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Title:

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Submitted to:

International Workshop on Accelerator-Driven Transmutation Technologies

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Form No. 836 R5
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Radiation Damage Effects at Spallation Neutron Sources

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Radiation damage effects at accelerator-based spallation neutron sources correspond to a new regime, distinct from that encountered at fission reactors. We characterize the radiation environment of a spallation neutron source in some detail, and review the major issues associated with radiation damage at these sources. As a first step toward better understanding of radiation damage effects at such sources, we are trying to link together radiation transport codes with a variety of other codes such as CINDER'90 for activation calculations and the Jülich code SID for the calculation of basic radiation damage parameters. Several examples of the application of these codes to the determination of radiation effects at spallation sources are presented.

INTRODUCTION

Accelerator-driven spallation neutron sources (SNS) are becoming popular for a variety of applications ranging from cancer therapy and the production of medical radio-isotopes to accelerator production of tritium (APT), from accelerator transmutation of nuclear waste (ATW) and accelerator transmutation of plutonium (ABC) to accelerator-based neutron sources for neutron scattering studies of materials. Such projects are currently under study in the US, Europe, Japan, and the former Soviet Union. There are several spallation sources in operation at Los Alamos National Laboratory at the Los Alamos Neutron Scattering Center (LANSCE), the Los Alamos Meson Physics Facility (LAMPF), and the Weapons Neutron Research group (WNR). All these neutron sources have in common certain elements, such as an accelerator, a neutron production target, and a blanket system surrounding the target. The nature of the blanket is specific to the application considered, but it generally includes a reflector and/or moderator.

In view of the extreme conditions under which many materials used at SNS operate, it is hardly necessary to emphasize the importance of understanding and controlling the change in physical properties these materials undergo in the spallation radiation environment. These effects have usually a dramatic impact on material properties, and ultimately determine important characteristics of the SNS such as failure modes or component lifetime, among others. While these effects have received considerable attention for fission reactors, there is a lack of theoretical and experimental studies devoted to radiation damage in the radiation environment of an accelerator-driven spallation neutron source, particularly for high-power applications.

The complexity of the radiation environment at a SNS, coupled to the desire to do most of the design calculations within a coherent framework lead us to try to couple well-known radiation transport codes such as LAHET and MCNP to other codes more directly relevant to radiation effect calculations such as CINDER'90 for activation calculations and SID for gas production and microstructural damage parameters.

COMPUTER CODES

For the benefit of the reader unacquainted with LCS and SID, this section contains a very brief description of both code systems.

The LAHET Code System (LCS) is, broadly speaking, a set of analog Monte-Carlo radiation transport codes. It has two main components: LAHET and MCNP. LAHET is based on the Bertini

intranuclear cascade/evaporation model and deals with the spallation physics and the transport of most of the particles born in spallation reactions. Photons and low energy (< 20 MeV) neutrons, however, are passed to MCNP for transport. MCNP uses ENDF/B-VI continuous cross sections, and a large number of complex laws to transport photons and neutrons. Both codes have a sophisticated geometry interface that allows the modelling of arbitrarily complex geometries. The interested reader will find more details in Refs.[1,2].

The Spallation-Induced Damage code was developed in Jülich, and implements the ideas of Lindhard et al.[3] to calculate a few basic radiation damage parameters. It makes use of a history tape dumped by LCS and containing detailed transport histories to calculate quantities such as damage energy cross sections, atom displacement cross sections, recoil spectra, spallation products, gas production, etc. SID is also capable of providing the contribution of various particle types and interaction mechanism (elastic scattering, non-elastic interactions, and fission events) to the various damage parameters calculated. We refer the interested reader to the complete manual [4] for more information.

Many essential features in these codes have been extensively benchmarked by comparison with other codes or with experimental results, see Refs.[4,5].

RADIATION ENVIRONMENT OF A SNS

Several important features distinguish the radiation environment of a spallation neutron source from that of a reactor, namely:

- The presence of high-energy (hundreds of MeV) charged particles from the accelerator (typically protons);
- The neutron spectrum resulting from spallation reactions is harder than a typical fission reactor spectrum;
- The spallation reactions induced by high-energy particles produce a large number of impurities in the form of spallation, fission, and transmutation products that alter the physical and chemical properties of the materials.

