AN ABSTRACT OF THE THESIS OF

STEPHEN H. SHEPHERD for the degree MASTER OF SCIENCE

in Nuclear Engineering presented on July, 30, 1976

Title: An Experiment for Accurate Determination of Time-Dependent Fission Product Energy Release After Thermal Neutron Fission of ^{235}U

Abstract Approved: Redacted for Privacy

Bernard I. Spinrad

An experiment is described which utilizes thermometry to establish an improved average decay scheme, per fission, for the determination of nuclear reactor afterheat. It will measure decay heat from the moment of scission to a period several hours later.

The calorimetric apparatus is to be placed adjacent to the core of a pulsed research reactor where the ^{235}U sample in a bismuth dilatometer is irradiated. Absolute, integral, and differential energy release will be measured in situ. The expansion of the thermometric fluid, bismuth, will be determined by weighing the quantity of fluid expelled through a capillary on a precision electrobalance.

High accuracy is obtained through the use of extensive calibration which allows systematic errors to be neglected. This high accuracy is expected to supplement and validate computer studies of decay heat for short times after fission.

An Experiment for Accurate Determination of Time-Dependent Fission Product Energy Release After Thermal Neutron Fission of ^{235}U

by

Stephen H. Shepherd

A THESIS

submitted to

Oregon State University

in partial fulfillment of
the requirements for the
degree of

Master of Science

Completed July 30, 1976

Commencement June 1977

APPROVED:

Redacted for Privacy

Professor of Nuclear Engineering

Redacted for Privacy

Head, Department of Nuclear Engineering

Redacted for Privacy

Dean of the Graduate School

Date thesis is presented July 30, 1976

ACKNOWLEDGEMENT

In designing this experiment, it has proved helpful to "bounce" ideas and concepts off of several persons. Their restraint when confronted by my exuberance, and their critical analyses contibuted to this thesis. Additional contributions were made by those faculty here at OSU and elsewhere whose teaching I regard as exceptionally superior, and whose influence, although not specifically incorporated in the thesis, is evident.

My gratitude is extended to Frederick H. Mattes, Arthur G. Johnson, Arthur D. Payton, Walter D. Loveland, Robert L. Purbrick, Sanderson M. Smith, and, Bernard I. Spinrad.

I give additional thanks to the TRIGA reactor operations staff, Terry Anderson, Bill Carpenter, and, Steve Bennett.

However, the majority of my appreciation is reserved for my wife whose encouragement and technical advice led to the completion of this project. Thank you.

TABLE OF CONTENTS

List of Illustrations

List of Tables

An Experiment for Accurate Determination of Time-Dependent Fission Product Energy Release After Thermal Neutron Fission of ^{235}U

There is considerable need for a reliable experimental determination of the decay heat after reactor scram for safety analysis related to a loss of coolant accident (LOCA). Previous experiments have established standards with associated errors ranging upwards of seven percent.[42] Computer modeled calculations of shutdown heating have been determined to have lesser errors.[32a] These studies cannot be validated by such poor experiments. This thesis details an experiment which would utilize thermometry to provide the required information within an uncertainty range of 2% for the integral heat, as shown below:

Source of Uncertainty	Magnitude
Normalization of fission product gamma escape from dilatometer	0.25 %
Normalization of delayed neutron escape from dilatometer	0.13 %
Calibrational uncertainty (See Table 3.)	1.9 %
Net Uncertainty	1.92 %

A small sample of ^{235}U is to be alloyed with natural bismuth (^{209}Bi) to form a small pellet which is then placed at the center of a sphere of solid bismuth inside an adiabatic dilatometer. The entire mass will then be melted and have its level adjusted to a suitable initial position. The

dilatometric apparatus will undergo a pulse irradiation of neutrons. This neutron bombardment causes heating to occur in the sphere due to both the coulombic and prompt γ energy of fission (prompt or direct heating), and the β and γ heating due to fission product decay (afterheat). The diameter of the sphere will be about 15 cm (6 inches) so that 99% of the fission product gamma ray photons will be attenuated in the bismuth. All beta emissions will be absorbed in the sphere since betas are charged particles will a low power of penetration.

Bismuth has a very low heat capacity (similar to that of mercury) and consequently will experience a significant temperature rise due to this heating. This rise in temperature causes an expansion which will expel liquid bismuth from the sphere through a capillary, increasing the mass in a conical bismuth reservoir. (See Figure 1.) As the sample cools, the bismuth will contract and return back into the sphere. Thus, the temperature of the bismuth sphere is directly proportional to the mass of bismuth in the reservoir. By the use of a sensitive electrobalance, in situ, and a remote chart recorder, the quantity of bismuth expelled may be displayed as a function of time. Through suitable calibration, this may be converted into a plot of temperature and thus energy release, both integral and differential, over time.

The calibration for the number of fissions in the sample

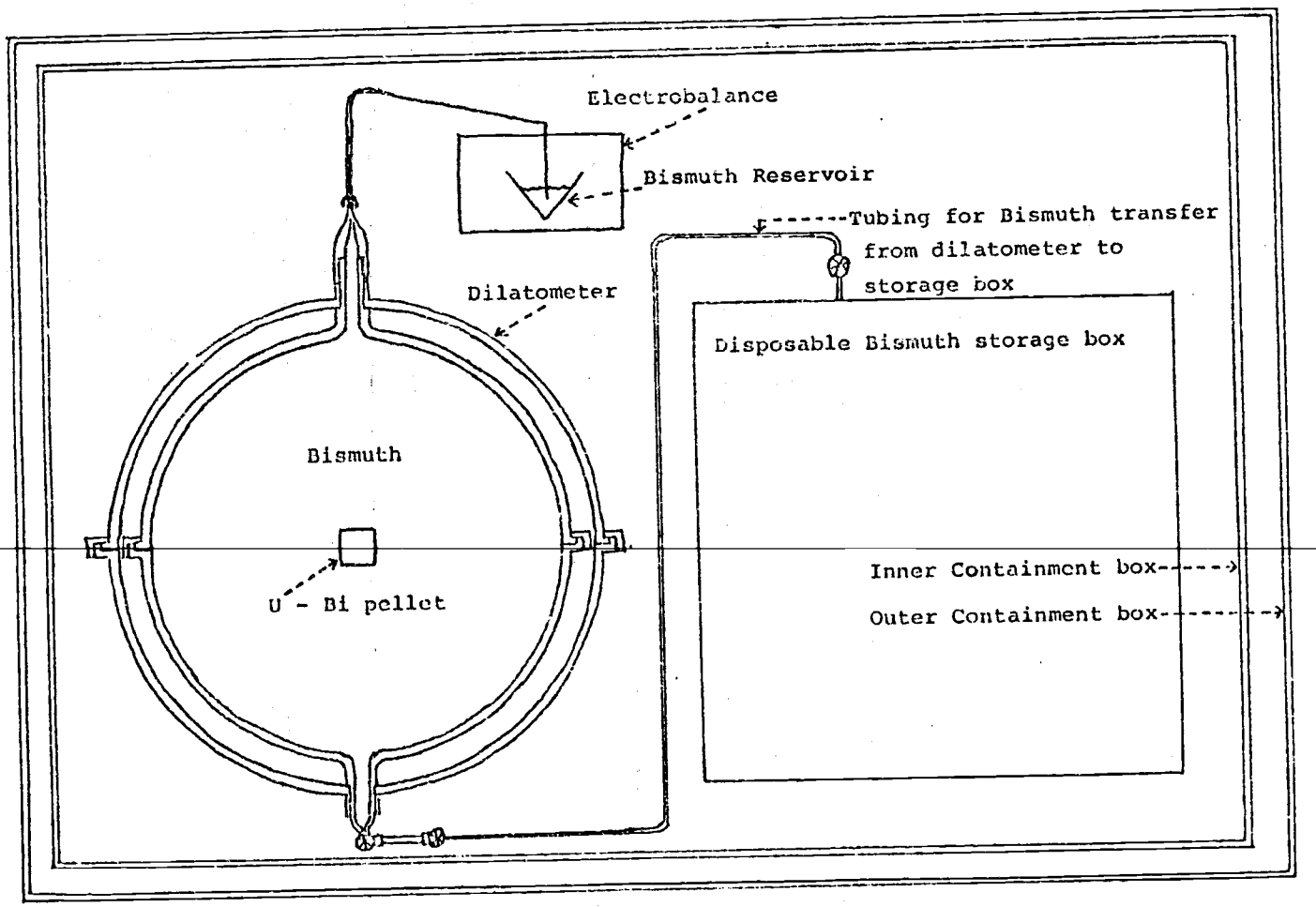


Figure 1

The Bismuth Calorimeter for Decay Heat Measurement

is available immediately. The prompt energy release from fission causes a temperature spike on the chart recorder. The size of the spike is proportional to the energy input. This determination is separated in time from afterheat and can be ascertained independently.

The neutron source for the experiment will be the Oregon State University TRIGA Reactor (OSTR). A short burst of neutrons is obtained from the reactor by rapidly removing the transient control rod. While the reactor is licensed for a 4000 MWt peak power pulse, pulse operation is routinely conducted at about 2600 MWt. The pulse duration is approximately 10 msec. This makes available a high neutron fluence, in excess of 5×10^{13} neutrons/cm^2, adjacent to the core during pulse operation. Any other pool type research reactor, with pulse mode capability, could be used if the requisite flux can be achieved.

The apparatus is located close to the reactor core to take advantage of the locally high neutron flux. Therefore, the experiment needs to be shielded from gamma ray photons originating in the core to avoid their heating effects. If not attenuated, they will be mistakenly included into the afterheat energy measured. Shielding can easily be performed by interposing lead bricks between the experiment and the core. The lead bricks will shield the gamma flux but will not substantially reduce the neutron flux.

Adiabatic isolation from the surrounding environment is

accomplished by the use of silvered vacuum regions (similar to a dewar flask). Extensive calibration of the system will decrease the error range for the heat losses due to the imperfectly adiabatic apparatus. Replicate volumetric technique should yield an error of less than 0.1% in determining the temperature of the bismuth. Blank runs, ie., without ^{235}U can be performed in or out of the reactor to ascertain the 'background' characteristics of the apparatus. These can then be subtracted out from an actual run to yield the true energy release of the run.

The experiment is sufficiently broad in scope of application that, using the same apparatus, further decay heat studies of other fissile radionuclides can be undertaken. A logical followup to this study would examine the energy release from thermal neutron fission of ^{239}Pu. Since about 50% of the fissions in light water reactors occur in ^{239}Pu at the end of fuel life, this particular secondary analysis should be of great interest. Other determinations could include the decay heat from thermal fission of ^{233}U and ^{241}Pu. This thesis will address only the energy released from fission of ^{235}U. The feasibility calculations, detailed herein, would be approximately the same for other fissile nuclides. Radiological hazards would not.

The experiment provides some novel problems for a university scale health physics department. It involves both a thermally and radiologically hot sample. Direct contact with

the sample can not be tolerated. The design of the transfer box (see Section IV), together with standard radiation precautions, will minimize the release of the sample from the contained volume so as not to require elaborate and expensive remote handling facilities. The sample is to be irradiated within a precision, reusable, dilatometer and then transferred to a disposable storage container. The storage boxes from the several runs will be allowed to decay and then shipped to a government repository. (^{235}U is special nuclear material and must be safeguarded and accounted for.)

The precise data which this experiment will yield will enable regulatory agencies to analyze more exactly a loss of coolant accident. Current practice is to accept only the worst possible (greatest) heat source and then to provide an additional, liberal safety margin when designing emergency core coolant systems. If the error range is reduced, then, for the same mean value of decay heat, there will be a lower maximum heat source. This could result in an uprating of the power level of operating reactors, leading to an additional source of electric power not now available.

Necessity for the Experiment and a Review of the Berkeley Experiment

The Physical Nature of Fission Energy Release

The majority of energy liberated from the fission process is released promptly (within 10^{-15} seconds)[21] after scission through the slowing down of the fission fragments. This release requires two conversions of energy. First, the particles repulse each other due to potential, coulombic, energy which is then transformed into kinetic energy of motion. Second, this kinetic energy is dissipated as heat energy through the electromagnetic interaction of these highly charged particles with the adjacent atoms.

The other contributors to the energy transmission from binding energy to released heat energy are prompt gamma photons, prompt and delayed neutrons, neutrinos, and, beta and gamma emission from the decaying neutron rich fission fragments. Prompt neutrons and neutrinos are also released in the same 10^{-15} second time frame during which coulombic energy is dissipated.[21] The prompt fission gammas are emitted within 10^{-11} seconds of the fission split.[21] These time ranges are so narrow that they may be considered coincident to fission for all purposes except detailed nuclear structure analysis.

Once the control rods are inserted into a nuclear reactor, stopping the chain reaction, the only heat sources remaining will be decay heat and delayed neutrons.

Delayed neutrons arise from fission products which are particularly unstable against neutron decay, such as those nuclei with one neutron more than a neutron "magic number." (19,25)

They will be emitted from the parent radionuclide with a characteristic half-life.(21) The longest half-life of a delayed neutron precursor is ^{87}Br with a 55.6 second half-life.(17) Clearly, delayed neutrons will not contribute to the afterheat for more than the first few minutes after fission.

For ^{235}U, 0.65% of the total neutron population is delayed.(19) Since only about 2.5% of the energy of fission is manifested in neutrons, this means that delayed neutrons represent less than 0.0163% of the total energy of fission. As the fission product α, β, and γ afterheat is about 3.5% of the total energy of fission, delayed neutrons make up less than 0.47% of the afterheat.

This amount is quite small and will not substantially affect the determination of total afterheat energy if it is not included in the determination due to leakage. It should be noted that this induces an error of one-half of one percent in the the proposed experiment if it is not normalized.

There is a wide range of half-lives exhibited by the fission product radionuclides due to their varying nuclear stability. Nuclides which have long half-lives such as ^{90}Sr or ^{137}Cs (28.9 and 30.2 years, respectively) (17) are relatively stable against beta decay as they both lie at one of

the unusual "magic numbers" of the shell model of nuclear structure.[15,25] Those species which are greatly removed from islands of nuclear stability such as ^{110}Tc or ^{146}Cs have half-lives on the order of tenths of a second (0.83 and 0.19 seconds, respectively).[17] It is these short lived nuclides which create difficulties in modeling decay heat by computer.

A computer decay heat simulation, such as CINDER,[11] while difficult to program, is actually very simple arithmetic. It is necessary only to feed in the characteristic decay scheme of all fission products, give the computer the proper mix of the ingredients, and they ask it to 'decay' the radioactive materials for a given time interval according to the well validated exponential recipe. At that point, the computer prints out the new mix of nuclides, sums up the products of the individual nuclide's decay energy and radioactivity, and thus yields the instantaneous decay energy. A number of such calculations, at different times, will produce a well documented decay heat curve.

Unfortunately, the accuracy of such a calculation depends solely on the accuracy of the input decay schemes. The computer will retain any input errors throughout its calculations. Therefore, if speculative data is given to the computer, only speculative data will be output.

The magnitude of the experimental errors in determining the decay schemes varies as the inverse of the half-life.

Those nuclides which have long half-lives lend themselves easily to sophisticated and time consuming analysis. Such analysis is generally quite accurate. On the other hand, when the nuclide of interest exists for one second or less, only relatively quick and dirty observations may be made. These measurements may typically have an error of an order of magnitude or more. Indeed, for many radionuclides there can be no direct experimental observations of their properties using today's state-of-the-art equipment.

In these cases, quantum mechanical models of nuclear structure and decay are proposed. Information obtained in this manner not only carries with it the inaccuracy of such mathematical calculations, but, also the possible intrinsic error of the model. Indeed, these calculations are at the current frontiers of human knowledge. Logically, and historically, research in nuclear structure can modify or outdate the antecedent knowledge. One precendent for such an intrinsic error was the outdating of many predictions of the shell model when large electrical nuclear quadrupole moments were detected in nuclei which are far from closed shells.[15] The collective model was then proposed as rectification. Further refinements have been evinced by the developement of the unified, the optical, the statistical, and the Brueckner—Bethe—Goldstone models of nuclear structure.[6,15,25,26] Calculations had to be revised with the introduction of each theory.

Thus, characteristic data about such transitory constituents of nature has been difficult to obtain and is often of dubious accuracy.

Because of such inaccuracies inherent to the computer simulation, it is necessary to experimentally validate the decay schemes. Until science and technology advances, it is not possible to verify each scheme individually. The thrust of the experiment detailed in this thesis, and those currently being undertaken elsewhere, is to provide a quantitative assessment of the total time-dependent decay energy of all fission product radionuclides.

Relation of Energy Release to Nuclear Reactor Safety

A loss of coolant accident (LOCA), one hypothetical reactor design basis accident, has the following basic scenario: 1) A double ended, 'guillotine', pipe break occurs, interrupting the normal flow of cooling water to the reactor core. 2) The reactor is scrammed by a rapid insertion of control rod neutron poisons (absorbers) into the core. 3) An auxiliary coolant system, emergency core coolant system (ECCS), is actuated to deliver water to the core in sufficient quantity to avert a potential core meltdown due to the continuing heating caused by radioactive decay (afterheat).

