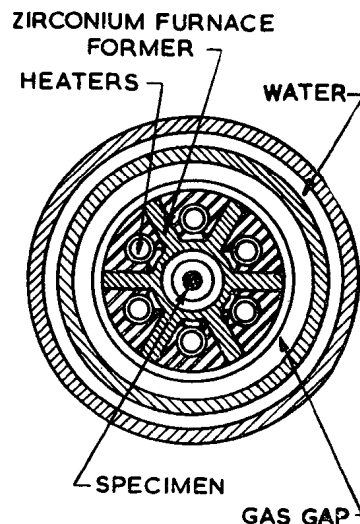


FIGURE 1: Reactor experiment cross-section.

aluminium jacket by a helium-filled gas gap. Nichrome furnace windings of 800-1000 watts rating were supported between the zirconium fins by ceramic supports. During fission, the heat was conducted down the zirconium fins and across the gas gap. On shut-down, a controller increased the furnace input to maintain the required temperature.

To cope with rapid reactor shut-downs a fast response furnace controller was required. After investigating several systems, a magnetic amplifier unit operated from a potentiometric recorder controller was adopted. The system has been found to work very satisfactorily, and even at "scram" shut-downs, when the pile power is reduced to a low value in a few seconds, temperature cycles of only 10-20°C were observed, and samples could be irradiated at controlled temperatures in the range 400-800°C.

#### LOW TEMPERATURE IRRADIATION

For basic investigations of irradiation dam-

age in metals where some induced changes anneal out at ordinary pile temperatures (50-150°C), it is often desirable to carry out the irradiation at reduced temperatures. Irradiations at liquid nitrogen temperatures have been carried out by A.E.R.E., Harwell, in BEPO, and by Oak Ridge National Laboratories in their X10 reactor, and recently Oak Ridge have developed a helium cryostat for irradiation at temperatures down to 20°K (Coltman et alia, 1957). Nuclear heating in these reactors is low—the order of a few milliwatts per gram. However, it is considered that the problems involved in carrying out these experiments in HIFAR, whilst more difficult than the above cases, due to the higher nuclear heating, are not insurmountable.

#### Materials of construction

There are three main limitations on the use of materials for irradiation apparatus:—

##### NEUTRON ABSORPTION

Excessive neutron absorption in the apparatus will not only reduce the effective flux in the sample, but also will reduce the excess reactivity available in the reactor. Wherever possible, materials with a low neutron capture cross-section should be used in the construction of apparatus, but this requirement has to be related to their suitability in other respects, e.g., strength at elevated temperatures. Aluminium and magnesium are useful for low temperature work. For higher temperatures, hafnium-free zirconium can be used, but it is in limited supply and must be protected from oxidation, and often stainless steels or Nimonic alloys must be used for part of the assemblies.

##### INDUCED ACTIVITY

To facilitate handling of the experiments after irradiation and to reduce the problems of disposal of the assemblies, the induced activity in the apparatus should be kept to a minimum. Even very small quantities of some materials with high capture cross-sections and medium to long half-lives, e.g., cobalt and tantalum, can result in high induced activities. For this reason, alloys should be chosen with care, and wherever possible high purity grades of metals, e.g., aluminium, should be used. For instance, cobalt impurities in the nickel in stainless steel account for a large proportion of the long-lived activity induced in stainless steel, and special low-cobalt stainless steels are now being manufactured in the U.S.A. for use in reactors.

##### STABILITY UNDER IRRADIATION

Generally, the only limitation this places on the use of materials is on those plastics and organic materials which are unstable under irradiation. This is particularly important in the case of insulating materials, and pure ceramics should be used for this purpose.

In some special cases, other factors must be considered, e.g.,

- (i) The change under irradiation in the brittle-ductile transition temperature of

a steel used in a high pressure water loop may cause failure of the pressure shell.

- (ii) The large change in dimensions of graphite precludes the use of this material where close tolerances must be maintained.

#### Safety

Safety of the research reactor is of prime importance at all times, and the designs of all experiments must incorporate precautions against every foreseeable hazard.

