

NEUTRON-IRRADIATION FACILITIES AT THE  
INTENSE PULSED NEUTRON SOURCE-I FOR FUSION MAGNET MATERIALS STUDIES

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## ABSTRACT

The decommissioning of reactor-based neutron sources in the U.S.A. has led to the development of a new generation of neutron sources that employ high-energy accelerators. Among the accelerator-based neutron sources presently in operation, the highest-flux source is the Intense Pulsed Neutron Source (IPNS), a user facility at Argonne National Laboratory. Neutrons in this source are produced by the interaction of 400-500 MeV protons with either of two  $^{238}\text{U}$  target systems. In the Radiation Effects Facility (REF), the  $^{238}\text{U}$  target is surrounded by Pb for neutron generation and reflection. The REF has three separate irradiation thimbles. Two thimbles provide irradiation temperatures between that of liquid He and several hundred degrees centigrade. The third thimble operates at ambient temperature. The large irradiation volume, the neutron spectrum and flux, the ability to transfer samples without warm up, and the dedication of the facilities during the irradiation make this ideally suited for radiation damage studies on components for superconducting fusion magnets. Possible experiments for fusion magnet materials are discussed on cyclic irradiation and annealing of stabilizers in a high magnetic field, mechanical tests on organic insulation irradiated at 4 K, and superconductors measured in high fields after irradiation.

## INTRODUCTION

For magnetic fusion reactors the containment of the highly energetic reacting particles requires high magnetic fields. Economic considerations dictate that the plasma-containment magnets (as well as other magnets in the various fusion-reactor designs) will be superconducting. These magnets will be required to operate under extreme conditions: low temperatures, high internal mechanical stresses, large asymmetric forces, pulsed magnetic fields, and neutron and gamma irradiation. There have been no irradiations of large superconducting magnets under fusion-reactor conditions, and it is doubtful if data from small magnets irradiated in the superconducting state are of any engineering value. This is because changes in the operating parameters of a magnet depend not only on changes in the various components, but also on the design details. Therefore, the radiation effects

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in the various components of a superconducting magnet must be experimentally determined and the magnet designers must then incorporate the results of these effects into their design.<sup>1,2</sup>

It is well known from many radiation-damage experiments that the details of the property changes can depend strongly on the type and energy of the irradiating particle, the irradiation temperature, and the metallurgical condition of the starting material. Since neutrons are the only damaging particles incident on the magnet at significant fluxes, only neutron irradiation experiments are of direct interest for fusion magnet studies and, additionally,  $\gamma$  irradiations of insulators.

Since the magnet will be irradiated during operation, the property changes for the various components must be determined during irradiation at cryogenic temperature. The role of irradiation temperature is related to defect mobility. If the irradiation takes place above the migration temperature of a defect, then the defect can move about during the irradiation and possibly annihilate or agglomerate. In most cases this defect state leads to property changes which are different from those caused by the immobile defects produced during low-temperature irradiation. The difference can be in magnitude if the property change is proportional to total number of defects or in character if the property change depends on the details of the defect structure. Therefore, only irradiations performed near the operating temperature of the magnet ( $\lesssim 10$  K) are unambiguous and, thus, useful for predicting changes in magnet operating characteristics. Higher-temperature irradiations are useful only to the extent that they help explain results of or serve as guidance for low-temperature irradiations. Radiation effects should be considered for the four major components of the magnet: superconductor, stabilizer or normal conductor, insulator, and structural material.

## FACILITY

A new neutron source has been recently built at Argonne: IPNS-I, which is a national facility for condensed matter research using neutron scattering and radiation damage techniques. The layout of IPNS-I is shown in Fig. 1. Pulses of H ions are accelerated to 50 MeV in a linear accelerator (LINAC). The electrons are stripped off and the H<sup>+</sup> ions (protons) are accelerated to 400-500 MeV by a rapid cycling synchrotron (RCS). These protons are then directed to a heavy metal target where approximately 20 neutrons/proton are produced (for a <sup>238</sup>U target) by the processes of spallation and fission. Because the synchrotron delivers the protons in bursts (at 30 Hz), the neutrons produced are likewise delivered in high intensity pulses.

