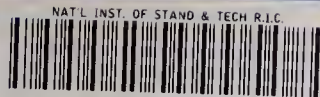


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*Practical Applications of
Nuclear Research Reactors*

Boualem Hammouda

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PREFACE

This report has been put together for a series of lectures for non-experts in the nuclear field; it is based on material compiled by this author over the last few years from different sources. Subject matters are covered at a basic descriptive level and require familiarity with introductory nuclear physics notions only (elementary particles, elements of the periodic table, etc.). The intent, here, is to introduce the reader to the various practical uses of nuclear reactors with no attempt for thoroughness.

The following scientists have been very helpful in discussing various parts of the subjects covered here: Drs. R. Brugger, G. Ehrhardt, J. Farmer, M. Glascock, and F. Ross from the University of Missouri-Columbia, Drs. G. Downing, G. Lamaze, and M. Rowe from the National Institute of Standards and Technology, Dr. W. Cunningham from the U.S. Food and Drug Administration, and Dr. Y.T. Cheng from the Smithsonian Institution.

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TABLE OF CONTENTS

Preface	iii
Acknowledgment	iv
Introduction	1
-Nuclear Reactors	2
-Nuclear Research Reactors	3
-Highlights	5
Neutron Activation Analysis	7
-The Neutron Activation Analysis Technique	7
-A Few Approximate Detection Limits	8
-Biomedical Applications	8
-Geological/Archeological Applications	10
-Forensic Science/Art History	11
-Industrial/Environmental Applications	12
-Last Word About NAA	12
Radioisotopes Applications	13
-Medical Applications	14
-Industrial Applications	16
-Agricultural/Environmental Applications	17
-Other Radioisotopes Applications	18
Co-60 Applications	19

-Food Sterilization	19
-Other Co-60 Applications	20
Neutron Interrogation	22
-Neutron Radiography	22
-Neutron Gauging	25
-Neutron Depth Profiling	26
Neutron Processing/Radiation Effects	28
-Track Etching	28
-Transmutation Doping of Silicon	29
-Coloration of Gemstones	29
-Neutron Capture Therapy	30
-Other Beneficial Applications of Radiation Effects	30
Neutron Scattering	32
-Condensed Matter Science Applications	32
-Macromolecular Morphology in Plastics	33
-Super Structures and Transitions in Microemulsions	34
-Neutron Scattering in Biology	34
-Engineering Materials/Industrial Applications	34
-Structure Determination and Porosity in Minerals, Clays and Ceramics	35
-Last Word About Neutron Scattering	35
Some Conclusions	37
References	39

INTRODUCTION

Most people associate nuclear energy with weapons programs and electrical power production. While it is true that these two applications are the most important ones when it comes to spending money (for the former) or making money (for the latter), there is a host of other very beneficial applications of nuclear energy that are not as well known. Some of these applications have passed the research stage and entered into a routine-use mode while others are still highly experimental. All of the applications covered here have something in common; they involve the use of a nuclear research reactor for neutron/gamma irradiations.

The appellation "research" reactors is taken, here, to cover nonweapon-material producing and nonelectrical-power producing reactors. Nuclear weapons (as well as nuclear waste) have given the field of nuclear energy a split (mostly bad) reputation while applications such as those described here bring great common benefits that people can easily relate to. The word "Phoenix" has been often used to name nuclear reactors in an attempt to resuscitate the nuclear field out of its own ashes by emphasizing such nonweapon applications. The "Atoms for Peace" program started by President Dwight D. Eisenhower in December 1953 had as its main purpose for the nation "to devote its entire heart and mind to find the way by which the miraculous inventiveness of man shall not be dedicated to his death (through the use of nuclear weapons) but consecrated to his life (through other uses of nuclear energy)."

It is difficult to draw a clear line between what constitutes a "practical" application (i.e., which is of direct use and therefore involves applied research/development) and more fundamental research for which the practical return is not so direct. Most of the applications included here have direct practical aspects. Some borderline applications (especially in neutron scattering) have also been included. A whole host of interesting fundamental physics research applications (such as interferometry, neutron lifetime measurements, etc.) that use nuclear research reactors have not been included here.

Nuclear Reactors:

The most common type nuclear reactor is the Pressurized Water Reactor (PWR) which uses U-235 as fissile fuel and water as both moderator and coolant. The moderator slows the 2 MeV neutrons produced by fission down to thermal energies so that they can be absorbed again in the fuel, therefore feeding a continuous chain reaction. The slowing down process is maintained through collisions with light elements (such as the hydrogen in water or deuterium in heavy water) while neutron leakage is minimized by surrounding the core with a reflector blanket (also light elements such as beryllium or graphite are used). Cooling down of the reactor core is done by circulating water (either light or heavy water) inside a primary loop which is isolated from a secondary loop through a heat exchanger.

There are other nuclear reactor concepts that have been used successfully. They are based on the use of different nuclear fuels (thermal neutron fission in U-235, fast neutron fission in U-238, Pu-239, Th-232 cycles, etc.), different moderators (solid such as graphite, liquid such as water or liquid sodium, or gases such as CO₂ or helium) and different coolants (liquids or gases). Some of these concepts are: the Boiling Water Reactor (BWR), the High Temperature Gas cooled Reactor (HTGR), the Liquid Metal Fast Breeder Reactor (LMFBR), the Pressurized Heavy Water Reactor (PHWR), etc. An interesting "inherently safe" design, still at an experimental stage, uses spherical fuel pellets coated with ceramic material that remain intact up to temperatures of 1800 °C and therefore withstand loss of coolant accidents.

Most fission neutrons appear instantaneously during a fission event, and are called "prompt" neutrons. Less than 1% of the neutrons appear after an appreciable delay time because they come from the subsequent decay of radioactive fission products. Although these "delayed" neutrons are a very small fraction of the neutron population, these are vital to the operation of nuclear reactors and to the effective control of the

nuclear chain reaction by "slowing" the transient kinetics. Put in simpler words, if these delayed neutrons did not exist, the neutron population would grow so fast following a small increase in reactivity that it would be impossible to operate a reactor safely. This is truly a gift of nature.

In a nuclear fission event, 200 MeV of energy is released mostly as kinetic energy of the fission fragments which in their turn decay to yield beta and gamma radiation. Two to three fast neutrons are emitted per fission. Neutrinos and neutron-capture nuclear reaction products are also produced in the core, but these have no bearing on the applications discussed here.

Fast neutrons and gamma rays are the most penetrating forms of radiation and therefore those that have the strongest effect on matter. Even though charged particles (α , β , β^+) are less penetrating and deposit their dose close to the surface of the material that they penetrate, these can produce substantial damage if in contact with sensitive organs.

