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ABSTRACT

Materials samples were irradiated in the Los Alamos Radiation Effects Facility (LASREF) at the Los Alamos Neutron Science Center (LANSCE) to provide data for the Accelerator Production of Tritium (APT) project on the changes in mechanical and physical properties of materials in a spallation target environment. The targets were configured to expose samples to a variety of radiation environments including high-energy protons, mixed protons and neutrons, and predominantly neutrons. The irradiation was driven by an 800 MeV 1 mA proton beam with a circular Gaussian shape of approximately $2\sigma = 3.5$ cm. Two irradiation campaigns were conducted in which samples were exposed for approximately six months and two months, respectively. At the end of this period, the samples were extracted and tested. Activation foils that had been placed in proximity to the materials samples were used to quantify the fluences in various locations. The STAYSL2 code was used to estimate the fluences by combining the activation foil data with calculated data from the LAHET Code System (LCS) and MCNPX. The exposure for each sample was determined from the estimated fluences using interpolation based on a mathematical fitting to the fluence results. The final results included displacement damage (dpa) and gas (H, He) production for each sample from the irradiation. Based on the activation foil analysis, samples from several locations in both irradiation campaigns were characterized. The radiation damage to each sample was highly dependent upon location and varied from 0.023 to 13 dpa and was accompanied by high levels of H and He production.

I. INTRODUCTION

To support the APT project, the LANSCE accelerator was used to perform materials irradiations aimed at determining the changes in mechanical properties in high-energy proton and spallation neutron environments *[1]*. The details of the LANSCE irradiation and determination of the fluences from activation foils for some targets have been published previously *[2,3]*. The current work follows this methodology for fluence analysis and applies it to samples located some distance away from the primary proton beam as well as to samples irradiated in-beam from a separate, two-month irradiation campaign. Results were obtained for groups of neutron furnaces that were in two locations in the irradiation and also for tubes that were located a W target insert. Both of these areas saw primarily a neutron flux. Also analyzed were the exposure conditions from an in-beam insert ("new" 17A) irradiated from Oct. to Dec. 1998. Use was made of the both the LAHET Code System *[4]* and MCNPX *[5]* transport codes both for modeling of the target region and to provide data on the dpa and H/He gas production cross sections. Determination of the fluences and appropriate exposure parameters are performed using activation foils and these results are then interpolated to the samples in the irradiation. That information is used to interpret the impact of radiation on the properties of the candidate materials.

II. METHODOLOGY

A. FOIL COUNTING

At the conclusion of the irradiation campaign, the activation foils were extracted from the target containers. Specific foils were selected for analysis, sent to the CST-11 group at LANL, and gamma counted using high-purity germanium detectors. The major isotopes were identified and their activities were extrapolated back to the zerotime (shut-off of the accelerator). Using the beam history of the irradiation, the average production rate of the major isotopes was calculated.

Initial results from the foil counting provided input on the precise location of the targets in Area A with respect to the beam, shown in Fig. 1. Activation foil data from the 1998 irradiation of the 17A target revealed the samples had offsets specific to particular tubes, a trend clearly visible from the location of the Gaussian curves from each tube, shown in Fig. 2. In this figure, the location of the peaks in the Na22 activity indicate discrepancies in the proton beam between the four tubes. The data from the Al-Na22 reaction was confirmed by the Nb-Se75 data, another reaction sensitive to the high-energy protons. Offsets in the individual tubes that were consistent with the foil data produced relative positions shown in Fig. 3. The offset information was used to revise the as-irradiated transport code models. A similar methodology was applied to the tubes in the 18A insert. Although there were no in-beam activation foils, the beam extent was still sufficient to verify a small offset in the insert location relative to the proton beam that had been initially seen with autoradiography. In this case, the insert (18A) was approximately 1.3 cm offset and the transport code models were adjusted accordingly.

The transport codes (MCNPX 2.1.5 and LCS) were then used to establish spectra for the protons and neutrons in the locations of interest, the starting point for the foil analysis. Using the production rate (saturation activity) from the isotopes and the estimated fluences from the transport codes, the STAYSL2 package provided estimates for the neutron and proton spectra and fluences.

