LA-UR- 96-3665
CONF-9605223--6

USING SPALLATION NEUTRON SOURCES FOR DEFENSE RESEARCH

Roger Pynn, Stephen M. Sterbenz, and Daniel J. Weinacht

Los Alamos National Laboratory, Los Alamos, NM 87545

Advanced characterization techniques and accelerated simulation are the cornerstones of the Energy Department's science-based program to maintain confidence in the safety, reliability, and performance of the U.S. nuclear deterrent in an era of no nuclear testing. Neutrons and protons provided by an accelerator-based facility have an important role to play in this program, impacting several of the key stockpile stewardship and management issues identified by the Department of Defense. Many of the techniques used for defense research at a spallation source have been used for many years for the basic research community, and to a lesser extent by industrial scientists. By providing access to a broad spectrum of researchers with different backgrounds, a spallation source such as the Los Alamos Neutron Science Center is able to promote synergistic interaction between defense, basic and industrial researchers. This broadens the scientific basis of the stockpile stewardship program in the short term and will provide spin-off to industrial and basic research in the longer term.

1. Introduction

In his July 1993 announcement of the extension of the moratorium on nuclear testing, President Clinton reaffirmed the importance of maintaining confidence in the enduring U.S. nuclear stockpile. He clearly acknowledged the need to explore other ways of maintaining confidence in the safety, reliability, and performance of weapons in the stockpile. By Presidential Decision Directive and Congressional action (Public Law 103-160), the Department of Energy (DOE) was directed to "establish a stewardship program to ensure the preservation of the core intellectual and technical competencies of the U.S. in nuclear weapons".

This deceptively simple directive immediately raises a number of difficult questions. How is the country to certify the performance and reliability of its weapons if they cannot be tested? How will we be able to predict degradation of performance, reliability, or safety of weapons that result from the aging of components well beyond their design lifetime? How do we ensure the capability to rebuild or replace worn out components (including tritium)? And perhaps most difficult of all, how do we make sure that the scientists and engineers doing this work have all the knowledge and ability needed to address such complex issues when they are denied the ultimate experimental test of their ideas?

The DOE responded to these challenges by putting in place its Science-Based Stockpile Stewardship and Management program. The central tenet of this program is that confidence in the stockpile can be maintained if we have a detailed scientific and engineering understanding of the properties of weapons components and their evolution in time, and if we can simultaneously develop the tools to simulate performance accurately on the basis of our understanding.

The Stockpile Stewardship and Management (SSM) program is not an easy challenge. It involves understanding at a deep level physical problems that the likes of Fermi, Bethe and Feynman chose to design around during the Manhattan Project because they found them intractable. Clearly, to succeed on a path that such luminaries refused to tread, the weapons program will need sophisticated tools that were not available 50 years ago. In addition to powerful computers, experimental facilities are needed to probe a multitude of different

DISCLAIMER

physical properties on which a nuclear weapon depends. One such facility that provides an important subset of the required information is a spallation neutron source, such as that at the Los Alamos Neutron Science Center (LANSCE).

In this article we describe some of the attributes of LANSCE that are important to the SSM program. Perhaps more importantly, we show that an accelerator-based facility like LANSCE can bring additional benefits to the weapons program by embedding it in the broader national scientific enterprise. In this way, not only does the weapons program maintain an important touchstone for the quality of its scientific understanding, but the national scientific agenda in areas of basic and applied research are also strengthened.

2 What can LANSCE do for Stockpile Stewardship?

In February 1995 a workshop was held at Los Alamos [1] to identify the contributions that LANSCE could make to the SSM program. In addition to scientists from the three weapons laboratories (Livermore, Los Alamos, and Sandia National Laboratories), the meeting was attended by researchers who had experience using accelerator-based facilities for basic and industrial research using neutrons and protons. What emerged was an impressive list of areas in which such a facility could provide important information for the SSM program.

LANSCE Research Areas ↓	**Key Stockpile Stewardship and Management Issues**				
	Annual Certification of Performance and Reliability	Safety	Stockpile Aging	Rebuild/ Limited Production Capability	Tritium Supply
Dynamic Radiography for Hydrotests	●	●	◒	◒	
Static Neutron Radiography for Nondestructive Surveillance	◒		●		
Weapons Nuclear Data	●	◒			◆
Weapons Manufacturing Process Studies	◒	◒	◒	●	

Weapons Materials Characterization	●	◒	◒		●
High-Explosives Characterization	●	●	●		
Dynamic Materials Response	●	◒	◒		

Table 1: Projected impact of LANSCE on the Stockpile Stewardship and Management (SSM) program. A filled diamond indicates critical importance, a filled circle indicates high importance, and a half-filled circle indicates medium importance

Table 1 is an matrix of the current research areas at LANSCE and an assessment of their importance to the central questions which the SSM program is designed to answer. The following paragraphs provide more detail about some types of research performed.

