

# PREFACE

The symposium commemorates the 50th Anniversary of the Phoenix Memorial Laboratory. Originally it was seen as an opportunity to briefly review the many radiationrelated enterprises in which Nuclear Engineering alumni have been or are now engaged. We believe that for many of these the beginning interest can be traced back, at least in part, to the existence of the Phoenix Laboratory and the Ford Reactor. In large measure the abstracts herein have indeed captured that opportunity. However, the symposium has been expanded somewhat to include technical programs carried on by other University of Michigan scientists and engineers. In addition, the symposium addresses several programs which are of current and future importance at the FNR and indeed at all medium power research reactors, but which have had less historic attention at Michigan. Taken together, we hope the symposium does adequately celebrate the past, present, and future of the Phoenix program, and does show that its original charter has been preserved.

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Honorary symposium chair: William Kerr Program committee chair: Jacob I. Trombka Co-chair: John S. King Co-chair: Ronald F. Fleming Committee: Hadi Bozorgmanesh Edward Ficaro David Gilliam Brent J. Heuser

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Lee A. Feldkamp, Ford Motor Company, Ford Research Laboratory

# **Historical Perspectives on PML/FNR**

Session Chair: John C. Lee

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## The Early Years of the Ford Nuclear Reactor

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This presentation will emphasize the design, construction, and early operation of the Ford Nuclear Reactor. It will pay tribute to some of the pioneering investigators who used the reactor to develop methods for measuring reactor characteristics, to some who used it as a neutron source to develop neutron scattering as an investigative tool, to some who used it as a tool in developing methods for measuring cross sections for neutron interactions with nuclei, and to some who used it to further investigate the characteristics of elements associated with the fission process. Lastly it will mention the reactor's use as an important part of formal laboratories in which students are introduced to methods that are used to ensure safe operation of devices that make use of critical nuclear systems for various purposes.

# Promoting the 'Beneficent' Use of Atomic Energy through Research: A Brief History of the Phoenix Project

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Looking back at the recently ended World War, UM Alumnus Fred Smith noted that there was "an unlimited amount of work to be done in the world to make it safe, free, and a place in which to live .... " His suggestion that the University of Michigan's memorial to those who served in the War should be a program to explore the peaceful uses of atomic energy provided the inspiration for the Michigan Memorial Phoenix Project. As established just over fifty years ago, MMPP was dedicated "to explore the ways and means by which the potentialities of atomic energy may become a beneficent influence in the life of man." After briefly describing the early history of MMPP, this paper presents an overview of the MMPP research program, looking particularly at funding distribution, levels, and trends. Based on this analysis it is suggested that consideration should be given to refocusing the MMPP research program more specifically on the initial goals of the Project. Given the current uncertain state of the future of nuclear energy and nuclear research more broadly, there is as much need today to explore and thereby to demonstrate to the public the "beneficence" of atomic energy as there was when MMPP was established.

## Days and Nights of Neutron Scattering Research at the FNR

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During the interval of approximately 1960 to about 1980, the FNR was host to the birth, life, and then slow decline of a wonderful confluence of excited people and exciting experiments. The excited people included NE faculty and visiting scientists, and, over the years, some 26 remarkable graduate students. From ground zero spectrometers were designed, built, and put into operation at A, F, G, H and J ports. A few of the students were theorists; but most had to master daunting experimental techniques and at the same time be able to exploit the new generalized neutron scattering theory<sup>T</sup>. PML and the FNR beam port floor were lighted 24 hours a day, and the population was about the same at 2:00 AM as at 2:00 PM. The confluence brought together an unusual collaboration between theorists and experimentalists because the neutron data were complex and depended strongly on theoretical calculation. Excellent contact developed with an international community of spectroscopists, three UM international symposia were orchestrated by NE's Paul Zweifel in Michigan, and for many years a graduate seminar course pulled in an audience of 20 to 25 people to learn how to use quantum mechanical and group theoretical techniques to interpret scattering data. The best lectures were often those prepared by our students! It is fair to say that in that atmosphere, the UM-NE/FNR program took its place among the leading world laboratories in neutron spectroscopy.

Why was there such excitement? I believe there were three key reasons. Foremost was our recognition, along with the rest of the world, that thermal neutrons possessed unique properties for probing condensed matter, which conventional probes, i.e. Raman scattering, x-ray scattering, infra-red absorption, electron spectroscopy, and NMR- did not have. Those properties will be recalled briefly in this talk. Second, the necessity to manage both instrumentation and complex theory at the same time had great intellectual appeal. Third, new experimental methods, particularly TOF phased choppers and the "constant-Q" technique for triple-axis crystal spectrometers were just being published, and we were eager to emulate them. I might add a fourth reason which I am sure will beg some disagreement: our choice of experiments was "wide open"; there were not many restrictions required by "programmatic" or "strategic" research funding in those days.

What scattering experiments were done? A broad spectrum of experiments were undertaken. Elastic and inelastic scattering measurements were made on pressurized molecular gases, on molecular liquids, and on crystalline and glassy solids. Crystal structures for simple metals and for complex molecular crystals were observed by Bragg diffraction. Both coherent and incoherent inelastic scattering was done on semiconductors and on polymers. Aniferromagnetism was characterized in chromium and in metal oxides. Inelastic experiments at a 1.0 to 2.0 Mw light water reactor took blind faith and heart. We had some of both.

What did we accomplish? A number of physical or chemical parameters, which have withstood the test of time, were established for the first time by these experiments. A number of important experimental innovations were created as well. This paper will attempt to list them. But surely the greatest contribution was to give 26 great students an extraordinary education.

1. A. C. ZEMACH and R. J. GLAUBER, *Phys. Rev.*, **101**, No. 1, 118 (1956)

#### **Developments in Radiation Instrumentation**

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Over the period of time represented by the existence of the Phoenix Project, there are some interesting contrasts in the evolution of instruments used to detect and record the properties of ionizing radiation. Some types of sensors have changed very little, and an experimentalist transported from the late 1950s would not have great difficulty using many modern instruments. These would include air-filled ion chambers, traditional proportional counters, Geiger tubes, and sodium iodide scintillators for gamma rays. together with BF3 tubes, proton recoil detectors, and activation foils for neutrons. Much more significant changes have taken place in other types of detectors, and there are some new categories that were entirely unknown even one or two decades ago. In common with other areas of technology, the most dramatic changes are seen in semiconductor-based instruments. The emergence of germanium detectors in the early 60s revolutionized gamma ray spectroscopy overnight. Silicon diodes have replaced older instruments as the standard approach to charged particle spectroscopy. New cryogenic detectors offer energy resolution unprecedented in traditional sensors. But perhaps the most important changes have occurred in the electronics available to process and record the signals from radiation detectors. The multichannel analyzer first available in the early 60s made spectroscopy routine. Today, a quiet revolution in pulse processing electronics is taking place that may have similar effect. Fast analog-to-digital converters now permit digital techniques to replace traditional analog methods for the shaping and recording of spectroscopic information from detectors. The advances in microelectronics have also led to readout chips that can process the outputs of detectors with many individual channels. allowing unprecedented advances in position-sensing and imaging detectors. Some examples from our research will be shown in which this new capability can also be used to overcome inherent limitations in specific detectors.

#### How Things were in the Early Days

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Gamma Radiation. In the early days before the start up of the Ford Nuclear Reactor much of the activity at the Michigan Memorial Phoenix Project involved the use of gamma rays from spent fission products or from Cobalt-60 radiation sources. First work was done at the Fission Products Laboratory (FPL) where a 1000 Ci and a 9250 Ci source were used. Systems examined included the chlorination of benzene and of toluene, polymerization of ethylene and the sterilization of foods accompanied by small animal feeding studies. As the Phoenix Memorial Laboratory (PML) building became available a Cobalt-60 facility was installed with a nominal 3500 Ci source placed on an elevator and stored under 17 feet of water. Color photographs show the preparation underway for a new source. The holders will contain 35 aluminum pencils into each of which are inserted nine 0.25" by 1" Cobalt-60 pellets encased in aluminum. The Cobalt-60 was prepared by neutron irradiation at the Chalk River, Ontario reactor. Insertion of the pellets and closure of the pencils by cotter-like pins is shown. The irradiation room where material is exposed contains a 21x21inch opening to the wall. When samples are arranged in known locations, the room is evacuated and the source elevator is raised for the desired radiation time and then lowered. The shielding by the water in the well makes it safe to reenter. Gamma dose rates in and around the source were determined by the Fricke ferrous sulfate dosimeter where oxidation to ferric sulfate was measured by optical density change using a Beckman DU Ultraviolet Spectrophotometer.