B. CROSS SECTIONS

Proper characterization of the exposure conditions required computing the dpa, H and He production for each material. Cross sections for the APT candidate materials for these parameters were computed using the LAHET Code System (LCS) *[6]*, version 2.83. The Bertini intranuclear cascade model was used with pre-equilibrium turned on in conjunction with the Gilbert-Cameron-Cook-Ignatyuk (GCCI) level density model *[7,8]*. These physics options, employed in LCS as the standard APT settings, were chosen primarily to optimize the correct n/p ratios for high-Z targets such as tungsten. The accuracy in He production for mid-Z (i.e. Fe, Ni, Cu) elements is known to be poor *[8]* for these parameters. In most cases, the measured helium levels in the Fe and Ni-based alloys are considerably higher (typically about factor of 2) than calculated. The values measured *[9]* are in agreement with that seen in earlier high-energy proton irradiations in LAMPF *[10]*. He gas measurements will be applied to adjust he cross sections and establish more accurate He production estimates in future reports.

Estimates of the H production are also subject to uncertainties that in this case depend on the long recoil lengths of the secondary particles produced in spallation. Up to 50% of the secondary protons (H) are born with sufficient energy to escape a typical sample. Characteristic dimensions of the samples are usually from 0.1 to 2 mm. The values reported are also strictly production cross sections minus recoils; no attempt was made to estimate the diffusional losses.

From the results of the activation foil analysis, the proton and neutron spectra and the resultant exposure parameters for the major materials (Alloy 718, 316L and 304L SS, modified 9Cr-1Mo and Al6061) were computed at each foil stack location. It was then necessary to apply this knowledge to the sample locations.

C. INTERPOLATION TO SAMPLES

To obtain the final results for the proton fluence, neutron fluence, dpa, He and H production, the data were interpolated to the sample positions by using a mathematical fitting process. The process is congruent with the recommendations in ASTM E 798 "Standard Practice for Conducting Irradiations at Accelerator-Based Neutron Sources." Foils were grouped together by tubes and a two-dimensional fit was made of the data. For example, tubes 1, 2 and 3 were assumed to be co-planar and the foil locations mapped onto a plane. Two-dimensional Gaussians were then fit to the final exposure data and interpolated to the samples using the best-fit Gaussian. This was done for all exposure parameters, including the neutron fluence, which was divided into four energy groups. This method was applied to the tubes in 18A, 18C and to groups of foils from the neutron furnaces on the 18B and 9B inserts.

III. RESULTS

Multiple locations from the Area A irradiations were characterized with the previously described methodology, examples of which are presented here. For example, the setup of the 18A insert consisted of tubes containing Al mechanical test samples adjacent to tubes containing W rods. The W rods occupied the center portion where the proton beam was located. The sample tubes were above and below the W rods and, as such, saw primarily a neutron flux from the secondary particles. Figure 4 shows a map of the H production in Al6061 from tubes 8 and 10. This map is drawn from data in out-of-beam locations and is only valid adjacent to those points. Given the lack of data from the center (in-beam) location, it should not be considered reliable to extrapolate to these regions.

The proton fluence map for the neutron furnaces on the 18B insert, which straddled the proton beam is shown in Fig 5. In this figure, the outlines of the furnaces are shown and the activation foil locations are marked with dots. The total proton fluences for samples located in the furnaces varied from $2x10^{18}$ to $1x10^{19}$ p/cm^2.

Neutron furnaces located further downstream of the irradiation, were also modeled, as shown in Fig 6. These furnaces were positioned parallel to the path of the proton beam and were exposed to primarily secondary particles from spallation events upstream of their location. The He generation in Al6061 samples was computed in these furnaces and mapped. The dots show the locations of the foil stacks that were distributed among five furnaces.