Nuclear Research Reactors:

There are 304 nuclear research reactors in the world, 124 of which are in the United States. These vary in power, ranging from the small "piles" (less than 1 MW) used for training and some high neutron capture cross section irradiations to the advanced reactors (10 MW-100 MW) used for large-scale materials testing, neutron activation analysis, routine radioisotopes production, thermal and cold neutron beam research, transmutation doping, etc. Note that the U.S. Nuclear Regulatory Commission uses the terminology of "research reactors" (power equal to or less than 10 MW) and "test reactors" (power more than 10 MW). The main design concept of research reactors is similar but much simpler than that of power reactors. For instance, some small research reactors generate so little heat that they do not have a pressure vessel or even a closed-loop primary cooling system (the fuel elements are cooled completely

by the pool water).

In research reactors, no use is made of the power produced; in fact the (low grade) thermal heat generated to cool down the core is thrown out in most research reactors while the radiation produced (neutrons, gamma-rays) in the core is used for irradiation or scattering purposes. For that reason, research reactors have much lower operating powers than their industrial power producing counterparts whose typical powers range from 500 MW to 3000 MW. Electricity generating stations use low fuel enrichment (about 2% U-235) and have very large cores while research reactors use highly-enriched fuel (93% U-235) and have compact cores to maximize the neutron flux.

A short list of the highest power research reactors in the United States follows: HFIR-Oak Ridge (100 MW), HFBR-Brookhaven (60 MW), NBSR-National Institute of Standards and Technology (20 MW), MURR-University of Missouri (10 MW), MITR-Massachusetts Institute of Technology (5 MW), GTR-Georgia Tech University (5 MW), FNR-University of Michigan (2 MW), RIAECR-Rhode Island University (2 MW), SUNY-State University of New York (2 MW), UVAR-University of Virginia (2 MW), etc. The first three are national laboratories while the others are university based.

A clear distinction should be made between neutron scattering (which is a "non-destructive" technique) and the other research reactor applications where the irradiated samples are subject to permanent change (nuclear mutations and/or radiation damage). Neutron scattering uses thermal or cold neutrons that come down beam tubes and/or guides while the other applications involve irradiating samples in the core or reflector region. Samples are prepared for irradiation in Al cans (activated Al decays with a short half life) or put in small plastic containers to be sent to the irradiation spot inside the core or reflector region using pneumatic (or hydraulic) tubes. Some applications (such as in neutron capture therapy) require the use of epithermal/fast neutron beam tubes, while other irradiation applications (such as for prompt-gamma neutron activation analysis) use thermal beam tubes.

Highlights:

Neutron activation is an analytical technique used to detect and quantify minor and trace levels of elements for a wide variety of applications encompassing many fields of physical, natural and life sciences. It consists of irradiating a sample and measuring the gamma activity emitted from it. NAA is used for various biomedical, geological, archeological, forensic science, art history, industrial, and environmental applications. This technique has existed as long as reactors have existed.

The need to use radioisotopes has been growing ever since they were discovered. Natural radioisotopes (Po, Ra) have been known since the turn of this century while artificial radioisotopes were discovered slightly after the discovery of the neutron (1932). Artificial radioisotopes are produced from stable isotopes present in a sample when it is irradiated. The radioisotopes then decay and emit charged particles and gamma rays. The two most common ways to produce isotopes are particle accelerators and nuclear reactors. Particle accelerators produce mainly positron (β^+) emitters while nuclear reactors produce mainly β emitters. Particle accelerators inherently produce small amounts of radioisotopes compared to nuclear reactors. Practical applications in medical, industrial, agricultural, and environmental fields are covered. Co-60 applications are singled out in a chapter of their own because of their wide usage.

Three kinds of applications (neutron radiography, neutron gauging and neutron depth profiling) are sometimes referred to as "neutron interrogation." Neutron radiography is another very much unknown application (compared to x-ray radiography, for example). Yet, this technique is very useful for visualizing contrasts between various elements as well as in following real time dynamics of hydrogen-containing fluids. Neutron gauging is used in mining operations to produce indirect imaging of geological layers, in industrial bulk process streams for the grading of ores, and in commercial applications to determine the hydrogen and nitrogen contents of various food stuffs (nuts, grains, etc.). Neutron depth profiling is a recently developed technique for the characterization

of the density profile of some prompt charged particle emitters following neutron absorption (B for example) close to a sample's surface.

The radiations produced in reactors (fast neutrons, gamma rays) have strong "effects" on materials. Note that the word "damage" is sometime used in this context with a negative connotation, whereas these effects can be useful such as in track etching (a form of age determination over a range of 10 billion years), in the transmutation doping of Si to produce semiconductors, in the coloration of gemstones through the introduction of color centers, and through radiation hardening of various hard working mechanical components (drill bits for example).

The field of neutron scattering uses thermal or cold neutrons as a diagnostic probe to investigate the structure, phase transitions and dynamics of a wide variety of materials. It yields information that advances our general scientific knowledge about matter and does usually provide immediate specific help in meeting our needs. There are some very practical aspects of neutron scattering such as in the monitoring of microstresses in engineering materials. Many applications that are borderline between being of a practical or more fundamental nature are included here.

NEUTRON ACTIVATION ANALYSIS

The goal of Neutron Activation Technique (NAA) is to detect and measure the amounts of trace elements in a sample. NAA (which detects nuclear transitions) is a complementary technique to other analytical techniques such as x-ray fluorescence and atomic absorption (that detect atomic transitions). These other techniques, however, suffer from interferences that do not affect NAA, so in most cases it is much more accurate and can detect extremely low trace levels.

The Neutron Activation Analysis Technique:

NAA involves irradiating samples in the reactor core or reflector region, and measuring the gamma activity that results from the newly produced radioisotopes. Samples are enclosed in special plastic cells (called rabbits) which are transported to the irradiation spot and back using pneumatic or hydraulic tubes.

This technique depends on the accurate knowledge of the neutron flux at the irradiation spot at the time of the irradiation which is usually measured by irradiating a gold foil or any other known standard at the same time as the sample. The sensitivity of the technique is based on the availability of high neutron fluxes for irradiation and on the amount of sample used. Limits of detection depend on interferences from the various elements present in the sample and from natural background radioactivity. Many of the elements that are heavier than Oxygen ($Z > 8$) can be detected at the ppm level or lower.