Texaco Radiation Laboratory. The Texaco radiation laboratory contained a 29,100 Ci Cobalt-60 source, obtained from the Chalk River Reactor. The 156 pencils arranged in a cylindrical rack permits insertion of any object up to 21 inches in diameter and 10 inches high into the center of the array. Employing methods and techniques learned at the Phoenix Laboratory the source was use to qualify greases and lubricants for the Nuclear Ship Savannah and was used for study of gamma ray effects on catalysts for a number of hydrocarbon processes. The principal use was the development of a high level dosimeter effective in the range of  $1 \times 10^6$  to  $6.5 \times 10^8$  rads using the radiolysis of purified acetonitrile. The laboratory also had a linear accelerator which provided 3.5 to 4 million electron volt electrons. Radiation of acetonitrile using calorimetric measurements of dose show in spite of an instantaneous dose rate from the electrons that is 484,000 times that from the isotope source.

<u>The Ford Nuclear Reactor</u> Photos are of the Ford Nuclear Reactor before and after criticality in 1957. The bridge and the control room are shown. Pneumatic sample delivery tubes and the access ports and tubes for samples of equipment placement near the reactor core are seen. A chemical pilot unit for insertion in the tubes for operation in the neutron and gamma ray field is shown. The control console and the instrument recorders direct and manage the reactor operation. Gold foil placement by the reactor crew was used to determine reactor dose rate around the core and the foil activation is measured in the counting room. The gamma ray field was measured by the ferrous sulfate dosimeter which was also used for the relative neutron measurements by wrapping cadmium foil around the sample vials and the resulting neutron-gamma reaction gave incremental oxidation of the ferrous sulfate proportional to the neutron flux. The Cerenkov radiation provided an unexcelled view of the operating reactor core.

# Utilization of Research and Test Reactors – Part I

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Session Chair: David K. Wehe

# History of Radiopharmaceutical Development for Nuclear Medicine Imaging and Therapy

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For the past three decades the University of Michigan's Division of Nuclear Medicine has conducted research aimed at developing radioiodinated tracers for diagnosis and treatment of disease. The Phoenix Memorial Laboratory has played a central role in this effort. This working relationship began in the early 1970's when a radioiodinated derivative of cholesterol was shown to visualize tumors of the adrenal gland. PML staff produced iodine-131 labeled cholesterol for the first clinical studies and their commitment expanded during the 70's to providing this investigational tracer routinely to medical centers in the U.S. and Canada. This scenario was repeated a decade later on a much larger scale with MIBG, a mime of norepinephrine developed by the Division's basic research group. Subsequently five pharmaceutical preparations of radioiodinated MIBG using three different isotopes (I-131, I-125, I-123) were produced at PML for diagnostic imaging and therapy of diseases such as neuroblastoma, a virulent childhood cancer. Increasing demand for MIBG saw it emerge from orphan drug to commercial product following adoption by CIS-US, Inc. Presently, there is world-wide use of MIBG, especially in Japan where the I-123 tagged tracer is used to determine regional nerve loss in the heart. More recent research directed at Alzheimer's disease produced a tracer that maps brain decreases in the transporter of the transmitter acetylcholine.

This presentation will be a very personal, biased account of some of the more memorable events highlighting this unique and productive 30-year collaboration.

## Research Activities at Sandia National Laboratories' Reactor Facilities

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Sandia National Laboratories has operated nine nuclear reactors and critical facilities over the last four decades. Currently Sandia has two operational reactor facilities. The Annular Core Research Reactor Facility is a pool-type reactor with two types of fuel that can be arranged in a variety of core configurations for pulse and steady state capability. The Sandia Pulsed Reactor Facility houses two bare-metal fast-burst reactor cores that can be operated interchangeably.

The Sandia reactors are multi-program facilities with a variety of missions. Typically the reactors are utilized to provide a specified radiation environment over a large experiment exposure volume. Their strength is the flexibility to tailor configurations to meet unique test requirements.

The primary mission for both reactors is to simulate hostile radiation environments that materials and systems will encounter in space or military applications. The SPR Facility produces a hard neutron energy spectrum that is important to radiation effects in electronics. The facility is widely utilized by Sandia organizations developing and certifying electronics for military applications and similar work for Defense Contractors from the private sector. Other recent applications involve reactor-pumped lasers, calibrations for criticality dosimetry for Oak Ridge National Laboratory and placing desired displacement damage in commercial semiconductor wafers.

The ACRR Facility produces a moderated fission spectrum important to energy deposition and structural integrity for materials. The facility is utilized by Los Alamos National Laboratory and Lawrence Livermore National Laboratory for the testing and certification of materials and nuclear stockpile components. Past applications include safety research for the Nuclear Regulatory Commission and the Department of Energy simulating reactor cores under accident conditions, reactor-pumped lasers, and currently the capability to produce radioactive isotopes for nuclear medicine applications.

# Use of Nuclear Research Reactors in Multidisciplinary Research and Education

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Neutrons produced in nuclear research reactors are widely used in science and engineering. Since neutrons are highly penetrating they can be used to probe deep within the materials to characterize structure or to obtain analytical chemical analysis. Most commonly used reactor-based nuclear methods are: i) Neutron Capture Methods; Neutron Activation Analysis (NAA), Cold-Neutron Prompt Gamma Activation Analysis (PGAA), Neutron Radiography (NRad), Neutron Depth Profiling (NDP), and Boron Neutron Capture Therapy (BNCT) and ii) Neutron Scattering/Diffraction Methods; Small Angle Neutron Scattering, (SANS) Neutron Reflectometer (NRef) and Powder Diffraction (PD). Among these techniques, NAA, Cold Neutron-PGAA, NRad, NDP and BNCT are of interest at the Ward Center for Nuclear Sciences at Cornell University. Brief descriptions, new developments, and current and planned applications of these techniques at Cornell University will be presented. Utilization of Cornell TRIGA research reactor for multidisciplinary teaching and research will be discussed.

# Reactor Noise Research at the Ford Nuclear Reactor

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One person's noise is another person's music! Reactor noise is the fluctuation of the neutron density in a nuclear reactor. From the characteristics of the reactor noise, it is possible to determine some of the key parameters of a nuclear reactor. In the early days, the most common parameters measured were the prompt neutron generation time and the subcritical reactivity. UM research at the FNR and elsewhere contributed to measurement of these parameters plus to the understanding of the role of delayed neutrons in reactor noise. Reactor noise research spread all over the globe and it was found that other key nuclear reactor properties could be determined by reactor noise measurements. Properties of most interest are core barrel fluctuations, fuel vibrations, voiding, xenon oscillations, coolant jetting, reactivity, loose parts detection, feedback parameter determination.

# Utilization of Research and Test Reactors – Part II

Session Chair: Brent J. Heuser

#### The Role of the Michigan Memorial Phoenix Project in Irradiation Material Science

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The Ford Nuclear Reactor (FNR)/Michigan Memorial Phoenix Project (MMPP) is playing a key role in solving the problem of irradiation embrittlement of reactor structural steels, which is one of the key issues in the broad field of irradiation materials science. Following a brief introductory remarks on irradiation effects on materials as manifested by the embrittlement phenomena, the talk will focus on: a) the unique irradiation facilities that have been developed at the FNR/MMPP; and b) how they are being used to support NRC funded embrittlement programs led by researchers at ORNL and UC Santa Barbara. The Heavy Section Steel Irradiation (HSSI) Program at ORNL currently operates the facilities that were designed by ORNL in collaboration with UCSB. The combination, UCSB's irradiation variable (IVAR) facility and HSSI's Irradiation Annealing Re-Irradiation (IAR) facility provide capabilities that are unique on a worldwide basis. Indeed, they are supporting increased international participation in the irradiations at MMPP. The enabling role that FNR neutrons provide to cutting edge materials science is also described.