2.1 Dynamic Radiography for Application to Hydrotests

Hydrodynamic testing with radiography — imaging the flow and density changes of materials subjected to very high rates of distortion — is the only tool available for studying the integral performance of a material during implosion. Hydrodynamic testing has been widely regarded as the single most important experimental effort in the nuclear weapons program apart from nuclear testing. Traditionally such imaging is performed with intense x-ray beams, in spite of the fact that most of the x-rays are scattered by the imploding object and do not contribute directly to the image. An intriguing possibility for improving these measurements is being explored by LANSCE scientists who are using protons for radiographic imaging.

At high enough particle energies, the principal interaction between protons and matter is very similar to the interaction between neutrons and matter, so one might imagine that high energy protons could be used for radiography. The problem is, of course, that protons suffer from multiple Coulomb scattering in addition to nuclear collisions so their trajectories are devitaed by passage through a solid object. However, by using a system of magnetic quadrupolar lenses, it is possible to refocus a beam that has passed through an object and image features within the object with sub-millimeter resolution provided the proton energy is high enough. Since protons also lose energy when they pass through matter, causing chromatic aberration in the lenses, practical radiography is likely to require proton energies as high as 20 GeV for application to full-scale hydrotests. Nevertheless, this new technique seems to have great promise for measuring both the density and type of material in an object. The latter information can be obtained because the degree of multiple scattering depends on atomic charge, Z. By choosing a particular aperture through which to create a radiographic image, one is therefore able to choose essentially the maximum value of Z that is imaged.

Proton radiography for hydrotests appears to offer not only the required spatial resolution and sensitivity to density variations, but also capabilities that are not available with x-rays. For example, the lower intrinsic background of proton radiography, coupled with the ease with which proton beams can be rapidly pulsed and steered, makes this technique better suited to time-sequenced tomographic recording. light and heavy materials, simply by selecting various apertures for the radiograph. Experiments using this technique have been performed using 800 MeV protons at LANSCE and 10 GeV beams at the Alternating Gradient Synchrotron (AGS) at Brookhaven National Laboratory. Figure 1 depicts results from the latter experiment.

Figure 1: Proton radiograph taken with 10 GeV protons at the AGS. The upper panel shows a schematic of the object radiographed. The lower panel is the radiograph imaged through a magnetic lens on a phosphor-scren detector.

2.2 Static Neutron Radiography for Surveillance

Neutron radiography, principally with thermal neutrons, is used for non-destructive testing in a variety of industrial and military settings. However, a recent advance at LANSCE has widened the applicability of this technique by using the fact that high energy (10 to 100's of MeV) neutrons provide a method for imaging defects in materials with low atomic number when they are hidden behind materials with high atomic numbers. This important capability for non-destructive surveillance is not available using any other technique. The physics behind this new methods rests on the observation that the scattering cross section for x-rays is proportional to Z^2 while that for neutrons is more closely proportional to the number of neutrons and protons in the nucleus. Using Computer-Aided Tomography, it is expected that structure could be reconstructed using this technique with a resolution of about 1 mm.

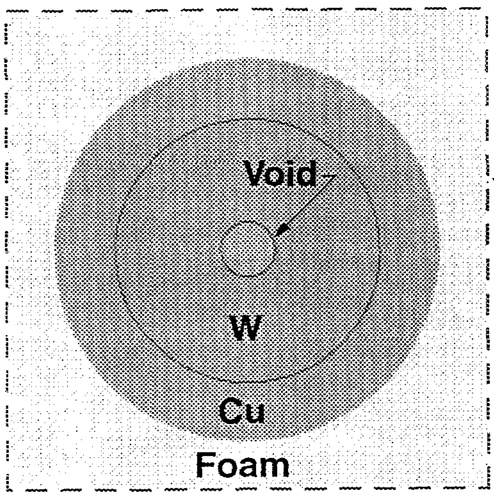
Although more conventional, cold neutron radiography also has a part to play in surveillance of nuclear weapons. This technique is excellent for imaging hydrogenous materials in the presence of polycrystalline metals. Neutrons with wavelengths greater than about 4 Å are beyond the Bragg cut-off of most crystalline materials and thus penetrate them without being strongly scattered. On the other hand, neutrons in this same wavelength range are scattered incoherently and very strongly by hydrogen, providing a large imaging contrast. Experiments underway at LANSCE are aimed at establishing the additional information that would accrue to such measurements if neutron wavelengths could be separated by time-of-flight. Such separation is provided "free" at a pulsed source such as LANSCE and may enhance radiographic images by allowing them to be obtained in a manner analogous to photography with different colored illumination of the subject.