There are four prime considerations in assessing the possible consequences of the accident when performing safety analysis. First, will the pipe rupture? Will the scram

circuit operate in a timely manner? Will ECCS actuate? And lastly, will the quantity of cooling water delivered to the core be sufficient to avoid meltdown? Wash-1400 contains an extensive analysis of the first three questions.[38] The last question requires that the decay heat source be known. This is the subject of this thesis.

Previous experiments have set standards for afterheat on which regulatory judgements as to the adequacy of cooling water flow rates for a rated power level have been made. These experiments have had large associated error ranges.[42] Analytical, or computer studies, have been demonstrated to have lesser errors, but, the previous experiments do not yet give empirical validation of the studies.[32a] Because of this, the conservative regulatory judgements have prompted large design safety margins to ensure that the 'worst possible' value of afterheat is much less than cooling capacity.

If the basic data on radioactive decay heating is improved, with a reduction in the error range, the design margins can be safely and justifiably reduced. Since decay heat is directly related to the reactor power level (strictly a function of the number of fissions), the immediate consequence of this change would be to allow reactors to operate at a higher power level than they are currently rated for. Such an uprating of reactors would produce more electric power. This certainly would help alleviate present and future energy shortages.

A Review of the Decay Heat Experiment at UC Berkeley

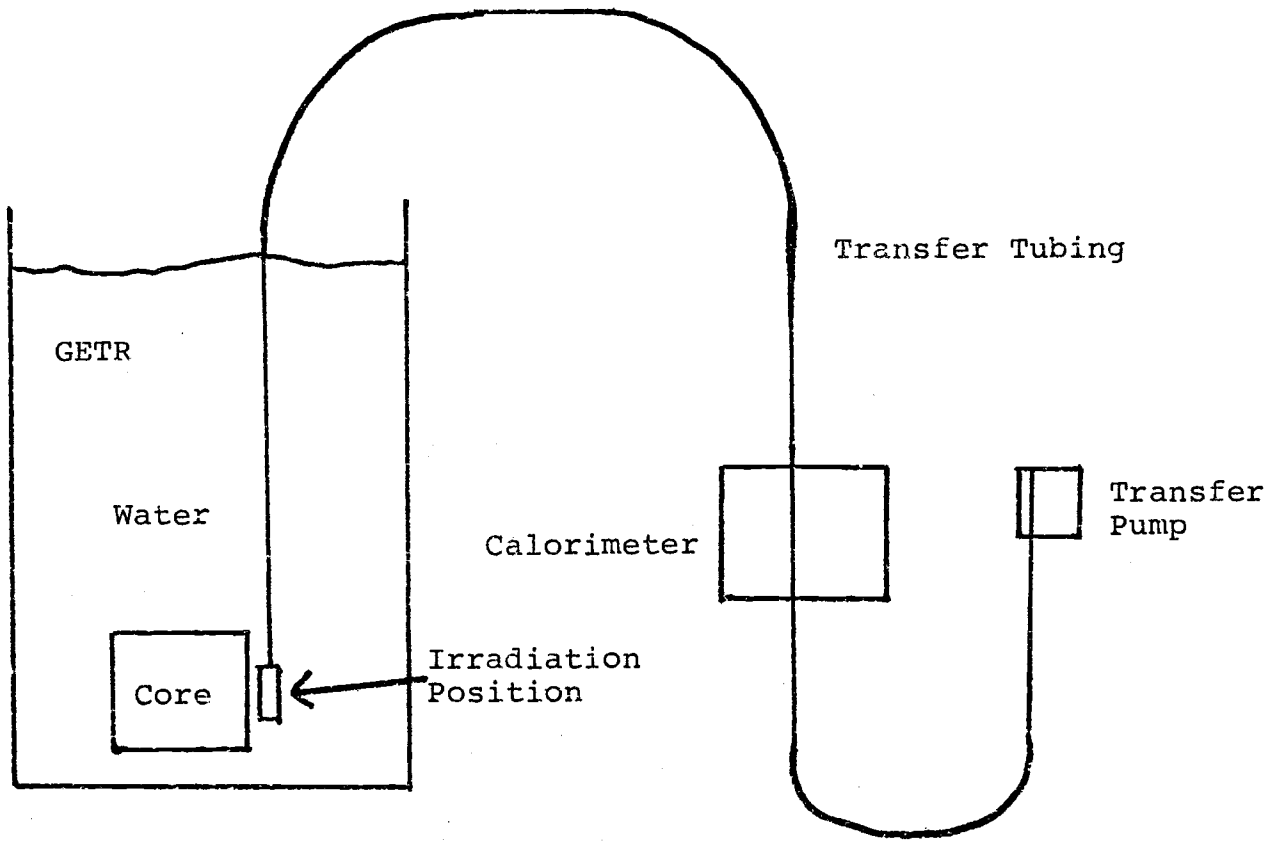
Three categories of experiments have been or are being performed or designed to measure fission product afterheat. These are: 1) Measurement of the fission product beta release rates.[3,18,23,24,34] 2) Measurements of the fission product gamma release rates.[4,12,27,40,44] 3) Calorimetric determination of total fission product heat release.[39,43] The first two methods have been analyzed elsewhere.[42] This paper designs one calorimetric method. Another calorimetric method is being conducted at the University of California at Berkeley (UCB) under the direction of Prof. Virgil E. Schrock.[39] Their experiment, while similar in physical description to the proposed OSU project, has a different philosophy of execution.

Figure 2 shows a general schematic of the Berkeley experiment. The fissile sample, a 24 mg uranium wire, is enriched 40% in ^{235}U. It is clad in a tapered aluminum housing.

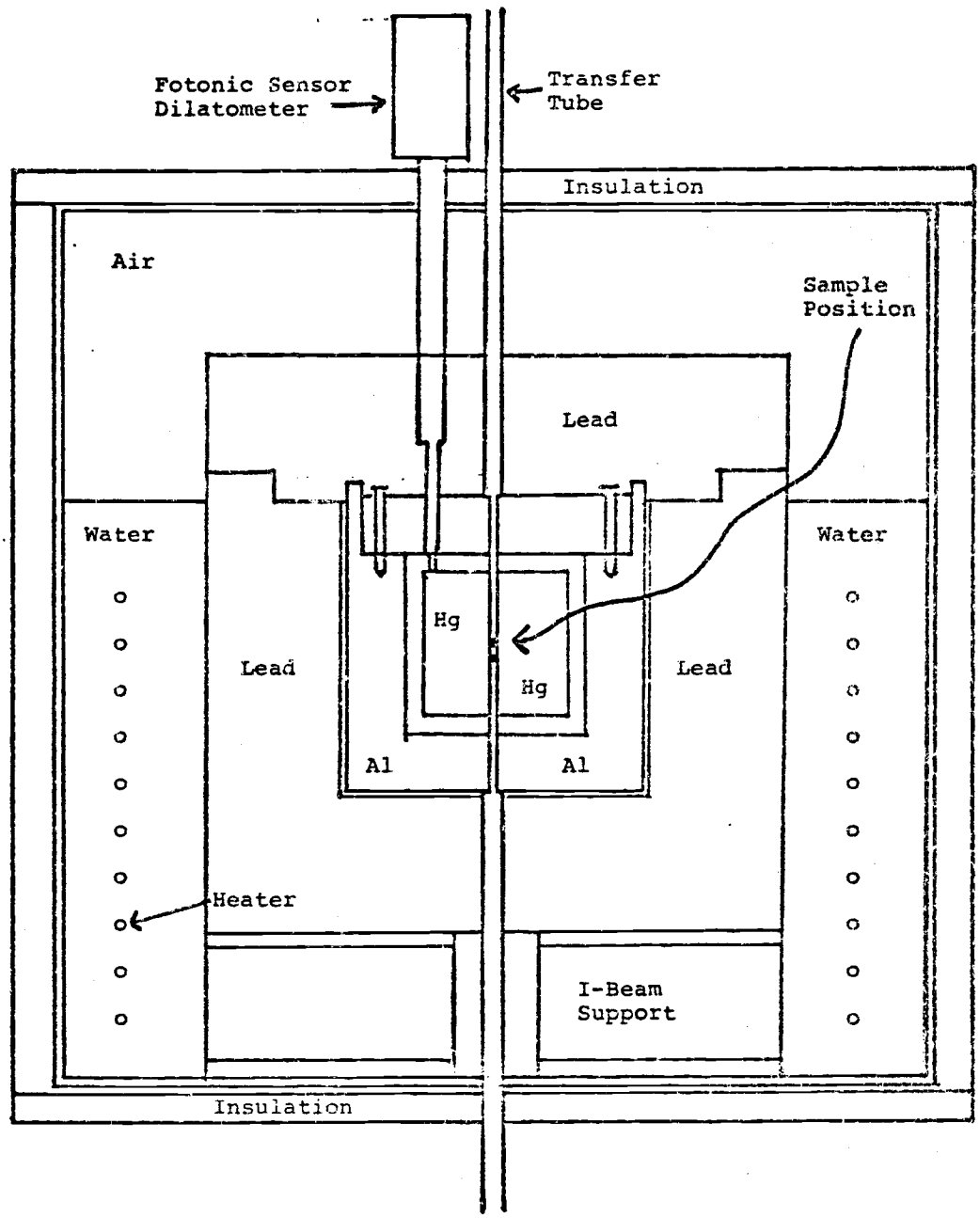
Irradiation will be varied from 1 to 11 days adjacent to the core in the General Electric Test Reactor (GETR) at a flux of 10^{14} neutrons/cm^2 second. Cooling water will be supplied to the sample while in the core to remove the heat of fission. After the desired irradiation, the sample will be hydraulically transferred by water to a calorimeter external to the reactor. The calorimeter is shown in Figure 3.

The calorimeter utilizes mercury as the thermometric

Figure 2



Schematic of the Berkeley Experiment



Berkeley Calorimeter for Decay Heat Measurement

Figure 3

fluid. It will be heated by absorbing 99% of the fission product gammas as well as by the transfer of heat from the aluminum-uranium capsule which is caused by beta and alpha decay. Due to delays in heat conduction, the beta energy time constant will be 0.5 seconds.

The detection of the level in the UCB dilatometer is accomplished by a fiber optic visual scan device, a "fotonic sensor." It will have a precision of 2.5×10^{-8}m (1 μ-in). Readings will continue for 28 hours.

Four parameters are used to calculate the heat source. A constant time base recording will be made of: 1) The "fotonic sensor" (volts); 2) The output of the thermopile (mv); 3) The five bridge thermistors of the calorimeter (mv); and, 4) The pressure transducer (mv). Thus, the rate of heat loss from the dilatometer can be added back in to the registered heat of the mercury. The volume of expelled mercury can be corrected from experimental conditions to standard conditions of temperature and pressure. It is then a relatively simple task to obtain the absolute heat input.

There are self-powered neutron detectors at the terminus of the hydraulic system in core. These are useful in assuring a constant power history during the irradiation. With suitable calibration, they should also serve as a secondary determination of the number of fissions which took place in the sample. The primary method is radiochemical γ ray spectroscopy of the wire after removal from the calorimeter

(presumably analyzing for ^{99}Mo whose fractional fission yield is well known). Gamma spectroscopy of ^{99}Mo is routinely performed with 99% accuracy.

The most significant difference between the UCB project and the OSU project is the time span of the irradiation. Nuclear decay of the fission products will occur during the UCB irradiation. Therefore, the heat source data as determined in the calorimeter will not contain all the afterheat of all the fissions. Some afterheat will be dissipated during the neutron bombardment. Since irradiation at OSU will be during a 10 msec pulse (as opposed to the 1 to 11 days at UCB) all fissions are restricted to essentially one point in time and thus total afterheat is recorded.

Because the afterheat will be dependent on the history of the reactor power level during the irradiation period at UCB, it is important that a near constant power level be maintained. This is particularly vital during the last hour before sample transfer. The sensitive, self-powered neutron detectors should keep extremely good track of this.

There is a short time delay in removing the fissile sample from the GETR and obtaining temperature information from the calorimeter. The OSU calorimeter, however, is designed to operate in situ adjacent to the core. There is no time delay in recording afterheat. In fact, the primary determination of the number of fissions is directly available due to the prompt energy release temperature spike (see Section

V). UCB will rely on the somewhat more difficult and less accurate radiochemical γ determination.

Both thermometric fluids, mercury and bismuth, will absorb 99% of the gamma ray energy. Although the experimental apparatuses are approximately the same size, there is a basic difference in geometry. The OSU concept is a near spherical ball of bismuth. UCB's is a right cylindrical annulus of mercury. This second version will allow gamma escape vertically through the transfer tube: however, the additional mercury in the diagonal directions will provide extra shielding so as to yield a net 99% absorption of the gamma rays.

Since the "fotonic sensor" fiber optic device will be of high precision, little difficulty should be experienced in determining the temperature of the mercury after appropriate calibration is performed. The OSU electrobalance would work equally well in the UCB project.

One last important difference in the approaches: At OSU, the uranium is in an intimate mix with the thermometric fluid so that there will be no time delay in registering beta energy. The UCB delay of 0.5 seconds should be of little importance, however, as their experiment will not yield data for 10 seconds after the end of bombardment due to the settling time required for the capsule after its hydraulic transfer. One-half second out of 10 is not a very great error and it quickly becomes insignificant at longer

times.

The experiment proposed in this thesis has several advantages over the other experiments. 1) There will be no time delay betwee irradiation and the measurement of afterheat. 2) It measures both beta and gamma energy release simultaneously. 3) There is a well defined reference time of fission. 4) Because it is self contained, with the exception of electrical connections, it can be moved to any pool type reactor which can supply the requisite or higher neutron flux. On the negative side, it will be more difficult to work with or near the irradiated bismuth. See Section VI for a detailed analysis of this problem.

The elucidation of the average decay power, per fission, will enable sophisticated calculations of reactor fuel temperature due to afterheat. These calculations can utilize integration over the power history curve to provide an expected fuel temperature for any desired power history and coolant flow rate.[16] It will provide needed data which can be used as input by regulatory agencies such as the US Nuclear Regulatory Commission when evaluating LOCA safety. It is both a desirable and needed project.

Feasibility of the Experiment

When an experiment is designed, it is necessary to know what its purpose is, what data it will produce, how it yields that data, and how reliable its conclusions are.

This project will determine the time dependent energy release of fission. It will detect all energy in the form of fission fragments, all beta and alpha decay energies, and 99% of gamma photon energies. It will not detect energy given up in the form of neutrinos. Neutrino energy, however, does not contribute to afterheat.

One of the oldest ways of measuring temperature, and thus indirectly, heat, is to use a thermometer. This experiment is essentially a big, fancy thermometer.

Bismuth is used rather than mercury as the thermometric fluid because mercury has much too high of a neutron capture cross section, 370 barns,[17] to be used. Bismuth's cross section is 35 millibarns,[17] approximately one-ten thousandth that of mercury. It is important to use materials with low thermal neutron absorption cross sections because they will not substantially attenuate the neutron flux before it reaches the fissile material.

The fissile material, ^{235}U, is placed at the center of the thermometric liquid. When irradiated by thermal neutrons it will fission. The heat of fission and its afterheat causes heating in the fluid. The fluid expands through a capillary and we have our thermometer.

The harsh radiation environment at the irradiation position precludes direct, visual monitoring of the height of the capillary column—the traditional manner of reading a thermometer. Indirect visual methods such as closed circuit television are sensitive to radiation and are too expensive to buy and regularly replace. They would give poor (or nonexistent) data during the pulse and doubtful data thereafter. The purpose of visual methods is to determine how much fluid has been expelled from the bulb reservoir. The same task, with very much greater accuracy, can be done by using an electrobalance to weigh the expelled bismuth.

The output from the electrobalance transducer is analog voltage. It is quite easy to utilize electronic differentiators so that integral, instantaneous, and differential energy release can be supplied to the experimenter. These three quantities can be simultaneously displayed on a chart recorder to provide time synchronized records. Alternatively, or additionally, an analog to digital converter could be used to store the results on magnetic tape for subsequent analysis.

Weighing is an exceptionally precise and accurate means of analysis. A commercial balance suitable for this determination is accurate to ± 0.1 μg. As the lowest expected weight change will be on the order of milligrams, this leads to a possible error of only 0.01% in determining the weight and therefore the temperature of the bismuth, much better

than the total calibrational accuracy which is expected to be 1.9%. The net relative error is 1.9%.

The dilatometer has been chosen to be constructed out of Invar, a steel, because of its low linear coefficient of thermal expansion: 2×10^{-5} cm/cm^{o}C[1] for the temperature range of this experiment. Such an expansion can be easily cancelled out by the procedures given in the calibration section. There is, additionally, only very minor solvation between bismuth and iron (the major constituent of Invar) for the planned experimental durations and temperatures.[2] Invar will not appreciably affect the performance of the apparatus as it will not change size or react with the thermometric fluid.

Bismuth is a good choice for a thermometer above 280^{o}C. It melts at 271.3^{o}C.[41] Its low heat capacity, 0.167 joule/g^{o}C,[8] is similar to mercury so that even a minor input of heat will result in a measurable temperature rise. The thermal expansion coefficient is 1.8×10^{-4}/^{o}C[1] which means that a 2000 jould heat input will raise the temperature 0.65^{o}C and thus expel 2.15 g bismuth from the 1850 cm^{3} dilatometer. (The 2000 joule input would be approximately the energy from 1 g of ^{235}U undergoing a 1300 MWt pulse on the OSTR.) Again, such a quantity of expelled bismuth is easily and accurately measured by the electrobalance.