Much of the accelerator system was obtained by IPNS-I from a discontinued High Energy Physics program. This plus many available buildings, beam line magnets and shielding materials were utilized by IPNS-I to build a facility for \$8.8 M that is estimated to be worth \$50 M.

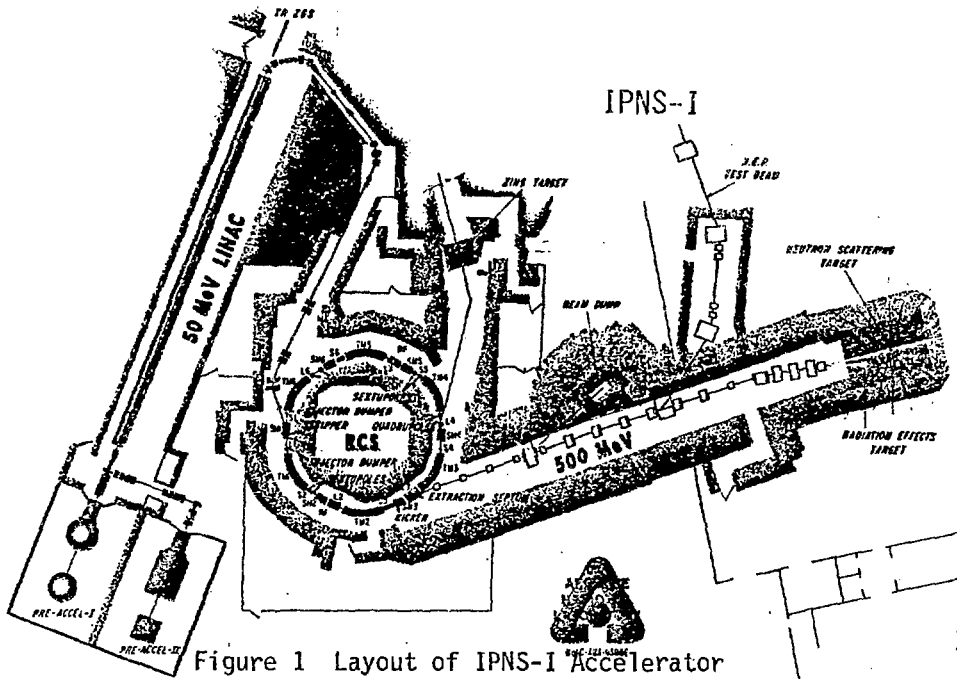


Figure 1 Layout of IPNS-I Accelerator System, Proton Transport Line & Neutron Generating Systems