The measured gamma activity depends on the incident total fluence on the sample, the activation cross section and the number density of each element, the half life of the decay and the nuclear scheme branching ratio. Each gamma transition in the activated sample yields a photopeak in the spectrum that provides the basic measurement

information. Peak energies tell what elements are being measured and peak areas tell how much of the elements are present.

A Few Approximate Detection Limits:

A few detection limits (for a flux of 1×10^{14} neutrons/cm²s, a 1-hour irradiation time and a 1-hour counting time) are included here:

-- Mn, In, Eu: 10^{-12} - 10^{-11} g

-- Cl, Ti, Zn, Pt, Hg, Th: 10^{-9} - 10^{-8} g

-- Si, S, Fe: 10^{-6} - 10^{-5} g.

Half lives vary from a few seconds (O, F, etc.) to years (Zn, Au, etc.). When the half lives are too short for the conventional NAA technique to be used, activation and prompt gamma detection are performed simultaneously. This technique is called the Prompt Gamma Neutron Activation Analysis (PGNAA) and is used for elements such as: H, B, C, N, Si, S, Cd, Gd, and Hg.

Biomedical Applications:

NAA can be used to monitor the presence of many elements (e.g., Br, Cl, Fe, K, Na, P, Rb, Zn), and their functions in living systems as well as the role that they may play in connection with disease processes. One procedure, for example, consists in drying of the specimen (this is therefore definitely a destructive technique!) before activation and counting. This technique has found its use in clinical medicine, toxicology, public health, etc. Examples of studies follow:

-- Evidence of the loss of Ca resulting from bone disease such as osteoporosis (bone degeneration makes them brittle) and from chronic renal disease.

- Knowledge of body Ca has provided data for the diagnosis and management of a variety of metabolic bone disorders.
- Observation of reduced levels of Na in patients with osteoporosis, metastatic breast cancer and Cushing's disease.
- Observation of a chlorine depletion that results from vomiting or diarrhea as well as from various metabolic disorders.
- Correlation of iodine levels with diseased thyroid.
- Determination of Cd levels in the liver and kidney to search for possible correlations between the Cd body burden and hypertension.
- Total body N measurements indicate muscle mass and protein content and are useful in studying the roles of diet and nutrition. Also N content in the soil assesses the nutritional value.
- Studies of Cu levels in biliary cirrhosis.
- Quantification of the amount of trace elements (Se-75, Cd-111m, Hg-199m, Pb-204m) have been used for nutritional studies to look for correlations with occurrences of some diseases (such as cancers).
- Assessing the importance of the role of selenium (by Se-77m) in animal reproduction.
- Identification of Al in brain tissue in relation to Alzheimer's disease.

Specimen "banking" combined with NAA is useful for assessing slowly occurring changes to the environment and their impact on various species. For instance, Oysters from the Chesapeake bay have been "banked" (i.e., conserved) for many years for present and future comparative studies.

Sometimes, stable isotopes are entered into natural biocycles and activated later in order to determine their distribution:

- Introduce Cr labeled red cells to follow blood kinetics.
- Introduce Br to study secretional pathways in nursing mothers.
- Introduce Mn to monitor the propagation of pollen in the case of labelled pine trees.

NAA has many toxicological applications:

- Monitoring of airborne contamination (occupational and environmental exposure).
- Hg poisoning (detection in hair) as for example the case of fishermen in Japan during the early 1970's.
- During the 1970's there was also a case of food poisoning in Spain (300 people died) that was traced back to the presence of chlorine in olive oil. It turned out that large amounts of seed oil were mixed with olive oil in order to increase profits.
- In the mid 1980's, a few people died in the United States after taking Tylenol capsules. Trace elements of arsenic were detected (by NAA as well as other techniques) in some remaining capsules and traced back to a batch that was tampered with.

Geological/Archeological Applications:

In geology, NAA is important for hydrogeochemical surveys, for sedimentation distribution of various elements and for oil and mineral mapping. It helps in the understanding of the origin of geological systems (on earth, in meteorites and on the moon) and to analyze minerals to gauge their values as ores. In one instance, NAA was used to identify five DNA bases (which are found in the genes of all living creatures) which were discovered in a meteorite collected in Australia in 1969. This fact raised the odds that life exists elsewhere in the universe. Also a thin Ir rich layer of clay exposed in many places around the world has been connected to a very old (65 million years ago) dust cloud that covered the earth following the impact of a huge meteorite. It is conjectured that this dust cloud was the cause of the death of the dinosaurs.

In archeology, NAA has been useful to identify the source and time origin of artifacts in order to reconstruct past history (routes of trade, civilization characteristics, etc.). Obsidian glass used by prehistoric people of Mesoamerica to manufacture tools, weapons, and decorative objects has been traced back to its volcanic origins in

Mexico. Medieval french limestone reliefs coming from various parts of the country were also examined by NAA and their origin traced to the Perigord region in the south of France. "Reading" pottery as to its provenance is another very attractive application of NAA: pottery found far from its homeland is a sign of trade, a sudden alteration of pottery style is a sign of outside invasion, etc. Mesoamerican jades that were made between 600-900 AD were traced back to the Motagna River valley (now part of Guatemala) which was Mayan territory then.

Forensic Science/Art History:

NAA is widely used for evidence gathering by law enforcement agencies. Elemental analysis (of glass, soil, fibers, paints, etc.) used to match a suspect to a place or object is now accepted in court (since 1959) as legal evidence. Traces of a gunshot can be detected on the hand of a person for up to 2 days after firing a gun and even if the subject has washed his/her hands with soap in the meantime. This is done by taking a wax imprint of the hand and using NAA to find residuals from gun powder. In this instance, it should be noted that the micellar electrokinetic capillary electrophoresis (not a reactor-based application) is being looked at as a potential alternative to NAA. It allows the separation and detection of 26 major components contained in gunpowder and explosives and can be done in under 10 minutes.

Another nondestructive application of NAA is the authentication of art pieces (paintings, objects, etc.). X-ray films are placed in contact with a painting at various times after neutron irradiation (autoradiography), so that brush strokes can be seen and underneath painting layers can be uncovered. In one instance, a hidden picture turned out to be the self portrait (the only one available) of the renown artist Van Dyck who had apparently decided to reuse the same canvas (that contained his self portrait) to paint his famous masterpiece "Saint Rosalie Interceding for the Plague-stricken of Palermo." In another instance, autoradiography showed that the American painter Thomas Wilmer Dewing performed extensive modifications to his own paintings after he adopted the surrealist style (for instance, he eliminated the room furniture around

his famous portrait of a nude making her seem to float in the air).