Highly enriched, 93.15+ w/o, ^{235}U will be the fissile material for the initial afterheat determination. Future

studies can consider other materials. Uranium is soluble in bismuth. A temperature of 300°C will allow 1% uranium to be dissolved in bismuth.[2] The alloy is then cooled and allowed to solidify as a pellet.

This is one area in which preliminary research needs to be performed. For small quantities of fissile material, 0.1 gram or less, the pellet size is quite small. If however, a 1 gram sample of ^{235}U is desired, the pellet needs to be 10 cm^3, that is, it would have a diameter of 2 cm and a length of 3 cm. This size would reduce gamma ray absorption by the bismuth a bit, allowing heat energy to escape the dilatometer. Uranium is, however, 20% soluble at 1000°C.[2] The pellet size would return to small dimensions.

The problem is that the bulk bismuth in the reservoir would still melt at about 272°C. The pellet remains intact until the 1000°C temperature is reached. During the intervening time, it would be free to move away from the center of the sphere. Although bismuth is less dense when solid than when liquid, the 20% uranium would create a pellet with a higher density than the surrounding fluid so that the pellet would sink to the bottom of the sphere before it melted. The gamma absorption by bismuth is now nonexistent.

Several courses of action are available to remedy the problem. First, if the melting migration problem is not solvable, a low temperature pellet can be made and placed in an enlarged dilatometer which would absorb 99% of the gammas.

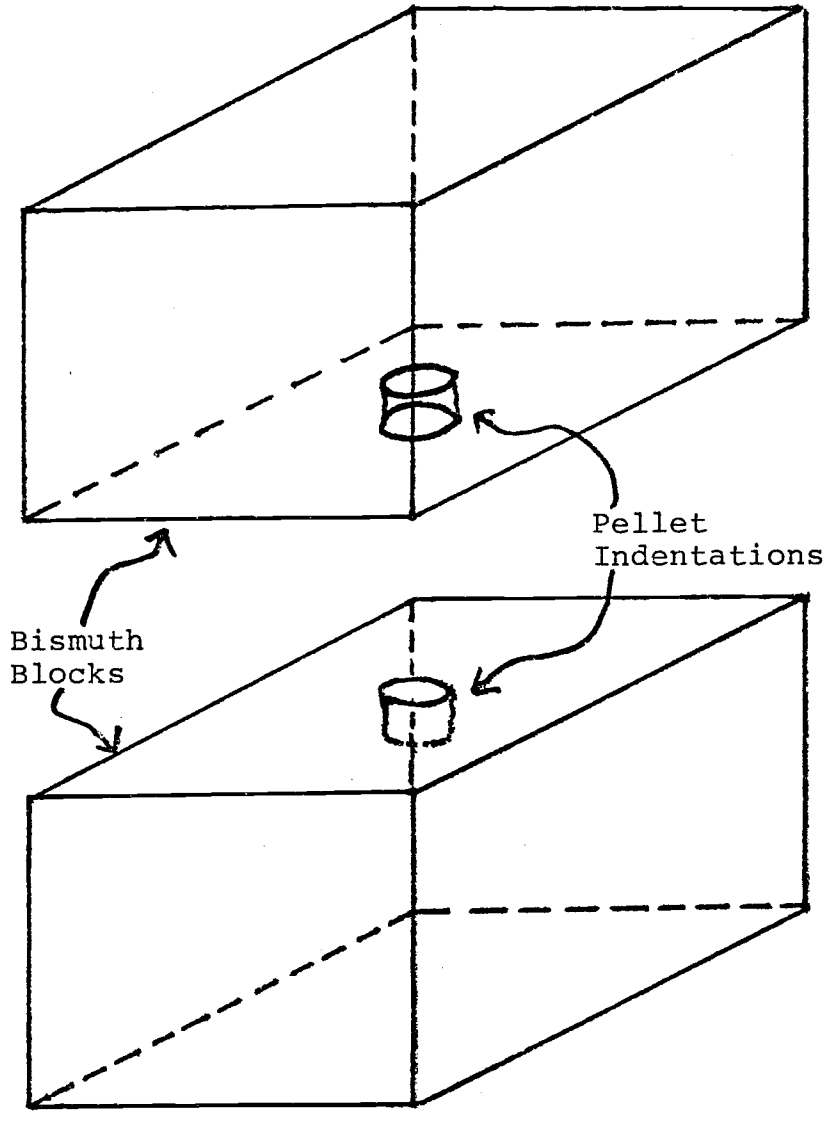
This would mean that smaller samples would have an attenuation of greater than 99%. Such a change can be calculated so that all runs may be normalized.

A second action would be to keep the size of the dilatometer constant and normalize the change of attenuation for the low temperature pellet. That is, accept attenuation of about 97% of the gammas. This method should be equal in accuracy to the foregoing example. It would require less bismuth and thus have lower radiological and dimensional consequences. On that basis, it is the preferred action should the following attempts at preventing migration fail.

It is quite simple to determine if migration occurs. Simply mock up the bismuth-uranium system, melt it, and allow it to solidify as shown in Figure 4. (Caution: Do not do this in the calibrated dilatometer for reasons specified in the procedure section.) Then, take samples at various positions in the block, dissolve in nitric acid,[5] and measure the radioactivity of ^{238}U to test the hypothesis of migration. If the hypothesis is incorrect (which would be very surprising), then there is no problem and no corrective action need be taken. An alternative detection method to the counting method is to utilize neutron radiography. Prepare a cross section and radiograph using an indirect activation transfer method. If the cross section can be easily cut, then this should speed up the determination.

If, as expected, there is migration, serious considera-

Figure 4



Mockup of Bi-U System to Ascertain

Severity of Pellet Migration

tion should be given to some sort of wire support inside the dilatometer which would hold the pellet in place until it melted. While the contribution to heating from the gamma decay in the wires can be calibrated out in a blank run, it would be regrettable to complicate the apparatus to any great extent.

The decision as to a preferred method of dealing with samples of ^{235}U in greater than 0.1 gram weights will need to be based on an assessment of the severity of the migration problem, and, the difficulty of installing wire supports. The decision cannot be made, a priori, in this thesis. Experimental data and experience with the apparatus is required. It should be noted, however, that 0.1 gram samples appear adequate for valid results and that normalization should be of sufficient accuracy for the small dilatometer, large pellet method to yield defensible data if 1 gram samples are desired.

The containment and bismuth storage boxes are to be made out of stainless steel. This material, like Invar, is inert to bismuth. It also has the strength necessary for a vacuum container and is cheap. Its thermal neutron cross section, about 3 barns,[17] is somewhat high. However, the thickness of the material will be small enough so that only a minor attenuation of the flux will occur.

A Mark II or III TRIGA reactor can provide a maximum neutron flux of about 10^{16} neutron/cm^2 second during pulse

operation. The annular core pulse reactor (ACPR) TRIGA at Sandia should be capable of about 10^{17} neutrons/cm^2 second. These high flux levels are required to obtain the number of fissions during the pulse necessary for a measurable temperature rise in the dilatometer. Pulse operation is advantageous because it essentially restricts all fissions to one reference point in time (10 msec width). Irradiation times in excess of 100 seconds would be required in steady-state operation at 1 MWt to yield the same neutron fluence. The free volume and ease of access of the TRIGA tank allows the apparatus to remain adjacent to the core before, during, and after the pulse. Data taking can begin immediately. Other experiments require at least a 10 second delay in receiving information due to the transit time necessary for the sample to leave the core and arrive at the detector position and for the initial oscillations to be damped out.

A support stand will need to be constructed and place next to the core so that the weight of the apparatus will not rest on the reflector, but rather on the floor of the reactor tank. Such an arrangement will easily fit in a TRIGA reactor tank.

The limit to the weight range of ^{235}U which can be used in the various runs is established by the expected weight change in the conical bismuth reservoir and the maximum allowable error. We can solve for the energy E released by the sample during irradiation by the following equation for

fission energy release:[10]

$$E = c \cdot f \cdot \sigma_f \cdot N \cdot \tilde{\Phi} \quad , \quad \text{(III-1)}$$

where c is a constant, 1.6×10^{-13} joule/MeV;[19] f is the prompt energy release during fission, 180 MeV;[35] σ_f is the fission cross section, 580 barns;[17] N is the number of ^{235}U atoms; and, $\tilde{\Phi}$ is the neutron fluence. The change in temperature of the bismuth, ΔT, is found from the relation:

$$\Delta T = \frac{E}{C_p \cdot W} \quad , \quad \text{(III-2)}$$

where Cp is the heat capacity of bismuth, 0.167 joule/^{o}C;[8] and W is the weight of the thermometric fluid, 18500 g. Therefore, we can solve for the change in weight of the bismuth in the conical reservoir by utilizing the thermal expansion coefficient of bismuth, $L_t = 1.8 \times 10^{-4}/^{o}C$,[1] in the equation:

$$\Delta W = W \cdot L_t \cdot \Delta T \quad . \quad \text{(III-3)}$$

Table 1 lists various aspects of the irradiation for weights of ^{235}U ranging from 1 g to 1 mg. The error in determining the weight will be no more than 0.03% for all cases. As noted in the table, the flux level is increased for samples of 50 mg or less. This will increase the temperature change over that of the lower flux and thus decrease

TABLE 1

Weight	Flux n/cm^2sec	Fluence n/cm^2	Minimum Pellet Volume (cm^3)	Energy (Heat) (joules)	ΔT (^{o}C)	ΔW (g)	Error (%)
1.0 g	5×10^{15}	5×10^{13}	10	2088	0.676	2.25	4.4×10^{-5}
0.5 g	5×10^{15}	5×10^{13}	5	1044	0.338	1.13	8.9×10^{-5}
0.1 g	5×10^{15}	5×10^{13}	1	208.8	0.0676	0.225	4.4×10^{-4}
50 mg	10^{16}	8×10^{13}	0.5	167.0	0.0541	0.180	5.6×10^{-4}
10 mg	10^{16}	8×10^{13}	0.1	33.40	0.0108	0.0360	2.8×10^{-3}
1 mg	10^{16}	8×10^{13}	0.01	3.340	0.00108	0.00360	2.8×10^{-2}

Energy Release and Bi Weight Change in Dilatometer for a Range of U Weights

the error. Errors for the heavier samples are sufficiently small with the lower flux; there is no need to activate the bismuth more strongly. This lower flux will help expedite the radiological handling of the sample.

The valid temporal region of the experiment is almost unlimited. It begins at the moment of scission. The experiment must end when bismuth approaches its solidification temperature. The exact time of this will be a function of the adiabaticity of the dilatometer. It can not be determined, a priori, as it will be dependent on the quality of the construction of the apparatus. However, as noted in the calibration section, additional heat can be fed into the dilatometer by means of the wire wrap heater to compensate for the energy loss. Since the heat loss rate of the apparatus is also known (see the calibration section), the afterheat, for a constant temperature, is then the difference between the heat loss and the wire wrap input energy.

For reasons of convenience, namely reactor scheduling and personnel work loads, it is expected that the heat will be monitored for a maximum of twelve hours. In any case, afterheat data beyond this time has been well established. The nuclides contributing to afterheat at such a long time after fission have relatively long half-lives and thus their decay schemes are well known.[32] It is for the short times after fission, 0 to 1000 seconds, that adequated data is lacking.

A tentative budget for the proposal requires less than

$45,000. Details are noted in Table 2. The major expense is in preparation and acquisition of the equipment. Subsequent analyses of other nuclides could be performed rather cheaply—probably under $1,000 per run including reactor use charges.

The experiment has a very low associated error range for integral heat. This is because precise methods of calibration are available. These will cancel out all systematic errors through the use of blank runs. Potential errors such as decay heating from bismuth activation or from reactor core gamma ray heating can be determined separately from fission decay heat so that they can be subtracted out of an actual run.

TABLE 2

Item	Cost
Instrumentation	$ 6,600
Apparatus	$ 7,500
Reactor Use	$ 4,000
Chemicals, Computer, Travel	$ 3,300
Salaries	$15,000
Indirect Costs (overhead)	$ 7,700
Total	$44,100

Proposed Budget Outline of the Experiment

Experimental Procedure

The experimental apparatus is shown in Figure 1. The key components are the dilatometer and its dewar region, the electrobalance and its conical bismuth storage reservoir, the disposable bismuth storage box, transfer tubing between the dilatometer and the box, and the two containment boxes which are separated by a dewar region.

The dilatometer, see Figure 5, will be constructed out of Invar, a low expansion steel. Two threaded domes are joined to form a spherical reservoir in which the sample and the bismuth can be melted by means of an external wire wrap heater.

Two hemispheres of pure bismuth will be molded into a configuration which is of the same diameter as the dilatometer reservoir. A small indentation will be made, as shown in Figure 6, which will allow theemplacement of the pelletized U-Bi alloy at the center of the joined hemispheres. This solid metal sphere will then be placed into the dilatometer and the primary wall of the dilatometer will be closed.

The capillary will be sealed and the sphere evacuated. Enough molten bismuth will then be added to the vessel through the access penetration (again, see Figure 5), so that voids in the apparatus are filled.

The access penetration is then closed and the secondary

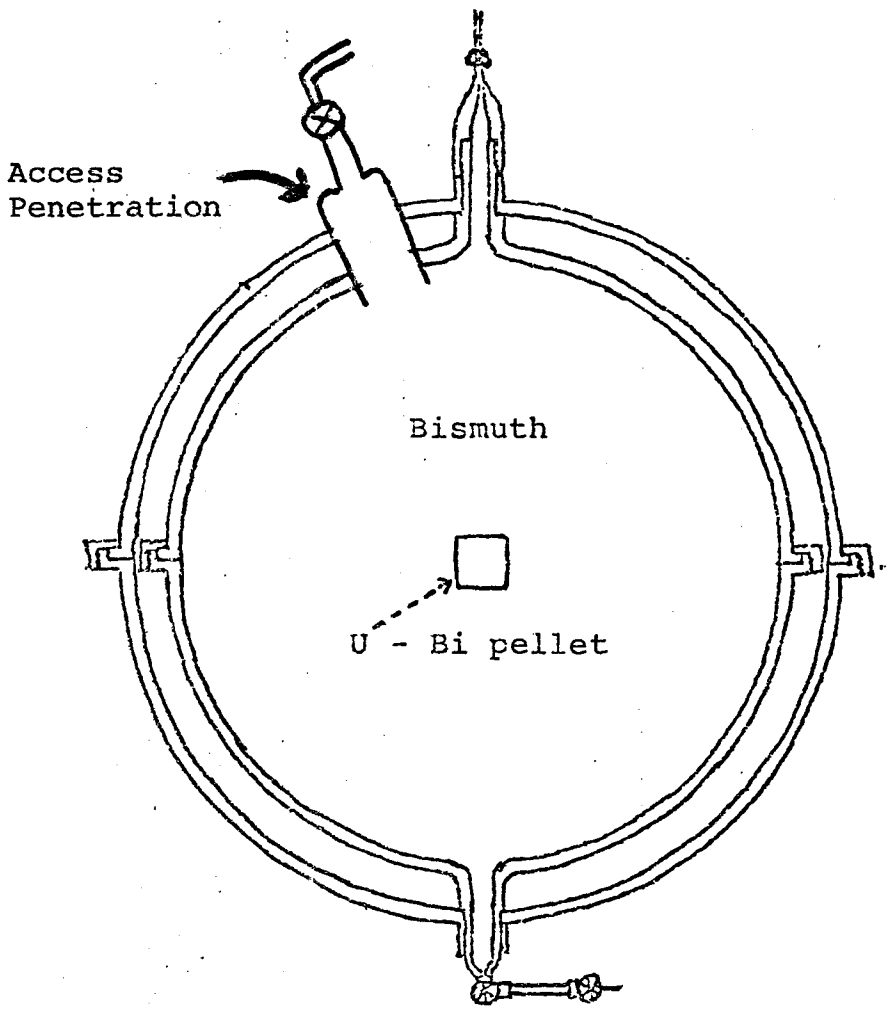


Figure 5

Diagram of Dilatometer and Access Penetration

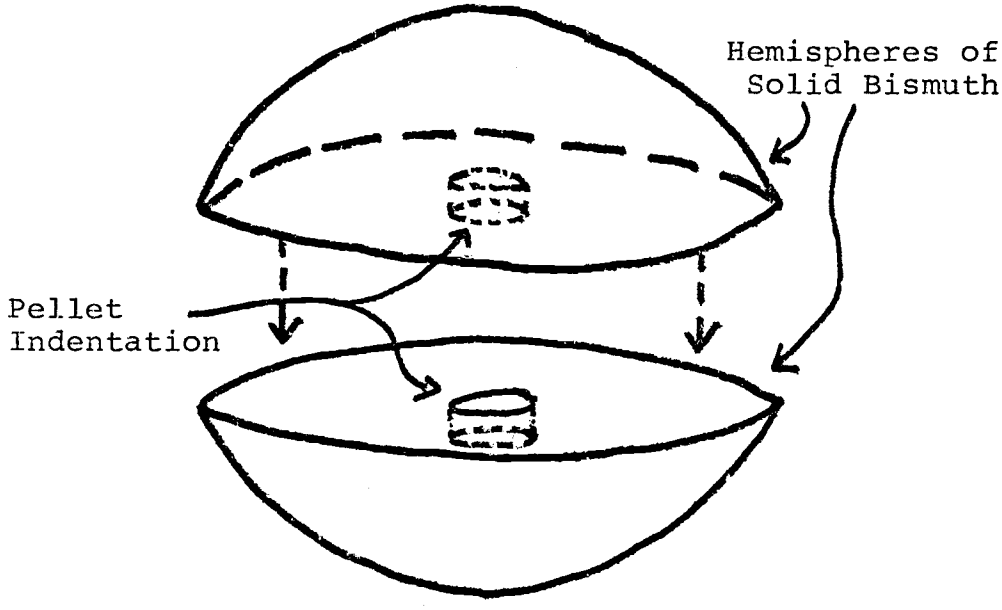


Figure 6

Bismuth Hemispheres with Pellet Indentation

wall of the dilatometer is attached and closed. The wire wrap heater is now energized to melt the entire mass of bismuth.

