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IRRADIATION EFFECTS ON THE PHYSIO-MECHANICAL PROPERTIES OF SUPER-ALLOYS CHARACTERIZED BY LOW THERMAL EXPANSION*

N. Simos#, H. Kirk, BNL, Upton, NY 11973, USA
K. McDonald, Princeton University, Princeton, NJ 08544, USA
N. Mokhov, FNAL, Batavia, IL 60510, USA

Abstract

In an effort to address the limitations on high power accelerator target performance prompted by the elevated dose levels and the associated irradiation damage, an experimental study has been undertaken to evaluate the potential applicability of super alloys characterized by low thermal expansion over certain thermal regimes. The intriguing properties associated with materials such as super-Invar and the "gum" metal (Ti-12Ta-9Nb-3V-6Zr-O) are observed in their un-irradiated state. Irradiations were performed using the 200 MeV protons of the BNL Linac and/or a neutron flux generated by the stopping of the primary 112 MeV protons upstream of the exposed super-alloys. The paper presents the post-irradiation analysis results which reveal interesting damage reversal by the super-invar and unexpected low threshold of radiation resistance by the "gum" metal.

INTRODUCTION

The irradiation damage of certain super-alloys possessing low thermal expansion such as such as super-Invar and the "gum" metal (Ti-12Ta-9Nb-3V-6Zr-O) has been evaluated through a series of irradiation experiments using the BNL accelerator complex and in particular the Isotope Production Facility which receives a 200 MeV, 90 μA proton beam on target and allows the exposure of materials to high fluences. The primary interest in these super alloys stems from the low thermal expansion property α which enters the metric that determines the behavior of a target material under beam shock according to the relations below.

$$\Delta P = \Gamma \rho \Delta E_m \quad \text{where } \Gamma = [E/(1-2\nu)]\ \alpha/(\rho\ c_v) \qquad (1)$$

Γ is the Gruneisen parameter; $\boldsymbol{\Delta P}$ represents the change in hydrostatic pressure and is related to the energy density change $\boldsymbol{\Delta E_m}$ through the equation of state ν is the Poisson ratio, ρ is the densit and c_v is the constant volume specific heat. Clearly, the lower the thermal expansion in the material, the smaller the resulting shock stresses which will result in longer target lifetime.

The study was focused in these two alloys because both exhibit extremely low thermal expansion at certain temperature ranges. Specifically the super-Invar exhibits very low thermal expansion up to ~150 C at where there it transitions to a "typical" material behavior. The gum metal on the other hand also possesses the invar-like property but with extended temperature range (~ 400 C). Beyond this point the material undergoes a phase transition with rapid increase of the thermal expansion coefficient. On the mechanical side, the material exhibits low Young's modulus and inelastic behavior followed by extreme ductility while maintaining high strength. The physio-mechanical properties of the gum metal [4] are shown in Figure 1.

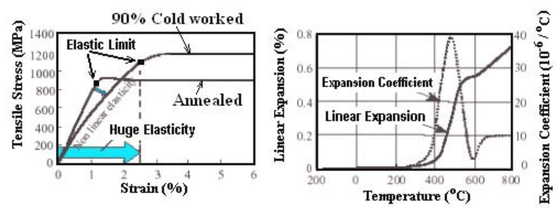

Figure 1: Un-irradiated Gum material properties [4]

The goal of the experimental effort was to assess how radiation affects these attractive properties that the super-invar and gum metal possess. A series of experiments were conducted and the post irradiation analyses led to a series of conclusions regarding the resilience of these alloys to radiation. The experimental effort details and the results are presented in the following sections.

EXPERIMENTAL EFFORT

Using the BNL accelerator complex and the Isotope Production Facility, shown schematically in Figure 1, two types of irradiation were achieved. The initial phases focused on the effects of direct protons on the properties of these materials. The 200 MeV proton beam from the BNL Linac was used to irradiate an array of water-cooled targets according to the scheme shown in Figure 2. The first of proton irradiation phases explored the effects on super-Invar. Radiation damage levels of ~0.25 displacements –per-atom (dpa) were achived. Following the post-irradiation analysis and the interesting annealing behavior observed, two additional proton irradiations were conducted looking into the behavior of the gum metal under higher fluences and the behavior of super-invar following annealing of the damage accumulated.