Industrial/Environmental Applications:

PGNAA has various industrial applications such as: real time process monitoring and control of bulk process stream, grading of ores under bulk handling conditions, etc. It is routinely used for online bulk analysis of coal for sulfur and moisture contents and weight fraction of ashes.

Environmental monitoring of air and airborne particulate is also performed routinely. NAA is highly sensitive to rare earth metals (In, Lu, La) which can be signs of industrial pollution. For example, in order to get rid of Pb in car exhausts, catalytic converters containing Pt or Pd are used. Studies assessing the content of these metals (Pt and Pd) in lung tissues being conducted are showing low amounts so far, but should be pursued for monitoring purpose. Environmental B is also important to monitor because it enters the environment quite easily and can have detrimental effects on many agricultural plants, most of which have a narrow tolerance range between deficiency and toxicity.

Last Word About NAA:

In conclusion, one can say that NAA is a very useful and sensitive technique with wide applications in many fields. It should be noted, though, that like any other technique, NAA has its own limitations; for instance, many elements cannot be detected and it cannot directly yield information about chemical forms and structures.

RADIOISOTOPES APPLICATIONS

There are about 300 stable isotopes and 1300 known radioisotopes. Some heavy elements (Th-232, U-233, U-235 etc.) and a few other light ones (K-40, V-50, Rb-87, Cd-113, In-115, etc.) are naturally radioactive while many more are made radioactive through irradiations by neutrons (in nuclear reactors) or by charged particles (using accelerators). Radioisotopes decay by emitting either charged particles (α , β , β^+) or photons (γ rays) or both. Their production depends on the capture cross section of the parent element, on the irradiation flux and on the half life of the product. Most reactor produced radioisotopes are β and/or γ emitters, but a few are β^+ emitters. A wide number of applications exist for artificial radioisotopes in medical, industrial, and agricultural uses.

A few applications make use of natural tracers such as C-14 produced by the $N-14(n,p)C-14$ reaction in the atmosphere. C-14 may be trapped in a form which ceases to exchange with atmospheric carbon (C-12). The ratio of C-14 to C-12 (determined through either counting the radioactive decay of C-14 whose half life is 5700 years or through precise mass spectroscopy) can give estimates of the age of organic materials on a scale of 10 000 years. For ages longer than 1000 years, though, this method requires corrections that are performed through a calibration using the tree ring method (the age of a particular ring in a tree trunk that is a few thousand years old is determined by counting how many rings are larger in diameter than it). A similar dating method is based on the determination of Uranium-Thorium ratios (U-234 decays to Th-230 by emitting an α particle with a 75 000 year half life).

Medical Applications:

Radioisotopes are used mainly as tracers (γ emitters) for the diagnostics of organ functions and as high dose carriers (mostly β emitters) for specific radiation therapy. So many radioisotopes (mostly β/γ emitters) have been studied and isolated to date, that it is possible to curtail their use (as to the kind, energy, and half life of the radiation emitted) to fit a specific need or application. The number of alpha emitters available for radionuclide therapy is limited (At-211, Bi-212, etc.).

A few examples of the use of radioisotopes follow:

- Co-60 is used in the radiation therapy of various forms of cancers.
- Mo-99 is a β emitter (1.2 MeV energy, 66 hour half life) with high specific activity. It is attached to monoclonal antibodies to seek and destroy targetted cancer cells.
- Tc-99 tagged agents are able to pass the blood brain barrier to evaluate brain function.
- I-131 is used for radioimmunoassay and for thyroid treatment.
- Y-90 microspheres are injected into the blood stream to treat neoplasm. They are also used in the treatment of metastatic liver cancer. In this case, the microspheres are injected into the aorta artery which leads them through the lungs into the liver where they get trapped (because of their size) and dump most of their dose.
- Sr-89, Sm-153 and I-131 are used in the treatment of metastatic bone cancer.
- Na-24 and K-42 are used in the treatment of hypertension. K-42 is also used to study cystic fibrosis (disease of the muscle fibers).
- Re-186 phosphate bone agent is used to relieve the intractable pain associated with bony metastases following breast, lung, and prostate cancer. Moreover, it can help reduce the size of such bone tumors.
- P-32 is used for imaging in human organs using a standard Anger scintillation camera. P-32 labeled glass microspheres are also used as a radiotherapeutic agent for liver malignancies.

-- Implanted β sources of Pd-103 are used for the therapy of prostate and breast cancer.

-- Re-188 labelled white cells are incorporated into heavily hydrated gels that allow them to diffuse freely and reach the bone marrow where they ablate the lymphoblast (abnormal white cells) in leukemia patients. Re-188 is also used in the treatment of rheumatoid arthritis of the knee.

Sometimes, the daughter product used in the medical application has a half life too short to be produced directly in the reactor. In this case, a radioisotope generator containing the parent element (this one must have a long half life) which is continuously decaying and therefore producing the needed short half life radioisotope. Washing down of the daughter product (called elution process) to get it pure and suitable for injection is performed just prior to use. Such radioisotope generators are sometime referred to as "cows" that can be "milked" on site. Many generators are available:

-- Cd-115/In-115m is used to label blood platelets.

-- Mo-99/Tc-99m used for brain imaging and diagnostics purposes.

-- W-118/Re-186 is used for the treatment of rheumatoid arthritis.

Besides imaging and radiotherapy treatments, radioisotopes are also useful as tracers for biological studies. They can give insight into the function and perturbations of the fine structure of biological systems (for example, to help the physician evaluate important physiological functions, assess the patient's response to a treatment, etc.). Gamma activities are measured locally after tracers are administered. A few examples follow:

-- Zn-70 and Se-74 are ingested by humans to study metabolic processes. Activity is measured in the blood, feces and other body fluids.

-- Na-24 is used to study ion transport across membranes.

-- Pt-195m is used for measuring chemotherapeutic uptake in brain tumors. It is also

used as a tracer in patients with high grade astrocytoma following either intravenous or intra-arterial drug administration.

-- Tracers such as H-3, P-32, Cr-51, Fe-55 or I-125 used along with high resolution autoradiography are valuable to study cytological structure and function. Subcellular structures can be correlated during cell cycle and cell proliferation. For example, the effect of various chemotherapeutic agents on the killing of leukemic cells (in laboratory animals) can be quantitatively measured.

-- Ir-192 used as tag for wild salamanders.

--Also, Ir-192 is used in Anger Gamma cameras for whole body counting.

It should be noted that some radioisotopes have harmful side effects (higher incidence of cancer, genetic damage to the fetus, etc.) because the ionizing radiation emitted not only destroys the cancerous cells but also healthy ones indiscriminately.