Because bismuth, like water and gallium, expands on solidification, it will contract on initial melting.[41] As greater heating is applied, it will expand, expelling air from the reservoir region. Eventually, bismuth will begin to flow out of the capillary. At this time, the capillary should be submerged into the conical, molten bismuth weighing reservoir. If the dilatometer cools, bismuth will flow back into the dilatometer.

Two caveats are appropriate at this point. First, once the bismuth has melted, it is imperative that it not be allowed to solidify inside the dilatometer. The expansion of the phase change would ruin the apparatus. (Requiring a new dilatometer and total re-calibration of the system.) The bismuth must be transferred to the storage box before it colls. The storage box is not a precision piece of equipment and is disposable.

The second warning is about accuracy. Essentially all air must be removed from the dilatometer or the expansion of this air will alter the results. As an example, an air bubble in the capillary could interrupt the capillary back flow during cooling. This stoppage would falsely lead the experimenter to conclude that the bismuth is at a higher temperature than it really is.

There are three methods which can alleviate or eliminate air pocketing. The method of choice is to alternately heat and cool the bismuth (always in the liquid phase), so that any air bubbles will become entrained in the flow of bismuth out of the capillary and thus be removed. Should this fail, the dilatometer may be redesigned as shown in Figure 7 so that there is less of an opportunity for air to collect near the head of the capillary. Lastly, the interior surface of the dilatometer primary wall could be coated with a surfactant such as oil which would expedite the movement of air bubbles during the 'exercise period' of the first method mentioned.

Next the wire wrap heater on the capillary and the conical reservoir need to be hooked up and energized. These are necessary to prevent localized solidification which would interdict the flow of bismuth to and from the weighing point. These heaters will remain on during the experiment. Their contribution to the background can be determined in a blank run and subtracted out.

The pressurization schematic of the experiment is shown in Figure 8. Note that the dewar region around the dilatometer, the space between the containment boxes, and the bismuth storage box will all be at 10^{-5}torr or less. The bulk region cannot be as strongly evacuated or the bismuth might boil. It should be at 1 to 0.1 torr.

The pressure differential between the storage box and

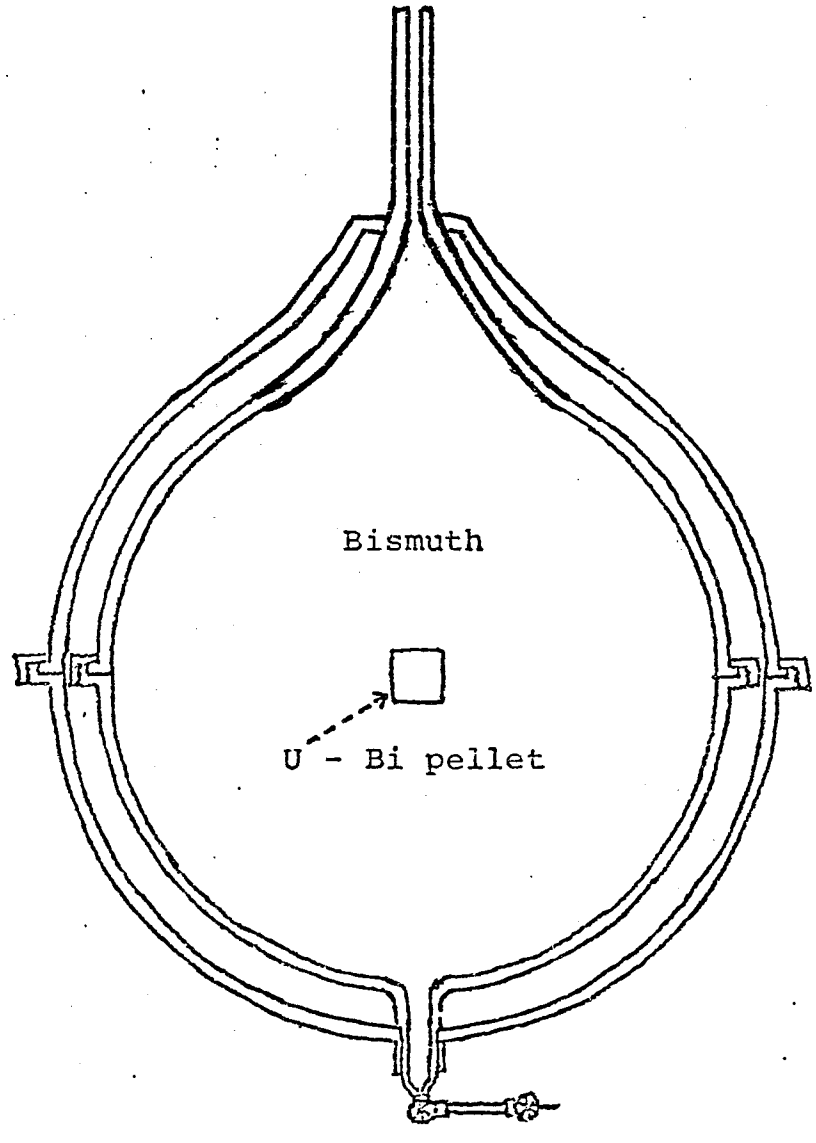
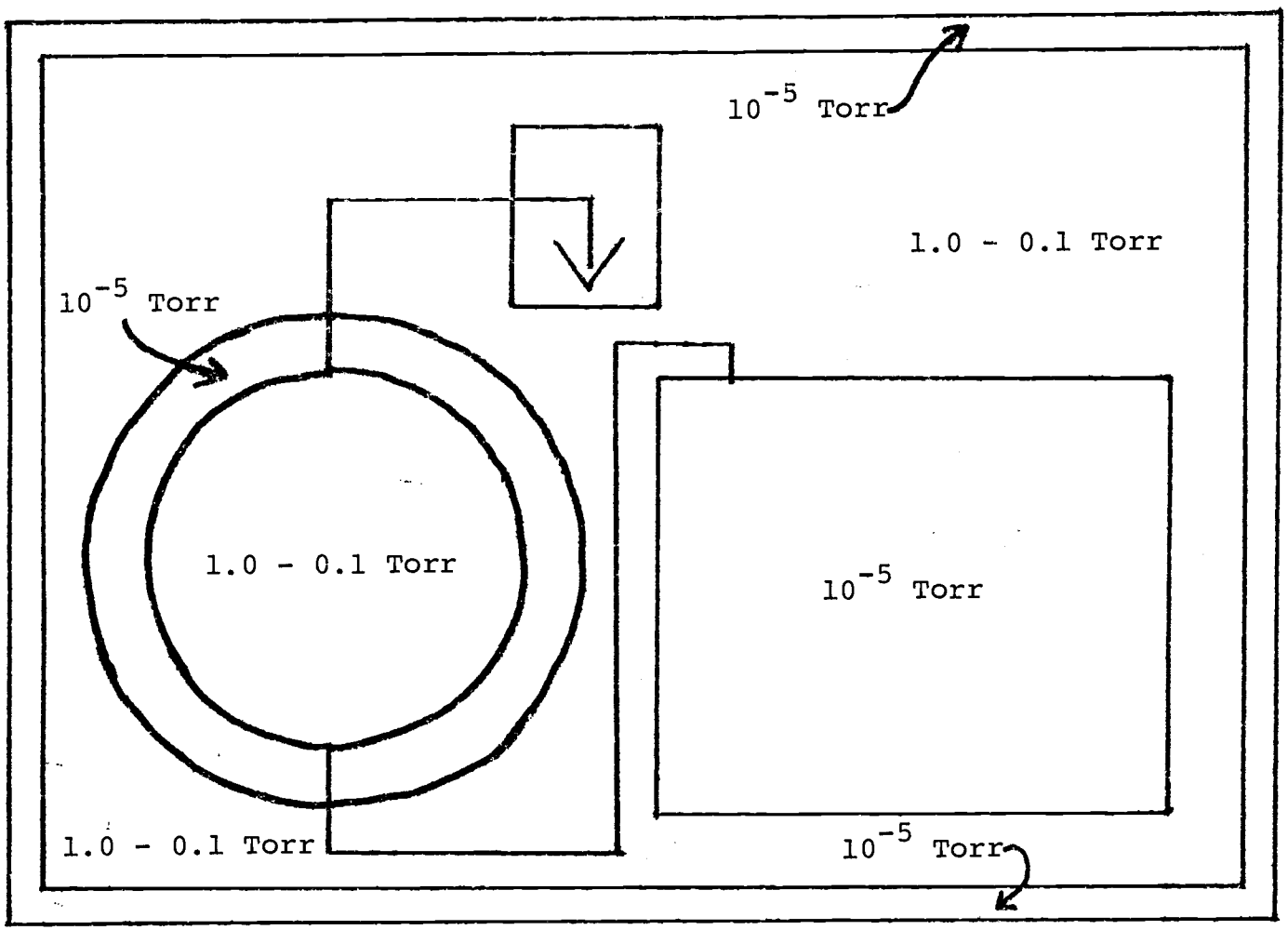


Figure 7

Alternate Dilatometer Design to Alleviate Air Pocketing

Figure 8



Pressurization Schematic of the Experiment

dilatometer will be sufficient to cause almost all of the bismuth in the dilatometer reservoir to flow into the storage box when the connecting valves are opened. This maneuver must be accomplished under the safety precautions detailed in the radiological safety section.

The electrobalance should be similar to a Cahn model RG which will handle a 1 g weight change with a maximum capacity of 2.5 g. There is also a larger Cahn balance which will handle up to 50 g. The sensitivity of each balance is 0.0001 mg and they are operable up to 2540°C (much higher than needed for this experiment). It should be equipped with a three pen chart recorder and two electronic differentiators. The single derivative yields the instantaneous heat and the second derivative yields the rate of heat input. If ultimate accuracy is desired, an analog to digital converter could be used to store the data on magnetic tape for subsequent computer analysis. The electronic differentiators would not be needed if computer analysis is performed. Only the balance will be in the tank: the electronics will remain topside.

The total system should now be assembled and the dewar regions evacuated. The temperature of the bismuth needs to be initialized to 300°C (ie., just slightly above its melting point). This can be done by adjusting the rheostat of the wire wrap heater to the setting corresponding to 300°C which was determined during the calibration procedures.

(see the calibration section). The closed system is allowed to equilibrate for two hours. At that time the boxes will be re-pressurized and opened. The level of the conical bismuth reservoir will then be adjusted, if necessary, so that it will not overflow during the run or run dry when the bismuth cools. The boxes are again sealed and evacuated, and the experiment is placed into the reactor tank over the graphite reflector atop lead bricks. It will be left to equilibrate overnight.

Actual irradiation will take place the following morning. A neutron flux of 5×10^{15} neutrons/cm^{2}second has been located adjacent to the core of the OSTR. (See Flux Determination Appendix.) This flux level can be maintained for 10 msec, yielding a neutron fluence of 5×10^{13} neutrons/cm^2. A similar fluence should be available in other Mark II or Mark III TRIGAs and in most other pulsed research reactors.

Just before the transient rod is fired for the pulse, the ohmic heating should stop and the data acquisition should begin. There will be an immediate temperature spike due to the prompt energy of fission. After this, the temperature will continue to rise until the heat loss rate from the calorimeter exceeds the heat input rate of beta and gamma heating.

The experiment can continue to yield defensible data until the temperature of the bismuth begins to approach its solidification temperature. Then, the wire wrap heater must

be re-energized to avoid breaking the dilatometer. Actually, this need not end the data taking as the input of heat is known and can be subtracted out as backqround. (See Section III.)

The vessel must finally be withdrawn from the reactor tank and be handled in accordance with the radiological precautions detailed in Section VI.

While the prompt spike will yield the number of fissions directly, as noted in the calibration section, it will be convenient to have a backup method. In this secondary method, a number of foils will be activated around the dilatometer. By locating them at several (at least ten) positions around the sphere, a reasonably accurate estimate of the flux at the center of the sphere may be made. This flux calibration should use both bare and cadmium covered foils. It will then yield values for both the thermal and the epithermal fluxes. An accounting for fissions from both fluxes can be made by using the appropriate energy dependent fission cross sections of ^{235}U. The foils need to contain long lived radionuclides as the foils will not be counted for at least one day after irradiation. Gold foils might be used. Counting will be done by gamma ray spectroscopy using a Ge(Li) Crystal. This is a high resolution, low efficiency method. The efficiency is, however, easily calculated so that the activity values may be normalized. This method will serve as a check on the much more precise prompt energy calibra-

tion.

The actual construction of the apparatus should be contracted out to a metal working shop. It is important that the system be strong and rigid so that it need not be recalibrated after each run.

The experiment is capable of great precision. However, much attention needs to be paid to the tedious details in setting it up. It can not be a rush job.

Calibration of the Apparatus and Propagation of Errors

The most important aspect of the experiment lies in the precise calibration of the apparatus. It is important that the experimenter be familiar with volumetric calibration as routinely performed in chemical analysis.[14,30]

Since the heat capacity of bismuth is known[8] the energy input to the bismuth can be derived if both the temperature and mass of the bismuth are also known. This is illustrated in the equation for energy input:

$$E = \left(T - T_o\right) \cdot C_p \cdot W \quad , \quad (V\text{-}1)$$

where T_o is the initial temperature, T is the final temperature, C_p is the heat capacity, and, W is the mass of the bismuth.

First, let us consider the determination of the mass of the bismuth. In a normal volumetric calibration, one would need only to weigh the empty dilatometer and then re-weigh it when filled with bismuth to get the net weight. Although this could be done in this experiment, it would require weighing the entire apparatus. There would be much difficulty in working with such a hot sample as the dilatometer. In order to avoid thermal fluctuations on the balance, the entire apparatus would have to be assembled. This would make for a rather bulky apparatus on the balance.

The bismuth in the conical reservoir would not be affec-

ted by afterheat because it is external to the dialtometer and will not be heated by either the beta or gamma decay in the sphere. Care must be taken not to include the weight of bismuth in the reservoir in the weight of bismuth in the dilatometer. This can be accomplished by making sure that there is the same weight of bismuth in the reservoir for both weighings. It would not be a simple or precise prodedure to do this.

Fortunately, there is a more valuable and more accurate method. If the volume of the dilatometer and the temperature of the bismuth are known, the mass of bismuth can be determined by temperature-density data according to the following equation:

$$W = \rho(T) \cdot V \quad . \quad (V-2)$$

Consider the following example: The volume of the dilatometer is 1850 cm^3. Temperature is 300°C. Density of bismuth at 300°C is 9.9 g/cm^3.[41] Thus, the mass of the bismuth is 18.315 Kg. Because Invar has such a low coefficient of thermal expansion, 2 x $10^{-5}/°C$,[1] the volume of the dilatometer will be essentially constant over the temperature rang e of the experiment. In any event, the temperature dependent volume of the dilatometer can be readily determined by a white oil calibration.

Since the density of white oil varies greatly over the temperature range of the experiment (really, from 0-400°C),

it can be used to ascertain the volume change of the dilatometer over the temperature range.[41] Thus, volume and mass can be easily determined by the use of tabulated density values for bismuth and white oil.

The advantage of this density calibration of mass is that is can be done continuously over the temperature range so that the mass of bismuth is always known. The bulk weight method is valid only for one point at a time. The entire apparatus would have to be re-weighed, both empty and full, at every temperature. Moreover, it would require extensive juggling to remove from consideration the weight of the conical reservoir each time if a range of data was to be generated by that method. Because of the difficulties, it will not be performed. The density calibration is the method of choice.

The calibration for temperature seems almost like the reverse, namely, weigh the expelled bismuth and then convert that to temperature. However, no density correlation will be used. In a blank run, the experimenter will insert a thermometer, as shown in Figure 9, through the access penetration and note the temperature for a give rheostat setting in the wire wrap heater circuit. Voltage and current to the heater are also recorded. The rheostat is then adjusted to yield a temperature of 300^{o}C. This will be the null point and the reading on the electrobalance may be considered to be zero. Essentially, we have just tared the balance.