2.3 Nuclear Data

Past nuclear tests have provided an impressive archive of diagnostic measurements that will be a cornerstone of the SSM program. As simulation codes achieve greater sophistication and incorporate deeper physical understanding and better data, they will need to be benchmarked against data from past integral tests. Prompt diagnostic and radiochemical

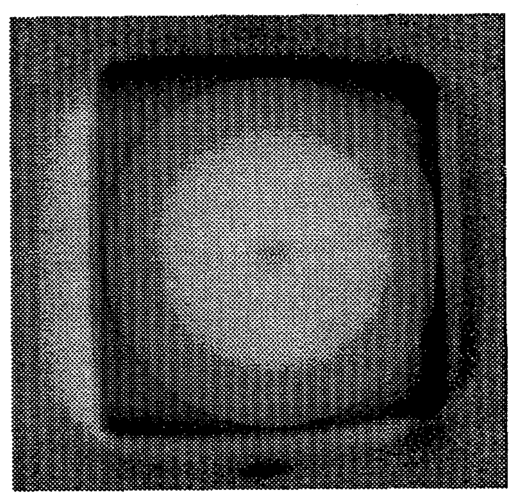
Anita Flores 5-1537

French Test Object (FTO Schematic)



Void - 1 cm
Tungsten - 4.5 cm
Copper - 6.5 cm
Foam - 22.5 cm

FTO Proton Radiograph



- Imaged through magnetic lens system on phosphor-screen detector
- Exposure less than 1/1000 of available single-pulse capability
- Particle tracking data were also taken. Presently under analysis

Figure 1

measurements from past tests provide a potential for detailed inference of nuclear performance that has not been fully exploited, in part because of the lack of basic nuclear data governing many of the underlying nuclear processes. In addition, many uncertainties remain in the nuclear data used in weapons design codes and the impact of many of these uncertainties on predictive capability has not been determined.

A primary focus of work at LANSCE is the measurement of nuclear cross sections for various neutron-induced reactions at energies relevant to a nuclear device (thermal to 30 MeV). The recent acquisition by LANSCE of a multi-million dollar, high-efficiency gamma ray detector from Lawrence Berkeley National Laboratory's nuclear physics program, has provided greatly enhanced capability in this area. One issue that is being addressed at LANSCE using this detector is the rate of ^{238}Pu production by neutron bombardment of ^{239}Pu, the fissile isotope used in nuclear weapons. Radiochemical measurements of plutonium abundance after a nuclear test have always been a critical diagnostic of nuclear yield, but data were normalized to reproduce earlier tests results. Isotopes of more than 20 elements that have been used as radiochemical detectors will be the subject of future research at LANSCE because of their importance in characterizing the neutron energy spectrum in the region of the thermonuclear fuel.

2.4 Quality Signatures for Manufacturing Processes

If mechanical components of a weapon need to be remanufactured, there will be a need for quality assurance which, because of the limited scope of the remanufacturing, will likely not be based on methods such as statistical process control. Rather, direct quality signatures will be required. One example of such signatures applies to welded or brazed joints, whose lifetime and performance is strongly affected by residual stresses induced during fabrication. These stresses can be affected by microstructural modification and phase transformations during production as well as by corrosion, thermal cycling, and exposure to radiation during storage.

Neutron scattering provides a powerful tool for assessing elastic strains deep within an engineered component. Using known constitutive relations these strains can be converted to stresses [2]. Because the technique is based on the measurement of interatmoic distances by neutron diffraction, it is non-destructive. Further, strains in different phases can be distinguished because each phase has a unique pattern of Bragg peaks in its diffraction pattern. Unlike x-rays, which do an excellent job of measuring surface strains, the neutron's ability to penetrate many materials makes it suitable for mapping strains deep within large components.

