Radiation-Damage Robust, Engineered, Self-repairing Meso-Materials

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ABSTRACT

Nuclear power is one of the most compact, long lasting and might be the most safe and environmentally friendly energy source if it might be done right, by achieving the harmony between the nuclear reactions inside and the material structure these reactions took place.
The structural materials used inside a nuclear power source, mainly stainless steel, zircalloy, etc., are suffering the radiation damage, and achieving high burnup factors, or near perfect burning is practically impossible using the present metallic alloys, due to safety reasons.
Producing robust materials with micro-structure shape memory, like SiN, Ti, composites, etc., whose properties to be constant with neutron fluence, or dose after impaired by radiation damage, come back to the initial structure and recover, process also known as self-repairing mechanism.

The application of these materials in nuclear reactors will make possible the increase of reactor lifetime by a factor of 5, allowing burnup factors up to 50% based on breed and burn technology, reducing the need for fuel reprocessing, and may have good applications in space technology. Good understanding of these processes involved in self-repairing, may be applied to electronic devices for space and radioactive environment, and many other applications.

Keywords: hetero-structure, micro-hetero-structure shape memory, fission products separation, transmutation, meso-structure. Shape-memory alloy

1 INTRODUCTION

The advanced cladding made by the nano-composite sinter structure makes the harmony between nuclear recoils dislocation and the structure creating a dislocation tolerant structure that exhibits self-repairing properties [1].

Radiation may cause the materials to become radioactive by neutron activation, that may also induce nuclear transmutation of the elements within the material changing its physical properties, and induce chemical changes, as radiolysis (breaking chemical bonds) weakening the material make it swell, polymerize, promote corrosion, cause belittlements, promote cracking or otherwise change its desirable mechanical, optical, or electronic properties [2].

The electrical discharges initiated by the ionization events by the particles result in plasma populated by large amount of free radicals [3]. The highly reactive free radicals can recombine back to original molecules, or initiate a chain of free radical polymerization reactions with other molecules, yielding compounds with increasing molecular weight. Liquids lack fixed internal structure, therefore the effects of radiation is limited to radiolysis that is altering the chemical composition of the liquids, one of the primary mechanisms is formation of free radicals.

All liquids are subject to radiation damage, with few exotic exceptions as liquid metals like molten sodium or LBE, where there are no chemical bonds to be disrupted.

Liquid water or hydrogen fluoride, produces gaseous hydrogen and oxygen respectively fluorine, which spontaneously react back to releasing the reaction energy.

Absorption of neutrons in hydrogen nuclei leads to buildup of deuterium and tritium in the water.

Two main approaches to reduce radiation damage are:

-reducing the amount of energy deposited in the sensitive material (e.g. by shielding, distance from the source, or spatial orientation), or

-modification of the material to be less sensitive to radiation damage (e.g. by adding antioxidants, stabilizers, or choosing a more suitable material).

In addition to the electronic device hardening mentioned above, some degree of protection may be obtained by shielding, usually with the interposition of high density materials (particularly lead, where space is critical, or concrete where space is available) between the radiation source and areas to be protected...

2 TYPES OF RADIATION DAMAGE

In order to understand how the materials have to be made we need to understand that the types of radiation that can alter structural materials consist of neutrons, ions, electrons and gamma rays, because have the capability to displace atoms from their lattice sites, and this is the fundamental process that drives the changes in structural metals.

2.1 Defects in material

The primary event is the interaction between radiation and a particle from the solid lattice triggering its displacement from its lattice site. Irradiation displaces an atom from its site, leaving a vacant site behind and the displaced atom eventually comes to rest in a location that is between lattice sites, becoming an interstitial atom, also called Frenkel pair (FP). The presence of the Frenkel pair and other consequences of irradiation damage determine the physical effects, and with the application of stress, the mechanical effects of irradiation and the occurrence of interstitial, phenomena, such as swelling, growth, phase change, segregation, etc [4]. Therefore the radiation damage event is defined as the transfer of energy from an incident projectile to the solid and the resulting distribution of target atoms after completion of the event. This event is composed of several distinct processes:

-The interaction of an energetic incident particle with a lattice atom

-The transfer of kinetic energy to the lattice atom giving birth to a primary knock-on atom (PKA)

-The displacement of the atom from its lattice site

-The passage of the displaced atom through the lattice and the accompanying creation of additional knock-on atoms

-The production of a displacement cascade (collection of point defects created by the PKA)

-The termination of the PKA as an interstitial

The result of a radiation damage event is the creation of a collection of point defects, vacancies and interstitials, and clusters of these defects in the crystal lattice, creating the so-called spike affected volume[5].