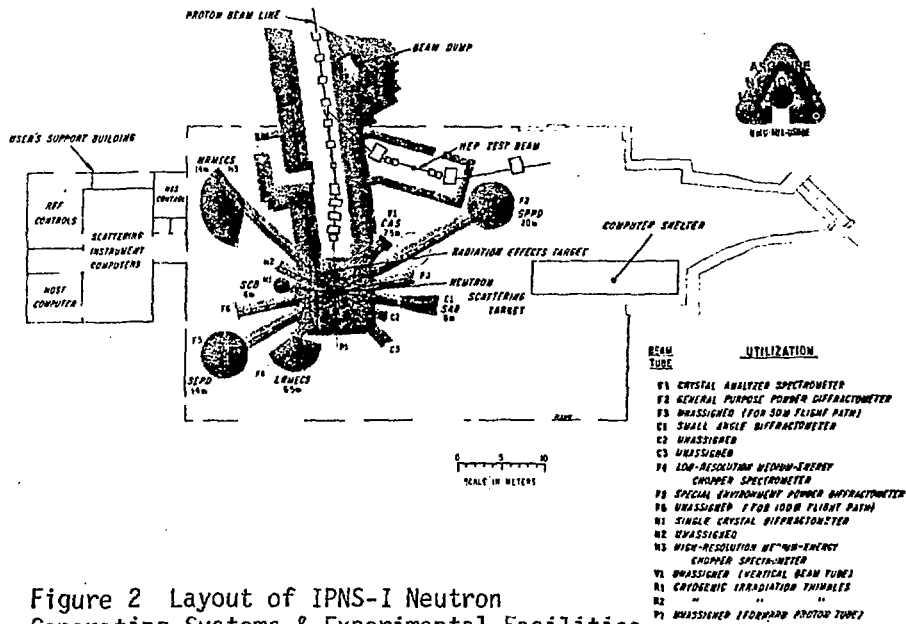


Figure 2 Layout of IPNS-I Neutron Generating Systems & Experimental Facilities

The experimental area is shown in Fig. 2. The proton beam can be sent to one of two identical  $^{238}\text{U}$  targets. The neutron scattering target, which receives the beam  $\sim 75\%$  of the time, is surrounded by Be reflector and cryogenic moderators to produce neutron beams scattering at instruments that are shown in Fig. 2.

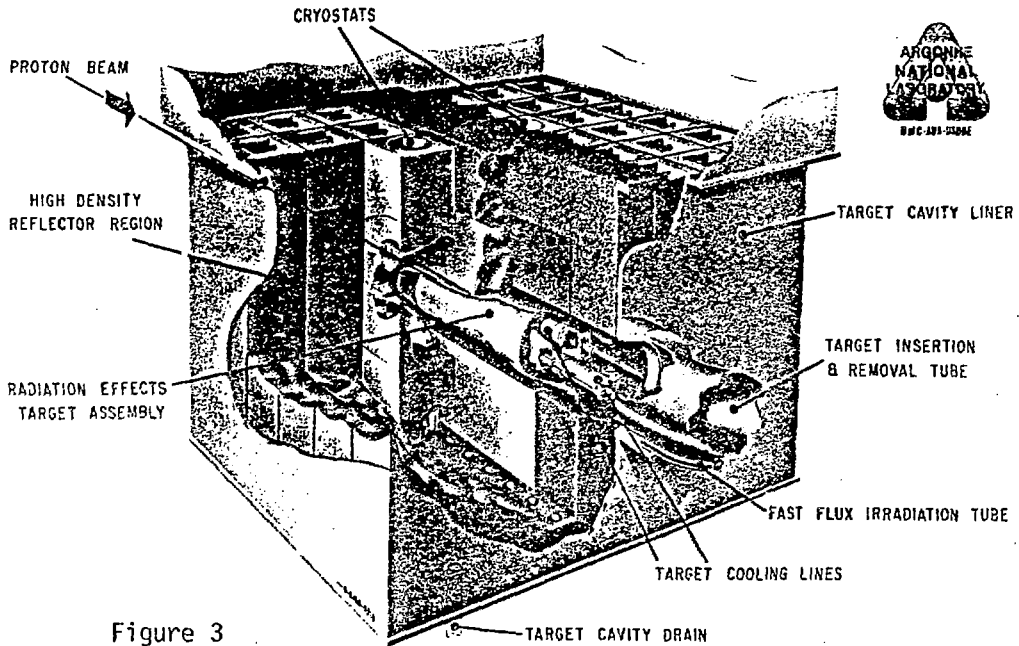


Figure 3

### IPNS-I Radiation Effects Experimental Assembly

The REF, shown in Fig. 3, consists of the  $^{238}\text{U}$  target, two vertical irradiation thimbles, and a horizontal irradiation thimble, all surrounded by a Pb neutron reflector. The target material was chosen to maximize the conversion of protons to neutrons.<sup>3</sup> There is some gamma production associated with the fission process in  $^{238}\text{U}$ , although much less than in a reactor-based facility where all neutrons are produced by fission. Should the gamma flux pose an experimental problem, it is possible to change to a Ta target, from which there would be a greatly reduced gamma flux. Lead was chosen as the reflector material based on the results of experiments and calculations on a prototype facility.<sup>3</sup> The neutron reflection and high-energy (n,2n) reactions in Pb increase the neutron flux in the irradiation facilities and reduce the flux gradient in a direction perpendicular to the target axis. The Pb reflector alongside the target is in the form of removable sections 10 cm on a side and 45 cm long. For specialized needs, reflector sections can be removed to increase the irradiation volume or to allow replacement with a different reflector material. Such a change

in reflector or target material could change the energy distribution of the neutrons within the irradiation facilities.