An example of work performed at LANSCE is the study of the failure of welds in beryllium, a long-standing problem. Processing conditions affect the failure of these welds and it is believed that the residual stresses in the region of the weld are sufficient to enhance cracking and failure. A step weld with an aluminum shim is one solution to this problem, but the mechanical performance of the interpenetrating beryllium and aluminum microstructure that develops around the shim is poorly understood. Since this is a composite mixture, neutron diffraction is uniquely placed to study the mean phase strains during a load test. Elastic strains were measured with material representative of the weld in a unixial compression test [3]. Strains for the two phases *perpendicular* to the applied load are shown in Figure 2. In a single-phase material one would expect a tensile strain as a result of perpendicular expansion, governed by Poisson's ratio. Indeed, this is true at low stress for both Be and Al — curves for both constituents start at the origin of figure 2 with a negative slope. At higher applied loads, however, the aluminum develops a compressive (not tensile) strain perpendicular to the load. Why is this? The answer appears to be that the very small Poisson's ratio of beryllium, coupled with its high elastic modulus constrain the aluminum and prevent it from expanding in the direction perpendicular to the load. When this essential physics is built into a simple finite element model consisting of cylinders of aluminum embedded in a beryllium matrix, the predicted strains show the same qualitative shape as those observed by neutron diffraction. However, without the data that neutrons can provide, such a simple model would have been suspect because it leaves out so many phenomena which could, in principle, affect the induced strain.

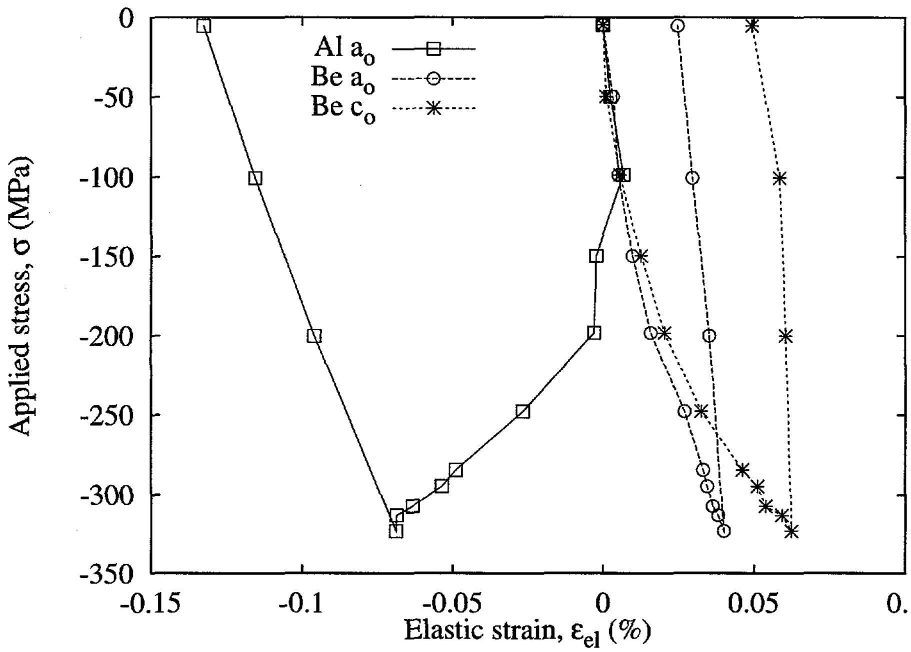
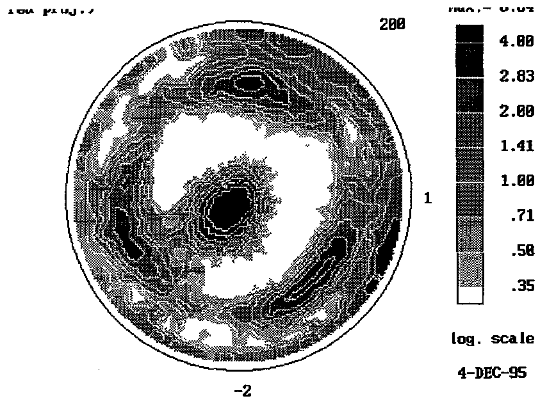


Figure 2: Elastic strain measured in a Be-Al composite in a direction perpendicular to the applied compressive stress. The measurement was on the Neutron Powder Diffractometer at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC) at Los Alamos.

Another important quality signature that is directly related to the strength and toughness of polycrystalline metals is the materials texture, which describes the distribution of the orientations of crystal grains within the material. Once again this can be measured non-destructively using neutron diffraction and expressed as a so-called pole figure, which is a stereographic representation of the orientation distribution of a single crystallographic direction.