# Focusing of Cold Neutrons by Capillary Optics

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The principle of multiple mirror reflection from smooth surfaces at small grazing angles enables the transport of high intensity thermal neutrons beams to locations of low background for neutron scattering experiments and to provide facilities for multiple instruments. Miniature versions of neutron guides, glass polycapillary fibers with thousands of channels, can be used to transport, bend and focus cold (~0.4 nm) neutron beams. The advantage of the narrow (~10  $\mu$ m) channels is that the neutron beam can be bent more sharply than for the wide (~50 mm) guides. The neutron trajectories have more reflections through narrow channels, so that the coefficient of reflection must be high for a reasonable transmission through the channels.

Neutron absorption techniques in analytical and materials research such neutron depth profiling, NDP and prompt gamma activation analysis, PGAA) do not require a monochromatic neutron beam, nor a small angular divergence. Consequently, a high intensity beam focused by a neutron capillary lens onto a small sample area can be useful for improving both the detection limits of individual elements and the spatial resolution of the measurement.

A polycapillary fiber lens containing 1763 fibers of outer diameter 0.5 mm and length 125 mm, each with 1657 hollow channels of diameter 10  $\mu$ m has been constructed. The lens compresses a polychromatic neutron beam of cross section 50 mm by 45 mm and current density 6.5 108 cm-2 s-1 onto a focal spot of diameter 0.53 mm (FWHM) with a transverse profile with a Gaussian distribution at a distance of 52 mm from the exit of the lens, and with an average gain of 80 in neutron current density. The transmission characteristics depend strongly on the divergence of the incident beam. PGAA measurements have demonstrated a gain in gamma count rate of 60, a gain in signal-to-noise of 7, and an improvement in detection limits by a factor of 20, as well as submillimeter spatial resolution for boron, cadmium and gadolinium. These increases are obtained despite the fact that only 3.5% of the neutron beam incident on the lens is delivered to the focus.

The background from unfocused neutrons can be reduced and the lens efficiency increased by capturing a larger fraction of neutrons at the entrance of the lens. This can be achieved in angular space by the use of metal capillaries with larger critical angles than glass, and in real space by the use of a monolithic lens consisting of tapered capillaries, such that a greater fraction of the entrance area is used. Suitable metal capillaries cannot be produced presently. A prototype monolithic lens with entrance and exit diameters of 4 mm and 2.5 mm respectively, and length 40 mm, has been tested for NDP measurements. The lens gives a factor of 8 increase in neutron current density within the focal spot of diameter 0.21 mm (FWHM of the Gaussian distribution) at a plane 22 mm from the exit of the lens.

Another alternative to reduce the background from unfocused neutrons is to create the focus outside the direct path of the incident beam, enabling the sample under investigation to be placed in a region of lower background. A glass polycapillary lens that both bends and focuses a cold neutron beam has been constructed. The bender-focuser guides part of the incident beam away from the line of sight and focuses it to a spot of width 0.65 mm at a distance 95 mm from the lens exit and 20 mm below the bottom edge of the beam path, with a gain of 20 in neutron current density.

# Small Angle Neutron Scattering: Retrospect and Prospects

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Over 50 years ago, Paul Flory predicted on intuitive grounds that polymer chains would adopt random-walk configurations in the amorphous state. For several decades, only indirect evidence was available to support this assertion, until the application of small-angle neutron scattering (SANS) techniques to polymers in the early 1970s. The marked difference in scattering power between hydrogen and deuterium atoms means that isotopic labeling may be used to "color" polymer molecules and make them "visible" in the condensed state. The first experiments provided dramatic direct evidence for Flory's original prediction, and over the past two decades SANS has developed into an extremely powerful method for the study of polymer structure. It has also found wide application in the study of colloidal materials, detergents, high-temperature superconductivity etc.

The new HB4 Cold Source and Guide Halls at the ORNL High Flux Isotope Reactor (HFIR) will provide greatly increased SANS intensities (see HFIR website at "http://neutrons.ornl.gov/NSatHFIR/Upgrades/HFIRUpFac.HTML") and the instruments will be available to external users from the general US scientific community (universities, industry, Government laboratories etc.). This lecture will give a brief review of the role of neutron scattering in polymer and colloid science and will concentrate on practical applications of the technique.

#### Neutron Detector Developments at the Phoenix Memorial Lab

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Neutron absorbing and reactive materials, when used in the solid state, can be manipulated to produce compact detectors for thermal and low energy epithermal neutrons. Four salient reactions are presently under investigation for use as semiconductor based thermal neutron detectors, those being the <sup>10</sup>B(n, $\alpha$ )<sup>7</sup>Li reaction, the <sup>6</sup>Li(n, $\alpha$ )<sup>3</sup>H reaction, the <sup>113</sup>Cd(n, $\gamma$ )<sup>114</sup>Cd reaction and the <sup>157</sup>Gd(n, $\gamma$ )<sup>158</sup>Gd reaction. Two basic detector forms are being investigated, those being solid state semiconductor neutron detectors and thin film coated semiconductor detectors.

The ideal material for a neutron detector is a substance that acts as both the neutron absorber and the detection medium. Semiconductors composed of one or more neutron reactive materials are excellent choices. Of the previously mentioned neutron reactive isotopes, only <sup>10</sup>B and <sup>113</sup>Cd can presently be produced as compound semiconductors. BAs and BP are semiconductors that offer many attractive properties, such as low gamma ray background interference, relatively high neutron absorption efficiency, high energy reaction products and chemical ruggedness. Unfortunately, due to intrinsic material problems, samples of BP and BAs acquired to date have not shown positive results for neutron detection. Cd compound semiconductors such as CdTe and CdZnTe are commercially available. Measurements in A port of the FNR have shown that CdZnTe can be used for neutron detection, in which prompt gamma rays (558.6 keV and 651.3 keV) were clearly observed. The main advantage of using Cd is the rather large absorption cross section for <sup>113</sup>Cd. Disadvantages include low detection efficiency of the prompt gamma ray emissions and high background gamma ray interference. Detection efficiency is presently calibrated at approximately 5% for a 3mm thick CdZnTe sample.

An alternative method of detecting neutrons with semiconductor detectors is by coating thin diodes with neutron reactive materials. GaAs based thin film coated detectors have been under investigation at the PML for many years, and three variations are under investigation. The earliest form is a radiation hard Schottky barrier design that utilizes the "truncated field" effect naturally occuring in SI bulk GaAs. The basic form is a bulk GaAs diode with a thin film of <sup>10</sup>B coated over the front. Maximum detection efficiency achievable is calculated to be approximately 4.8%, although the entire device (GaAs and <sup>10</sup>B film) is only 200 microns thick. Stacked variations can increase the overall efficiency. Self-biased designs have also been developed in an on-going effort with SPIRE Corporation. The detectors are composed of high purity n-type and p-type GaAs layers that, when grown onto each other, produce enough internal electrical potential to operate the device. Hence, <sup>10</sup>B coated detectors operate without an external voltage applied. The self-biased GaAs detectors are a new development, and advanced forms for pixellated arrays are presently being designed. Compound GaAs neutron detectors, being developed in collaboration with Argonne National Laboratories, utilize <sup>10</sup>B films over which a coating of HDPE is applied. Devices are presently being tested for sensitivity to both thermal and epithermal neutrons.

## Neutron Radioscopy and Radiography as a Quantitative Investigative Tool

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<u>Introduction</u>. Neutron Radioscopy and Neutron Radiography have been developing rapidly over the past few years into a major means of non-destructive testing. These include primarily, situations where the investigator wishes to follow the movement of hydrogenous fluids inside metallic structures or determine the alignment or presence/absence of internal parts or contaminants. In many cases the situation calls for the simple determination of whether the contaminant or the flow of the hydrogenous fluid is present or not. In other situations, the question is how much is there. This can be as simple as what is the thickness of the investigated parameter to what is the flow rate of the fluid.

