Final Report

for

ERDA Contract
E(11-1)-2282 Modification No. 2

for

Fuel Acquisition and Design

of A

Fast Subcritical Blanket Facility

Submitted by

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I. INTRODUCTION

The Contract No. E(11-1)-2282, Modification No. 2 was for three phases of work leading to the design of a subcritical facility for the study of fast reactor blankets. The first phase was the acquisition of the necessary enriched and natural uranium fuel for the proposed facility. The second phase included the design of the components of the facility and shielding, based on previous calculations. The third phase of the contract called for the planning and development of experiments to be performed with the facility.

II. FUEL ACQUISITION

The acquisition of the nuclear fuel identified in the proposal for use in the facility has been completed. License amendments to licenses SNM-142 and SUD-296 issued to Purdue University for the storage of the fuel were received. The 4.8% and 1.3% enriched fuel were received at Purdue University on February 19, 1974 and March 7, 1974 respectively. The natural uranium was received on September 24, 1975. The fuel is currently in storage at Purdue University.

III. DESIGN OF FACILITY

Based on the preliminary concepts and design calculations reported in the proposal for contract, the individual components of the facility and shielding were identified and preliminary specifications, designs and drawings were made. Most of the design work and the drawings were done by the Purdue Physical Plant and Central Machine Shop. Based on the work covered in this phase of the contract, cost estimates for the completed facility were obtained and used as the basis for a new proposal for the construction of the facility.

IV. EXPERIMENT DESIGN

The first step in the experiment design was to identify the types of information that would be useful and would lead to a meaningful comparison between experiment and theory. Three types of information were identified: integral reaction rates, neutron energy spectra; and gamma ray energy deposition. The second step in the experiment design was to identify and design experiments which could determine the desired quantities with the necessary accuracy and with the neutron flux and gamma radiation levels anticipated in the proposed subcritical designs.

Four different experimental techniques were selected for development for the subcritical facility. These were neutron activation of foils, solid state fission track recorders, hydrogen recoil neutron spectra measurements using proportional detectors and gamma ray dosimetry experiments using TLD (thermoluminescent) detectors. These four techniques were chosen on the basis of their sensitivity, ease of obtaining reliable results and/or the degree of expertise in the area already existing at Purdue. The first two techniques will be used to determine integral reaction rates as well as to supply experimental input to computer codes for determining the neutron spectra. The hydrogen recoil data can be used directly to determine neutron spectra. The TLD measurements will supply information on the rate of energy deposition from the gammas. As the facility is operated, other techniques may also be developed and employed to gain information.

Neutron Activation of Foils

A careful study of the effects of neutron cross section certainties in the blanket regions of both a typical liquid metal fast breeder and the subcritical facility was made. Comparison of the two sets of results showed that the effects were similar in both cases and defined the limits of

accuracy with which experiments must be made if meaningful results are to be obtained. The results of the sensitivity studies were prepared as a Purdue Report, Review of Nuclear Physics Uncertainties in Fast Reactor Blankets, PNE-75-103 (March, 1975) by Ott, Boldt and Clikeman.

One particular activation reaction, the $^{238}\text{U}(n,\gamma)$ reaction, was selected for detailed development. This neutron capture reaction was selected because of its importance in the fast reactor neutron flux calculations, the breeding characteristics of the blanket and because experimentally it will be one of the most difficult reaction rates to measure. Experimentally, the determination of the capture rate in ^{238}U is complicated by the fact that both the natural activity of ^{238}U and the decay activity of fission products from both fast fission in ^{238}U and in ^{235}U , interfere with the detection of the radiation from the ^{239}U decay chain. Review of the literature indicated and experiments verified that the best method of detecting $^{238}\text{U}(n,\gamma)$ events is to detect the 228 and 277 keV gamma rays from the ^{239}Np decay in the ^{239}U decay chain. Minimum detection levels using both a 3" dia. NaI(Tl) well type scintillation detector and a small 6 cm³ Ge(Li) detector were determined. From these measurements, the lower detection limit for a 10% efficient Ge(Li) detector has also been determined. These results show that the ^{238}U neutron capture rates can be determined with acceptable accuracy in the subcritical blanket facility. The results show, however, that highly depleted ^{238}U foils should be used with a 10-12% efficient Ge(Li) detector for the best results.

The sensitivity study for the ^{238}U capture rate and the development of the technique for determining the capture rate have been the subject of a M.S. thesis research project. The research has been completed and the thesis is currently being written. The other neutron activation rates that are planned are considered to be straightforward experiments since the parent

material for the reaction will not be naturally radioactive and there will be no competition from a fission reaction in the foil. Foils for most of the desired reactions were obtained and preliminary measurements verified the above conclusions. The only problem will be caused by the small activation cross sections of some of the materials and the lower limit with which the activities can be determined (M.S. Thesis by Kenneth R. Boldt).

Fission Reaction Rates

Original plans called for using mica solid state fission track recorders for determining the fission reaction rates in the blanket facility. After an extensive literature survey and some preliminary experiments, it was decided to use a plastic foil, Makrofol, instead of mica for the fission track recorder. Two main reasons for the choice of Makrofol were the low background of the foils compared to mica and the fact that Makrofol could be used with a spark counter to count the fission tracks.