An important step in enhancing the tools for this type of measurement has recently been made by scientists at LANSCE who have expanded the computer code that is used to analyze powder diffraction data to enable texture to be extracted as part of this analysis [4]. Thus, pole figures no longer involve the measurement of a single Bragg peak at many sample orientations, but can be obtained from all of the diffraction peaks at once, even if many of them partially overlap their neighbors. Not only does this permit much faster measurement, but it also opens up the possibility of measuring

X-Ray Diffraction



Neutron Diffraction

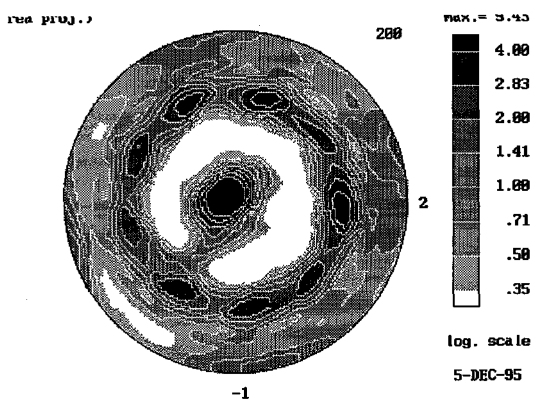


Figure 3: X-ray and neutron pole figures of upset-forged tantalum showing the difference between the surface and bulk textures.

texture easily in materials with complex crystal structures.

There are many examples of successful texture measurements at LANSCE, ranging from comparison of the textures of deep-drawn, spin-formed and cold-rolled uranium, to a study of textured titanium matrix compounds that will be used in the engine of the Advanced Tactical Fighter aircraft and will reduce its weight by over 40 pounds. In the former case, neutron scattering showed that a proposed, low-waste process for forming uranium produced similar texture to a less benign manufacturing protocol, while in the latter it demonstrated that the texture of Ti wires was retained when the wires were rolled together to form a plate. In both of these cases, retention of a particular texture during processing is key to achieving desired mechanical performance.

In a very detailed study of tantalum processed by a technique known as upset forging [5], neutron scattering was able to obtain texture information which, when combined with known elastic constants, allowed a Los Alamos computer code (known as the Los Alamos Polycrystal Plasticity code [6]) to predict the form of the anisotropic yield surface. The development of texture during processing could be followed for this high-temperature corrosion-resistant metal, and correlated with improvements in hardness. One point which emerged clearly from this study was the advantage of the neutron's ability to penetrate and probe the structure of bulk materials. Figure 3 shows x-ray and neutron pole figures for the (001) direction. The x-ray figure clearly shows only three (111) directions while the corresponding neutron results have 9 peaks of this sort, indicating that the average bulk texture is not the same as the surface texture. In fact, texture measurement at Los Alamos uses neutron, x-ray, and electron diffraction as complementary probes to sample bulk, surface and local texture in a way that gives a much more complete picture than any of these techniques could give alone.

2.5 Characterization of Nuclear Materials

Even though a detailed knowledge of the response of plutonium to high-pressure shocks is essential for predicting criticality in a weapon, there are few direct measurements of the equation of state of shocked plutonium. Another way to approach this problem is to study the interatomic potentials and to use this information to infer macroscopic properties. The technique of inelastic neutron scattering has been the method of choice for obtaining information about interatomic potentials since its first use by Brockhouse in the early 1950s [7]. Full exploitation of this method requires a sample in the form of a large single crystal, which is not yet available for plutonium in any of its many phases. Less detailed information can be obtained from polycrystalline samples by measuring either Debye-Waller factors or phonon densities of states. Measurements of the former type have been carried out at LANSCE on plutonium alloys at ambient pressure. In the future, these measurements will be extended to pressures up to 300 kbar (30 GPa) using a diamond-anvil pressure cell especially designed for research using neutron beams.

2.6 Structural Characterization of High Explosives

Damaged high explosives have been demonstrated to be much more sensitive than recently manufactured material. Damage can result from aging, exposure to extremes of temperature, or impact. Small angle neutron scattering (SANS) allows the microstructure of high explosive to be studied on length scales between a few tens of Ångstrøms and a few thousand Ångstrøms as a function of age, temperature, and impact history. The limit of the SANS intensity as the momentum transfer of the scattered neutrons tends to zero is proportional to the total internal surface area of pores and cracks, a quantity that has been found to correlate well with HE sensitivity. Like other neutron scattering techniques, SANS is a bulk, non-intrusive probe that allows measurement of internal microstructure without special surface preparation.