The two vertical irradiation thimbles, located on either side of the target at the positions of maximum flux, contain liquid helium cryostats (5cm inner diameter) that can operate at temperatures between 4 and 1000 K. The liquid helium is supplied by a single 400-W refrigerator (CTI model 2800 R). The two cryostats have separate vacuum systems, which allow the temperature to be controlled independently in each cryostat. The horizontal irradiation thimble (2 cm inner diameter) is located on an axis parallel to and directly below the target. The majority of the  $^{238}\text{U}$  target-cooling water is between the target and this thimble. The horizontal thimble operates at ambient temperature and is designed to permit short irradiations with sample removal while neutrons are being produced. A transfer cryostat has been designed that permits removal of irradiated samples without warmup above 4.2 K for transfer to other cryostats for more complete measurements. This permits maximum use of the irradiation time. The neutron flux is fairly constant ( $\pm 10\%$ ) over a 10 cm length of the center of the cryostat which results in a 50 cm<sup>3</sup> irradiation volume.

The temperatures, pressures and other variables of the cryostats are constantly monitored by a computer system in an adjoining building. Continuous data acquisition can be done automatically by a computer system which can then be connected to a large host computer for more complex data analysis. The characteristics of the REF are summarized in Table I.

Table I

Radiation Effects Facility at IPNS-I

Flux ( $E > 0.1$  MeV)  $\sim 1 \times 10^{12}$  n/cm<sup>2</sup>s  
at proton beam current of 8  $\mu\text{A}$

Irradiation temperature, 1000 K  $> T \geq 4.2$  K

Cryostat inner diameter = 5 cm

Cryostat irradiation volume  $\sim 50$  cm<sup>3</sup>

400 watts of refrigeration at 4.2 K

Transfer capability without warmup above 4.2 K

Short term irradiations in "rabbit" tube

The  $^{238}\text{U}$  target in the Neutron Scattering Facility (NSF) is surrounded by reflectors which are penetrated by 12 neutron beam lines. Moderators

for producing the thermal-neutron beams are located directly above and below the target. Two unused horizontal beam lines have been modified to contain irradiation thimbles ( $\sim 1$  cm diameter). These two thimbles radially approach within 4 cm of the target axis at the position of maximum neutron flux. The majority of target-cooling water is between the target and these thimbles. Both NSF irradiation thimbles operate at ambient temperature.

#### NEUTRON SPECTRUM AND DAMAGE ENERGY

The study of radiation effects has reached the stage where the neutron flux cannot be adequately described by a single number; rather, the energy distribution of the neutrons is required to fully understand and compare experiments. A multiple-foil activation technique<sup>4</sup> was used to determine the neutron energy spectrum and has been described in detail.<sup>5</sup> More than 36 different neutron-activation reactions were measured at approximately the maximum flux location in each thimble. The foil activities were measured with Ge(Li) detectors over several decay- $\gamma$  half-lives for each reaction.