The "new" 17A insert from the 1998 irradiation was very similar to the previous insert used in that position, although the materials contained within it differed and the total time of exposure was much lower. The dpa distribution for tubes 1, 2 and 3 are given in Fig. 7. The distribution of the dpa can be modeled as a 2-D Gaussian and follows the proton beam dimensions. The maximum dpa for 316L SS samples in this target was around 3.5 dpa.

IV. DISCUSSION AND CONCLUSIONS

The results shown here summarize the methods used to evaluate the irradiation exposure for the LANSCE irradiation campaigns and how different areas of the irradiation were handled. Determination of the neutron spectra and fluence was of major importance with both the in-beam and out-of-beam samples in calculating the final exposure parameters.

Analysis of foil activities illustrates the importance of verifying as-built and as-irradiated conditions. Adjustment of tube locations to conform to the irradiated condition was made possible by careful examination of Al-Na22 and Nb-Se75 activities.

With the precise locations relative to the proton beam, the fluences for the 18B, 9B and "new" 17A targets in LANSCE Area A were calculated by the combination of transport codes, activation foils and mathematical interpolation.

Cross sections were used to estimate the dpa, H and He production in the materials. The gas production values are subject to some uncertainty. In the case of the He, systematic errors are present in the LAHET physics model. For the case of H, the recoil escape fraction is unknown and in some cases is dependent on the target geometry configuration.

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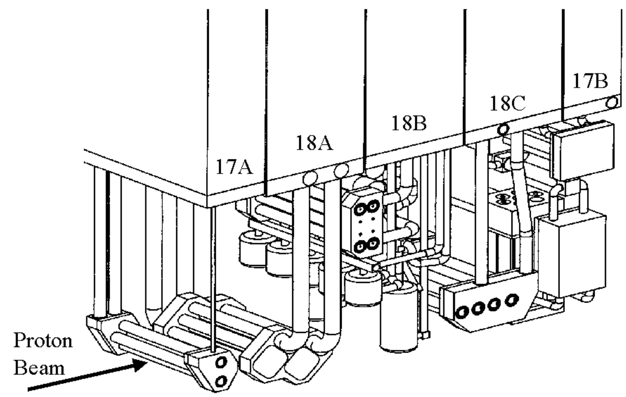

Figure 1. Graphic of the 1996-1997 target configuration in LANSCE Area A-6

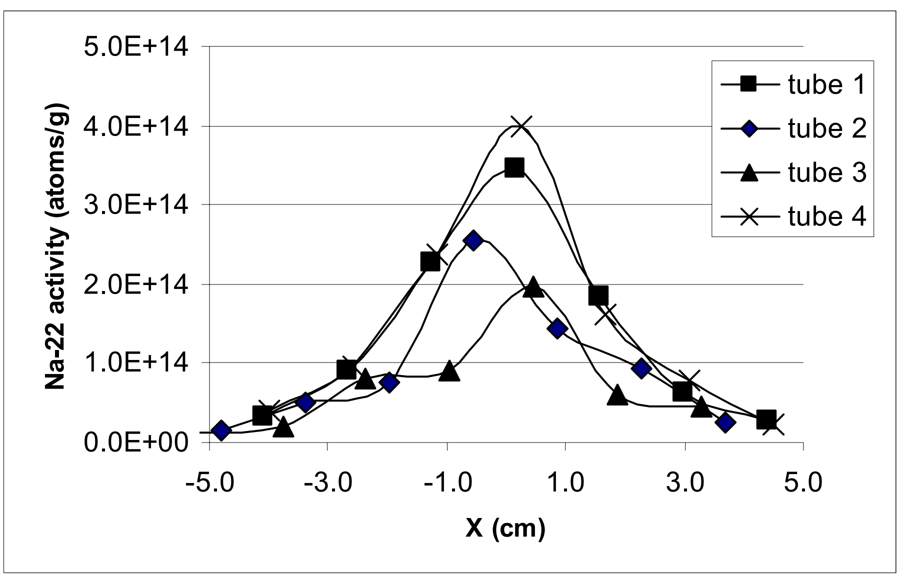

Figure 2. Curves for Al-Na22 activity in foils from "new" 17A.