Industrial Applications:

Radioisotopes have also found many uses as tracers and imaging agents in industry. Some examples follow:

-- Ir-194 is used for industrial (on site) radiography to detect machine wear, locate leaks, examine welds in metals, etc. It has the advantage of portability and convenience.

-- Fe-55 is an excellent x-ray source (10 keV energy) with no hard γ rays. Cr-51 and Co-57 also emit x rays in the 4 keV-9 keV range.

-- Co-60 and Cs-137 are used in many applications (food sterilization, sterilization of disposable medical supplies, sewage treatment, chemical processing, etc.).

-- Ni-63 is used to power remotely located devices such as smoke detectors, etc.

-- Pu-238 provides a convenient, continuous low power supply for heart pacemakers.

-- Po-209 (stable isotope) is used in ionizing air guns to control static electricity and remove dust from product containers.

Another industrial application of radioisotopes is in gauging; β and γ emitters are used (along with radiation detectors that are properly positioned) to determine thicknesses, fluid levels, etc., as part of on-line factory operations.

Agricultural/Environmental Applications:

Radioisotopes play a major role as tracers used to follow the fate of agricultural chemicals and to understand metabolic interactions between climate, soil, air, water, plants, and animals. Moreover, they are useful in nutritional studies as well as for autoradiography.

- As-76 has been used as metabolic tracer in bacteria.
- Ca-45, Mg-28, Fe-59 have been used for plant nutritional studies.
- H-3 and C-14 have been used to follow sucrose gradients in plants.
- Beta emitters such as H-3, C-14, P-32, I-125 are used to produce autoradiographies. This technique consists in introducing tracers into the studied system (plants, etc.) which is then prepared (sectioned, dried, mounted on tape, etc.) and imbedded into a photographic emulsion. The emitted β 's produce a latent image (autoradiography) which is then related to what is known about the system (health, nutritional intake, etc.).

Radiotracers are also used for environmental monitoring and hydrological studies. For instance,

- Pt-195m, Pt-191, Pd-103, Ir-192 have been used as tracers in studies of precious metals in sea water.
- H-3, C-14, S-35 and Cl-36 have been used to follow combustion products (such as H_2O , CO_2 , SO_2 , Cl_2 , HCl) in the environment.

Other Radioisotopes Applications:

Radioisotopes are used in many other scientific (research) applications such as sources of coherent radiation (γ rays) for scattering/diffraction research. When cooled down to liquid nitrogen temperature, some of these sources (W-185, Ta-183) yield highly monochromatic γ sources (with a very sharp spectral line) suitable for Mossbauer spectroscopy which detects energy changes with very good resolutions.

Co-60 APPLICATIONS

The Co-60 radioisotope is used in so many applications that it deserves a chapter of its own. The main applications include: the sterilization of food on an industrial scale, the routine sterilization of disposable medical supplies, the treatment of sewage, the bulk processing of some chemicals, the reduction of pest infestation by suppressing their immune system, the doping of Si chips, the shrinking of plastic tubing, etc.

Co-60 is produced in nuclear reactors by neutron capture of Co-59, it decays into Ni-60 by beta emission followed by two delayed gamma rays (1.17 MeV and 1.33 MeV in cascade) with a half life of 5.26 years. Typical activities for Co-60 facilities vary from 1000 to 100 000 rads depending on their use. A rad corresponds to an energy deposition of 100 ergs/gm of materials.

Food Sterilization:

The sterilization of foods is essential for their long term preservation and storage. The spores which cause spoilage in food are sometimes unaffected by the conventional methods of preservation (drying, freezing, addition of salts or acids, etc.). Ultraviolet radiation works better, but it tends to change the color, flavor and texture of food. Food irradiation is being more and more accepted as an adequate means of sterilization. The U.S. Food and Drug Administration approved irradiation of wheat and potatoes more than 20 years ago, of spices and pork in the early 1980's and fruits and vegetables in 1986. Twenty five other countries have approved irradiations of various food stuffs.

Co-60 irradiation kills insects, bacteria and parasites that tend to hasten food spoilage. It is being considered as an alternative to the controversial pesticides, fumigants and preservatives. It can extend shelf life (barbecued chicken can sit on the

shelf for up to 8 months) and is of low cost (it costs \$0.05 per pound to irradiate food stuffs). "Radurization" (technical word for radiation treatment of foods) can be applied on the finished (even packaged) product. Astronauts, and recipients of organ transplants, rely heavily on irradiated foods for their diet. Common people, on the other hand are reluctant to try them. Opponents of food radurization argue that radiation also kills the odors that can warn about spoiled food therefore increasing the chances for food poisoning. They also claim that radiation can create cancerogens such as benzene, formaldehyde, etc., in foods. The U.S. Department of Energy is planning to build demonstration plants in six states within the next 10 years. The Japanese industry has been irradiating 10 000 tons of potatoes per month to a level of 10 000 rads to prevent sprouting and therefore increase the shelf life for several months.

Other Co-60 Applications:

When one thinks of Co-60 irradiations, the first application that comes to mind is radiation therapy for various forms of cancer treatment. Co-60 has a convenient half life and gamma energies to be used in hospitals as the principal form of radiation therapy delivering either localized (e.g., to the head) or more extended (e.g., to the spinal cord) doses.

Up to 30% of disposable medical supplies (syringes, bandages, etc.) are being sterilized by Co-60 irradiation in the United States. This method avoids the use of very toxic ethylene oxide gas for sterilization. Co-60 irradiation can reduce pest infestation by suppressing their immune system and inducing chromosomal changes. A dose of 700 rads is enough to inactivate the immune system in mice (and probably in humans too). Radiation is also used to remove endophytes from fescue feed stocks.

Co-60 radiation can reduce the pathogenic organisms in domestic sewage making it suitable for use as compost or fertilizer. Moreover, small doses of radiation permit sewage to be dewatered more readily (plants exist in Boston and Munich).

Co-60 radiation enhances the reaction rates (and yields) of some chemical processes of great industrial importance which otherwise proceed slowly at room temperature and pressure. It is cheaper and much more convenient to use radiation than elevated temperatures and/or pressures in chemical reactors. This enhancement of some chemical reactions happens because of the production of excited molecular and atomic states upon irradiation of the feedstock. The Dow Chemical Company has a Co-60 driven production facility of ethylbromide.

Co-60 can also be used for solid state polymerization of some plastics which are otherwise synthesized in solution using catalysts. This procedure has the advantage of polymerizing after crystallization and is therefore the only means to obtain single crystals of macromolecular materials.