2.7 Dynamic Response in Reacting High Explosives

It is important to be able to predict and control the lifetime of high-explosives used in nuclear weapons. In particular, better equations of state are needed for reacting explosives to enable the accurate description of the interaction of explosive products with their confinement for a wide range of shock states. Internal temperature distributions and particle velocities have not been measurable in the past, but, thanks to a technique known as resonant neutron radiography may soon be known [8]. In these experiments, neutron resonances in dopant materials are observed during a single neutron pulse, while a high explosive is detonated. To obtain a reasonable signal in this case, a very high peak flux of neutrons is required, for which LANSCE is uniquely suited.

In one scoping experiment performed last year, the dopant was a 500 μm indium foil sandwiched between two pieces of high explosive. Clear Doppler broadening of the 9.07 eV resonance in indium was observed (cf Figure 4) as a result of the high temperature generated during the explosion. Indeed, with the high peak neutron intensity at LANSCE, temperatures can be measured with an accuracy of about 30° using this method. In another similar measurement, a tantalum foil was blown towards the neutron source by an explosion, allowing its 4.28 eV resonance to be observed twice in the time-of-flight spectrum — once immediately before the explosion and once, Doppler shifted, after the explosion (cf Figure 5). From the interval between the explosion and the second observation, the velocity of the tantalum — about 3.9 km/sec — could be deduced. Quite apart from the importance of measurements like this to the nuclear weapons program, they open up a whole new domain of fundamental shock wave physics for study.

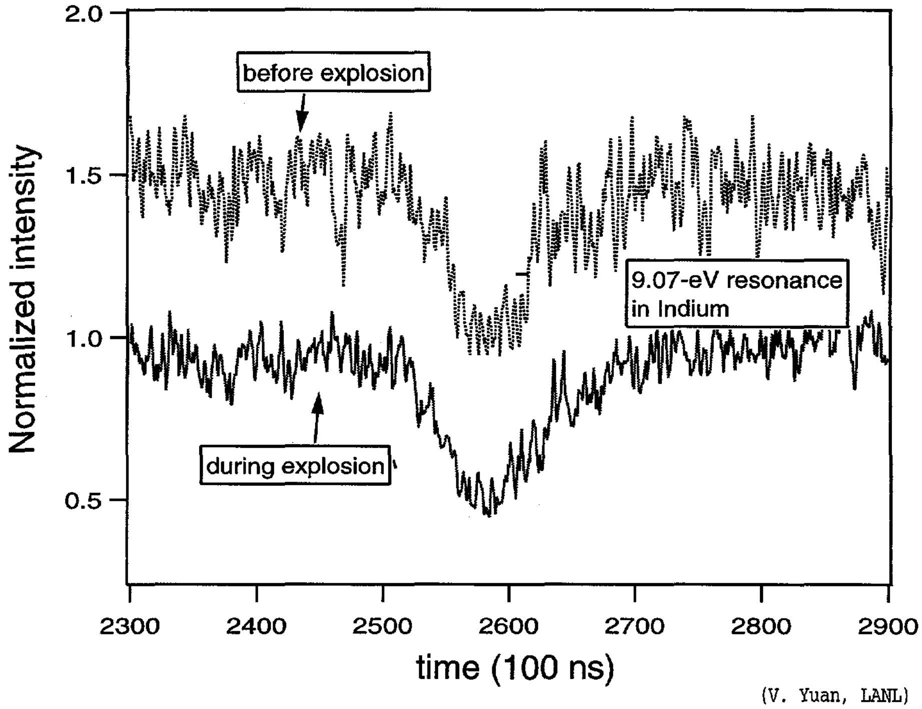


Figure 4: Neutron time-of-flight spectra obtained with a 500 μm indium foil during a single neutron pulse, before and during an explosion that compresses the foil. The broadening of the resonance recorded during the explosion is evident to the naked eye.