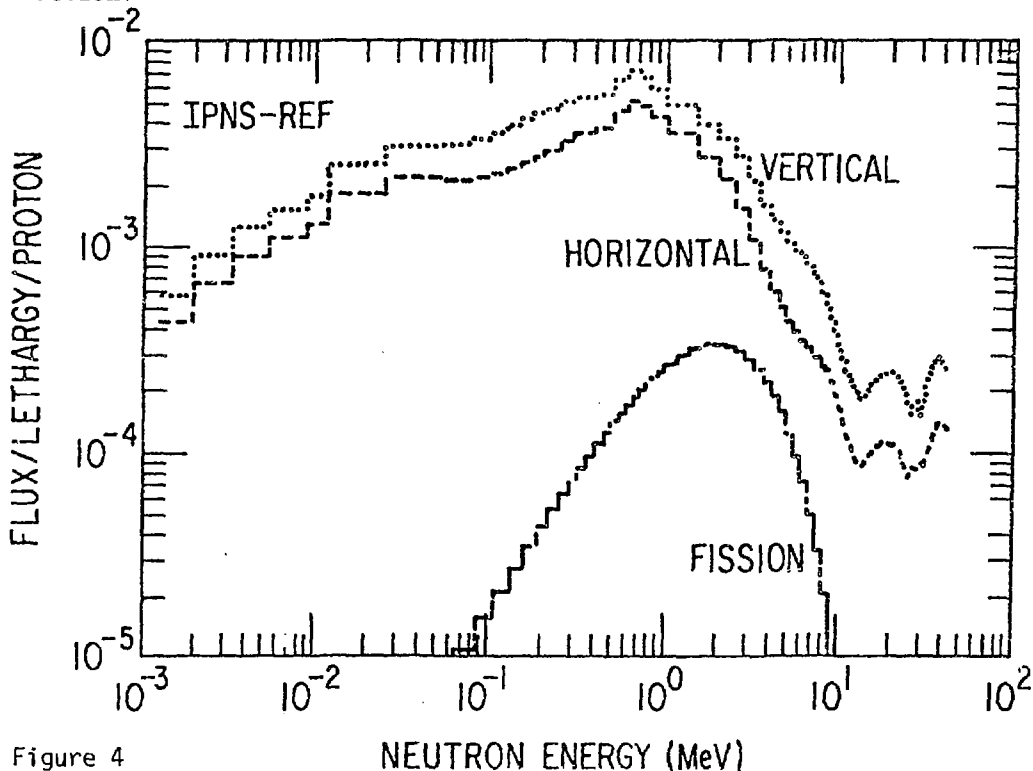


Figure 4

Neutron spectra produced in the vertical and horizontal thimbles of the REF by 500-MeV protons incident upon the  $^{238}\text{U}$  target; a pure fission neutron spectrum is shown for comparison.

The energy distribution of the neutrons at the position of maximum flux along the center of the REF vertical thimble is shown in Fig. 4 along with the energy distribution for fission neutrons. The neutron flux measurements were made with 1 atm of He gas in the irradiation thimble, and only minor changes are expected if the cryostats contain liquid helium. The neutron flux is plotted as the flux per unit lethargy ( $d\phi/d\ln E$ , or equivalently,  $E d\phi/dE$ ). The REF neutron spectrum can be characterized as a degraded fission spectrum with a high-energy component. The flux of neutrons with  $E > 0.1$  MeV is  $184 (n/m^2)/p$ , and the ratio of thermal to "fast" ( $E > 0.1$  MeV) neutrons is 0.014 for 500 MeV protons incident upon the  $^{238}\text{U}$  target. The secondary proton flux is estimated to be  $0.7 \pm 0.5 (p/m^2)/p$  or 0.4% of the flux of neutrons with  $E > 0.1$  MeV. The neutron energy distribution for the REF horizontal thimble is also shown in Fig. 4.