<u>Dimensional Measurements</u>. When the desired information is how wide is a foreign substance or how wide is the gap between two components parallel to the beam direction, the problem has two difficult aspects. These include the correction for magnification of the object and blurring of the edges caused by beam unsharpness. The second difficult aspect is determining exactly where the edge of the object is. If the edge of the object is sharp, such as a cube with the edge parallel to the beam direction, it is easier to determine the edge provided the unsharpness has not overridden the resolution. On the other hand, if the edge is rounded, such as a round ball, the edge becomes extremely difficult to determine and the determining factor is the skill of the radiographer making the measurement. When the desired information is how thick is the fluid or substance of interest perpendicular to the beam direction standards must be developed using the substance of interest and placed in the beam at the same time as the measurement is being made.

<u>Flow Rates</u>. More and more, the information that is desired, is the flow rate of the fluid and is a very difficult process. If one is following a hydrogenous fluid and the channel is completely full, the flow rate can not be directly measured and neutron transparent fluids and tracers must be used.

# Industrial Applications of PGNAA and DGNAA

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Neutron activation analysis has made significant impact in the industrial community over the last 5 decades. NAA is not only an analytical tool but a strategic technique used by key industries in order to gain competitive advantage. The industrial role of NAA has risen not only because of the fundamental characteristics of the technique, also because of the improvements in radiation detectors, gamma-ray data acquisition systems and data manipulation capabilities. NAA methods are now capable of competing with other analytical tools in three key areas: 1) areas where provision of results can be achieved with appropriate precision, accuracy and sensitivity, 2) areas where NAA results can be obtained within reasonable times and 3) with reasonable cost. Traditionally, NAA was used in such industrial problems where all other conventional analytical methods are not applicable. This unfair position existed out of public sentiments and concern for radiation safety. Years of experience, research and methodology developments combined with the unique nature of neutrons, (the ability to penetrate and interact with the nucleus of most elements and yielding characteristics radiation), have proved that neutron activation analysis has several competitive advantages over other analytical tools. NAA also has a unique advantage of less prone to matrix type, unlike other analytical tools such as ICP-MS and XRF where significant sample preparations are necessarily done before application. The following are a few examples to illustrate successful industrial applications of NAA.

<u>Material Characterization</u>: At Dow Chemical company, NAA is one method of choice for the elemental analysis of many Dow materials. Characterization of chlorine content of several products manufactured with chlorine, measurement of residual bromide, assay of additives, measurement of trace impurities or monitoring of residual catalysts in both Dow products and user's products are routinely done using NAA.

<u>Quality control</u>: NAA analysis results are used to calibrate X-ray fluorescence (XRF) online analyzers that are used for monitoring bromine contents in plastics. This quality control measurement is made possible because NAA can be used to determine bromine content of plastics with higher degree of precision than on-line XRF.

<u>New product or method development</u>: The role of NAA in new product or method development is achieved through experimentation. The ability to analyze materials to some degree of precision and sensitivity, for several elements and in the presence of others is the main advantage.

<u>Process Control</u>: PGNAA is now a popular principle in the sensors of online bulk material analysis instrument. By incorporating PGNAA instrument online, industries such as cement have seen throughput improvement of over 10% while meeting the high quality standard expected of their products. Coal power plants and coal preparation plants have optimized their productivity and decreased pollution by using PGNAA instrument. Two advantages here are the penetrability of the neutrons into the bulk of the material thereby allowing analysis of kg size material without sampling errors and the speed of analysis.

# **Nuclear Power Technology**

Session Chair: Hadi Bozorgmanesh

#### Nuclear R&D Challenges for the New Millennium

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Nuclear energy today contributes about 20 percent of the world's electricity generation and will have to continue to play a major role in meeting the future energy demand. Throughout the world, in both developed and in developing economies, electricity is one of the most primary inputs to economic output. If economies are to grow, electricity will have to be supplied in unprecedented quantities in the future.

The alternative sources of this expanding electricity supply are few. Fossil fuels bring the uncertainty of atmospheric  $CO_2$  buildup. Fusion remains a long way off. Renewables are making only a painfully slow penetration in the market, and the ability to deliver an inexpensive product in the future is uncertain. Even if today's light water reactors could overcome public perception issues of waste disposal and safety, the ultimate contribution of this generation of reactors is limited because of their inability to use more than a small fraction of the uranium resource.

As we approach the new millennium, nuclear R&D challenges will be to develop advanced reactor concepts that will maintain nuclear as a long-term energy option. We should aim to utilize all the uranium resources. We need technological innovation to aid the waste disposal problem. The safety of plants should be based on the intrinsic characteristics of the technology, with less reliance on engineered systems and highly trained operators. The nonproliferation regime must be given primary attention so that we can avoid both separation of purified plutonium and unmitigated and dispersed buildup of plutonium stocks in spent fuel. And we must produce affordable nuclear electricity, comparable in price to the best of whatever viable alternatives can produce.

#### Nuclear Fuel Management Optimization: In Search of Cost Savings

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With an annual fuel cycle cost of about \$45M for a 1,000 MWe nuclear power plant, there is considerable economic incentive to reduce this cost, particularly in a deregulated electric utility environment. Nuclear fuel management for LWRs involves the following design decisions: lattice design, bundle design, number of fresh fuel bundles, burnt fuel to reinsert, core loading pattern (LP) and control rod patterns (CRP). Lattice design involves the specification of pin-wise fissile enrichments and burnable poison loadings. Bundle design involves the specification of the axial span of various lattice designs. The LP specifies where fresh and burnt assemblies (*i.e.* bundles) are to be placed in the reactor core. Finally, for BWRs the CRP specifies the position of control rods as a function of cycle exposure. Collectively, the nuclear fuel management optimization problem can be characterized as a large, combinatorial optimization problem. Given the size of the decision space, number of active constraints, non-linearity and lack of derivative information of objective functions and constraints with regard to decision variables, and computational effort to evaluate the objectives and constraints, the complete optimization problem cannot be solved at this time. For example, just the combined LP and CRP decision space involves greater than 10<sup>50</sup> combinations. This has necessitated the subdivision of the complete optimization problem into sub-optimization problems, specifically the following: lattice design, bundle design, out-of-core fuel management (*i.e.* number of fresh fuel bundles, and burnt fuel to reinsert), and in-core fuel management (*i.e.* LP and CRP).

Three advancements have taken place to make possible the solution of real-world nuclear fuel management sub-optimization problems, those being increase in computational power, development of highly efficient core simulators, and advancement of stochastic optimization. Stochastic optimization techniques, such as Simulated Annealing (SA) and Genetic Algorithms (GA), are generally viewed as necessary to employ due to problem attributes associated with nuclear fuel management optimization. Unfortunately, these methods require many evaluations of alternative decision variable values. Given the computational effort required to complete the associated neutronic evaluations, the need for high computational power and efficient core simulators follows.

Through NCSU's Electric Power Research Center (EPRC), research on sub-optimization problems has been conducted for BWRs and PWRs. The results of EPRC research and others are summarized in a recent review article<sup>1</sup>. This presentation will focus on in-core nuclear fuel management optimization. Potential objective functions and constraints will be presented. The basis of the SA optimization method will be explained and performance demonstrated. Formulation of a higher-order Generalized Perturbation Theory (GPT) core simulator will be developed and results demonstrating fidelity and computational efficiency summarized. Finally, a conjecture on the future direction of nuclear fuel management optimization will be made.

<sup>1</sup>P. J. Turinsky & G. T. Parks, "Advances in Nuclear Fuel Management for Light Water Reactors," Advances in Nuclear Science and Engineering, Editors: J. Lewins and M. Becker, V26, 137, Kluwer Academic/Plenum Publishers, New York, 1999.

#### Use of Spallation Neutrons to Drive and Control Subcritical Nuclear Systems

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The stability, control, and ultimately the safety of most nuclear reactors trace largely to physics characteristics such as delayed neutrons and negative reactivity feedbacks that lead to reduced power production when reactor temperatures increase. Such favorable characteristics are likely in thermal spectrum systems with uranium-based fuels, but are more difficult to achieve when fuels are based exclusively on transuranics. Such a fuel choice becomes desirable in missions focused on destroying plutonium or minor actinides, e.g., in efforts to reduce the inventory of these materials in waste repositories or other long-term storage (reducing concerns regarding the diversion of nuclear materials).