To address the combined effect of neutrons photons and electrons, a special arrangement in the target station was introduced that allowed the generation of high neutron fluxes. Specifically, the upstream isotope targets were utilized and enabled the stopping of the primary protons resulting in a uniform flux of neutrons, photons, and electrons at the location of the accelerator targets of

* Work supported by the U.S. Department of Energy
# simos@bnl.gov

interest (super-invar and gum metal). Figure 4 depicts the schematic of the target arrangement as generated by the MARS15 Monte-Carlo code [5] which was used to generate the flux and estimate the integrate dose received. Figure 5 depicts the particle tracks which show that the primary protons are stopped by the last isotope target situated upstream of the gum metal and super-invar materials. Figure 6 are MARS15 estimation of the neutron, photon and secondary proton fluxes in the entire target configuration. Figure 7 are neutron and proton energy spectra at the super-Invar location.

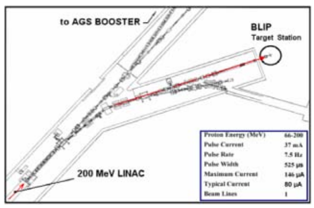

Figure 2: Schematic of the BNL BLIP irradiation facility

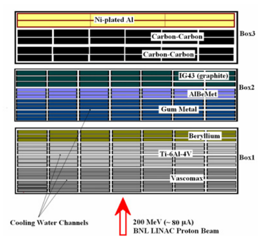

Figure 3: Target assembly for 200 MeV proton irradiation

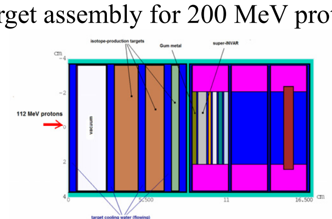

Figure 4: Un-irradiated Gum material properties

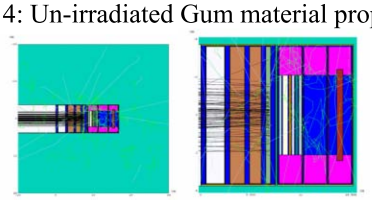

Figure 5: Un-irradiated Gum material properties

POST-IRRADIATION RESULTS

Initial evaluation of the irradiated super Invar revealed that even modest levels of irradiation (~0.20 dpa) resulted in dramatic increase of its CTE, as shown in Figure 8. As shown in Figure 9 the material exhibited good resistance in the ductility loss due to irradiation while also showed dependence on the temperature. Prompted by findings of annealing behavior of other special materials and composites the irradiated super-Invar was annealed using different thermal cycles. The results of the process are shown in Figure 10. It was found that the material reverses the damage completely when annealed to a temperature above 600° C which appears to be a characteristic threshold.

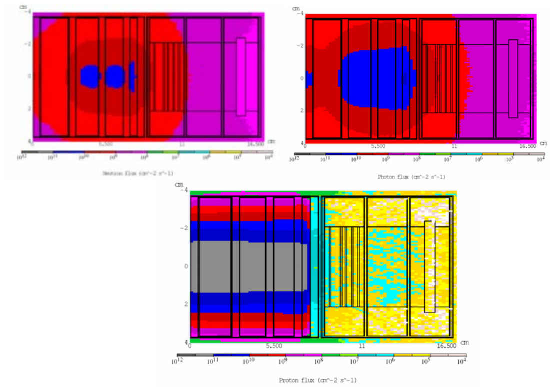

Figure 6: Estimated neutron, photon and proton spectra in the target irradiation arrangement (MARS15 code)

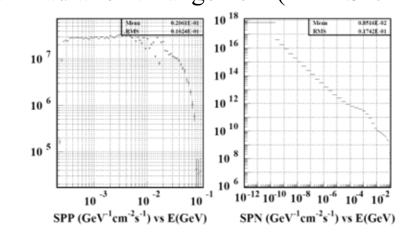

Figure 7: Estimated proton and neutron energy spectra

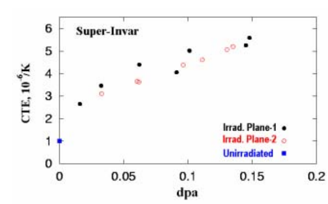

Figure 8: Proton irradiation effects on super-Invar CTE

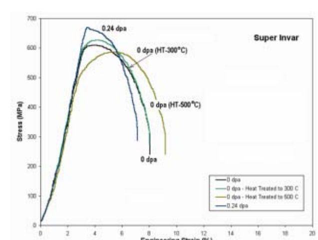

Figure 9: Proton irradiation effects and temperature effects on the stress-strain relation of super-Invar