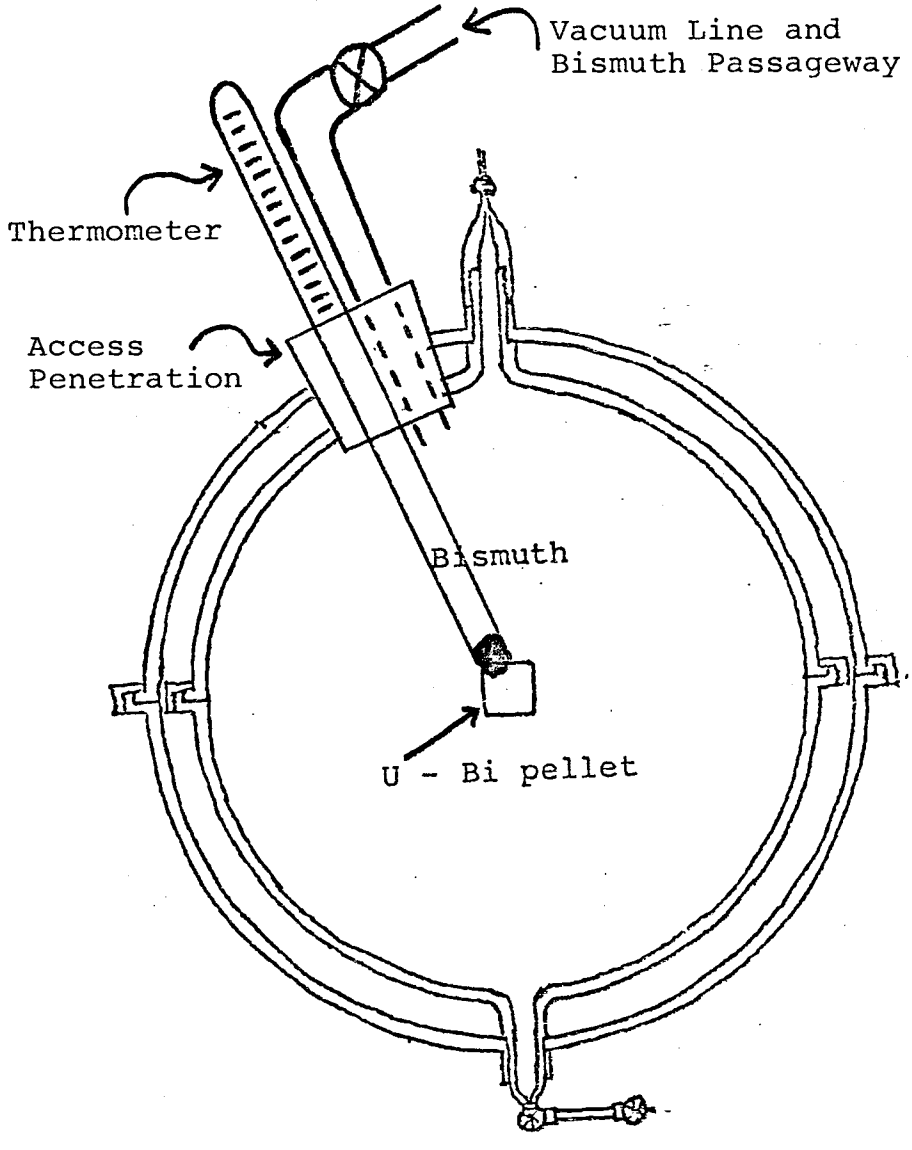


Figure 9

Cross Section of Dilatometer with Thermometer in Place for Temperature Calibration

The voltage is then increased to raise the temeprature a tenth of a degree. This temperature and the change in the weight are recorded. The procedure is continued until there is a well established curve of weight of bismuth expelled versus temeprature. This curve would be similar to the hypothetical curve of Figure 10.

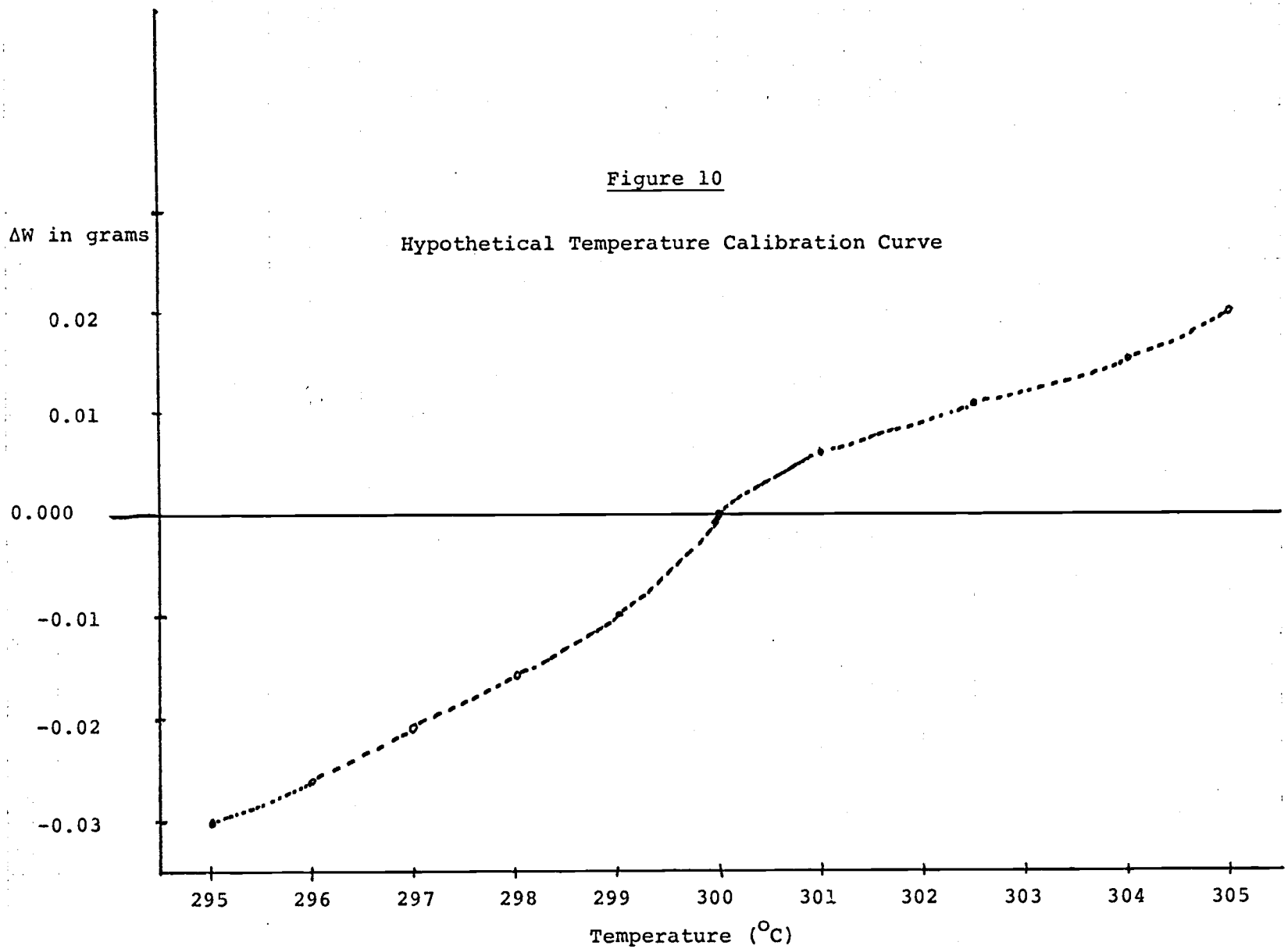
Now, in order to begin a run without the thermometer in place, the rheostat on the wire wrap heater is adjusted to the same position (as measure by voltage and current data) at which it was for the original 300°C run (which had the thermometer). This heat input will match the heat loss rate which is a particular function of the temperature difference between the bismuth and the ambient temperature in the reactor pool. Thus, if the pool temperature is the same, and the heat input is the same, the temperature will be 300°C. This can be checked by the small thermistores (temperature sensitive resistors) affixed to the outside of the inner wall of of the dilatometer.

The weight of the electrobalance is then recorded as tare. As shown in the hypothetical Figure 10, a weight change of +0.02 g would indicate a temperature of 305°C. This can be used, together with the mass of bismuth at 305°C, in equation (V-1) to obtain the energy input to the bismuth.

It is important that thermal equilibrium be established at all calibration points. It will probably be necessary to wait an hour to assure that such equilibrium is attained be-

Figure 10

Hypothetical Temperature Calibration Curve



fore taking data.

The experiment will have less than a 10°C temperature rise. In order to construct a reliable calibration curve, data should be taken in 0.1°C steps. This calibration process may be expected to require one week of hourly readings to complete. This is why it is essential not to allow solidification of bismuth in the sphere. The previous work would be wasted.

For a temperature calibration run, the normal experimental procedure needs to be varied a little. The access penetration plug must not only allow the addition of molten bismuth, it also needs to hold the thermometer. The dilatometer can be initially half filled. This reduced quantity of bismuth is then melted and the thermometer inserted. Filling then proceeds as before in the manner described in the experimental procedure section. See Figure 11, 12, and, 13 for a schematic representation of the process.

The thermometer should have scale readings of less than 0.01°C increments. A calibrated Beckman thermometer would be ideal as it is precise to 0.001°C. Such precision is necessary to determine whether equilibrium has indeed been achieved.

When the calibration is performed, a bismuth-depleted uranium pellet will be used intead of the normal ^{235}U-Bi pellet. Since ^{235}U and ^{238}U will react chemically the same (they have the same number of protons and electrons), there

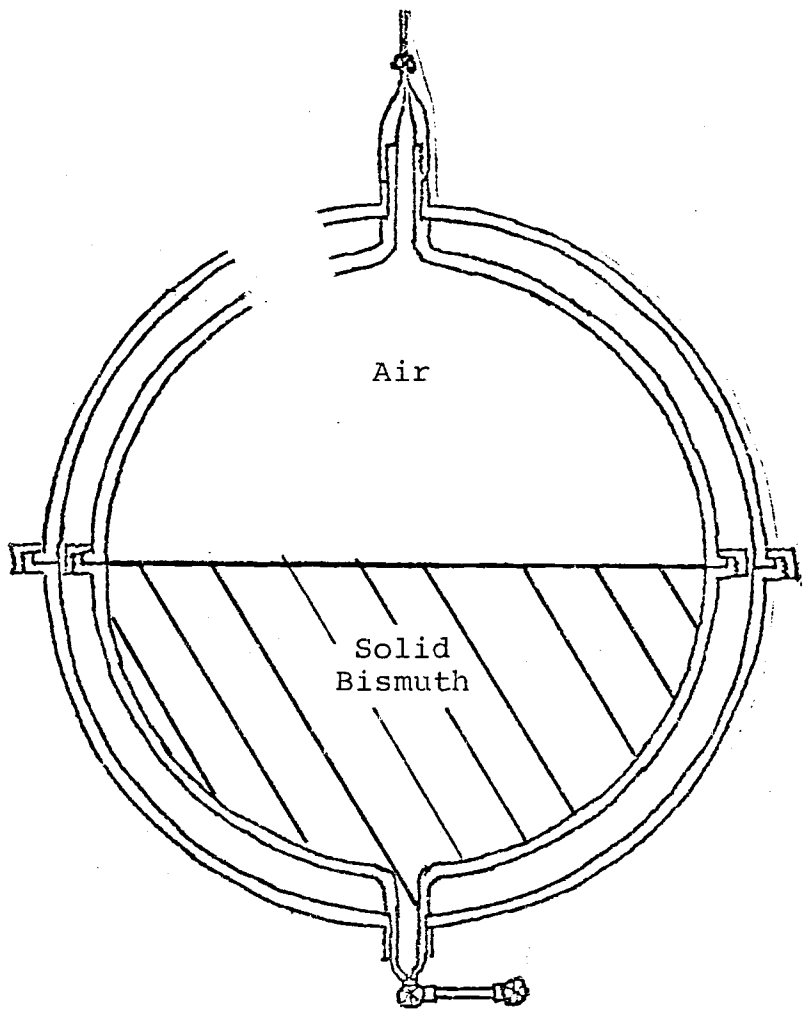


Figure 11

Dilatometer Half Full of Bismuth

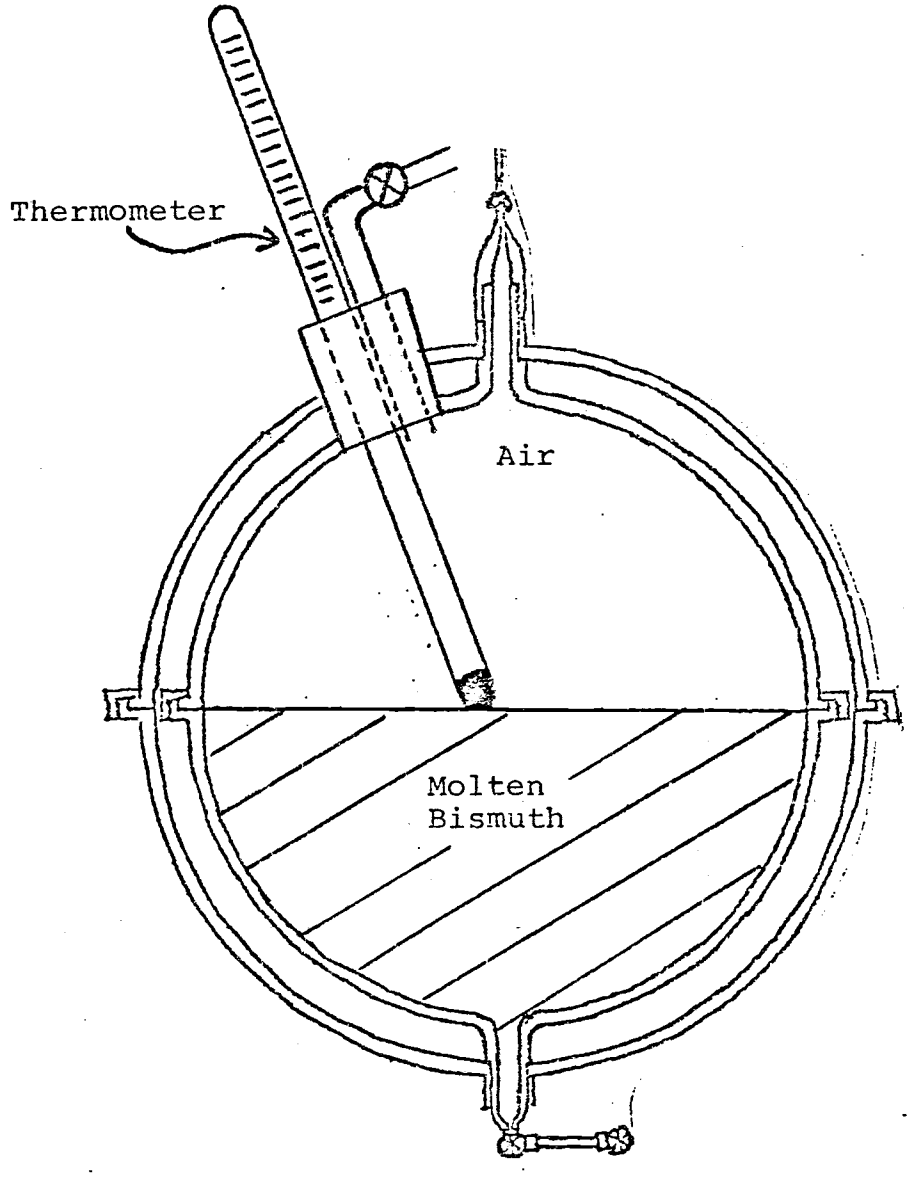


Figure 12

Insertion of Thermometer and and Preparations for Filling Dilatometer

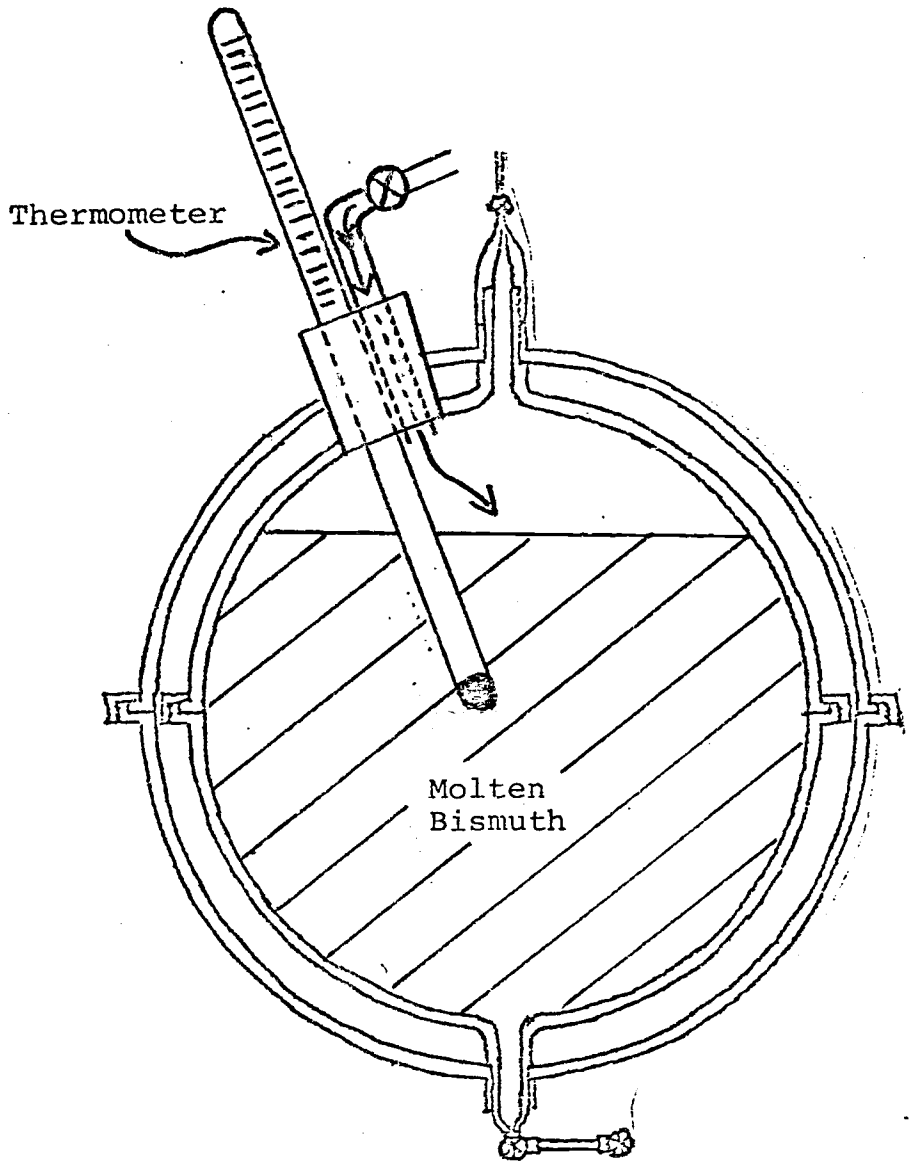


Figure 13

Filling of Dilatometer with Molten Bismuth

is no need to waste the more valuable ^{235}U.