Any materials likely to lead to a hazard must be adequately contained, e.g., liquid sodium and quantities of fission products are generally contained inside a double-walled can.

Any conditions likely to endanger the reactor or the operating personnel must be adequately covered by instrumentation, and may need to be wired into suitable alarms, and sometimes into the reactor trip circuit. To minimise the reactor trips which cause loss of operating time, the number of circuits tied with the reactor trip circuit should be kept to the absolute minimum consistent with safety.

The need for complete reliability in these circuits, and indeed for all equipment associated with in-pile experiments, cannot be over-emphasised. The problem is often made more difficult because non-standard components have to be used, and also because standard components are often found to have insufficient reliability for use in this work. Hence it is essential that all components should be thoroughly tested before use, and that the completed assemblies should be tested, as far as possible, before insertion in the reactor.

#### Post-irradiation examination

It should always be borne in mind that the irradiated apparatus will have to be handled remotely after irradiation, both during removal from the reactor and during removal and examination of the samples. Hence, the design should include all possible features which will allow easy handling and dismantling by remotely operated tongs or manipulators.

#### ASSOCIATED TECHNIQUES

##### Flux measurement

It is usually desirable that the integrated flux which an irradiated sample has received should be known fairly accurately. This can be obtained in three ways:—

- (i) Calculation based on a knowledge of the undisturbed flux in the irradiation position before insertion of the experiment. As mentioned earlier, this is not very satisfactory owing to variations in the flux over a period of time with movement of control arms and changes in the loading of the reactor, and to uncertainties in the calculation of the flux depression caused by the experiment.
- (ii) Analysis of the irradiated samples. In the case of fissile material this is easily carried out using gamma-spectrometric techniques to measure the quantities of

some fission products whose yields are known fairly accurately. The method can also be used for non-fissile samples by measuring induced activities or the quantity of a particular isotope produced by the irradiation.

- (iii) Use of a suitable flux monitor placed adjacent to the sample. For thermal flux measurements cobalt in the form of wire can be used—the induced  $\text{Co}_{60}$  activity being measured after irradiation. Sometimes in order to obtain an easily handled sample of low activity, the cobalt is diluted by dispersion in another material, e.g., high purity aluminium.

For monitoring fast flux, no really suitable technique has been developed. The reaction  $\text{S}^{32}(\text{n}, \text{p})\text{P}^{32}$ , which has a threshold of about 1 MeV, has been used (Martin, 1956), the activity of the beta emitting  $\text{P}^{32}$  being measured. However, the short half life (14.3 days) of the  $\text{P}^{32}$  means that the induced activity approaches saturation after a few weeks' irradiation and is not suitable for longer irradiations.

The fast fission reaction which occurs in  $\text{U}^{238}$  at neutron energies greater than 0.6 MeV is another possibility which is being investigated. The monitor must be shielded from thermal neutrons by a suitable material, e.g., cadmium or boron, to prevent interference by thermal fission of  $\text{U}^{235}$  or  $\text{Pu}^{239}$ .

Care must be exercised in the placing of the monitor with respect to the specimen. Ideally, the monitor should actually be inserted in the specimen, but often this is not possible and an uncertainty is introduced if the monitor is placed adjacent to the specimen, because it is not known to what extent the flux at the

monitor was affected by the flux depression due to the sample.

#### Temperature measurement

Thermocouples have been found quite satisfactory for temperature measurement provided suitable insulating materials are used—mineral-insulated couples such as "Pyrotenax" are the most satisfactory. There is no evidence that the neutron flux affects the characteristics of the thermocouples in any way.

#### CONCLUSIONS

Although a considerable amount of experience has been accumulated on the utilisation of experimental space in low flux reactors, and to a lesser extent in high flux reactors, many problems remain unsolved. Many of the irradiation investigations carried out to date have produced conflicting results, because the experimental conditions have not been precisely known or controlled, and there is a wide field for improvement in this respect.

This paper has served to outline the more important problems encountered in the design of experiments and some of the techniques which have been used to overcome them, and it is hoped that it has given potential users of HIFAR an idea of the factors which need to be considered in the planning of an experiment.

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