One option for dealing with fuels that have small delayed neutron fractions (minor actinides and plutonium), and near-zero (positive or negative) Doppler and/or coolant temperature coefficient is to drive and control the reaction use large quantities of source neutrons, driving the system significantly sub-critical. The resulting power production is proportional to the number of source neutrons and inversely proportional to the degree of subcriticality (S/(1-k)). The subcriticality can provide a significant safety margin should the reactivity creep upwards, although strongly positive reactivity feedbacks could still pose problems.

Spallation neutrons are produced when high energy particles (sufficient to overcome repulsion from the nucleus) are able to penetrate and interact with individual nucleons, effectively dislodging them via an intranuclear cascade process. A 1000 MeV proton will dislodge 20 to 30 neutrons from a heavy target of lead or tungsten, and even more from an actinide target.

As an example, consider an accelerator-driven transmutation system with fuel composed of 90% plutonium and 10% minor actinides. For a fast-spectrum blanket (surrounding a spallation target) producing about 1000 MWt of fission heat, the delayed neutron fraction will be around 0.25%, the Doppler feedback will be very close to zero, and the coolant temperature feedback could be positive in central regions of the target/blanket. If a k, effective of 0.97 if sufficiently subcritical to provide adequate safety margins, fundamental scaling laws indicate a proton beam power of about 13 MW would be needed. In contrast, if k, effective must be at or below 0.95 to assure adequate margins, a beam power of nearly 22 MW would be needed. Although proton beams in the range of several tens of megawatts are very credible using linear accelerators, the cost of providing 22 MW of beam power is at least 50% greater than the costs of providing a 13 MW beam. As a result, the design of accelerator-driven subcritical nuclear systems utilizing fuels with unfavorable reactivity characteristics (e.g., plutonium and minor actinides) requires extensive work in designing the system for favorable characteristics and then determining the necessary safety margins through careful analysis of the anticipated performance throughout the burnup cycle.

# Advancements in Nuclear Power Technology and Experience in Asia

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The increasing use of nuclear power as an energy source in Asia has been spurred by well recognized regional factors – the need for energy cost stability, lack of fossil resources particularly in northeastern Asia, and the emerging international consensus to limit the growth of greenhouse gas emissions while sustaining economic development. In the most developed Asian countries, including Japan and the Republic of Korea, existing nuclear generation as a percentage of total electric power generation is approximately double the worldwide average of 17%. The continued commitment to nuclear power in Asia is evidenced by the active programs of these countries and the planned expansion of nuclear power construction, notably in China.

The Republic of Korea's nuclear power program illustrates successful achievement of industry development and incorporation of advanced technologies in present day nuclear plant construction projects, which may serve as model for other development programs in Asia. This presentation will summarize major aspects of this program.

<u>Industry Development</u> The development of industrial infrastructures for nuclear power is guided by individual national goals for economic development, technology capability and security in energy supply. The Republic of Korea has pursued a policy to develop broadly based localization of design, manufacturing, and construction of nuclear power plants. This development over more than 20 years has achieved desired goals for self-reliance in supply and construction. Further development is focusing on international cooperation in research and development, and export capabilities in the field of nuclear power.

<u>Design Standardization</u> The Republic of Korea has conducted a program of design standardization based upon evolutionary advanced PWR technology. This program has produced the 1000 MWe class Korean Standard Nuclear Power Plant (KNSP) with four units in operation and currently four additional units under construction. The 1400 MWe class Korean Next Generation Reactor (KNGR) is under development for first unit operation in 2010.

<u>Operations Experience</u> The success of the KSNP program is indicated by the record of construction and startup and the performance of operating units. Key measures including nuclear capacity factors, unplanned shutdowns, and radwaste generation show that the KSNP has achieved desired improvements in plant operations in comparison to nuclear plants worldwide.

# Artificial Intelligence and Nuclear Engineering

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The field of Artificial Intelligence (AI) originated in 1956 when researchers met at Dartmouth College to discuss their work in pursuit of making machines behave more intelligently. The objective then was to develop *general purpose thinking machines*, which would have the ability to think like humans do. Over the years, however, the objectives and the definition of AI have changed as new and revived information processing technologies were incorporated into the field. This presentation will briefly review the four major AI technologies and describe their applications and future trends in nuclear engineering (NE).

<u>Expert Systems (ESs)</u>. ESs were the first practical application of AI. By processing symbolic information in a narrow but deep knowledge domain, ESs are able to perform at levels comparable to human specialists. Initially proposed as on-line reactor operator decision support systems, the use of ESs in NE has mushroomed over the years with applications ranging from control of crane movement during fuel shuffling to optimization of reactor loading patterns and to non-destructive testing.

<u>Fuzzy Logic (FL)</u>. Based on fuzzy set theory, which allows for the classification of a proposition into multiple classes with different degrees of membership, FL provides a formal mathematical framework for dealing with uncertainties. The most prominent use of FL has been in fuzzy controllers, which employ the structure of ESs for the control of processes through fuzzy linguistic descriptions. In NE, fuzzy controllers have been proposed, among other uses, for the control of the water level in steam generators and reactor power output.

<u>Neural Networks (NNs</u>). Inspired by the structure of the human brain, NNs are generic, powerful, nonlinear inductive modeling systems used for both classification tasks and regression tasks. This popular technology has been widely used in NE with applications ranging from power plant equipment monitoring and diagnostics to the prognosis of severe accidents and from plant performance analysis to on-line prediction of plant safety margins.

<u>Genetic Algorithms (GAs)</u>. Based on the concepts of biological evolution, GAs provide a robust, generic, and computationally simple procedure for systematized random search and optimization. In NE, it has been applied mainly in combinatorial optimization problems, such as the optimization of fuel loading patterns, reactor core design, and burnable poison design.

Due to stringent regulatory requirements in the nuclear industry, practical applications of AI in NE are not as ubiquitous as in other fields. AI systems that are amenable to V&V and that provide statistical measures of the reliability of their inferences will need to be developed before a significant increase in their practical applications in the nuclear industry occurs.

# **Applications of Nuclear Science and Technology – Part I**

Session Chair: David Gilliam

#### **Exploring the Universe with Nuclear Radiation**

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Our concept of the Universe through most of history has been strongly influenced by our knowledge gained through observations made in the visible portion of the electromagnetic spectrum. The first region of the spectrum beyond the optical to be explored was the radio range first observed in the 1930's. Detailed observations were not carried out until the 1950's. With the coming of the space age in the late 50's and 60's, observations were first carried out in the infrared, ultraviolet and x-ray regions of the electromagnetic spectrum.

Gamma-ray astronomy was the last major wavelength range to yield its wealth of information. This late development grew from a combination of factors, including the need to place telescopes and remote sensing systems above the Earth's atmosphere, the requirement to develop rather complex instruments and the relatively low intensity of gamma-rays photons, particularly in relation to the charged-particle cosmic-ray intensities. Even though the photon intensity is low, the energy emitted in the gamma-ray range may be quite high because each photon carries a very large amount of energy over this very broad spectral region.

Observations made in the gamma-ray region permits the direct study of the largest transfer of energy occurring in astrophysical processes, including rapid expansion processes, explosions, high-energy particle acceleration, gravitational accretion unto superdense objects, the fundamental process of the building of the elements, particle-antiparticle annihilation. Gamma-ray astronomical observations also find important application in studies of the evolution of our solar system, and the nature of the high-energy processes in Sun's atmosphere, and their relation to solar activity. Analyses of the characteristic x-rays and gamma-rays from planetary surfaces can be used to infer global distribution of major and, in some cases, minor and trace elements. Knowledge of the overall elemental composition of a given body can be related to the mechanism of condensation and accumulation of materials from the primordial solar nebula.