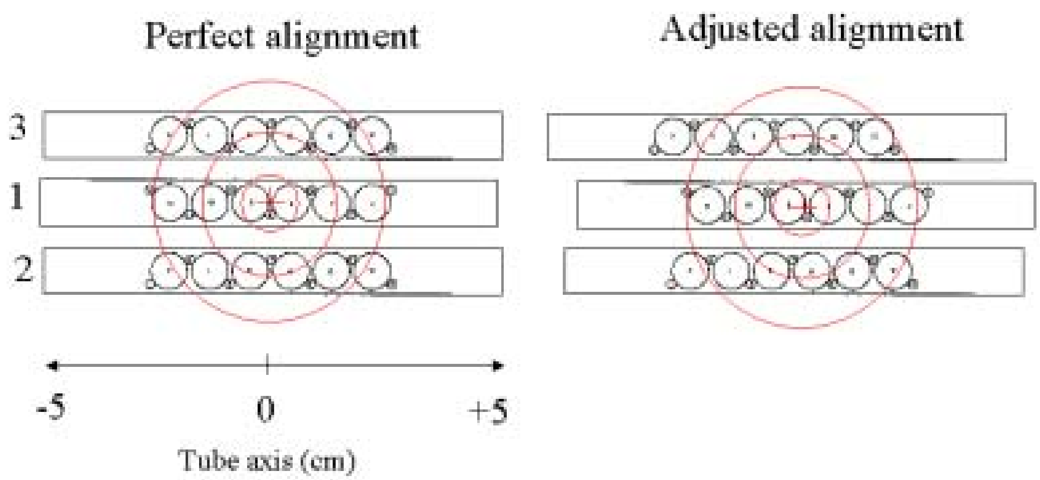

Figure 3. Relative offsets for the tubes and the proton beam – “new” 17A.

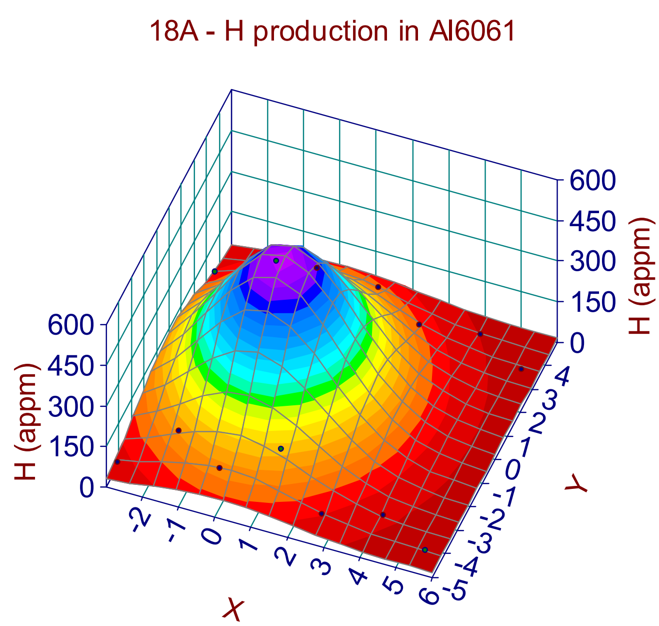

Figure 4. H production for Al6061 in 18A tubes. The foil stack locations are the points distributed above and below the peak.

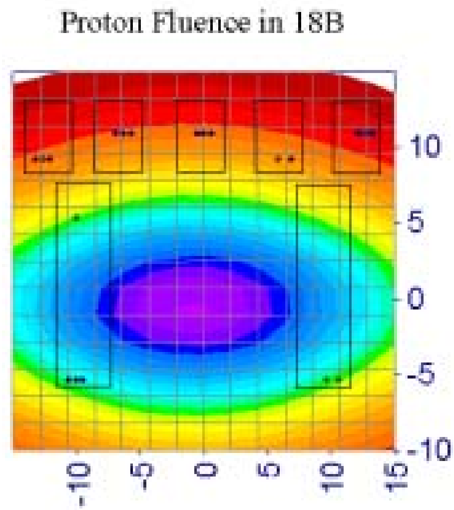

Figure 5. Proton fluence contour plot showing neutron furnaces, foil stacks and distribution of protons around the proton beam. The proton fluence in the furnaces ranged from $2x10^{18}$ to $1x10^{19}$ p/cm^2. In this figure, the peak proton fluence is about $1.34x10^{19}$ p/cm^2.