We now have a calibrated system from which all parameters may be determined by knowing the weight of the expelled bismuth. To illustrate: If the weight is X grams, then the temperature is Y. Since the temperature change is Y, then the mass of bismuth in the sphere is Z.

The only remaining calibration task is to determine the background of the system. By this, we mean the heat loss rate, any delayed effects, and other systematic errors.

In such a typical background determination, the position previously occupied by the thermometer is now taken up by a small, high wattage immersion heater. All initial concitions are set up just as in a real run with the exception that there will be no uranium present. The apparatus is in position in the reactor tank, the temperature (as determined from current-voltage data from the heater) is 300^{o}C. A large, known surge of electricity is fed to the immersion heater. The size of the resultant temperature spike is directly proportional to the input electrical energy.. Since this was measured, the coulombic energy input in a real run can be determined from the size of the spike. The number of fissions necessary to cause this energy input can be calculated from the known energy deposited by fission fragments and prompt gammas.

The prompt, recoverable energy of fission is 179.9 MeV.[35] By converting the electrical power in joules to MeV, we can

determine how many fission events took place.

Further calibration continues so that systematic errors may be canceled out. The dilatometer is again initialized to 300°C and placed in core. When this is done, all ohmic heating is discontinued and the reactor is pulsed. Since there is no uranium present, the only input of heat to the dilatometer is the decay of activation products and the gammas from the reactor core which were not attenuated. These quantities are the background of the experiment. Since they are now known, they may be subtracted out of the data in future runs.

There is one last item to be concerned with, and that is the heat loss rate from the calorimeter. We can reasonably assume that the heat loss rate is a linear function of the temperature difference between the calorimeter and the ambient temperature of the pool. Thus, if the temperature difference is twice as great, then the rate of heat loss is twice as great. This can be easily checked. After accounting for conversion efficiency, the electrical power necessary to keep a constant temperature must be equivalent to the heat loss. Thus, there should be an equality in the relation:

$$\frac{E_{T_1}}{E_{T_2}} = \frac{T_1}{T_2} \quad . \quad \text{(V-3)}$$

Should the relation not be valid, we can make a calibraion curve which can cancel out this anomaly. We merely require knowledge of the temperature of the system (obtainable from the weight of the expelled bismuth) to utilize the correction data.

This section shows why calibration is so important. All perturbations which might otherwise affect the accuracy of the final data, integral energy release, may be subtracted out (or, added back in as in the case of heat loss). The only assumption is that the calibration is precise.

Propagation of Errors

Extensive use of normalization will further decrease the uncertainty range for the decay energy beyond what it was lowered through the use of blank runs (which eliminated background).

Because gamma ray attenuation follows well documented physical laws, calculations can be performed which estimate attenuation to a high degree of accuracy. For this experiment, it is estimated that 99% of the gamma photons will be attenuated in the bismuth.

The 1% which escape do not necessarily create an error of 1% in the determination of decay power. The quantity of energy which escapes is itself known to an error of less than 25%. By adding back in this escaped energy, ie., normalizing, we obtain a net uncertainty of 25% of 1% or 0.25% in the

gamma energy contribution to decay power.

The normalization for delayed neutrons is similar as they too follow established physical laws. The uncertainty in the computer calculation of energy release is about 25%. Thus, the propagated error is the product of this error and the fraction of decay energy which escapes the dilatometer as delayed neutrons. In this project, it has been assumed that the leakage probability of neutrons is high and therefore they all escape. Thus,as shown in Section II, 0.5% of the afterheat energy escapes. 25% of 0.5% is 0.125%. This is the net uncertainty in the delayed neutron contribution to decay power.

As can be seen in Table 3 and in the Introduction, the uncertainty in prompt energy release after fission is the determining uncertainty for this project. If this quantity were known to great precision, the total uncertainty of the experiment would drop to about 0.4%. Given the state of current technology, there is, unfortunately, little hope of decreasing the prompt energy release uncertainty.

There are other contributors to the calibration uncertainty, although they are overshadowed by the prompt release. The weighing error will directly affect the energy release determination and thus the maximum value, 0.01%, is listed directly. The dilatometer temperature and prompt energy release errors are also propagated directly as independent errors in the uncertainty of the final integral energy re-

Table 3

Source			Magnitude
Weighing Error			0.01%
Accuracy of Prompt Energy Release			
Coulombic	167.57 Mev	±1.7%	
Neutron	4.69 Mev	±0.1%	
Photon	7.64 Mev	±0.75%	
Net	179.9 Mev	±1.9%	1.9%
Accuracy of Dilatometer Temperature			
Voltage-Current Error		≃0.01%	
Thermistor Error		≃0.05%	
Relative Error		≃0.01%	0.01%
Error in Determing Equilibrium (by Beckman Thermometer)			0.001%
Error in Determing Reactor Bulk Pool Equilibrium and Temperature			0.01%
Net Calibration Error			1.90%

Calibration Uncertainties

lease.

The uncertainty induced in using a Beckman thermometer to ascertain thermal equilibrium is an indirect error as it is dependent on the relation of temperature to the deviation from equilibrium. Equilibrium could be represented as the thermal gradient of the dilatometer. When it is flat, equilibrium is attained. The steadyness of the thermometer reading will indicate whether or not equilibrium has been reached. If the slope of the thermal gradient is large, the apparatus is not in equilibrium. The error induced in the decay heat due to non-equilibrium will be an unknown function of the maximum gradient for a given precision of the thermometer. As the thermometer is precise to $\pm 0.001^{\circ}C$, the assumption is that when it is steady, the gradient is flat. Thus, the propagated error is listed as 0.001%.

Similarly, the uncertainty in the reactor pool bulk temperature has been noted. In this case, due to the size of the fluid to be equilibrated, its surface area, and its localized heat source, it is not reasonable to assume that exact thermal equilibrium can be attained. It is estimated that this effect will propagate the uncertainty of the measurement, 0.001% by a factor of 10 to yield a net uncertainty of 0.01%. This is strictly an educated guess. (In comparison with the prompt energy uncertainty, it could be a factor of 100 and still have no impact on the total uncertainty.)

In both of these cases, the precision of the measurement

has to be converted, through a calibration curve, to the propagated uncertainty in determining integral energy release.

It is important to note that the uncertainty of the final result can be reduced by proper normalization. Additionally, some errors will propagate to the final result directly, while others, due to their compound nature, will need further calibration to be assured of their impact on the answer.

Errors have been considered only for the integral energy release. The impact of the instantaneous behavior of the apparatus on the true and differential energy release will be substantial. As an example, the temperature of the dilatometer may change by a small amount, say $0.01^{o}C$, in a given time frame, say 2 minutes. This leads to a nearly flat integral energy release and therefore a large uncertainty for the instantaneous energy release. There are two ways to reduce this error: Use a more sensitive electrobalance (which may not be commercially available). Or, irradiate the sample at a higher flux. This latter method would increase the temperature change for a given time period. (Heavier samples of ^{235}U would accomplish the same result, but, they would require much larger quantities of bismuth with the associated radiological problems.) Neither of these two options is available at OSU and so we will not consider them further. In any event, the accurate determination of the integral heat is sufficient for reactor safety analysis.

General and Radiation Safety

The experiment has several hazardous aspects which require caution and anticipation. While molten bismuth is not flammable, the experimenter will be severly burned if he comes in contact with it. Although there are many similar, conventional hazards in working with such a thermally hot sample, these are minor when compared with the potential radiological hazards of this project. If burned by the irradiated bismuth, not only will the experimenter be subjected to painful burns, but he would be also be subjected to an internal exposure to two alpha emitters, ^{210}Bi and ^{210}Po.(17)

There are several sources of radioactivity in this experiment. Before the pulse irradiation, the concern is solely with the alpha emitter, ^{235}U. One gram, the maximum quantity which is intended for use, has an activity of 2.15 μCi. Self absorption will lower the effective activity even further. As long as the material is kept from boiling, no unusual radiological precautions need be taken. Powered uranium is frequently pyrophoric so adequate fire equipment should be available.(5)

The pulse irradiation will generate two classes of radioactive material: fission products and activation products. In evaluating the potential hazards of the fissions products, it is necessary to subdivide further. Solid fission products, which remain in the bismuth, contribute to the total

radioactivity of the sample but are not really a source of individual concern. On the other hand, gaseous fission products must be calculated explicitly as they can escape the dilatometer and contaminate the inner box. Secondly, the evolved quantity of gas needs to be known to determine if there will be a significant pressure buildup. Because a convenient computer data file, CINDAT,[29] was available, detailed yields and activities were calculated for both classes of fission products. This should prove useful when the irradiated materials are sent for starage and/or processing. Moreover, it is good practice to have a detailed inventory of the types and quantities of radioactive materials which are present.

In order to determine these activities, it is necessary to determine the expected number of fissions in the sample. By evaluating the equation:

$$\#\ \text{of fissions} = N \cdot \sigma_f \cdot \tilde{\Phi} \quad , \quad \text{(VI-1)}$$

we find that there will be 7.4×10^{13} fissions for the experimental case where there is 1 g of ^{235}U, σ_f = 580 barns,[17] and $\tilde{\Phi}$ is 5×10^{13} neutrons/cm^2. As determinations of afterheat with uranium weights in excess of 1 g are not contemplated, this represents the maximum number of fissions, and thus, the maximum quantity of radioactive fission products.

Two convenient empirical relations for gross beta and gamma activity after fission allow solution for the activity

of the fission products at any time t.[13] These are:

$$\text{Activity}_{(\text{Beta})} = 3.8 \times 10^{-6} \cdot t^{-1.2} \beta/\text{sec} \cdot \text{fission} \quad , \quad \text{(VI-2)}$$

and,

$$\text{Activity}_{(\text{Gamma})} = 1.9 \times 10^{-6} \cdot t^{-1.2} \gamma/\text{sec} \cdot \text{fission} \quad . \quad \text{(VI-3)}$$

Summaries of the expected activities at various times after fission are given in Table 4 for zero to seven days and in Table 5 for zero to one hundred days. These were generated by the FISSACT program (see Appendix B).

It should be mentioned that the empirical equations are not valid for time close to or equal to zero. They falsely indicate that the activity at the end of bombardment (EOB) would be infinite. In fact, it is finite. For short times after irradiation, the following detailed summation method should be used.

The fission yields from CINDAT were used by the computer program ACTEOB (see Appendix C.) to determine the detailed inventories of 99.99994% of the fission products. CINDAT is a condensed version of the ENDF/B-4 nuclear file. It contains decay constants, fission yields, and information on decay modes and energetics. In using ACTEOB, it is found that we can expect to have 790.23 mCi of total fission products at the end of the irradiation period. Of this, 108.80 mCi are gaseous fission products. Independent yields

Table 4

Gross Radioactivities of Fission Products for 0 to 7 Days After Fission

TIME IN DAYS	BETA ACTIVITY (MILLICURIES)	GAMMA ACTIVITY (MILLICURIES)	TOTAL ACTIVITY (MILLICURIES)
0.000	0.	0.	0.
.125	9.2913E+01	4.4140E+01	1.3705E+02
.250	4.0001E+01	2.0024E+01	6.0025E+01
.375	2.7739E+01	1.3221E+01	3.0959E+01
.500	1.7447E+01	8.7320E+00	2.6179E+01
.625	1.3248E+01	6.6735E+00	1.9921E+01
.750	1.0795E+01	5.3694E+00	1.6064E+01
.875	8.9136E+00	4.4566E+00	1.3370E+01
1.000	7.5930E+00	3.7969E+00	1.1391E+01
1.125	6.5999E+00	3.2995E+00	9.8994E+00
1.250	5.8099E+00	2.9051E+00	8.7149E+00
1.375	5.1880E+00	2.5910E+00	7.7790E+00
1.500	4.6682E+00	2.3341E+00	7.0023E+00
1.625	4.2417E+00	2.1203E+00	6.3619E+00
1.750	3.8799E+00	1.9399E+00	5.8197E+00
1.875	3.5716E+00	1.7858E+00	5.3574E+00
2.000	3.3054E+00	1.6527E+00	4.9581E+00
2.125	3.0735E+00	1.5367E+00	4.6102E+00
2.250	2.8697E+00	1.4349E+00	4.3046E+00
2.375	2.6895E+00	1.3447E+00	4.0342E+00
2.500	2.5289E+00	1.2644E+00	3.7933E+00
2.625	2.3851E+00	1.1925E+00	3.5776E+00
2.750	2.2556E+00	1.1278E+00	3.3834E+00
2.875	2.1384E+00	1.0692E+00	3.2077E+00
3.000	2.0328E+00	1.0164E+00	3.0492E+00
3.125	1.9365E+00	9.6741E-01	2.9039E+00
3.250	1.8499E+00	9.2494E-01	2.7748E+00
3.375	1.7641E+00	8.8207E-01	2.6462E+00
3.500	1.6858E+00	8.4290E-01	2.5287E+00
3.625	1.6192E+00	8.0958E-01	2.4288E+00
3.750	1.5546E+00	7.7731E-01	2.3319E+00
3.875	1.4946E+00	7.4732E-01	2.2419E+00
4.000	1.4385E+00	7.1928E-01	2.1578E+00
4.125	1.3860E+00	6.9300E-01	2.0790E+00
4.250	1.3378E+00	6.6891E-01	2.0067E+00
4.375	1.2921E+00	6.4604E-01	1.9381E+00
4.500	1.2491E+00	6.2456E-01	1.8737E+00
4.625	1.2087E+00	6.0436E-01	1.8131E+00
4.750	1.1707E+00	5.8533E-01	1.7560E+00
4.875	1.1347E+00	5.6734E-01	1.7021E+00
5.000	1.1008E+00	5.5039E-01	1.6512E+00
5.125	1.0686E+00	5.3432E-01	1.6029E+00
5.250	1.0382E+00	5.1909E-01	1.5573E+00
5.375	1.0093E+00	5.0463E-01	1.5139E+00
5.500	9.8161E-01	4.9091E-01	1.4727E+00
5.625	9.5568E-01	4.7785E-01	1.4335E+00
5.750	9.3081E-01	4.6540E-01	1.3962E+00
5.875	9.0709E-01	4.5355E-01	1.3606E+00
6.000	8.8446E-01	4.4223E-01	1.3267E+00
6.125	8.6285E-01	4.3142E-01	1.2943E+00
6.250	8.4219E-01	4.2110E-01	1.2633E+00
6.375	8.2240E-01	4.1120E-01	1.2336E+00
6.500	8.0346E-01	4.0173E-01	1.2052E+00
6.625	7.8530E-01	3.9265E-01	1.1779E+00
6.750	7.6789E-01	3.8394E-01	1.1518E+00
6.875	7.5116E-01	3.7558E-01	1.1267E+00
7.000	7.3510E-01	3.6755E-01	1.1026E+00