#### **New Applications of Stable Isotopes**

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<u>Introduction</u>. Isonics has supplied stable and radioactive isotopes for a wide variety of research, development and commercial applications for almost ten years. In that time, we've participated in the dramatic growth of several applications, the short term "brilliance" of others and the promise of still others. I've chosen the following examples:

<u>Depleted Zinc.</u> EPRI sponsored testing showed the addition of zinc to reactor coolant reduces plant radiation fields, and in some cases, greatly mitigates environmentally induced cracking. The exposure benefit is significantly offset by the production of <sup>65</sup>Zn from <sup>64</sup>Zn (natural abundance of 48%). Development of the cost effective Depleted Zinc product (zinc depleted to <1% <sup>64</sup>Zn) in 1993 eliminated this detrimental side effect. At 400+ kg/year, this has become one of the largest applications of stable isotopes.

<u>HeCd lasers</u>. The use of an enriched (90+%) even isotope of cadmium results in a 30% increase in power output and a significant increase in coherence length. All three HeCd laser manufacturers offer this "premium" tube option resulting in a demand for these isotopes of several kilograms per year. The "brilliance" will be short lived - (1) the manufacturers now recover and recycle the cadmium, and (2) solid state lasers will make the HeCd laser obsolete in 3-4 years.

<u>Diagnostic Breath Tests.</u> The discovery that a bacterial infection is a major cause of gastric ulcers and other peptic disease has resulted in the commercialization of the first Diagnostic Breath Test - <sup>13</sup>C-Urea. Other DBTs are in development, a few in clinical trials. Applications include drug development, disease diagnosis, therapy monitoring, drug dosing and triage.

<u>Hyperpolarized Noble Gas Imaging</u>. Hyperpolarized <sup>3</sup>He and <sup>129</sup>Xe represent the most promising MR Spectroscopy imaging applications to date. <sup>3</sup>He is already in clinical trials as a lung ventilation agent, while <sup>129</sup>Xe holds great promise for multiple perfusion and brain function applications.

<u>Isotopically Pure Semiconductors.</u> Studies conducted at several institutions on isotopically pure carbon, silicon and germanium have shown thermal conductivity improvements over their natural counterparts. This property could translate into improved heat removal capability resulting in lower peak temperatures with implications for improved manufacturing yields, enhanced reliability and higher device speeds.

### Gamma-Ray Spectroscopy of Fission Products

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Gamma-ray spectroscopy has been applied to studies of the nuclear fission process in a variety of ways. During and immediately after the Manhattan project, most fission studies involved "counting" beta decays in radiochemically isolated samples of fission products. At about the time the Phoenix Project began,  $\gamma$ -ray spectrometers became the predominant tool for refining radiochemical methods and characterizing the radioactive products of fission. The following stages in applying  $\gamma$ -ray spectroscopy to fission will be described and illustrated.

<u>Monographs on Radiochemistry of the Elements</u>. Wayne Meinke helped design the Phoenix Memorial Laboratory (PML) and chaired the NAS-NRC Subcommittee on Radiochemistry, which managed the publication of a series of monographs. The first monograph, *The Radiochemistry of Cadmium*, NAS-NS 3001, was based on Ph.D. research conducted in PML. This and subsequent monographs showed the utility of NaI(Tl) detectors in determining the purity of radioactive samples.

<u>Gamma-ray Spectrum Catalogs</u>. The spectra of typical fission products were too complex and the resolution of NaI(Tl) detectors was too poor for  $\gamma$ -ray spectra to be equivalent to a list of energies and intensities. However, the spectra could be treated as patterns, and spectra of mixtures could be interpreted as combinations of the patterns of the components. This technique benefited from standardization in the sizes of NaI(Tl) detectors and publication of catalogs of spectra, such as Russ Heath's *Scintillation Spectrometry Gamma-ray Spectrum Catalogue* (IDO 16408) in 1957. When NaI(Tl) detectors were supplanted by Ge(Li) detectors, the catalogue was updated. Now intrinsic Ge detectors are useful at higher energies (large Ge crystals) and low energies (thin dead layers), and revisions are in process. Techniques for obtaining better statistics, lower energies, and purer samples will be described.

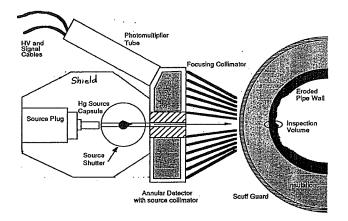
<u>Detector Arrays</u>. Large arrays of Compton-suppressed Ge detectors (e.g., Gammasphere) can resolve details of fission that cannot be sensed by any other means. Recent results for spontaneous fission of  $^{252}$ Cf will be reported.

#### An Erosion-Corrosion Monitor Using Gamma Ray Backscatter

E. H. Klevans, E.S. Kenney, R.L. Baxter, K. A. Burkert, B. Petrovic, B. Wilks Penn State University University Park, PA 16802

A new erosion-corrosion monitor (ECM) that is based on the use of gamma ray backscatter to determine metal wall thickness is discussed. This instrument facilitates inspection for flow-accelerated erosion and provides significant cost and functional advantages over ultrasonic testing for metal thicknesses up to 0.5 in. The ECM measures through the insulation, unlike UT where insulation removal and surface preparation are required. The device could be used in the power plant during operation. Wall thickness measurements up to 0.5 inches thick are accurate to within 10% uncertainty. Data is collected at points or grids and is formatted for analyses by EPRI CHECKWORKS and similar software that estimates wall erosion rates. The ECM works with empty or water filled pipes or tanks. Hole sizes of 0.5 in. diameter are readily detected.

The ECM is a 6 in. diameter cylinder, 8 in. long with two handles, and weighs 12 to 14 lbs. A schematic diagram is shown in the figure below.



The gamma ray source, less than one curie of Hg 203, is contained in a shield, and emits low energy gammas of 279 kev. Radiation fields at the surface of the ECM are 1 mr/hr or less, so no restrictive radiation precautions such as a rope-off areas are required. A gamma beam is produced when a thumb switch is triggered to activate a shutter. The backscattered radiation is focused onto the detector using a focusing collimator. The detector is a sodium iodide cylindrical crystal attached to a photomultiplier tube. A multichannel analyzer card in a portable computer, which is attached to the ECM, analyzes the signal from the backscattered gammas to produce a count rate. The actual thickness is calculated from this count rate by correlating with a calibration curve that is specific to each application. This calibration curve is developed by measuring certified plates of increasing thickness, from 0.0625 in. to 0.625 in., which are overlaid with representative insulation and jacketing. Results of field measurements at Commonwealth Edison's Dresden Units 2 and 3 for feedwater heater shells are discussed.

### Quantitative I-131 Single Photon Emission Computed Tomography

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Clinical trials of I-131 labeled radioimmunotherapy (RIT) are showing promise for the treatment of non-Hodgkin's lymphoma. Here at the University of Michigan, a phase I RIT trial of NHL using I-131 labeled anti-B1 MoAb has been completed and reports that out of 28 treated patients 79% achieved a response and 50% achieved a complete response. The success at this institution and others has renewed the interest in accurate quantitative I-131 Single Photon Emission Computed Tomography (SPECT) for dosimetric calculations. The most significant I-131 gamma-ray emission are at 364 keV (82%), 637 keV (7.2%) and 723 keV (1.8%) and collimator septal penetration by these medium and high-energy emissions has been recognized as one of the principal limitations in accurate SPECT quantification. In this work we propose the use of an ultra high-energy (UHE) parallel hole collimator for I-131 SPECT and accuracy of tumor quantification following RIT was investigated.

<u>Methods</u>: First both measurements and Monte Carlo simulations were carried out to compare the performance of the ultra-high energy collimator with a conventionally used high-energy collimator. Based on this comparison the UHE collimator was selected for the present investigation, which was carried out by simulation of spherical tumors in an elliptical phantom. Reconstruction was by a space alternating generalized EM algorithm that included scatter and attenuation correction. Keeping the tumor activity constant simulations were carried out to asses how tumor volume of interest (VOI) counts varies with background activity, radius of rotation, tumor location and size. Absolute quantification was carried out using a constant calibration factor determined from VOI counts corresponding to a sphere of known activity. Tight VOIs corresponding to the physical size of the spheres/tumors were used as in patient imaging where physical tumor sizes are available from CT-SPECT fusion.