These considerations underscore the importance of assessing radiation damage to the various components of high-power accelerator facilities for accelerator-driven transmutation technologies. Although many components of a SNS are subjected to radiation in various degrees, the target and its associated components (containment vessel, window, etc) are more likely to be affected by the conditions outlined above than other components. This is especially true in view of the fact that in many SNS applications, the neutrons are often moderated after leaving that target, thereby softening the energy spectrum to a more familiar regime similar to that encountered at fission reactors. Thus we will focus essentially on the neutron production target in what follows.

In order to illustrate these points, we calculated (with LCS) the neutron energy spectrum for a 1 GeV proton beam on a stopping-length solid tungsten target, and compared it to a typical fission reactor spectrum. Fig.1 clearly show that the spallation neutrons produced when an 800 MeV proton beam hits a tungsten target have, on average, a larger kinetic energy than fission neutrons.

There are several broad classes of radiation damage parameters of interest to metallurgists and material scientists: (a) The production of hydrogen and helium isotopes via (n, α) reactions.

Figure 1: Probability density of observing a nucleon with energy E in a W target bombarded by 800 MeV protons. The corresponding quantity for a typical LWR is shown for comparison. The curves are normalized to unity.

However, notice the possibility of (p, α) or (d, α) reactions at SNS. (b) Atomic displacements leading to the formation of microstructural defects, and corresponding changes in the mechanical properties of the metal or alloy. (c) The formation of chemical impurities (spallation, fission, and transmutation products).

The next two sections are devoted to various aspects of these problems in relation to the SNS radiation environment. It should be emphasized that, although essential, these parameters are not enough to determine radiation *effects*. Indeed, the kinetics of the defects formed during irradiation must be taken into account to calculate the effect of irradiation on the physical properties of metals. There are also other basic radiation damage mechanisms and effects worth mentioning. For instance, many components have to be actively cooled during operation. The coolant of choice -light or heavy water- undergoes decomposition in the radiation field with the formation of highly reactive atomic or molecular species that contribute to accelerated metal corrosion. Another example is the effect of irradiation on the kinetics of various transport processes (e.g., displacement mixing in alloys, segregation, enhanced-diffusivity, etc). These effects are out of the scope of the present work.

GAS PRODUCTION

Hydrogen and its isotopes are soluble, to various degrees, in many metals. Occasionally, hydrogen can form stoichiometric compounds with metal leading to drastic chemical and physical changes. Zircaloy is an example of a material that is important for nuclear technology where hydrogen effects must be controlled. The effects of hydrogen in metals have been documented extensively in the nuclear technology literature and will not be considered here.

Figure 2: ^3He and ^4He production cross sections for 660 MeV protons on a thin ^{58}Ni target.

Perhaps more important than hydrogen formation is helium production. Because of their large enthalpy of solution, helium atoms are largely insoluble in most, if not all, metals. They tend to migrate, congregate at defects, and form large bubbles that ultimately embrittle the irradiated metal, particularly at high temperature. Helium embrittlement of metals has been extensively studied. A review can be found in Ref.[6].

We will focus on the calculation of hydrogen and helium production rates, particularly at SNS, rather than the consequences of hydrogen and helium formation in irradiated metals. In principle, codes such as LCS are capable of calculating gas production rates *ab initio*. In order to evaluate the accuracy and reliability of the gas production rates calculated by LCS, we compared the results of detailed calculations with a variety of nuclear models to experimental results. Fig.2 is a representative example of our calculations. Generally speaking, nucleon production can be predicted rather accurately [5] (and is not very sensitive to the physics model used). The prediction of production rates for light composite particles, however, is much more sensitive to the particular nucleus model used. Furthermore, although the order of magnitude of the production cross section is, most of the time, within a factor of a few of the measured cross section, certain qualitative features are not reproduced by the calculation. Fig.2 is an example where reasonably good agreement was obtained between the experimental results and the LAHET calculation. Other isotopes did not fare nearly as well. In calculating the results shown in Fig.1, we used all the default physics parameters in LAHET, but varied systematically the nuclear energy level density parametrization. We also calculated gas production cross sections for other isotopes and elements (^{120}Sn , ^{209}Bi , U, ^{27}Al , ^{54}Fe , ^{58}Ni , ^{12}C , ^{197}Au). There appears to be no single combination of model options in LAHET that leads systematically to better agreement with experimental measurements. Worse yet, for a given

isotope, different sets of options lead to better agreement depending on the gas being produced. These results will be described in detail elsewhere.

