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Post Irradiation Dose Determination of 800 MeV Proton Irradiated Aluminum from LAMPF Experiment 407

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

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W. F. Sommer

POST IRRADIATION DOSE DETERMINATION OF 800 MeV PROTON IRRADIATED ALUMINUM FROM LAMPF EXPERIMENT 407

by

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ABSTRACT

Recorded here are the results of post irradiation radiochemistry analysis of 800 MeV proton irradiated ultra high purity aluminum. Standard gamma-ray counting equipment, including a Ge Li detector, a multichannel analyzer, and associated electronics, was employed to count the ^{22}Na activity in the activated aluminum. Since activation is proportional to proton fluence, relative dose levels can be determined. Also, use of a selected production cross section for ^{22}Na in aluminum and a calculated damage energy cross section, both for 800 MeV proton bombardment, allows determination of a calculated value for the number of displacements per atom (dpa) that the material received during an irradiation experiment at the Clinton P. Anderson Meson Physics Facility (LAMPF). It is felt that simple and sufficiently accurate post irradiation dose determination can be made when either the subject material is ultra high purity aluminum or if this material is provided as a monitor with other subject materials, the simplicity being that the transmutation product, isotope ^{22}Na, is relatively abundant and has a long half-life.

I. INTRODUCTION

A five μA beam of 800 MeV protons, generated by LAMPF, was used to conduct a materials science radiation damage experiment.[1] Figure 1 shows that the experiment was a differential one in that one sample received an applied cyclic stress while its companion was under no applied stress. Analysis of the elliptical Gaussian beam and a calculated minimal effect by multiple scattering[2] predicted that the sample arrangement shown would yield identical radiation histories in the two samples. Determination of the effect of a cyclic stress on radiation induced microstructural evolution can only be done at equivalent dose levels. Accordingly, the first goal of this exercise was to determine, by radiochemistry, that the desired equivalence in fluence had been achieved. This was accomplished by comparing the ^{22}Na activity in material taken from both of the samples from equivalent positions in the Gaussian beam. Secondly, since radiation damage phenomena are fluence dependent, comparison to other published work requires knowledge of the absolute value of the accumulated dose. This was done by relating the measured ^{22}Na levels to a published production cross section for ^{22}Na in Al[3], which gave the apparent proton fluence. In addition, the parameter displacements per atom (dpa) was determined by calculation using the results of the work of Coulter, et. al.[4,5]

A. Method

Standard equipment for gamma radiation detection was used; the system is shown schematically in Fig. 2.

The electronics were set to observe the 1.273 MeV decay gamma ray from the transmutation product ^{22}Na in our Al samples. The activity was found to be about 1.5 mCi/g; a sample to detector distance of 200 mm yielded a count rate of around 10 counts per second. The efficiency of this geometry was determined by also counting the ^{22}Na activity of a NBS standard source under identical conditions.

Samples were prepared from the 0.25-mm-thick irradiated material by a spark machining operation that gave 3-mm-diameter discs; these discs to be used later as transmission electron microscopy (TEM) specimens. The relation of the Gaussian beam spot to the samples was initially determined by autoradiography on Kodak Panchromatic X film. An example of an autoradiograph is shown in Fig. 3 and gives a pictorial view of the Gaussian beam. Photodensitometry measurements allowed determination of the apparent center of the beam. The discs were selectively removed in 3-mm increments along the length of the samples for the subsequent counting exercise (see Fig. 1).

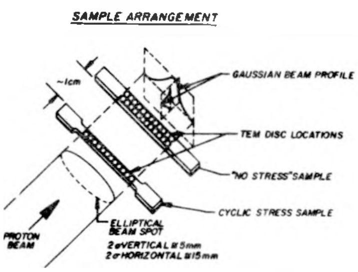

Fig. 1.
Sample arrangement for LAMPF experiment 407. TEM discs were spark machined from the sheet samples; post irradiation. The circles depict their location relative to the beam.

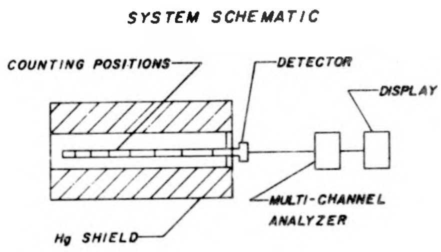

Fig. 2.
Schematic of counting equipment.

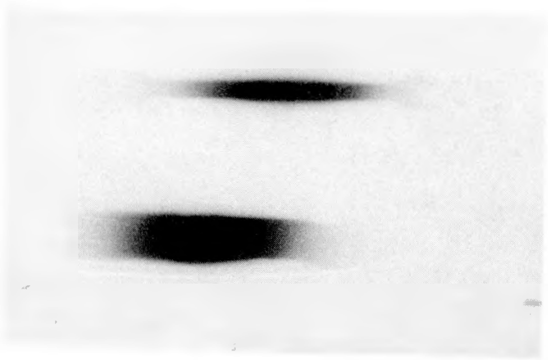

Fig. 3.
Autoradiograph of samples.