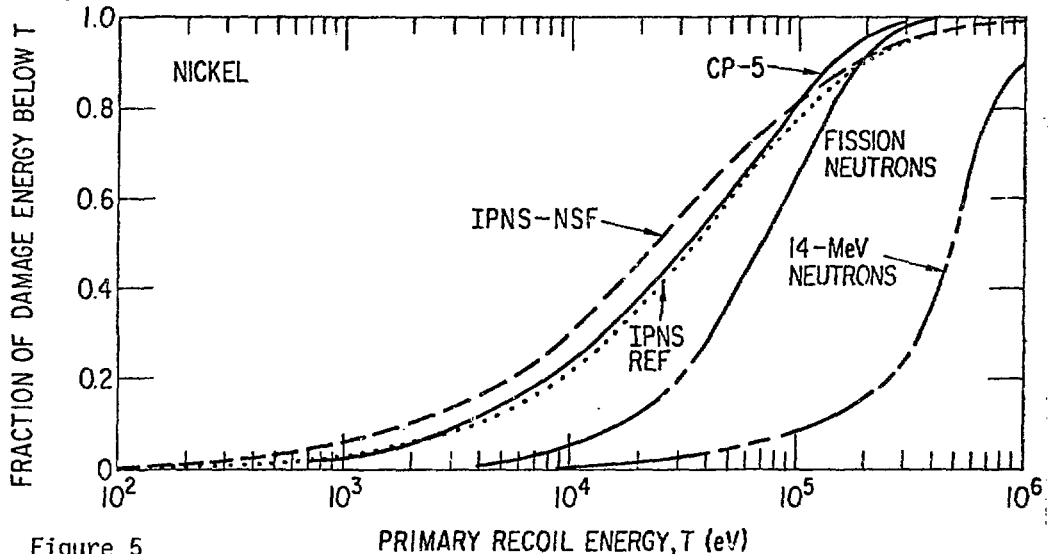


Figure 5  
Comparison of integral damage-energy distributions in Ni irradiated with NSF, REF, degraded fission (CP-5), fission, and 14-MeV neutrons.

The integrated damage energy as a function of primary atom recoil energy is shown in Fig. 5 for Ni irradiated with REF, NSF, fission, degraded fission (from the CP-5 reactor<sup>6</sup>), and 14 MeV neutrons. Such detailed calculations are needed to understand the different damages produced by different neutron spectra. The size and, perhaps, character of the defect cascades resulting from neutron irradiation will be a function of the neutron energy. Therefore, the energy spectrum and damage energy are necessary to characterize the damage state. The details of the damage energy spectrum will vary for different materials, but the general features will remain the same. In the REF, neutrons with energy above 10 MeV produce approximately 15% of the total displacements in Ni even though they comprise only 3% of the flux of neutrons with  $E > 0.1$  MeV. One method of comparing the different neutron spectra is the damage energy,  $T_{1/2}$ , for which half the damage is produced by higher- or lower-energy events. The

$T_{1/2}$  is 35 keV for the REF and 20 keV for the NSF thimbles. These compare to values of 500 keV for 14-MeV neutrons, 70 keV for pure fission neutrons, and 35 keV for degraded fission neutrons. The damage energy cross section,  $\langle\sigma T\rangle$ , the energy available to create defects per neutron, is given in Table II for Ni irradiated in the REF and NSF. Results of such calculations are available for most elements.

Table II

Neutron fluxes, and resultant damage parameters for Ni, at the IPNS with 8  $\mu$ A of 500 MeV protons incident upon a  $^{238}\text{U}$  target.

Radiation Facility	Neutron Flux			$\langle\sigma\cdot T\rangle^{\text{Ni}}$ , (keV-b)/n
	$E>0.1$ MeV, $10^{11}$ n/cm <sup>2</sup> ·s	$E>10$ MeV, % <sup>a</sup>	Thermal, % <sup>a</sup>	
REF Vertical Thimble (Central Plane)				
Near Center	9.2			
Max	16.0	2.9	1.4	63.4
REF Horizontal Thimble (Max. Axial)				
Bottom	6.1	2.4	1.4	61.1
Top	9.8			
NSF (Max.)	2.9	1.9	77	55.7

<sup>a</sup>Expressed as % of flux of neutrons with  $E > 0.1$  MeV.

#### EXPERIMENTS

The three areas of radiation damage of greatest uncertainty to fusion magnet designers for which the REF at IPNS-I can uniquely provide answers are low temperature fast neutron irradiations of insulators, and stabilizers and superconductors at high magnetic fields.

Recent work on gamma irradiations at 4 K of organic insulators showed that mechanical degradation occurs in the dose range of  $10^9$ - $10^{10}$  rads,<sup>7</sup> which is the range of lifetime dose in most magnet designs. Much of the energy deposited in the fusion magnet insulators (80% in some designs) will result from fast neutrons. Fast neutrons deposit their energy (ionization and displacement) over a much shorter distance than gammas and, therefore some low temperature irradiations must be performed with fast neutrons for comparison with the gamma results. The large irradiation volume permits



many samples to be irradiated simultaneously for screening purposes. Also, it is possible for mechanical tests to be performed in situ during the irradiation.

The original REF target is  $^{238}\text{U}$ , from which there are some gammas associated with fission processes, although still considerably fewer gammas than reactor based facilities. It is possible to change to a Ta target from which there would be a greatly reduced gamma flux. Therefore, the relative contributions of damage in the insulators, i.e., rads, from fast neutrons and gammas can be measured. Based on calculations the average weekly dose is  $\sim 0.6 \times 10^{18}$  n/cm<sup>2</sup> ( $E > 0.1$  MeV) or  $\sim 4 \times 10^8$  rad in epoxy.