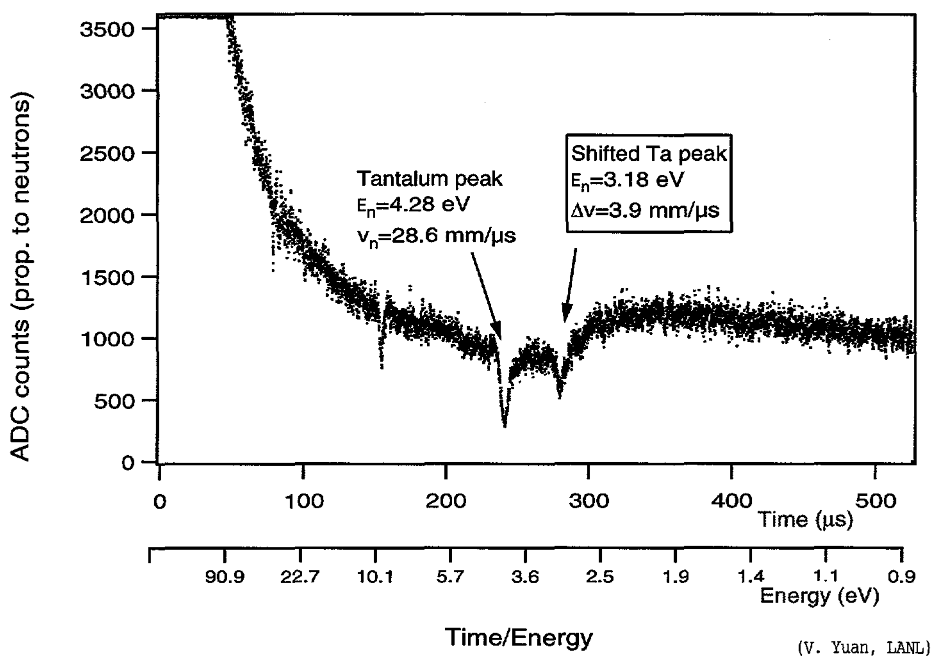


Figure 5: Neutron time-of-flight spectrum recorded during an explosion that forces a tantalum foil towards the neutron source. The 4.28 eV resonance is observed twice, allowing the velocity of the tantalum to be deduced.

3 The Synergy of Basic and Defense Research with Neutrons

Several of the experimental techniques now being employed at LANSCE for the SSM program have been used for basic research for many years. In particular, a variety of materials problems of fundamental and (to a somewhat lesser extent) industrial interest have been studied by neutron scattering. These experiments use common measurement techniques and data analysis methods and it is often difficult to determine whether the motivation for a particular experiment is scientific curiosity, product development, or the solution of a particular problem in the nuclear stockpile. The following three examples of neutron reflectometry experiments serve to illustrate this point.

Neutron reflectometry is a technique that is used to probe the atomic or magnetic density of materials close to a planar surface or interface. In this method, a well-collimated beam of neutrons is reflected from a planar surface at grazing incidence and the reflected intensity is used to infer the density variation close to the surface with a resolution of a few Ånstrøms. The technique has been used to probe buried interfaces in multilayer systems, diffusion between adjacent polymer films, lipid films on water, and a variety of other systems of interest in materials science.

The first LANSCE experiment of this type that we will describe involves the deposition of a block copolymer on a copper surface. The copolymer comprised imidiazole and amine blocks and was deposited on the smooth copper surface from two solvents — methanol, which is a good solvent for both polymer blocks, and a water / methanol mixture which is a poor solvent. In the first case, neutron reflection measurements showed that almost none of the copolymer was adsorbed on to the copper, while in the latter a thin film of copolymer was absorbed with the imidiazole block in contact with the copper.

In a second experiment, the sample was a thin film of adiprene on a molybdenum substrate. In this case, when the sample was exposed to D_2O, neutron reflection showed that the adiprene film swelled and that there was an extra layer at the adiprene / molybdenum interface. This layer was identified as water. When the molybdenum surface was silenated before the deposition of the adiprene film, exposure to D_2O did not cause this extra water layer to appear, according to the neutron reflection data.

The third experiment involved an examination of the structure of lipid (soap-like) molecules on the surface of water and the effect on this structure of attaching polymer molecules to a fraction of the lipid molecules. Because these molecules have a polar head group and an aliphatic tail, it is expected that they will form a monolayer on the water surface with the head groups in contact with water and the oily tails sticking into the air above the water (cf Figure 6). This is confirmed by neutron reflection measurements. When water soluble polymers are added to some of the lipid molecules, the structure of the surface layer depends on the fraction of lipid molecules "decorated" by polymers. Neutron reflection techniques tell us that at low polymer concentrations, the polymer conformation is more like an array of mushrooms (cf Figure 6, center panel), while at higher polymer concentrations the polymers are extended in a brush-like conformation (cf Figure 6, right panel). In addition, at high polymer concentration, the water surface becomes rough.