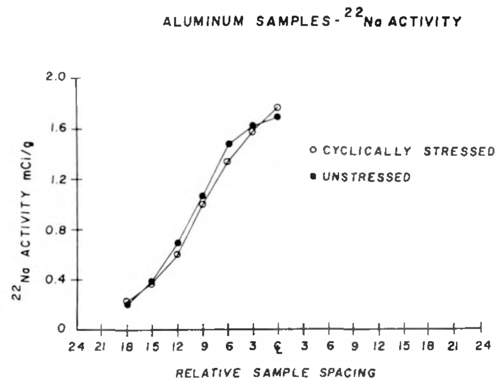

Fig. 4.
^{22}Na activity after a decay of 0.624y.

B. Results

Results of the radiochemistry analysis are shown in Fig. 4. The Gaussian beam intensity variation is noted by the fall-off in ^{22}Na activity for specimens away from the center. It is also noted that specimens removed from identical positions in the Gaussian beam, stressed and unstressed material, received a nearly identical dose, generally less than 5% different. To date only half of the specimens have been studied; the remainder are being held for study in the future.

Calculations gave the proton fluence as 3 x 10^{19} protons/cm^2 for the central, most heavily irradiated disc. This fluence translates to a calculated damage level of 0.045 dpa. A sample calculation is given in the included appendix.

C. Conclusions

1. Uniform dose was achieved for the differential radiation damage expeiment, as desired and predicated.

2. Maximum proton fluence was calculated to be about 3 x 10^{19} protons/cm^2.

3. Maximum damage level was calculated to be about 0.045 dpa.

4. Observation of ^{22}Na activity in Al is a simple, and to the extent of counting accuracy and the accuracy of the production cross section of ^{22}Na in Al, an accurate method of post-irradiation dose determination.

ACKNOWLEDGMENTS

Thanks are due to D. M. Parkin who assisted in the counting work, to B. J. Dropesky for the use of his counting facility at LAMPF, and to R. W. Martin who independently verified these results using his facility. Also, thanks are due to D. Shaffer for assistance in the autoradiography and photodensitometry work.

REFERENCES:

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2. C. A. Coulter and W. F. Sommer, "Flux Effect on a Aligned Second Foil due to Multiple Scattering of 800 MeV Protons in the Leading Foil," Los Alamos Scientific Laboratory Report, LA-7803-MS, April 1979.

3. Phy. Rev. C, 14, 1506 (1976).

4. C. A. Coulter, D. M. Parkin and W. V. Green, "Calculation of Radiation Damage Effects of 800 MeV Protons in a 'Thin' Copper Target," Journal of Nuclear Mat. 67 (1977) 140-154.

5. C. A. Coulter, unpublished results for Al.

APPENDIX

The following calculation is for specimen K which was taken from the maximum dose area of the material that received no applied cyclic stress.

A. Equipment Used

1. Cahn 21 electrobalance.

2. Canberra Industries detector, model LGCC.

3. NBS standard ^{22}Na source #4991-C-40, calibrated at 1000 EST on April 1, 1959 at $6.23 \times 10^4 \pm 1.53\%$ gammas per second, with a half life of $T_{\frac{1}{2}} = 2.603 \pm 0.002$ years.

B. Sample Physical Constants

1. Weight -- 4.59 mg

2. Molecular weight --26.98 g $mole^{-1}$

C. ^{22}Na Decay Constant

1. From

$$N = No \exp(-\lambda t)$$

and with

$$t = T_{\frac{1}{2}}$$

and

$$N = No/2$$

the decay constant;

$$\lambda = \frac{0.693}{T_{\frac{1}{2}}}$$

2. Here,

$$\lambda = \frac{0.693}{2.603} = 0.266 \text{ year}^{-1} = 8.44 \times 10^{-9} s^{-1} \quad .$$

D. NBS Calibrated Source Strength

1. Source calibrated on 4/1/69 @ 6.23×10^4 gammas per second = No_s

2. Elapsed time (12/19/79) is 10.69 y

3. Decay was

$$\frac{N}{No_s} = \exp(-\lambda t) = \exp -(0.266 \times 10.69) = 5.81 \times 10^{-2} \quad .$$

4. So the present source strength is

$$N_s = \frac{N}{No_s} No_s = 5.81 \times 10^{-2} \times 6.23 \times 10^4 = 3.62 \times 10^3 \frac{\text{gammas}}{s} \quad .$$