Many designs show that radiation induced resistivity increases in the stabilizer (and thermal conductivity decreases) will limit the performance of the magnets. Periodic anneals to near room temperature recovers either 85% or 100% for Cu or Al stabilizers, respectively. Therefore, a possible mode of operation would be irradiation to the limiting resistivity value followed by an anneal to 300 K. It is necessary to determine what the effect of a number of these cycles is on the successive percentage of recovery during anneals or the rate of resistivity increase during irradiation after an anneal. The REF is ideally suited for an experiment that would perform a number of irradiation and annealing cycles and measure in situ the total resistance (including magnetoresistance) for magnetic fields in the 7-10 T range. Since the defect state after an anneal depends strongly on the impurity and cold work state of the sample, many different samples that cover the range of possible magnet materials should be included in the experiment.

The critical current ( $J_c$ ) change in irradiated alloy or compound superconductors is presently of less concern than the insulators or stabilizers. However, some experiments can relatively easily be done at IPNS-I that test damage energy models and our ability to predict  $J_c$  changes for high magnetic fields. Measurements in magnetic fields up to 10 T and comparison to changes after 14 MeV neutron irradiation should give sufficient data to predict the  $J_c$  changes for lifetime doses on the superconducting magnets.

#### IPNS-I USER POLICY

IPNS-I is operated as a national user facility with experiments selected on the basis of scientific merit by a nationally constituted Program Committee. The Program Committee reviews proposals for experiments or experimental programs to be performed at IPNS-I and recommends acceptance or rejection to the Program Director. The Committee is to base its selection on its opinion regarding: a) scientific quality and timeliness of the proposed experiment; b) the match between available equipment and the proposed experiment; c) the likelihood of the experiment's being successfully completed, taking into consideration the experience of the named participants.

The IPNS-I facilities are provided without cost to qualified scientists if their research is suitable for performance at the facility, is of documented programmatic interest to DOE, is of high scientific quality and if the results are to be published in the open literature. Proprietary experiments may be scheduled with full cost recovery according to DOE policies. Outside users will have to provide their own travel and subsistence support, although Argonne Universities Association may be able to help in special cases.

IPNS-I is funded by the Materials Science Branch of the division of Basic Energy Sciences. Although proposals for irradiations of fusion magnet materials are considered, the Program Committee bases its selection of proposals on the criterion of scientific quality alone. The present budget permits source operation for approximately half of the year. Therefore, an appropriate and efficient way of doing irradiation experiments on fusion magnet materials is to fund the operation of IPNS-I for a period of time ( $\sim 1$  mo./yr.) sufficient for performing the types of experiments discussed above.

#### SUMMARY

The important aspects of the REF at IPNS-I are the dedication and flexibility of the facility. The beam intensity, frequency and periods of operation are completely dictated by the experimenter during his irradiation time. The irradiation facility not only permits irradiation down to 4.2 K, but there is also the capability of transferring samples at 4.2 K for measurement in other cryostats without warmup. This is extremely important when considering the effect of mobile defects on the measured property. The irradiation volume of  $\sim 50$  cm<sup>3</sup> allows the simultaneous irradiation of many samples. This is necessary for insulator irradiations since the present dearth of cryogenic fast-neutron irradiation data requires the simultaneous irradiation of a large number of samples for screening purposes. The large irradiation volume also permits unusual irradiation environments such as high magnetic field or stress.

Experiments that can be done at IPNS-I and that are needed for design data for fusion magnets include: 1) Multi-sample irradiations at 4.2 K of insulators to determine the contribution of fast-neutron damage. 2) The effect of periodic annealing (after fast neutron irradiation at 4.2 K) on the cumulative magnetoresistance of stabilizers with different initial resistivities. 3) Detailed dose and field dependence of various candidate superconductors during fast-neutron irradiation in the superconducting state.

#### ACKNOWLEDGMENTS

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