Neutron reflection techniques were used to study each of these systems. The data analysis methods were the same. The reader might like to guess which of these problems relates to the SSM program, which is motivated by industrial research, and which was done to satisfy the scientist's curiosity.

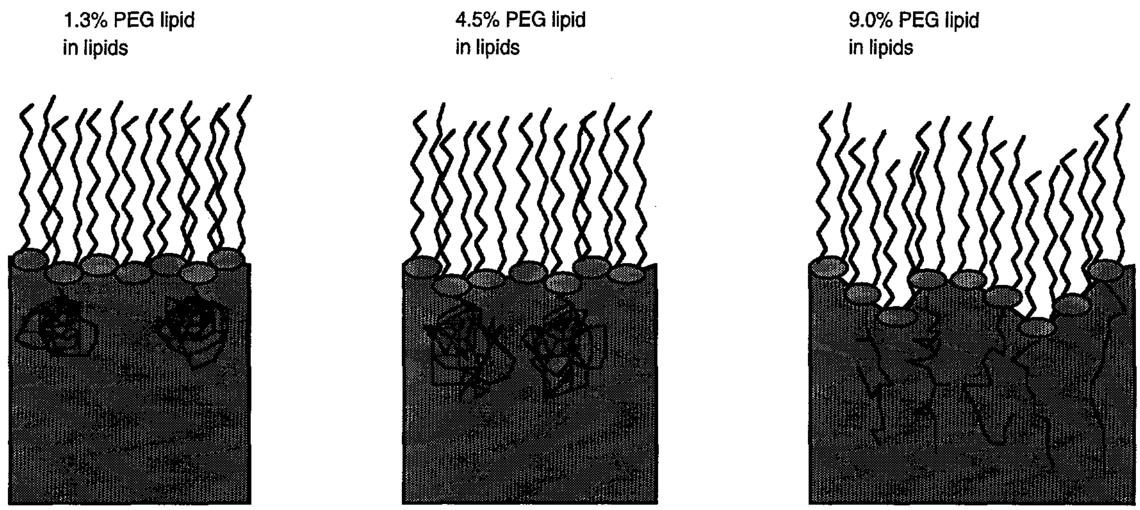


Figure 6: Scematic illustration of the structure of lipid molecules at the water / air interafce, as a function of the fraction of molecules decorated with a water-soluble polymer, polyethelyne glycol. These results were obtained by neutron reflection [9]

The answer is that the first two experiments were related to the SSM program. In the first case, the block co-polymer used was being considered as a better adhesive for circuit boards. The thinking was that since imidiazole is easily adsorbed onto copper and amines bond with epoxy, this system could form a very effective self-assembled adhesive to glue copper to epoxy in circuit boards. The experiment showed that the obvious strategy of using a good solvent for both polymer blocks would not work and that by making the solvent worse the desired outcome could be achieved. In the second case, adiprene was being used as an electrical insulator on a molybdenum part and not performing as well as expected. The guess was that water vapor was penetrating the plastic film and accumulating at the metal surface. This was confirmed by the neutron reflection experiment, which also showed that silanation of the molybdenum surface ought to be a good solution for the problem. In the third case there was both a strategic and a curiosity-driven motive for the experiment. The basic statistical mechanics of complex fluids (like polymers and lipids) at liquid / air interfaces is of fundamental interest, as is the mushroom to brush transition. However, the lipid / polymer system that was studied is also a model for a "membrane" that could contain drugs injectable into the human body and encapsulate them for long enough to reach their targets without being broken down in the blood stream.

The lesson from these three experiments is clear. The same expertise and techniques can often be applied to defense, basic and industrial research. A problem that has been resolved in one of these areas may well contain seeds of the solution of a problem in another area. In formulating a science-based stockpile stewardship program, it is important to benefit from the past experience and present research of scientists doing related basic research. Not only does this help ensure that the scientific foundation of the defense application is broadly based, but it will also provide new impetus for basic science. For example, techniques such as neutron resonance radiography or cold neutron radiography which are being developped for the nuclear weapons program, provide new imaging capabilities that are likely to find application in fields as diverse as the measurement of temperatures in operating engines and the uptake of water by plant roots.

A accelerator-based neutron source such as LANSCE is ideal for ensuring that the synergism between defense and basic research is realized because it is operated both for defense research and as a national user facility open to peer-reviewed access by university, industry and government scientists to do publishable research. Creative new ways to fund collaborative research at such a facility will lead to improved confidence in the nuclear stockpile as well as more productive basic research.

References