Table 5

Gross Radioactivities of Fission Products for 0 to 100 Days After Fission

TIME IN DAYS	BETA ACTIVITY (MILLICURIES)	GAMMA ACTIVITY (MILLICURIES)	TOTAL ACTIVITY (MILLICURIES)
0.000	0.	0.	0.
2.000	3.3054E+00	1.6527E+00	4.9581E+00
4.000	1.4388E+00	7.1938E-01	2.1581E+00
6.000	8.8446E-01	4.4223E-01	1.3267E+00
8.000	6.2626E-01	3.1313E-01	9.3939E-01
10.000	4.7914E-01	2.3957E-01	7.1871E-01
12.000	3.8499E-01	1.9249E-01	5.7748E-01
14.000	3.1997E-01	1.5998E-01	4.7995E-01
16.000	2.7259E-01	1.3630E-01	4.0889E-01
18.000	2.3667E-01	1.1833E-01	3.5500E-01
20.000	2.0856E-01	1.0428E-01	3.1284E-01
22.000	1.8602E-01	9.3009E-02	2.7903E-01
24.000	1.6757E-01	8.3787E-02	2.5136E-01
26.000	1.5223E-01	7.6114E-02	2.2834E-01
28.000	1.3927E-01	6.9637E-02	2.0891E-01
30.000	1.2821E-01	6.4104E-02	1.9231E-01
32.000	1.1865E-01	5.9327E-02	1.7798E-01
34.000	1.1033E-01	5.5164E-02	1.6549E-01
36.000	1.0301E-01	5.1507E-02	1.5452E-01
38.000	9.6543E-02	4.8272E-02	1.4481E-01
40.000	9.0780E-02	4.5390E-02	1.3617E-01
42.000	8.5617E-02	4.2809E-02	1.2843E-01
44.000	8.0969E-02	4.0484E-02	1.2145E-01
46.000	7.6763E-02	3.8381E-02	1.1514E-01
48.000	7.2941E-02	3.6471E-02	1.0941E-01
50.000	6.9454E-02	3.4727E-02	1.0418E-01
52.000	6.6261E-02	3.3130E-02	9.9391E-02
54.000	6.3327E-02	3.1663E-02	9.4990E-02
56.000	6.0623E-02	3.0311E-02	9.0934E-02
58.000	5.8123E-02	2.9061E-02	8.7184E-02
60.000	5.5806E-02	2.7903E-02	8.3709E-02
62.000	5.3653E-02	2.6826E-02	8.0479E-02
64.000	5.1647E-02	2.5824E-02	7.7471E-02
66.000	4.9775E-02	2.4887E-02	7.4662E-02
68.000	4.8023E-02	2.4012E-02	7.2035E-02
70.000	4.6381E-02	2.3191E-02	6.9572E-02
72.000	4.4840E-02	2.2420E-02	6.7259E-02
74.000	4.3389E-02	2.1695E-02	6.5084E-02
76.000	4.2023E-02	2.1011E-02	6.3034E-02
78.000	4.0733E-02	2.0367E-02	6.1100E-02
80.000	3.9514E-02	1.9757E-02	5.9271E-02
82.000	3.8361E-02	1.9180E-02	5.7541E-02
84.000	3.7267E-02	1.8634E-02	5.5901E-02
86.000	3.6230E-02	1.8115E-02	5.4344E-02
88.000	3.5244E-02	1.7622E-02	5.2866E-02
90.000	3.4306E-02	1.7153E-02	5.1459E-02
92.000	3.3413E-02	1.6707E-02	5.0120E-02
94.000	3.2562E-02	1.6281E-02	4.8843E-02
96.000	3.1749E-02	1.5875E-02	4.7624E-02
98.000	3.0973E-02	1.5487E-02	4.6460E-02
100.000	3.0232E-02	1.5116E-02	4.5347E-02

were used for this determination. This allows for the precise calculation of EOB activities which can not be determined by the previous empirical relations. Time dependent activities for one day or one week, while computed, are not particularly accurate. For shorter times, such as EOB, this detailed calculation is the method of choice. For longer times, the empirical relations should be used. These two methods are, to a large extent, complementary.

If greater precision is desired, on could alternatively use an involved computer program such as CINDER[11] with the cumulative fission yields to generate the data. Such sophistication is not necessary for the purposes of this preliminary examination.

From ACTEOB, we can examine a few nuclides of special radiological interest. Table 6 lists some isotopes of Kr, I, and, Xe with their yields and activities. It is interesting to note that the total volume of evolved gas will be less than $6.8 \times 10^{-6} cm^3$ under the operating conditions of this experiment ($T=300^{o}C$, P=1 torr). This is so low that it poses no problems due to pressure buildup and only a 0.003% error in the weight determination.

There are two groups of activation products: those from bismuth and those from the stainless steel and Invar containers. Bismuth activation poses the greates hazard due to the formation of ^{210}Bi and ^{210}Po, both of which are alpha emitters. Using the standard activation equation:[7]

Table 6

Nuclide	Half-life	Activity at EOB
^{85}Kr	10.7 years	95.45 pCi.
^{131}I	8 days	79.26 nCi.
^{133}I	20.8 hours	30.64 μCi.
^{133}Xe	5.3 days	19.74 nCi.

Summary of Activities of a few Radionuclides of Special Interest

$$A = N \cdot \sigma \cdot \Phi \cdot (1-e^{-\lambda t}) \quad , \quad (VI-4)$$

with 18.5 Kg ^{209}Bi at a flux of 5 x 10^{15} neutrons/cm^2 second with σ equal to 0.19 millibarns,[17] the bismuth activity will be 21.9 μCi at EOB. This will decay to 19.07 μCi of ^{210}Bi and 102.4 nCi of ^{210}Po after one day.

About 16 Kg (35 pounds) of steel will be in the apparatus. Table 7 summarizes the activation of the steel. These low activities whould present no problem in handling the apparatus after one hour delay, let alone after the planned one day delay.

All expected radioactivities for the project have now been calculated. What design features are needed to allow handling within all guidelines of 10CFR20.101 and 10CFR-20.103?[36] The experiment will be examined in a step-by-step manner in order to review all potential radiological hazards and health physics precautions.

The experiment is divided into four phases: 1) Preparation of the samples. 2) Irradiation of the apparatus. 3) Removal of the samples to storage. 4) Cleanup of the vessel in order to return to phase one.

In phase one, clean (non-irradiated) uranium and bismuth are melted to form the alloy. It is placed in the bismuth sphere and the apparatus sealed. The melting operation needs to be carried out in a hood to avoid bumping due to uneven heating and the uptake of radon gas. No special shielding

Table 7

Nuclide	Half-life	Activity at EOB	Activity at 1 Hour	Activity at 1 Day
^{51}Cr	27.7 days	9.98 µCi	9.96 µCi	9.73 µCi
^{55}Cr	3.6 minutes	71.3 mCi	685.8 nCi	-0-
^{55}Fe	2.7 years	12.4 µCi	12.4 µCi	12.4 µCi
^{59}Fe	44.6 days	6.59 µCi	6.59 µCi	6.49 µCi
^{65}Ni	2.5 hours	1.36 mCi	1.03 mCi	1.75 µCi

Summary of the Activation Products of the Steel Containers

will be necessary. Personnel should wear class D protective clothing (lab coats and safety glasses).[28]

During the irradiation, the TRIGA tank will be relied on as a primary biological shield. The apparatus will be monitored visually for signs of leakage. No additional precautions will be taken.

Phase three will be the most hazardous and the most difficult. It will be necessary to wait 24 hours after the irradiation before the box is removed from the tank. The activity at this time is estimated to be 12 mCi. Some sort of transfer cask will probably be necessary as the dose rate at one foot, from the 6CiEN law,[20] would be 72 mR/hr.

This is overstating the case. In actuality, the three inch radial thickness of bismuth would shield out 99% of the gammas leading to a reduced source of 120 μCi. The dose rate at one foot, if we assume a point source, would be 0.7 mR/hr. However, when valves are opened, the uranium and fission products will mix in the sphere so as to decrease the gamma absorption by the bismuth. This means that we will need to revise the dose rate upwards, again. It will, in any case, be less than 72 mR/hr. This is the dose rate if we consider the gamma source to be unshielded. If we then design the handling aspects of the experiment for this higher level, we can be assured that precautions are adequate for the hazard involved.

Because of this dose rate, the experimenter may not

approach closer than 15 cm (6 inches) with any extremity or 30 cm (12 inches) with his whole body. The dose rate to the extremities will be about 300 mR/hr at this distance. The dose rate to the whole body was noted above. Because of this spatial restriction, tongs will be required to manipulate latches and valves. It is estimated that less than 5 minutes will be necessary to accomplish transfer. This would lead to a total dose of 25 mR to the hands and 6 mR to the whole body.

Good radiological practice dictates that radiological exposure be minimized, even though it is below applicable limits. Additional shielding should be used between the experimenters body and the apparatus to further decrease the whole body dose. 10 cm (4 inches) of lead will reduce the dose by 99.95%.

Because of the dangers of loose alpha activity if the apparatus should break, spilling molten bismuth onto the floor of the glove box (or glove room), the experimenter must be clothed in class A or B protective garments. (Class A and B require that the personnel have a separate air supply so that they do not breathe in contamination.)[28] Additionally, he should wear thick asbestos gloves and boots.

A written emergency plan shall be devised, approved by the health physicist, and posted in the work area. It should include both evacuation and immediate counter-measures, such as cooling water, in addition to a plan to clean up the spill

after it cools.

All standard occupational safety guidelines shall also be followed.

The last phase involves the cleanup of the dilatometer and the isolation boxes. As there will still be some residual alpha activity, in addition to a moderate gamma field, personnel shall be in class A or B protective dress while they scrub and dissolve (in nitric acid)[5] the residual contamination. Work shall be carried out in a hood.

The experiment is well within the capabilities of a standard hot lab facility. Remote manipulations (other than tongs) or hot cell work is not necessary. While bioassay need not be routinely conducted, it would be wise to obtain pre-exposure bioassays so that if an accident did occur, the personnel would have an established history.

Because the bismuth can be transferred with complete isolation, there is very little chance of an accidental spill which would contaminate the work area. The bismuth will transfer to the transfer box due to the pressure differential. Wire wrap heaters can be used on all tubing to prevent clogging of the bismuth. There should be very little difficulty encountered in working with the apparatus.

Conclusions

As noted in the various sections, the experiment provides data which is of substantially greater accuracy than that of comparable experiments. This is made possible by the great care taken in calibration. There is a prevalent philosophy to the calibration and it is one of the blank run.[30] By this, the experiment can be simulated in a variety of ways to determine the background characteristics of the system. Since the background is the same whether there is uranium in the dilatometer or not, it can be cancelled out.

This greater accuracy will hopefully validate the computer studies for short times after fission in assessing a LOCA. This is the time span when there is the largest uncertainty over the individual decay schemes. By developing an average integral decay power curve, per fission, the problem is then reduced to one of multiplication to normalize it to a power rating and power history.

While the design of the dilatometer is complicated by the various valves and joints, it should be both easy to construct and reliable. The bismuth storage box, which is what complicates the assembly, is a concession for radiation safety. Open air handling of molten, irradiated bismuth is not within the expertise of a university health physics department.

The large thermal expansion of bismuth, coupled with the small thermal expansion of Invar, creates a prompt and accur-

ate dilatometer. Since the tip of the capillary is submerged in the conical bismuth weighing reservoir, both the flow into and out of the weighing pan can be noted. The precision and accuracy of the electrobalance is well suited to the task. This type of balance is regularly used for determinations of such minute phenomena as surface tension. magnetic susceptabiltiy and adsorption.

The project can accomodate a wide range in the weight of ^{235}U. This indicates, although not rigorously, that the experiment can operate with other fissile nuclides such as ^{239}Pu. These other determinations of time dependent fission product energy release will add greatly to our storehouse of nuclear data which is required for accurate safety analysis.

The experiment is capable of high accuracy if the construction and calibration procedures are performed with care and attention.

BIBLIOGRAPHY

1. American Society for Metals Handbook Committee, Metals Handbook: Volume 1, Properties and Selection of Metals, 8th Edition. American Society for Metals, Ohio, 1961.

2. American Society for Metals Handbook Committee, Metals Handbook: Volume 8, Metallography, Structures and Phase Diagrams, 8th Edition, American Society for Metals, Ohio, 1973.

3. Armbruster, P., and Meister, H., "Measurements of Chain Length for Beta Decay of Fission Products in Thermal Fission of U-235", Zeitschrift für Physik, 170, 274 (1962), or, AEC-TR-5655.

4. Bunney, L. R., and Sam, D., "Gamma-Ray Spectra of the Products of Thermal-Neutron Fission of U-235 at Selected Times After Fission", Nuclear Science and Engineering, 39, 81 (1970).

5. Cotton, F. Albert, and Wilkinson, Geoffrey, Advanced Inorganic Chemistry, 3rd Edition, Interscience Publishers, New York, 1972.

6. deShalit, Amos, and Feshbach, Herman, Theoretical Nuclear Physics, Volume I, John Wiley and Sons, Inc., New York, 1974.

7. DeSoete, D., et al, Neutron Activation Analysis, Wiley-Interscience, England, 1972.

8. Dean, John A., Lange's Handbook of Chemistry, 11th Edition, Mc-Graw Hill, New York, 1973.

9. Duderstadt, J. J., and Hamilton, L. J., Nuclear Reactor Analysis, John Wiley and Sons, Inc., New York, 1976.

10. El-Wakil, M. M., Nuclear Heat Transport, International Textbook Co., Pennsylvania, 1971.

11. England, T. R., CINDER, Doctoral Thesis, University of Wisconsin, 1970.

12. Fisher, P. C., and Engle, L. B., "Delayed Gammas from Fast-Neutron Fission of Th-232, U-233, U-235, U-238, and Pu-239", Physical Review, 134, B796 (1964).

13. Glasstone, Samual, and Sesonske, Alexander, Nuclear Reactor Engineering, Van Nostran Reinhold Co., New York, 1967.

14. Harris, W.E., and Kratochvil, B., Chemical Analysis, Barnes and Noble, New York, 1970.

15. Harvey, Bernard, Introduction to Nuclear Physics and Chemistry, 2nd Edition, Prentice Hall, New Jersey, 1969.

16. Holm, Jerald, A Simplification of Existing Methods of Calculating Decay Heat Following Reactor Shutdown and an Investigation of the Effects of Power History Upon Decay Heat, Masters Thesis, Oregon State University, August, 1976.

17. Knolls Atomic Power Laboratory, Chart of the Nuclides, General Electric, California, 1972.

18. Kutcher, J.E., and Wyman, M. E., "An Experimental Study of the Time Dependence of the Beta Energy Spectrum from U-235 Fission Fragments", Nuclear Science and Engineering, 43, 42 (1971).

19. Lamarsh, John R., Introduction to Nuclear Reactor Theory, Addison Wesley Publishing Co., Inc., Massachusetts, 1966.

20. Lapp, Ralph E., and Andrew, Howard L., Nuclear Radiation Physics, 3rd Edition, Prentice Hall, Inc., New Jersey, 1963.

21. Lederer, C. Michael, et al, Table of Isotopes, 6th Edition, John Wiley and Sons, Inc., New York, 1967.

22. Liaw, Jye R., Primary Fission Fragment Yields and Prompt-Neutron Emissions for After-Heat Analysis, Doctoral Thesis, Oregon State University, August, 1975.

23. MacMahon, T. D., et al. "Energy Released by Beta Radiation Following Fission: Partl, U-235 Data", Journal of Nuclear Energy, 24, 493 (1970).

24. McNair, A., et al, "A Measurement of the Energy Released as Kinetic Energy of Beta Particles Emitted in the Radioactive Decay of the Fission Products of U-235", Journal of Nuclear Energy, 23, 73 (1969).

25. Marmier, Pierre, and Sheldon, Eric, Physics of Nuclei and Particles, Volume I, Academic Press, New York, 1969.

26. Marmier, Pierre, and Sheldon, Eric, Physics of Nuclei and Particles, Volume II, Academic Press, New York, 1970.

27. Sakharov, V. N., and Malofeev, A. I., "Gross Gamma Activity of Fission Products of U-235", Soviet Journal of Atomic Energy, 3, 1161 (1957).

28. Shapiro, Jacob, Radiation Protection, Harvard University Press, Massachusetts, 1972.

29. Shay, Michael R., CINDAT, unpublished work, Oregon State University, 1976.

30. Skoog, Douglas A., and West, Donald M., Fundamentals of Analytical Chemistry, Holt Rinehart and Winston, New York, 1969.

31. Smith, Charles O., Nuclear Reactor Analysis, Addison Wesley Publishing Co., Massachusetts, 1967.

32. Spinrad, Bernard I., "The Sensitivity of Decay Power to Uncertainties in Fission Product Yields", unpublished work, Oregon State University, 1976.

32a. Spinrad, Bernard I., "Quarterly Report: Evaluation of Fission Product After-Heat", Contract IIA:RLR:74:4/OSU, July 1, 1975 - September 30, 1975, Oregon State University.

33. Stamatelatos, M. G., and England, T. R., "Fission-Product Gamma-Ray and Photoneutron Spectra", Conference of Nuclear Cross Sections and Technology, Washinton, D. C., March, 1975.

34. Tsoulfanides, N., et al, "Measurements of Time-Dependent Beta Spectra of Beta Rays from U-235 Fission Fragments", Nuclear Science and Engineering, 43, 42 (1971).

35. Unik, J. P., and Gindler, J. E., "A Critical Review of the Energy Released in Nuclear Fission", ANL-7748, Argonne National Laboratory, Illinois, 1971.

36. USAEC, 10CFR, General Services Administration, Washington, D. C., 1973.

37. US Health, Education, and Welfare, Radiological Health Handbook, Washington, D. C., 1970.

38. USNRC, Reactor Safety Study: An Assessment of Accident Risks in U. S. Commercial Nuclear Power Plants, Wash-1400 (NUREG-75/014), National Technical Information Service, Virginia, 1975.

39. University of California at Berkeley, "Decay Heat Experiment: Final Design and Analysis Report", NE76-1, unpublished work, California, 1976.

40. Warkentin, J. K. "Gamma Energy Release from Fission Product Decay Following a Short Reactor Transient", IDO-17161, (1966).