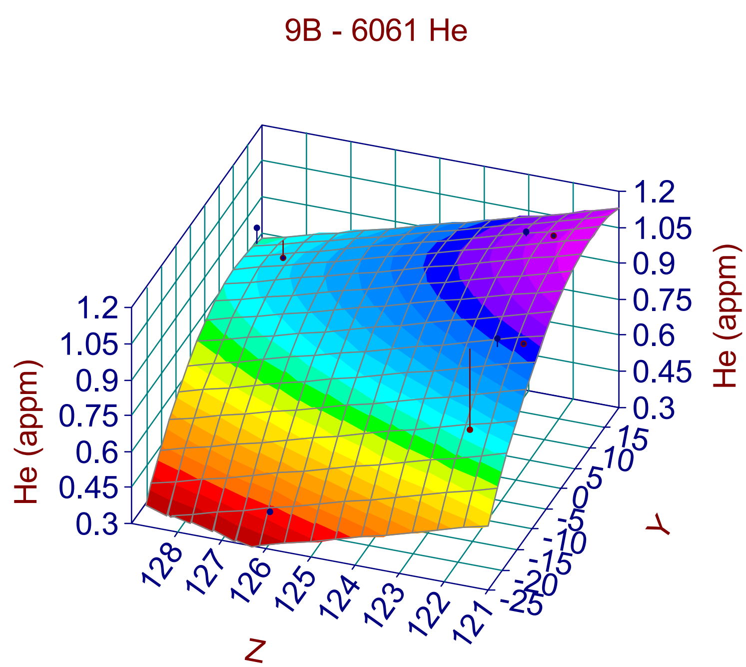

Figure 6. He production (in appm) in Al6061 for neutron furnaces located in off-beam location. There are five furnaces distributed over the Y-Z plane covered by this fit.

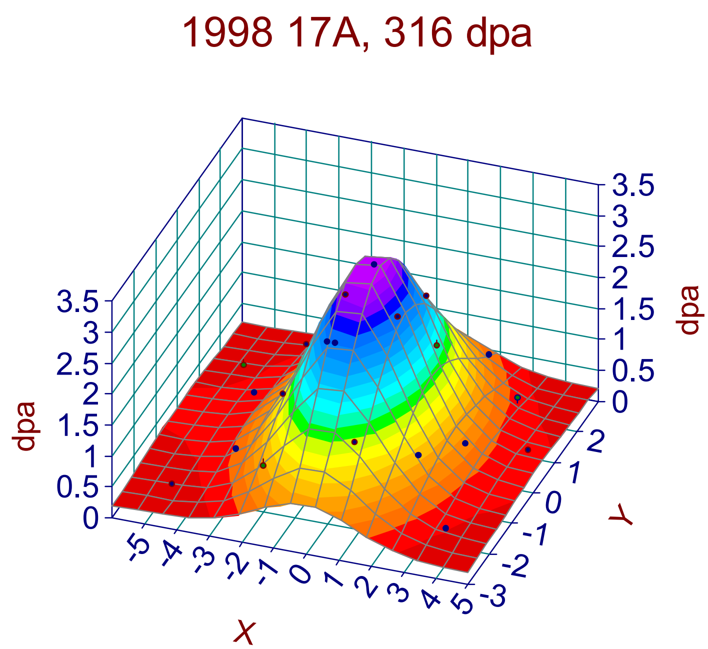

Figure 7. The 316L dpa from tubes in 17A insert from 1998 irradiation. The dots are activation foil locations. The tubes are located in the x direction.