NEUTRON INTERROGATION

Neutron interrogation consists of: neutron radiography, neutron gauging, and depth profiling.

Neutron Radiography:

Radiography of materials can be performed using neutrons similarly to the familiar x-ray radiography; the two techniques being characterized by different sensitivities to the various elements of the periodic table. X rays are sensitive to the electron cloud surrounding the atom (the x-ray scattering length increases with the atomic number) while neutrons "see" the nucleus (the neutron scattering length varies "randomly" from one element to another and even from one isotope to another). For instance, hydrogen containing materials (such as plastics and aqueous or organic fluids) are transparent to x rays while they appear opaque to neutrons because H does not scatter x rays but has a high neutron scattering cross section. In neutron radiography, a detection system is placed behind the radiographed object. This system is a neutron absorber (such as Gadolinium) called a converter plate that can emit gammas which are then recorded on a photographic film, or a converter that sends light to a scintillation detector or even a low light level TV camera for real time (dynamic) radiography. The imaging medium can also be an activable metal foil (such as indium or dysprosium) that produces a radioactive image upon neutron exposure. This image can be later transferred to an x-ray film away from any γ background; this technique is called transfer radiography.

Following are comparisons of element thicknesses needed to cut either a neutron or an x-ray beam down to 50% of its incident intensity (50% transmission) assuming 1.5Å wavelength in both cases.

Element	50% Thickness for Neutrons	50% Thickness for x rays
C	1 cm	7×10^{-2} cm
Al	7 cm	5×10^{-3} cm
Ti	1.5 cm	7×10^{-4} cm
V	1.25 cm	5×10^{-4} cm
W	0.66 cm	2×10^{-4} cm

From this simple table, it can be seen that neutrons are much more penetrating than x rays and can "see" some elements that are transparent to x rays.

The various modes of neutron radiography are: (1) static radiography (the simplest and most used); (2) dynamic radiography to observe moving parts and fluids (for example, lubrication fluids in working engines, etc.); (3) stroboscopic radiography to "freeze" in time the various phases of a periodic motion (for example one could look at only the ignition phase of the four -phase operating cycle of a combustion engine); and (4) resonance radiography which can be made sensitive to a particular atom or isotope by choosing the neutron energy to coincide with resonance absorption energies in the sample. The stroboscopic radiography mode of application involves dynamic recording (TV camera with a scintillation screen) as well as a synchronized gating of the detection process. The resonance neutron radiography method is sometimes used as a nondestructive high temperature thermometer (to measure between 1000 K and 3000 K with an accuracy of 10 K) because energy widths of neutron absorption resonances increase with temperature in a known and predictable way. The static and resonance modes do not require high fluxes while the other two

modes are possible only at medium and high flux facilities. Spatial resolution can be enhanced by the use of tight beam collimation before the sample.

Neutron radiography can be performed with most neutron sources, nuclear reactors being one of the most intense sources allowing short exposure times and good spatial resolution through the use of beam collimation devices to define the beam. Unlike in the case of neutron scattering, such collimators can be made divergent so that objects larger than the neutron source can be imaged.

A few examples of neutron radiography applications follow:

- Visualization of cracks, voids, air bubbles in synthetic materials as well as in metallic structures such as turbine blades, aircraft components, etc.
- Detection of the presence of corrosion in Al structures (corrosion product is $\text{Al}(\text{OH})_3$ and neutrons see H well).
- Visualization of flow, evaporation and condensation in metal tubes.
- Examination of operating characteristics of engines, carburetors, espresso coffee machines, hydraulics for robot systems, military aircraft components, etc.
- Special agricultural investigations (germination, plant water intake, etc.).
- Detection of U-235 which has a large thermal neutron cross section compared to U-238 making neutron radiography a highly isotope specific technique.
- Neutron radiography has also found wide use in uncovering the internal contents of bronze sculptures. For instance, Egyptian bronzes shaped in the form of birds were found to actually contain bird skeletons. Also early oriental (chinese) bronzes were found to contain an assortment of grains, nuts and fibers representing life. It should be noted that due to the religious and artistic value of such objects, these could not be temporarily opened to find out their content making radiography the only means to look into what is enclosed.

Neutron tomography consists in taking a number of (two-dimensional) neutron radiographs of the object, digitizing them on a computer that calculates a density

contour of slices through the object in order to reconstruct the full three-dimensional picture. Neutron tomography has found many applications in extending neutron radiography into a three-dimensional analog.

Neutron Gauging:

Neutron gauging has many applications in mining operations, in industrial bulk process streams and in the determination of hydrogen content in hydrocarbons and food stuffs.

Neutrons are used in mining operations to produce indirect imaging of geologic strata deep underground. The procedure consists of lowering a neutron source (Pu/Be or even a compact 14 MeV Van de Graff neutron generator) into a drilled hole (as small as 10 cm in diameter). Neutrons that are scattered back from the surrounding materials (rocks, mud, etc.) are detected along with gammas emitted following neutron capture. This technique is extensively used for oil well logging; for example, the presence of hydrogen (high incoherent scattering cross section) is a signature of the presence of oil, water or natural gas bearing rocks.

Neutrons are also used for real time monitoring and control of industrial process streams such as the grading of ores under bulk handling conditions (determined by exposing ore on a conveyor belt to neutron sources), online analysis of coal to determine the sulfur, moisture and energy contents, the online analysis of plastic filler to concrete relative amounts while the mixture is being poured (onto an airport runway for example), etc.

Hydrogen has one of the highest incoherent neutron scattering cross section making it very easily detectable, even when intimately mixed with other elements. Neutron transmission measurements are useful to determine the H content in hydrocarbons down to 1/1000 relative accuracy levels. H contents in Zr (a material used for fuel cladding in nuclear reactors) have been measured down to ppm levels. The H to C

fraction is important to know in order to assess how much H should be added in the cracking process (i.e., the breaking up of long petroleum macromolecules into smaller molecules) to produce a specific feedstock (monomer). These monomers are the raw material upon which the whole plastics industry is based. Neutron transmission measurements are sometime preferred to other methods (such as combustion analysis) used to determine the H content because it is a fast (result obtained within a few hours) and nondestructive technique to characterize new crudes. Moreover H content in nuts, grains and other foodstuff, and its presence relative to C and to N, helps determine the nutritional value of these products.