To assess whether the annealed and restored super-invar can resist radiation damage brought upon a new exposure (it has been found that other alloys with annealed damage suffer a greater damage after subsequent irradiation) the super invar was re-irradiated to about 1.0 dpa and was re-evaluated with post—irradiation analysis. It was found that while damage re-appeared in the form of increased thermal expansion in the range of 0-150 C, re-annealing was able to bring the material back to the original state. This remarkable behavior is shown in Figure 11. Restored invar material, following the second proton irradiation, were exposed to the irradiating field that is dominated by neutrons combined with photons, electrons and secondary protons. Following the completion of the exposure which achieved lower integrated fluence even though that target arrangement was exposed to much higher direct proton current (actual estimates still pending), the super invar was re-evaluated. As seen in Figure 12 the material experiences similar damage (departure from the low CTE in the 0-150 C) but through annealing it restores most of the loss. As it clearly seen some damage still remains. This is attributed to the different mechanism the governs the neutron interaction and to the presence of photons and electrons. Interesting to note that annealing restores the CTE of the irradiated material for the temperature regime above 150 ^{o}C. (parallel thermal expansion traces).

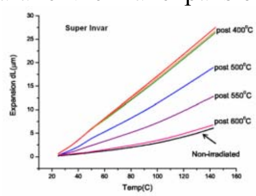

Figure 10: Damage reversal or CTE restoration in irradiated super Invar through thermal annealing

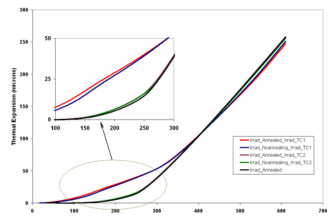

Figure 11: Damage reversal or CTE restoration in super-Invar following a second, higher dose proton irradiation

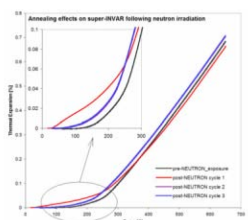

Figure 12: Damage reversal or CTE restoration in super-Invar following a third, neutron-dominated irradiation

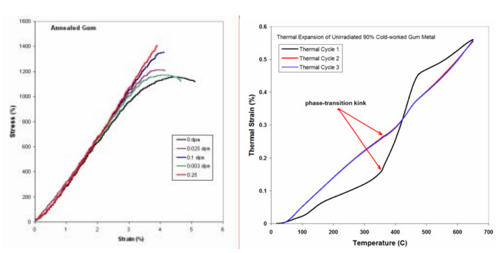

Figure 13: Irradiation-induced ductility loss and thermal cycle effects on Gum metal

The superior properties of the Gum metal [4] were also examined under proton and neutron irradiation in an effort to identify a material that possesses the right combination of physio-mechanical properties and is able to maintain them under irradiation. Irradiation of the annealed version of the material, however, revealed that even modest levels of irradiation damage remove its uncharacteristic ductility entirely (Fig. 13a). It was also found that a single pass of temperature above the phase transition temperature (~400 C) removes the low thermal expansion behavior exhibited by the 90% cold-worked material in the temperature regime up to 400 C. As shown in Figure 14 irradiation has minimal effect on this dramatic change. It is apparently temperature that dominates the behavior. Figure 15 compares the effect of proton irradiation with that of neutron, photon and electron combination. While similar in the general trend it appears the combination of irradiating species interacts with the material through a different mechanism (actually causes the lowering of the CTE shown by the un-irradiated material).

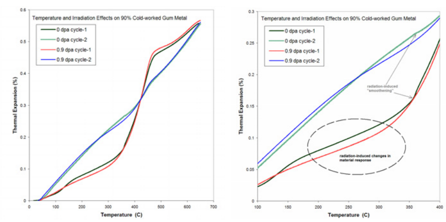

Figure 14: Proton irradiation and thermal effects on gum

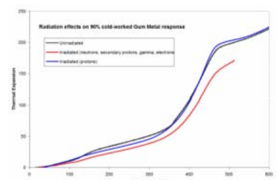

Figure 15: Proton and neutron irradiation effect comparison on the thermal expansion of gum metal

SUMMARY

The BNL experimental effort on the radiation effects on super-alloys with low CTE revealed that the super-Invar possesses the inherent ability of restoring proton-induced damage through thermal annealing when reaching a certain temperature threshold. Under neutron-dominated exposure however, the material reverses most of the caused damage. Further explorations are needed to address the difference in mechanism. The gum metal on the other hand was found to experience total loss of its ductility even under modest irradiation and is greatly affected by temperature that crosses the transition threshold of ~450 ^{o}C. The current assessment is that the gum metal is not suitable as high power target material.

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