System (target)	Proton beam	Power density MW/l	^1H appm/d (at/p)	^2H appm/d (at/p)	^3H appm/d (at/p)	^3He appm/d (at/p)	^4He appm/d (at/p)
ATW-aq. (W)	$E_p=1\text{ GeV}$ $I_p=0.2\text{ A}$ $W=200\text{ MW}$	~ 2	60 (2.7)	9 (0.4)	4 (0.2)	0.6 (0.03)	12 (0.5)
APT (W)	$E_p=1\text{ GeV}$ $I_p=0.2\text{ A}$ $W=200\text{ MW}$	~ 2	58 (2.7)	9 (0.4)	4 (0.2)	0.6 (0.03)	12 (0.5)
LANSCE-II (W)	$E_p=800\text{ MeV}$ $I_p=1.25\text{ mA}$ $W=1\text{ MW}$	~ 0.6	14 (3.0)	1 (0.3)	0.6 (0.1)	0.06 (0.01)	1.6 (0.3)
ISIS (Ta)	$E_p=800\text{ MeV}$ $I_p=0.2\text{ mA}$ $W=0.16\text{ MW}$	~ 0.07	2 (2.7)	0.2 (0.3)	0.1 (0.1)	0.006 (0.007)	0.3 (0.3)
LANSCE (W)	$E_p=800\text{ MeV}$ $I_p=0.08\text{ mA}$ $W=64\text{ kW}$	~ 0.04	1 (2.7)	0.1 (0.2)	0.04 (0.1)	0.003 (0.007)	0.1 (0.3)
FBR core	$W=2500\text{ MW}_t$					0.03	
Fusion reactor (first wall)	1 MW/m^2		1.5			0.3	
HFIR	$W=100\text{ MW}$		1.2			0.07	

Table 1: Gas production rates at various SNS. The numbers between parentheses are gas production per incident proton. For comparison, we give helium production rates at a typical fast breeder reactor and in the first wall of a typical fusion reactor. The power density is an average for the neutron production target.

In all fairness, we ought to emphasize that the experimental cross sections are measured at a fixed angle in the laboratory frame of reference, and thus part of the discrepancy observed may be due to the inability to calculate accurately the angular dependence of the cross section. Indeed, comparison with angle-integrated cross sections reveals, generally, better agreement [7].

We used LCS to study gas production in the neutron production target of a variety of SNS (listed in Table 1). SID was used to extract gas production rates and cross sections. The SNS listed in

Table 1 operate at a variety of power levels, but they all have in common a water-cooled tungsten (or tantalum in the case of ISIS) target. For comparison, we include in Table 1 typical numbers (when applicable) for other nuclear systems. It can be seen immediately that gas production at SNS is significantly higher than that at other nuclear facilities, especially at higher accelerator power. Thus swelling and embrittlement problems can be expected to be much more severe at SNS.

RADIATION-INDUCED DEFECTS

SID uses the well-known Lindhard formalism [3,8] to calculate a number of useful radiation parameters such as damage energy and damage energy cross sections, dpa's and dpa cross sections, recoil spectra, etc. It has important limitations that are not easily overcome. Going beyond these limitations requires a drastically different approach such as that adopted by MARLOWE or molecular dynamics codes. Nonetheless, SID can provide useful insight into the severity of the radiation damage problem under consideration.

System	dpa/yr
APT	24
ATW-aqueous	65
LANSCE-II	16
LANSCE	1
HFIR (SS-316)	41
Fusion reactor (first wall)	13

Table 2: Displacements per atom (dpa) in the W target of the SNS listed in Table 1. Typical dpa for other nuclear system have been included for comparison (see Ref.[10]).