Neutron Depth Profiling:

Neutron depth profiling is useful for the characterization of the content and distribution of some neutron absorbing elements in various materials. This technique has been applied to profile the density of many elements such as: B-10, Li-6, etc. This nondestructive technique is based on exoergic nuclear reactions of the type:



Other reactions involving N-14, Na-22, Cl-36 have also been performed. The two reaction products (such as Li-7 and He-4 in the case of B-10 profiling) are emitted with clearly separate kinetic energies (2.3 MeV total kinetic energy for the B-10 reaction). These nuclei slow down proportionally to their depth within the material, so that their emerging kinetic energies define their original (i.e., birth) location underneath the surface. Typical skin depths of microns or less are probed. This technique yields the density profile of the sensitive elements even when the material consists of multilayered structures (for example, B-10 in SiO₂/Si layers).

Boron ($B-10$ occurs at a 19% natural abundance) is an important element in the semiconductor industry with uses as a dopant in devices where it is implanted. It is important to find out the B density profile in order to adjust the implantation process parameters. B is also intentionally added to dielectric phosphosilicate glass to lower its melting point in order to minimize thermally induced diffusion. Lithium density profiles have been determined in optical waveguides and in ion implanted Boron in polymer films.

NEUTRON PROCESSING/RADIATION EFFECTS

Epithermal and fast neutrons have strong effects on materials and produce nuclear reactions (transmutation) as well as radiation damage (atoms are displaced from their lattice sites) in them. Many applications make use of such local changes which have sometime repercussions on the overall material properties. Some of these applications are: track etching, the coloration of gemstones, neutron transmutation doping of Si to produce semiconductors, radiation hardening, etc. Neutron Capture Therapy has also been included since it involves damage to cancerous cells through neutron irradiations.

Track Etching:

Heavily ionizing charged particles (such as fission products) leave a damage trail when they traverse materials. This track is a permanent record that can be used for dating purposes for example. U-238 fissions spontaneously at a rate of 10^{-16} per year. Each one of these decays leaves a track. Using NAA to estimate the levels of U-238 present in a material, and counting the number of fission tracks in that same material, one can estimate the time interval since the material was first created. Accuracy of this technique is obtained over a range of 10^{10} years.

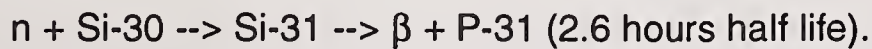
Microporosity produced by fission tracks in Uranium doped glass is essential in High Performance Liquid Chromatography. This technique measures macromolecular weight distributions by fractionation: heavier macromolecules get trapped more easily in the glass pores and therefore diffuse more slowly down the column.

Track etching is also used to produce plastic filters (Nucleopore filters) for chemical and pharmaceutical uses. A thin film of polycarbonate is passed parallel and close to

a fissioning plate to produce fission fragment tracks (holes of submicron size) in the film. The density of these holes is controlled by the irradiation time while their size is controlled by a subsequent chemical etching bath. These filters have proven to be of higher performance than conventional ones made out of cellulose fibers.

Transmutation Doping of Silicon:

Neutron irradiation of Si single crystals produces uniformly dispersed P-31 nuclei that act as electron donor impurities making the Si crystal a semiconductor:



The Si crystals are produced by a high temperature process called float zoning and have about 3% abundance in Si-30. Semiconductors using Neutron Transmutation Doped (NTD) Si have proven superior in electronic performance than those using accelerators to implant P-31 in Si crystals. The world production of NTD Si is around 50 metric tons/year.

Various uses have been found for these semiconductors: power rectifiers; particle, x-ray, and infrared detectors; avalanche diodes, etc.

Coloration of Gemstones:

Neutron radiation creates color centers which can give to some gemstones a nice "artificial" color. Topaz is the most known example whereby the stones turn from their original colorless state (when they are mined) to a nice blue color which enhances their value. This simple application has turned into a \$300 million international industry handling Topaz from its mining spots (Australia, Brazil, India, Nigeria, Sri Lanka) to its irradiation in nuclear reactors and then on to its wholesale and retail

distributions employing over 10 000 people just at the retail end alone. Irradiations to date are estimated at 3000 Kg total weight. This booming industry is driven by the high demand because of the low prices compared to naturally colored Topaz. It should be noted, though, that radiation processed Topaz loses its blue color if heated to 900 °C and has shown some chipping problems.

Neutron Capture Therapy:

Due to the high neutron absorption cross section of Boron-10, this application consists of injecting the patient with a compound highly enriched in B-10 which preferentially concentrates in the tumor. The patient is then irradiated with a beam of thermal neutrons producing energetic α particles through the reaction:



The emitted α particles have a limited range (10 μm) therefore killing selectively cancerous cells around the compound concentration. This method has been extensively tested on animals with various cancerous tumors and tried a few times on humans with bone cancer in Japan and at Brookhaven National Laboratory in New York. The technique depends primarily on the ability to find compounds that concentrate specifically at a tumor site (such as cancerous bones).

Other Beneficial Applications of Radiation Effects:

Many other practical applications of radiation damage have been identified. Some examples follow.

- By irradiating diodes with neutrons, it is possible to increase their electrical resistance under high AC frequency (100 Mhz) conditions. This happens because radiation damage increases the minority carrier lifetime.
- Fast neutron irradiation of memory units in bulk can reduce "soft" computer failure rates by a factor of 10. The radiation damage produced in the Si substrate reduces the intrinsic resistivity sufficiently to allow the rapid dissipation of any charge buildup created along the ionization path of any incident charged particle therefore lowering the probability of causing bit resets in computers.
- Fast neutron irradiation also creates lattice defects in materials that help release externally applied stresses along newly formed slip planes. This form of radiation hardening has been found to be a useful treatment to decrease wear rates in drill bits, etc.
- High irradiation doses can also introduce crosslinks in plastic materials making them tougher and more durable. Some frying pans are radiation treated.

NEUTRON SCATTERING

Most people are familiar with electron microscopy which can show the morphology of materials at a submicron scale. Neutron scattering can probe structures at a much finer (atomic) scale but scans what is called "reciprocal space," i.e., the Fourier transform of direct (configuration) space. Various applications of neutron scattering can be found in physical sciences, natural sciences, life sciences and in engineering/technology. These span many fields and topics such as condensed matter science, synthetic polymers and microemulsions, biology, engineering materials, minerals, clays and ceramics. Most of these applications are more of a fundamental research nature but have been included because of their indirect practical benefits to the engineering and technology of materials.