<u>Results:</u> Use of the UHE collimator results in a large reduction in I-131 penetration, which is especially significant in RIT where background uptake can be high. With the UHE collimator typical patient images show an improvement in contrast and no effects of holepattern visualization. For a 200 cc tumor VOI counts varied with background activity, location and radius of rotation by less than 3.2%, 3% and 5.3% respectively. Due to limitations imposed by resolution the variation in VOI counts with tumor size was much more significant (up to 22%) and was a function of the background level. Good quantification accuracy (<6.5% error) was achieved when tumor size was same as the sphere size used in the calibration irrespective of the other parameters. The activity was underestimated for the 50 cc (up to 23%) and 100 cc (up to 15.2%) tumors and recovery coefficients are needed to improve accuracy.

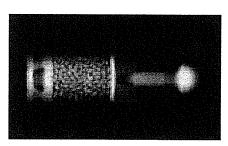
<u>Conclusion</u>: Reasonable quantification accuracy can be achieved for VOI quantification of I-131 using SPECT with an UHE collimator and a constant calibration factor. Of the factors evaluated difference in tumor size relative to the size of the calibration sphere had the biggest effect on accuracy.

#### Neutron and X-ray Microtomography

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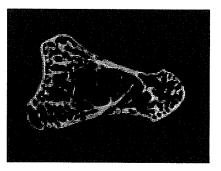
Microscopic inspection of a small specimens using penetrating radiation can reveal material properties that help define its micro-structure. When either neutrons or x-rays are used, projections images are obtained which depend on the relative transmission of primary radiation in different regions of a broad beam. The precise geometry of images obtained with parallel or orthographic projections permits reconstruction of the 3D material properties when many images are obtained at different object orientations. This can have particular value for medical, geological, biological, and other studies relative to optical or electron microscopy.

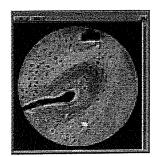
In 1969, a vertical beam port was installed at PML for neutron radiography. Collimators provided an effective source diameter of ~2cm resulting in low angular divergence of the beam (~1.5 milliradians) at the end of an evacuated flight tube (~6.5 meters length). This provided very high resolution for the examination of biological and industrial structures. The cross section of thermal neutrons compared with low energy x-rays (8 - 20 keV) produced very different image contrast.



For microscopic inspection of objects having ~1mm with a resolution of ~2 microns (1 micron pixels), images with 1 x 10<sup>6</sup> pixels are required with 10<sup>3</sup> detected radiation quanta per pixel. For tomographic reconstruction, about 720 images are needed at orientations differing by .5 degrees. A detected flux of 10<sup>9</sup> quanta/mm<sup>2</sup>/s is thus required for a total imaging time of 12 minutes. The exit flux of the PML vertical beam port of ~2 x 10<sup>4</sup> quanta/mm<sup>2</sup>/s limited the image quality of neutron microradiographs to objects of about 1cm size with 10 micron pixels (20 micron resolution) obtained with 500 second exposure times.

A sufficient flux of x-rays can be obtained from microfocus x-ray tubes to examine specimens of 1cm size in 12 minutes and reconstruct tomographs with ~20 micron resolution. Research instruments at Ford Motor Company, the University of Michigan, and Henry Ford Health System have used this method extensively over the last 20 years to study the structure of human and animal bone in relation to age and disease and in response to drug or mechanical stimuli.





Using synchrotron radiation and x-ray optic components, we have recently obtained microradiographic image of bone with submicron resolution (figure to left). With this method, a flux of  $4 \times 10^9$  quanta/mm<sup>2</sup>/s through a specimen of 0.5 mm size can be used to obtain a resolution of 1 micron or less in a few seconds. Work is now in progress to obtain microtomographs of bone with submicron resolution.

#### Advancements in Camera Technology for Nuclear Medical Imaging

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Nuclear medical imaging has been performed using large area, NaI(Tl)/photomultiplier tube, Anger cameras for over 25 years. The longevity of the Anger camera is due to its combination of relevant clinical performance and cost. Any new detector technology applied to nuclear medical imaging tasks faces a tough challenge in providing a significant improvement in clinical imaging, while remaining economical, in comparison to the NaI(Tl) detector.

A number of new candidate detector technologies are being developed as potential replacements for NaI(Tl) cameras. The advent of low noise, multichannel ASIC's is enabling pixelated detector concepts, both of semiconductor materials and scintillator/photodiode combinations. New detectors offer new performance tradeoffs that can be utilized to improve existing clinical imaging procedures and/or enable new clinical applications. Basic performance parameters include: intrinsic spatial resolution, energy resolution, sensitivity as a function of photon energy, count rate capability, and physical features such as size, weight, and modularity of the detector.

I will discuss the nuclear medical imaging system in terms of the relationship between detector performance and clinical imaging requirements. Trends in clinical imaging are changing, both in response to new clinical applications and to market demands. I will specifically discuss potential advantages and disadvantages of CZT arrays, pixelated CsI/photodiode arrays, the mini-Anger camera of CsI and drift photodiodes, and the hybrid PET/SPECT system of NaI and LSO, in light of current applications and market trends. Finally, I will speculate as to what performance goals must be achieved by new detector technology in order to supplant the NaI(TI) Anger camera.

# A Whole-body Monitor for Alpha Contamination Based on LRAD Technology – A Multidisciplinary R&D Project

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<u>Background</u>: About ten years ago, LANL pioneered long-range alpha detection (LRAD) technology. As opposed to directly detecting alpha particles which have a range of a few centimeters in air, LRAD is based on measuring the ionization produced by alpha particles (typically about 150,000 ion pairs per alpha). By introducing a controlled airflow, these ions can be detected by a simple ionization counter (two aluminum plates, a battery, and a picoammeter) at distances up to a few meters away from where the alphas produced them. Since the introduction of LRAD, this technology has been implemented in a broad spectrum of alpha detection applications ranging from counting swipes to characterization of contaminated soils.

<u>Personnel Monitoring</u>: An obvious large-scale extension of the LRAD technology is to monitoring of potentially alpha-contaminated personnel. Currently, personnel are typically monitored for alpha contamination by hand frisking of their entire body with a small ZnS scintillation probe. Due to the alpha particle range, this is a time-consuming and tedious process. Nevertheless, LANL employs about half a dozen full-time technicians to frisk personnel as they leave the plutonium-handling facility. For LRAD to compete with frisking and comply with regulations, contamination levels of 100 decays per minute (dpm) must be detectable.

<u>Results</u>: LANL constructed and tested a first-generation prototype personnel monitor. It was estimated that contamination levels of about 3,000 dpm could be detected using this prototype. Subsequently, we have collaborated with LANL to design, construct, and test a second-generation prototype. By taking the lessons learned from the first-generation prototype and incorporating a multidisciplinary approach, we have been able to reduce the detectable contamination level to about 350 dpm. The multidisciplinary approach has included involving wind tunnel, vibrations, and signal processing experts. As a result, a scaled model of the prototype was constructed to guide the design process in optimizing airflow. In addition, optimum fan speeds are being used and the sequential probability ratio test (SPRT) in being implemented to enhance the detection limit.

# Neutron-Photon Spectrum Modeling and the Composition Problem in Prompt Gamma Activation Analysis

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The Michigan Memorial Phoenix Project has a long history of expertise in reactor based Instrumental Neutron Activation Analysis, a technique especially prized for it ability to detect trace elements in very small samples. But increasingly Prompt Gamma Neutron Activation Analysis in being used to query the composition of bulk samples in areas such as in coal assay, concrete analysis, and environmental studies. Because of the large sample size in these applications, self-shielding of both the neutrons and the gammas are important, and the effects of such self-shielding are composition dependent. In principle then, there is a nonlinear relation  $\mathbf{A} = \mathbf{N}(\mathbf{w})$  between the photopeak areas  $\mathbf{A}$  and the composition  $\mathbf{w}$  (weight fractions of nuclides) of the sample. The *composition problem* is to solve this nonlinear problem for composition in terms of the measured photopeak areas. We have examined the application of nonlinear functional analysis to the composition problem.