Basic radiation damage parameters for medium- to high-energy proton irradiation of thin W targets have been studied extensively [9]. Coupling SID to LCS enables us to use the actual geometry of the system under study to calculate the radiation environment which is responsible for damage production. Table 2 shows the results of LCS/SID calculations for the W target of most of the SNS listed in Table 1. Typical numbers for other facilities have been included for comparison. The numbers quoted in Table 2 for SNS correspond to damage produced by the high-energy part of the radiation field, i.e., it includes everything except the effect of low-energy (< 20 MeV) neutrons.

CONCLUSION

The use of SID in conjunction with LAHET produces useful information for scoping, radiation damage calculations. Ultimately, more refined calculations (and experimental checks) are needed if one is to assess carefully the impact of radiation damage on component lifetime and failure modes. LCS and SID are useful to direct one's attention to potential problems for further study.

As illustrated by the gas production benchmark calculations more benchmarking and code validation is necessary to assess the reliability of primary yields of light composite particles. Clearly, improvements are needed. This implies an extensive reworking of the physics models

used. In all fairness, it should be mentioned that, to this day, the calculation of light composite particle yields remains a challenge to theoretical nuclear physicists.

We have shown that gas production rates are much higher at SNS than at fission reactors, and thus that the familiar embrittlement and swelling problems are likely to be felt more acutely at SNS, especially at higher power levels.

Similarly, dpa's are high at SNS, and it is likely that materials considerations will be an essential part of SNS design, particularly at higher accelerator power. Finally, it is worth emphasizing the importance of calculating damage parameters in the actual radiation environment of a SNS, with the appropriate geometry. It is thus highly desirable that LCS or similar codes be coupled directly to radiation effects computer codes.

ACKNOWLEDGEMENTS

This work was supported by the US Department of Energy. We thank R.E. Prael, L.S. Waters, and R. Kidmann for useful discussions.

REFERENCES

- [1] R.E. Prael and H. Lichtenstein, "User guide to LCS: The LAHET Code System", Los Alamos National Laboratory report LA-UR-89-3014 (September 1989); R.E. Prael, "A review of physics models in the LAHET code", submitted for Specialists' Meeting on Intermediate Energy Nuclear Data, Nuclear Energy Agency, OECD, Paris, May 30 - June 1, 1994, LA-UR-94-1796, Los Alamos National Laboratory (May 1994)
- [2] "MCNP - A general Monte-Carlo code N-particle transport code - Version 4A", Judith F. Briesmeister, editor, Los Alamos National Laboratory report LA-12625, (1993).
- [3] M.T. Robinson and I.M. Torrens, Phys.Rev.**B9**, 5008, (1974) and references therein.
- [4] T.W. Armstrong, P. Cloth, D. Filges, "SID - User's manual", Jülich internal report.
- [5] T.O. Brun, L.L. Daemen, E.J. Pitcher, G.J. Russell, C.A. Beard, and W.B. Wilson, "LAHET Code System/ Cinder'90 validation calculations and comparison with experimental data", Proceedings of the Twelfth Meeting of the International Collaboration on Advanced Neutron Sources, Rutherford-Appleton Laboratory (May 1993) RAL-94-025, p.T-26.
- [6] H. Ullmaier, in "Atomic defects in metals", Landolt-Börnstein (Springer-Verlag, Berlin, 1991), Chapter 3. See also, H. Ullmaier and H. Trinkaus, "Helium in metals: Effect on mechanical properties", in Materials Science Forum, Vol.97-99, G.Szenes Ed., (Trans Tech Publications, Zürich, Switzerland), pp.451-472.
- [7] S.L. Green et al., "Production of helium by medium energy protons.", J.Nucl.Mater., **155-157**, 1350 (1988).
- [8] M.T. Robinson, "The theory of radiation-induced defect production", in 'Radiation Damage in Metals', Papers presented at a seminar of the American Society of Metals, Nov. 9 and 10, 1975, N.L. Peterson, Ed., (American Society for Metals, Metal Park, 1975), p.1.
- [9] M.S. Wechsler, private communication.
- [10] T.A. Gabriel, J.D. Amburgey, N.M. Greene, "Radiation damage calculations: primary recoil spectra, displacement rates, and gas production rates", ORNL/TM-5160 (1976).

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