Condensed Matter Science Applications:

These include structure determination and dynamics characterization in metals, glasses, plastics, ceramics, liquids, biological substances, etc. Some examples follow:

- Crystallography (refining H positions to study H bonding, H positions in organometallic compounds, proteins, etc.) using mainly single crystal diffraction.
- Structure and dynamics of atomic, molecular and macromolecular liquids and their mixtures (phase separation and phase transitions, motions that contribute to the thermal diffusivity, basic understanding of microscopic interactions, etc.) using mainly wide angle and quasielastic scattering.
- Lattice dynamics investigations (phonons, spin density waves, plasmons, in crystalline systems, etc.) using mainly inelastic scattering.
- Structure/property relationship in amorphous glasses and glass mixtures using mainly wide angle scattering.
- Magnetism and superconductivity (magnetic and superconducting structures and excitations) using diffraction and inelastic scattering.

- Some surface studies using diffraction and inelastic scattering.
- Structural parameters and phase fractions in catalysts using diffraction.
- Diffusion of something in something else provided there is enough contrast and the time scale is right (10^{-6} sec- 10^{-14} sec) such as diffusion of H in metal hydrides, of ions in conductors, or self diffusion in plastic crystals using quasielastic scattering.

Macromolecular Morphology in Plastics:

Even though polymers (commonly referred to as plastics) can be categorized as condensed matter, they are usually included as a topic of their own because of wide differences in the structure, and uses of these materials. Partial deuteration (substitution of D atoms by H) is the primary asset used to create an "artificial" contrast for neutrons and therefore allows a monitoring of single chains (that appear as if they were colored differently) in the environment of normal (nondeuterated) macromolecules. Examples of studies:

- Morphology of amorphous (most of them are) plastic materials (observe conformations, their correlation to sample properties, and their changes upon various treatments such as the application of temperatures, stresses, etc.) using mainly small angle scattering.
- Assessment of the degree of crystallinity in semicrystalline polymers and its variation under various physical treatments (heat, solvent soaking, etc.) by wide angle scattering/diffraction.
- Phase separation in polymers blends (most polymers do not mix when blended) by small angle scattering.
- Crosslink density in rubbers and thermosets by intermediate angle scattering.
- Morphology of block copolymers (copolymerization is a way to force local mixing of binary polymer alloys) by small angle scattering.
- Molecular understanding of some mechanical properties and stress responses of polymer glasses (crazing, shear banding, etc.) by small angle scattering.

-- Adsorption of polymers used as coatings on cars, microsphere drug carriers, etc., by intermediate angle scattering.

Super Structures and Transitions in Microemulsions:

-- Micellar sizes and inter distances in homogeneous microemulsions by small angle scattering. Micelles are formed when oil/water and soap are mixed and are the basis for detergents.

-- Local ordering and order/disorder transitions in liquid crystals by intermediate angle scattering.

Neutron Scattering in Biology:

-- Determination of the structures of ribosomes, viruses, DNA's, tRNA's, proteins, etc., by diffraction.

-- Protein arrangements in membranes, their conformations in solution by small/intermediate angle scattering.

-- Fiber morphology in collagen by small/intermediate angle scattering.

-- Membrane bilayers ordering and their phase transitions by intermediate angle neutron scattering.

-- Dynamics of biological macromolecules by inelastic/quasielastic scattering.

Engineering Materials/Industrial Applications:

Neutron scattering is getting more popular as a "routine" characterization technique for "real life" (not specifically prepared for the experiment but taken from their normal use environment) industrial materials. The high penetration of neutrons makes them far superior to x-ray characterization. Some applications follow:

- Determination of void sizes and distributions in irradiated materials (steels, etc.) by small angle scattering.
- Measurements of texture and texture gradients in various kinds of crystalline materials by single crystal diffraction.
- Quantification of the amount of dislocation densities and their distribution by small angle scattering.
- Estimating grain sizes and microdomain morphology in metal alloys by small angle scattering.
- Measurement of residual and applied micro stresses as a function of depth, associated with welds, etc., in engineering materials by powder diffraction.

Explosive devices use lead or depleted uranium (heaviest naturally occurring materials) as the head tip in order to pierce (the armour of a tank, for example) and allow an explosive charge to penetrate. Neutron diffraction permits a detailed mapping of the microstresses right at the tip of these explosive devices.

Structure Determination and Porosity in Minerals, Clays and Ceramics:

- Size distribution and anisotropy of the petroleum bearing voids in shales by small angle scattering.
- Si/Al ordering in aluminosilicates (such as Topaz minerals) by diffraction.
- Location of water in hydrous granites, dehydration mechanism in gypsum, etc., by diffraction.

Last Word About Neutron Scattering:

Unlike most other applications of neutrons, neutron scattering is heavily dependent on modeling. Data analysis is done (1) either in a direct way by thinking up a model, calculating the scattering functions, then performing a nonlinear least squares fit to the

data to extract structural parameters or (2) by performing a Fourier transform of the data to obtain a density correlation profile in configuration space. Scattering from crystalline structures has become a sophisticated field (single crystal, powder diffraction), while scattering from liquids and amorphous systems is still at its infancy stage except for scattering from macromolecular systems where some powerful theories have helped make this field a routine characterization method to be performed along with other more conventional ones (Electron Microscopy, Nuclear Magnetic Resonance, Infrared Spectroscopy, etc.).

SOME CONCLUSIONS

Most of the practical applications of nuclear research reactors started as limited research projects. Some of them matured enough that they have become viable business ventures operated as a "service" to earn much needed funds in national laboratories and university based reactors. However, as yet, no research reactor has been built by an industrial firm for profit making only through the exploitation of such applications. Some industrial business involvement does exist through the use of university or government based reactors or through the use of small reactors that were built by industrial companies (for example General Atomics) for research purposes. For instance, most transuranic radioisotopes (such as Cf-252) are available only from government laboratories. The relative budget sizes (as of late 1980's) of the various applications in the United States are as follows:

- radioisotopes production and use: 100 million dollars
- neutron processing: 30 million dollars
- neutron interrogation: 30 million dollars
- neutron scattering: 20 million dollars
- trace elements detection: 15 million dollars.

These approximate figures are to be contrasted with the following ones for:

- electrical power production: 20 billion dollars
- nuclear defense: 10 billion dollars.

All U.S. national laboratory based and most university based reactors are government funded either at the Federal level (through DOE, DOC, NSF, NIH, etc.) or at the state level. All are suffering from very serious funding problems. National laboratories have managed to survive by limiting their programs and uptime while university based reactors are having real hard times. In fact, they are becoming a dying breed; not a

single major research reactor has been built in the United States for over 20 years, while a number of the existing ones have been shut down permanently (the Stanford University reactor has been shut down, Georgia-Tech, Berkeley and UC-Irvine reactors are contemplating shutting down). Recent upgrades (NIST) and the planned Advanced Neutron Source major reactor project (Oak Ridge) permit hope for the future.

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