41. Weast, Robert, C., Handbook of Chemistry and Physics, 50th Edition, The Chemical Rubber Company, Ohio, 1969.

42. Wei, Peter, A Review of Experiments Pertinent to the After-Heat Measurements of the Thermal Neutron Fission of U-235, Masters Thesis, Oregon State University, 1975.

43. Yarnell, J. L., and Bendt, P. J., "Fission Product Decay Heat Studies", Los Alamos Scientific Laboratory, December 15, 1975.

44. Zobel, W., et al, "The Spectra of Gamma Rays Associated with Thermal Neutron Fission of U-235", IAEA Symposium on Pile Neutron Research in Physics, Vienna, 1962.

APPENDICES

Appendix A

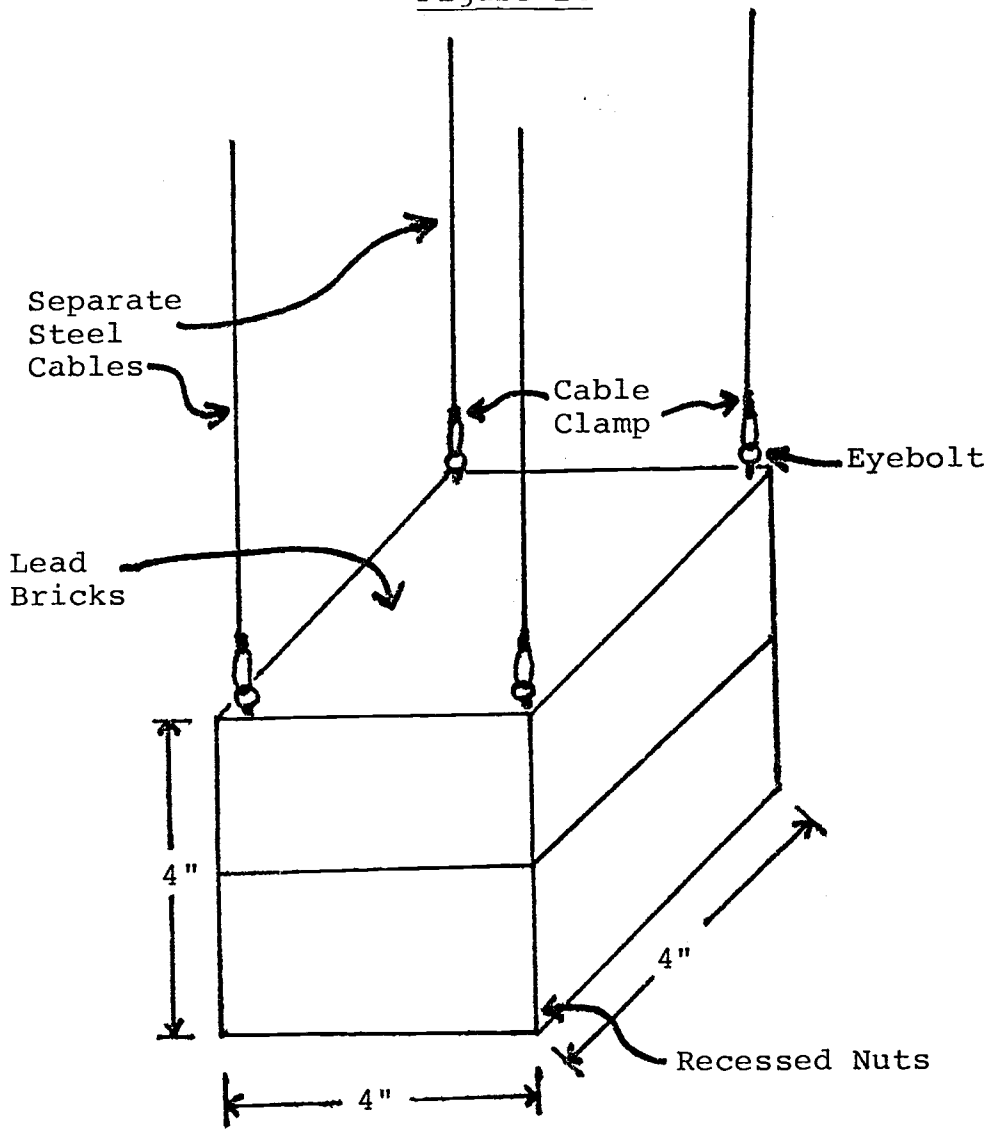
Neutron Flux Determination Adjacent to OSTR Core

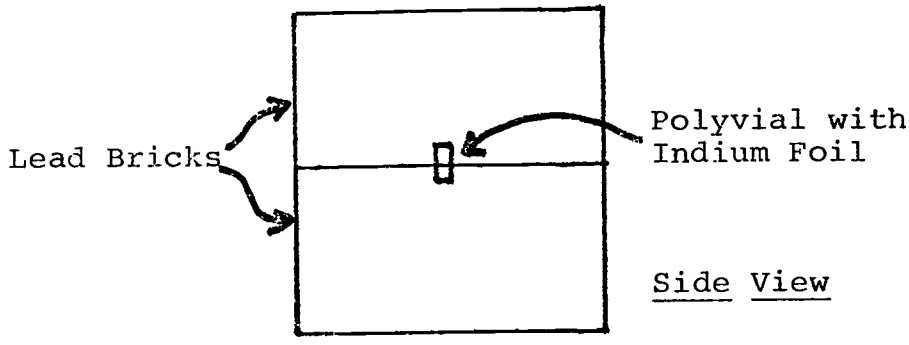
In order to be assured that the OSTR has a sufficiently high neutron flux to make the project feasible, a standard indium foil activation experiment was conducted on October 21, 1975.

The proposed size of the dilatometer apparatus is small enough that it can be placed atop the graphite relector of the OSTR. To simulate the absorption of neutrons by bismuth, the foil was encased in lead as shown in Figure 14. This mockup was lowered onto the reflector. Safety precautions, expected radioactivities, and safety analysis for this experiment are detailed in OSTR Experiment B-27.

The 0.2248 gram indium foil was activated at 09:27 during a pulse of 10 milliseconds duration at 1300 MWt peak power ($2.88 insertion). The foil was allowed to decay for 767 minutes and was counted at 22:14 using a Ge(Li) detector and the 4000 channel pulse height analyser. The activity of the 1.097 MeV gamma photon was 12.61 cps. The activity of the 1.293 MeV gamma was 14.97 cps. Through correlation with a known National Bureau of Stansards Standard Reference Material ^{60}Co gamma standard of comparable energy (1.173 and 1.322 MeV gamma's), and accounting for the isotopic abundances of the gamma photons, an average of 8.73×10^8 dps was obtained for the activity of the ^{116m}In at the end of bombardment. We are able to solve for the flux by re-writing the activa-

Figure 14





Lead Mockup of Bismuth Dilatometer for
Flux Determination Adjacent to OSTR Core

tion equation:

$$A = N \cdot \sigma \cdot \Phi \cdot \left(1 - e^{-\lambda t} \right) \quad , \quad \text{(A-1)}$$

as

$$\Phi = \frac{A}{N \cdot \sigma \cdot \left(1 - e^{-\lambda t} \right)} \quad . \quad \text{(A-2)}$$

Plugging in values,

$$\Phi = \frac{8.73 \times 10^{8}}{\left(\frac{0.2248}{115}\right)\left(6.022 \times 10^{23}\right)\left(70 \times 10^{-24}\right)\left(1 - \left(\exp\left(-\frac{\ln 2}{54.2} \cdot 16.6\right)\right)\right.}$$

$= 4.97 \times 10^{15}$ neutron/cm^{2}sec at 1300 MWt

$= 3.82 \times 10^{12}$ neutron/cm^{2}sec·MWt

$\simeq 10^{16}$ neutrons/cm^{2}sec at 2600 MWt.

As noted in the experimental strategy section, a flux level of 5×10^{15} neutrons/cm^{2}second will be adequate to make the experimental determination of energy release after thermal neutron fission of ^{235}U feasible. We actually have a reserve as the flux is in excess of 10^{16} neutrons/cm^{2}second for a peak pulse on the OSTR.

FORTRAN Program FISSACT

FORTRAN computer program FISSACT, as listed on the next page, was developed to yield the time-dependent gross fission product activity after thermal neutron fission of ^{235}U.

The program utilizes the empirical relation:[13]

$$A \propto t^{-1.2} \quad . \qquad (A\text{-}3)$$

This relationship is valid for times in excess of ten seconds. Naturally, if time equal to zero is input, the equation will incorrectly yield infinity for the activity. The computer will print 0.0 for time equal to zero.

This program has two options: 1) It will calculate the activity at a specific time, or, 2) It will generate an activity profile between two time values.

Pages faded, best available scan. Page: 83, 86, and 87.


```
      PROGRAM FISSACT(INPUT,OUTPUT,TAPE76)
C******  PROGRAM FISSACT -- CALCULATES CROSS FISSION  ***
C******  FISSION PRODUCT ACTIVITY USING THE EMPIRICAL ***
C******  TIME TO THE MINUS 1.2 RELATION.  THE CALCU-  ***
C******  LATION OPTIONS ARE AVAILABLE -- A SELECTED   ***
C******  TIME OR A TIME SCAN.  SINCE T TO THE -1.2 IS ***
C******  ZERO FOR T=0, THE FOR ACTIVITY IS NOT AVAIL- ***
C******  ABLE IN THIS PROGRAM.                        ***
C******  STEPHEN H. SHEPHERD, MAY, 1976               ***
      DIMENSION TIME(100),BETA(100),GAMMA(100),TOTAL(100)
      PRINT*,'NUMBER OF FISSIONS'
      READ*,FISNUM
      STEP=START=0.0
      CONST=-1.2
      PRINT*,'DO YOU WISH SELECTED TIME(OPT1),OR TIME SCAN(OPT2)'
      READ*,OPT
      IF (OPT.EQ.2) GOTO 50
      PRINT*,'TIME IN DAYS'
      READ*,TIME(1)
      INTERV=1
      GOTO 100
   50 PRINT*,'BEGINNING DAY (EG., 0,1,2,3,4...)'
      PRINT*,'** CAUTION, AT T=0, THE EQUATION DOES NOT HOLD **'
      PRINT*,'** IT WILL PRINT 0.0 FOR T=0 ACTIVITIES **'
      READ*,ISTART
   70 PRINT*,'ENDING DAY'
      READ*,END
      PRINT*,'NUMBER OF INTERVALS - LESS THAN 101'
      READ*,INTERV
      XINTER=INTERV
      STEP=(END-START)/XINTER
      INTERV=INTERV+1
  100 DO 200 I=1,INTERV
      XI=I
      J=I+1
      BETA(I)=FISNUM*3.2E-6*TIME(I)**CONST
      BETA(I)=BETA(I)/3.7E+7
      TIME(J)=START+(XI*STEP)
      GAMMA(I)=BETA(I)*0.5
      TOTAL(I)=BETA(I)+GAMMA(I)
  200 CONTINUE
      DO 300 I=1,INTERV
      WRITE(76,400) TIME(I),BETA(I),GAMMA(I),TOTAL(I)
  300 CONTINUE
      PRINT*,'  TIME        BETA ACTIVITY     GAMMA ACTIVITY'
     1,'   TOTAL ACTIVITY'
      PRINT*,' IN DAYS      (MILLICURIES)     (MILLICURIES)'
     1,'     (MILLICURIES)'
      PRINT*,' '
      DO 400 J=1,INTERV
      PRINT 400, TIME(I),BETA(I),GAMMA(I),TOTAL(I)
  400 CONTINUE
      END FILE 76
      PRINT 950
  950 FORMAT(1X,////////////1X)
      PRINT*,'THANK YOU, A COPY OF THE OUTPUT IS AVAILABLE ON TAPE76'
  401 FORMAT(2X,F7.2,2(2X,1PE11.4,2X))
      STOP
      END
```

FORTRAN Program ACTEOB

FORTRAN computer program ACTEOB, as listed on pages 86 and 87, determines the detailed fission product inventories necessary to devise radiological precautions for the experiment. As it uses independent fission product yields, from a computer data file such as CINDAT[29] its temporal validity is limited to short times after fission. However, the program will list out the radioactivities at one day and at one week after fission for informational purposes. While not strictly accurate, they should serve as general or ballpark estimates. A more sophisticated FORTRAN computer program, such as CINDER,[11] could be used to perform these calculations if they are desired.

The program reads the fission yields, calculates the number of atoms present at the end of bombardment, then converts to activity in curies. It reads alphanumeric characters of the chemical symbols from a sequential data file and then correlates these with the atomic number. Distinction is also made of the various meta-stable states which occur in the data file through the use of alphanumerics.

The total curies of activity, at the end of bombardment, and the total independent yield (in our case 99.99994%) are calculated and listed out for the user.

The program output is listed on a computer file grouped by isotopes of each element. If only one particular ele-

mental determination is desired, rather than all the fission products, the DO statement at line 40 can be replaced by as input request. This will yield a file of the isotopes of the input element.

```
      PROGRAM ACTFOR(INPUT,OUTPUT,TAPE25,TAPE26,TAPE30)
C******  PROGRAM ACTFOR -- CALCULATES RADIOLOGICAL ACTIVITIES ***
C******  OF FISSION PRODUCTS.  READS THE FISSION YIELDS FROM A ***
C******  DATA FILE SUCH AS CINDAT(TAPE25), DOES THE DECAY  ***
C******  AND ACTIVITY CALCULATION, AND THEN WRITES OUT THE DETAILS ***
C******  FOR EACH RADIONUCLIDE, GROUPED AS ISOTOPES.  IT IS     ***
C******  NECESSARY TO HAVE A SEQUENTIAL ALPHANUMERIC DATA FILE   ***
C******  OF ALL THE SYMBOLS OF THE ELEMENTS(TAPE30)              ***
C******  STEPHEN R. SHEPHERD, MAY, 1976                          ***
      DIMENSION ALPHA(100),ALMET(4)
      REAL LAMBDA,LAMTIM
      INTEGER A,Z,META
      ALMET(1)='  '
      ALMET(2)='M '
      ALMET(3)='M2'
      ALMET(4)='M3'
      TOTCI=0.0
      SUMYLD=0.0
      WRITE(26,7000)
      WRITE(26,1000)
 1000 FORMAT(2X,'RADIO-',4X,'DECAY',5X,'YIELD',5X,'ATOMS'
     1,3X,'ACTIVITY',2(2X,'ACTIVITY'),/1X,'NUCLIDE',2X,
     2'CONSTANT',2X,'FRACTION',2X,' AT EOE ',3X,' AT EOF ',
     23X,'AT 1 DAY',2X,'AT 1 YR'/)
      REWIND 25
      REWIND 30
C******  FILL THE ALPHANUMERIC ARRAY WITH ELEMENT SYMBOLS
      DO 25 I=1,100
      READ(30,3000) ALPHA(I)
   25 CONTINUE
 3000 FORMAT(A2)
      PRINT*,'NUMBER OF FISSIONS'
      READ*,FISS
   40 DO 999 INPUT=26,67
      CITOT=0.0
      SUM=0.0
C******  READ FROM CINDAT -- META=METASTABLE STATE NUMBER
   50 READ(25,4000) A,Z,META,LAMBDA,YIELD
      META=META+1
C******  CHECK FOR END OF CINDAT FILE
      IF (Z.EQ.68) GOTO 80
      IF (Z.NE.INPUT) GOTO 50
      ATOMS=FISS*YIELD
      SUMYLD=SUMYLD+YIELD
      SUM=SUM+ATOMS
      ACTOEF=ATOMS*LAMBDA
      EOECI=EOEACT/3.7E+10
      CITOT=CITOT+EOECI
      LAMTIM=LAMBDA*8.64E+4
      IF(LAMTIM.GT.150) GOTO 70
      DAYACT=EOEACT*EXP(-1*LAMTIM)
      GOTO 75
   70 DAYACT=0.0
      GOTO 100
```

```
   70  LAMTIME=LAMBDA*(CANCO
       IF(LAMTIME.GT.150) GOTO 90
       DAYTACT=FOTACT*EXP(-1*LAMTIME)
       GOTO 100
   90  DAYTACT=0.0
  100  DAYTCI=DAYTACT/3.7E+10
       DAYTCI=DAYTACT/3.7E+10
 7000  FORMAT(1H1,15X,'ALL ACTIVITIES ARE IN CURIES',/1X)
       WRITE(26,5000) A,ALMET(IMETA),ALPHA(Z),LAMBDA,YIELD,
      1ATOMS,FOTCI,DAYTCI,DAYTCI
       GOTO 50
 5000  FORMAT(1X,I3,2X,A2,6(1X,1PE9.3))
  200  WRITE(26,5100) ALPHA(INPUT),SUM,CISUM
       WRITE(26,7000)
       WRITE(27,1000)
       REWIND 25
       TOTCI=TOTCI+CISUM
 7000  CONTINUE
       PRINT*,'TOTAL ACTIVITY AT EOF IS',TOTCI
       PRINT*,'SUM OF YIELD IS',SUMYLD
 5100  FORMAT(1X,/1X,'SUM OF ',A2,' ATOMS IS ',1PE9.3,1X,
      1', TOTAL ACTIVITY AT EOF IS ',1PE9.3)
       PRINT*,'THANK YOU, OUTPUT IS REWOUND ON TAPE26'
       REWIND 26
 4000  FORMAT(1X,I3,I9,I3,1PE10.4,40X,1PE10.4)
       STOP
       END
```