Mica, being a mineral found in nature, has been exposed to cosmic radiation and hence has already recorded tracks due to the cosmic radiation. Makrofol is man made and has only a very limited exposure to cosmic radiation. It has a very low background, as was shown from our experiments.

The Makrofol also lends itself to use with a spark counter for counting the number of fission tracks. Mica is usually counted by an optical scanning system, the simplest of which is a microscope. Counting the fission tracks with a microscope is long and tedious and lends itself to errors. On the other hand, a spark counter is simple as well as fast and, as our experiments have shown, can give consistent results.

Three problems of the spark counting system were identified and experiments were conducted to solve each problem. A ^{252}Cf fission source was used to expose the foils used in these experiments. The first problem was to

develop a procedure for etching the Makrofol recorders so that the spark counter would give consistent results. This also involved modifications of the spark counter and its operating voltages.

The second problem was to determine the absolute efficiency of the spark counter system. For this purpose, a series of foils were counted on the spark counter and scanned visually with a microscope. The experiment gave consistent results and established the efficiency of this particular spark counter and operating procedure.

The third problem was to determine the density of tracks over which the system is linear. If the track density is large, then the probability is large that two tracks will be sufficiently close that the spark counter will not record both tracks, but rather give only one count. The range over which the spark counter would give linear results was determined. In addition, a correction factor was determined that could be applied to extend the useful range of the system.

The work of developing the spark counter and operating procedure was the basis of an M.S. thesis. The work is completed and the thesis is now being written (M.S. Thesis by Michael A. Hannah).

Hydrogen Recoil Neutron Spectrometer

The technique of determining neutron spectra by analyzing proton recoil spectra recorded in a hydrogen-filled proportional detector has been operational at Purdue University since 1970. The original spectrometer system was based on an early system developed by E. F. Bennett at ANL (ANL-6479 and ANL-6480). However, the spectrometer system used at Purdue was limited to a lower detection energy of about 30 keV. Two major modifications to the spectrometer system for improved discrimination against gamma rays were proposed to extend the useful range of the spectrometer down to

10 keV and below. Each of the two new ideas was the basis of M.S. thesis research. Both modifications were successful and experimental results showed that with a modest addition of equipment (as opposed to a two parameter, multichannel analyzer costing about \$20,000), the useful range of the spectrometer could be increased to the energy range below 10 keV. The details of the two spectrometer systems and the results obtained with each system will be given in the M.S. theses by James Klapproth and David Vehar.

Although the experimental results showed one technique slightly superior to the other, both of the systems required a great deal of time for data acquisition when compared to techniques using two parameter, multichannel analyzers. The major drawback of the long data acquisition time is the increased emphasis that must be placed on the long term stability of all of the components in the spectrometer system. The conclusions that have been reached from this research are that for proton recoil experiments in the fast blanket facility, a two-parameter, multichannel analyzer will be required. Several ideas for combining the existing gamma discrimination techniques with two parameter analyzer techniques have been proposed and work on these ideas is continuing.

TLD (Thermoluminescent) Detector Research

Work in the area of gamma-ray energy deposition centered around selecting a TLD dosimeter for making the gamma-ray dosimetry measurements. The best dosimeters for a measurement are usually those dosimeters that will have approximately the same Z (charge of the nucleus) as the material in which the measurement is to be made. Because most of the applications of TLD dosimetry have been in the area of personnel dosimetry, most of the development work has been on low Z TLD dosimeters. In the blanket of breeder reactors, most of the gamma rays will be interacting with steel or uranium,

medium and high Z material. After a thorough literature survey, $\text{CaSO}_4:\text{Dy}$ was selected on the basis of a material with thermoluminescent properties, medium Z, low neutron response and commercial availability. One disadvantage of $\text{CaSO}_4:\text{Dy}$ is the non-linear gamma-ray response of the material to gamma rays with energies below 100 keV. A series of experiments have been designed 1) to verify the reported energy response; 2) to investigate the properties of dosimeter coverings that might be used to make the response of the detector more nearly linear; 3) to verify the reported fading and annealing properties of $\text{CaSO}_4:\text{Dy}$; and 4) to establish the reproducibility of measurements utilizing the $\text{CaSO}_4:\text{Dy}$ dosimeters. These experiments are being conducted as part of the research for an M.S. thesis and are expected to be completed by the end of August, 1976 (M.S. Thesis by Janet Westbrook).

Continuing Experiment Development

The purpose of this portion of the contract was to supply equipment needed for the development of experiments for a fast blanket facility. The basic experiments have been identified, the major experimental problems identified and solved and the results used in preparing proposals to the Energy Research and Development Administration (ERDA) and the Electric Power Research Institute (EPRI). Work completed under this contract has also pointed to several areas where improvements in the experimental measurements might be obtained by improved equipment or techniques. This continued effort of improving the experiments will be carried out by students doing thesis research using equipment that either already existed or was acquired under this contract. When the contracts with ERDA and EPRI are signed, additional money for equipment and supplies will be available to implement the experimental design and equipment recommendations that have been developed under this contract.

Disposition of Government-Owned Property

We request that the nuclear fuel and experimental equipment acquired under this contract be transferred to the new ERDA contract No. E(11-1)-2826 subject to the contract's execution.

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