The composition problem is over determined, and in the presence of measurement errors can fail to have a solution. We have sought to develop a robust reformulation of the problem that is well posed (both has a solution and is stable against errors in measurement and modeling). In the absence of interferences between gamma lines, and in the absence of measurement and modeling errors: 1) the composition depends only on the ratio of photopeak areas, 2) a weight fraction is not zero if gammas were detected from the corresponding nuclide, 3) the weight fractions are positive and sum to one. These three properties can be preserved in a reformulation of the problem which is guaranteed to have a solution for every set of photopeak areas. In the absence of gamma line interferences the nonlinear function N can be written as N(w) = L(w)w where L(w) is a non-singular diagonal matrix with positive elements nonlinearly dependent on w. Inverting this linear operator on A and normalizing allows us to write the composition problem as a fixed point problem w = G(w, A), where G maps normalized weight fractions to normalized weight fractions. This later property implies that there is a solution, by the Brouwer fixed point theorem.

The actual solution of the composition problem requires an iteration to solve the nonlinear equation for  $\mathbf{w}$ , during each step of which a whole coupled neutron-gamma transport computation is required, for example to evaluate  $\mathbf{G}$ . Because such computations are expensive, a rapid iteration is important. We have examined the fixed point iteration  $\mathbf{w}^{n+1} = \mathbf{G}(\mathbf{w}^n, \mathbf{A})$  which is observed to converge to the solution of the composition problem rather quickly, for any initial guess of the composition. A proof of convergence is not in hand, but computational results suggest that the iteration is based on a contraction mapping, and hence rapidly convergent under the Banach fixed point theorem. Proving that the mapping is a contraction requires showing that the Jacobian matrix of  $\mathbf{G}$  is less than one in norm, and this same condition would ensure that the solution for  $\mathbf{w}$  is stable against modeling and measurement errors. In the computational tests we have performed, this condition holds, but a general proof is not available.

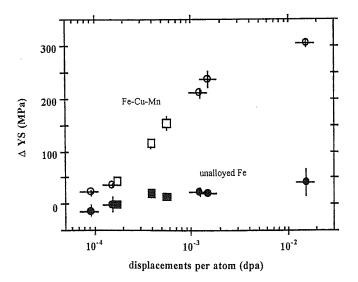
# Toward a Fundamental Understanding of Reactor Pressure Vessel Embrittlement: The Role of Radiation-Induced Primary Damage State

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A fundamental understanding of the phenomenon of radiation-induced embrittlement of nuclear reactor pressure vessels (RPVs) is key for continued safe and economic operation of commercial light-water reactors. As experimental understanding of specific embrittlement mechanisms has progressed, a "multi-scale modeling" approach to embrittlement has evolved which uses the very short time scale results of molecular dynamics (MD) simulations of radiation damage production (i.e. primary damage state) as input to longer time scale rate theory modeling.

Given the sensitivity of the modeling to this input derived from MD simulations, experimental validation of the impact of the primary damage state on embrittlement is desirable. Therefore, experiments have been undertaken to compare fast neutron and electron irradiation effects on embrittlement. Electrons produce displacement damage primarily by low energy atomic recoils, while fast neutrons produce displacements from considerably higher energy recoils. Yield strength (YS) changes induced by 10 MeV electron irradiation were compared with those induced by fast neutron irradiations in the Ford Nuclear Reactor. Identical materials, including an Fe-0.9 wt.% Cu-1.0 wt.% Mn model RPV alloy and unalloyed Fe, were irradiated at 300°C, the temperature of interest to commercial RPVs.

The figure below shows the results of these comparative experiments obtained to date. Whether irradiated with electrons (square symbols) or fast neutrons (round symbols), the figure shows that a clear enhancement of embrittlement is induced by small additions of alloying elements; consistent with previous work indicating the importance of Cu precipitation in embrittlement. Interestingly however, is the observation that electrons appear to be equally efficient (on a per dpa basis) at inducing embrittlement as fast neutrons. This is qualified however by the fact that the irradiations were performed at different damage rates. An analysis of the effect of damage rate on embrittlement efficiencies suggests that the efficiency for electrons to cause embrittlement is greater-than, or equal-to that of neutrons when extrapolated to the low damage rates of interest to commercial RPVs.



# **Applications of Nuclear Science and Technology – Part II**

Session Chair: John S. King

# **Multiscale Materials Modeling**

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As an illustration of how broad-based nuclear engineering training can prepare a person for a career in basic materials research, we reflect on the connection between neutron scattering and atomistic simulations, arriving at the present stage where modeling (theory and simulation) across several length scales holds promise of unprecedented advances in the analysis and prediction of mechanical behavior of materials. We point to specific current developments in this field to suggest the emergence of a new breed of scientists and engineers, those talented young men and women who are capable of bringing about the realization of the long-held goal of designing, synthesizing and using materials in a holistic fashion - freely crossing traditional disciplinary boundaries and exploiting without bias what theory, experiment, and simulation together can offer.

# The First Observation of Gravitationally-Induced Quantum Interference

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In most phenomena of interest in terrestrial physics, gravity and quantum mechanics do not *simultaneously* play an important role. Such an experiment, for which the outcome necessarily depends upon the gravitational constant and Plancks constant, was carried out at the FNR in 1975 by neutron interferometry<sup>1</sup>. Over the intervening years increasingly sophisticated versions of this experiment (now commonly called the COW effect) have been carried out with both neutrons and atoms. A description of this experiment now appears in most modern textbooks on quantum mechanics. In this talk I will discuss the physics of this historic experiment.

<sup>1</sup>R. Colella, A.W. Overhauser and S.A. Werner, Phys. Rev. Lett. 34, 1472 (1975).

## Neutron Powder Diffraction at Low and Medium Flux Sources

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The classic Cagliotti design of neutron powder diffractometer systems has been completely changed by the development of position sensitive detector systems and focussing monochromators. While each of these provides a substantial boost to instrument performance on its own, used together they produce astonishing gains in instrument throughput and resolution in comparison to naive (and traditional) designs. It is now possible to envision diffractometers at low and medium flux reactors that employ detector systems of more than  $1 \text{ m}^2$  in area measuring the diffraction pattern from samples of only a few tens of mg in mass in a few hours.

Linear position sensitive proportional counters provide resolution of 3 mm along the axis of the detector and when assembled into planar arrays, provide excellent area-averaged efficiency. By taking advantage of the two-dimensional nature of the detector array, it is possible to enlarge its vertical acceptance with only a small impact on the low angle diffraction peak resolution and asymmetric broadening while substantially increasing the data acquisition rate. These detector arrays can be flexibly configured for high resolution or high intensity applications.

Monochromators assembled from mechanically bent single crystal silicon slabs optimize the focussing of the neutron beam to the sample size and PSD resolution. The beam condensation, lack of soller collimator losses, high peak reflectivity and predictable performance of these monochromators make them preferable (and less expensive) than systems based on Ge wafers. The design and performance of the 14-element PUS diffractometer at the IFE-Kjeller, Norway, and the MURR PSD-II and PSD-III instruments will be described. A design study for the installation of a diffractometer at the 0.5 MW OSURR demonstrates that performance comparable to instruments at the highest flux sources can be obtained.

### A Random Walk Through Industry Research

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In the thirty years since I left the University of Michigan, the Department of Nuclear Engineering, and the Phoenix Memorial Laboratory, the world of industrial research has changed a great deal. When I joined the Ford Scientific Laboratory (as it was called then), it seemed a comfortable analog of a typical university department. General relativity and mathematical games were among the research topics of senior colleagues. This situation prevailed for a time, even as we heard of closings of other major industrial laboratories. As decreased emissions and increased fuel economy were mandated, and the technical challenges were recognized, it became clear that our lab too would change. Though it did not occur suddenly, research at Ford became much more directed toward topics relevant to the business.

In this environment, flexibility and the willingness to move fearlessly into new areas are the key to maintaining intellectual stimulation while working in areas that have the potential for significant practical impact. The years I spent in the Phoenix Lab, from tour guide to NE student, prepared me well for a life of continuous learning. In this talk, I will survey some of my former and present research interests – from phonons to electrons to x-rays to neurons to yacht race analysis. I will try to relate how my approach was formed by the environment at Phoenix and by the many wonderful people -- staff members, researchers, students, and faculty -- that I had the privilege to know.