E. Detector Efficiency at Counting Position 11

1. Calibrated source gave

$$\frac{1587 \text{ counts}}{14000 \quad s} = 1.13 \times 10^{-1} \frac{\text{counts}}{s} \quad .$$

2. Therefore the efficiency is,

$$\text{efficiency} = \frac{\text{recorded}}{\text{source strength}} = \frac{1.13 \times 10^{-1}}{3.62 \times 10^3} = 3.12 \times 10^{-5} \quad .$$

F. Specimen K activity (position 11)

1. $\frac{1908 \text{ counts}}{200 \ s} = 9.54 \frac{\text{counts}}{s} \quad .$

2. Specimens have decayed for 0.624y, therefore, relative to the end of the irradiation,

$$\left.\frac{N}{No}\right)_K = \exp - (0.266 \times 0.624) = 0.847 \quad .$$

3. And, therefore, the apparent count rate at the end of the irradiation was

$$\frac{9.54}{0.847} = 11.26 \ \frac{\text{counts}}{\text{s}} \quad .$$

4. And, applying the efficiency at position 11, the total activity at the end of the irradiation was

$$\frac{11.26}{3.13 \times 10^{-5}} = 3.59 \times 10^{5} \ \frac{\text{decays}}{\text{s}} \quad .$$

G. Population of ^{22}Na Atoms at the End of the Irradiation

1. For

$$dN = No \ \lambda \ dt$$

with

$$dN = 3.59 \times 10^{5} \text{ decays}$$

$$dt = 1\text{s}$$

$$\lambda = 8.44 \times 10^{-9} \ \text{s}^{-1}$$

2. Then

$$No = \frac{dN}{\lambda dt} = \frac{3.59 \times 10^{5}}{8.44 \times 10^{-9} \times 1} = 4.28 \times 10^{13} \ ^{22}\text{Na atoms} \quad .$$

H. Implied Proton Fluence

1. Production cross section

$\sigma_{^{22}Na}$ in Al under 800 MeV protons = 13.6 mb*

$= 1.36 \times 10^{-26}\ cm^{2}\ proton^{-1}$

and the published value

$\sigma_{^{22}Na} = 15.0$ mb**

We use 13.6 mb

*Private communication, B. J. Dropesky to D. M. Parkin

**Phys. Rev. C, 14, 1506 (1976)

2. Therefore,

$$N_{^{22}Na} = \sigma N_p N_{Al}/A \quad ,$$

where

$N_{^{22}Na}$ = number of ^{22}Na atoms

σ = cross section

N_p = number of protons

N_{Al} = number of aluminum atoms

A = area .

Here,

$$\frac{N_p}{A} = \frac{4.28 \times 10^{13} \text{ atom} \times 26.98 \text{ g mole}^{-1}}{1.36 \times 10^{-26} \text{ cm}^2 \text{ proton}^{-1} \times 4.595 \times 10^{-3} \text{ g} \times 6.023 \times 10^{23} \text{ atom mole}^{-1}}$$

$$= 3.07 \times 10^{19} \text{ protons cm}^{-2}$$

I. Implied Damage Level (dpa = displacements per atom)

1. For Al

σ_d = displacement energy cross section = 63 bkeV

E_d = 17 eV

2. And

$$\frac{\text{dpa}}{N_p/A} = \frac{0.8\sigma_d}{2E_d} \quad \frac{0.8 \times 63 \text{ bkeV} \times 10^{-24} \text{cm}^2 \text{ b}^{-1} \times 10^3 \text{ eV keV}^{-3}}{2 \times 17 \text{ eV}}$$

$$= 1.48 \times 10^{-21}$$

3. Then

$$\text{dpa} = \frac{\text{dpa}}{N_p/A} \times N_p/A = 1.48 \times 10^{-21} \times 3.07 \times 10^{19} = 0.0454$$

J. Sample Activity at the End of the Irradiation

1. With

1 mCi = 3.7×10^7 decays/s

2. And from F.4

$$\frac{3.59 \times 10^5 \text{ decays/s}}{3.7 \times 10^7 \frac{\text{decays/s}}{\text{mCi}}} = 9.7 \times 10^{-3} \text{ mCi}$$

3. And per unit weight

$$\frac{9.7 \times 10^{-3}\ \text{mCi}}{4.595 \times 10^{-3}\ \text{g}} = 2.11\ \frac{\text{mCi}}{\text{g}}$$

4. For Comparison to Figure 2, at

$$0.624\text{y},\ \left.\frac{N}{No}\right)_K = 0.847.$$

So, $2.11 \times 0.847 = 1.79\ \frac{\text{mCi}}{\text{g}}$.