The result is the total number of displacements in the target from a flux of particles with a known energy distribution. In radiation material science the displacement damage in the alloy (= displacements per atom (dpa) in the solid) is giving a better representation of the effect of irradiation on materials properties than the radiation (neutron, gamma, etc) fluence, but still irrelevant from the practical point of view.

2.2 Collision cascades

A collision/displacement cascade/spike is defined as a set of nearby adjacent energetic collisions of atoms, higher than local thermal energies, induced by an energetic particle stopping in that matter.

If the maximum atom or ion energies in a collision cascade are higher than the threshold displacement energy of the material ranging in tens of eVs, the collisions can permanently displace atoms from their lattice sites and produce defects. The initial energetic atom can be any moving particle or, an atomic recoil produced by a passing high-energy neutron, electron or photon, or can be produced when a radioactive nucleus decays and gives the atom a recoil energy [6].

Since the kinetic energies in a cascade can be very high, it can drive the material locally far outside thermodynamic equilibrium, producing remnant defects.

The defects can be point defects such as Frenkel pairs, ordered or disordered dislocation loops, stacking faults, or amorphous zones[7]. A material exposed to high radiation doses suffers amorphization.

The defects production can be harmful, such as in nuclear fission and fusion reactors where the neutrons slowly degrade the mechanical properties of the materials.

An apparently curious feature of collision cascades is that the final amount of damage produced is much less than the number of atoms initially affected by the heat spikes, process called recovery. In pure metals, and some ionic materials the final damage production after the heat spike phase can be orders of magnitude smaller than the number of atoms displaced in the spike.

When such cascade or spike is crossing an interface or a surface it leads to sputtering or material mixing, even for materials that are normally thermodynamically immiscible. Heat spikes near surfaces lead to crater formation.

The non-equilibrium natures of irradiation drives materials out of thermodynamic equilibrium, and thus form new kinds of alloys. This process of continuous transformation of material under irradiation, triggers materials continuous change in properties, generally called irradiation damage, because the initial material performances are better than the performances after irradiation.

2.3 Wigner effect

Discovered by E. P. Wigner, also known as the decomposition effect, is given by the displacement of atoms in a solid caused by neutron radiation.

All solids are affected by the Wigner effect, but the effect was of most concern in neutron moderators, such as graphite, that are used to slow down fast neutrons, and get dislocation defects.

An interstitial atom and its associated vacancy are known as a Frenkel defect.

In order to create the Wigner effect, neutrons that collide with the atoms in a crystal structure must have enough energy to displace them from the lattice; threshold displacement energy is approximately 25 eV for graphite.

A 1 MeV neutron with a significant amount of energy will create about 900 displacement cascade in a matrix via elastic collisions, but not all displacements will create defects because some of the dislocated atoms will find and fill the vacancies that were either small pre-existing voids or vacancies newly formed by the other struck atoms.

The atoms that do not find a vacancy come to rest in non-ideal locations; that is, not along the symmetrical lines of the lattice. These atoms are referred to as interstitial atoms, or simply interstitials. Because these atoms are not in the ideal location they have an energy associated with them, a kind of potential energy. When large amounts of interstitials have accumulated they pose a risk of releasing all of their energy suddenly, creating a temperature spike.

Accumulation of energy in irradiated graphite has been recorded as high as 2.7 kJ/g, but is typically much lower than this. This build up of energy referred to as Wigner energy can be released by heating the material. This process is known as annealing [8].

In graphite this exotermic annealing occurs over 250°C

Sudden unplanned increases in temperature can present a large risk for certain types of nuclear reactors with low operating temperatures and were the indirect cause of the Windscale Reactor fire that occurred during a controlled annealing [9].

It has recently been postulated that Wigner's energy can be stored by the formation of meta-stable defect structures in graphite. Notably the large energy release observed at 200-250°C has been described in terms of a meta-stable interstitial-vacancy pair. The interstitial atom becomes trapped on the lip of the vacancy, and there is a barrier for it to recombine to give perfect graphite.

Graphite is maintaining a high energy in defects, but anneals and repairs over 250 C while all the metallic clusters needs much higher temperatures.

3 MATERIALS ROBUST TO RADIATION DAMAGE

In fact, there is no way to protect a material against radiation damage, but material may be resilient to radiation damage and not modify its shape, dimension or properties with the absorbed dose. That, in generic terms means the achievement of a harmony between the damage and recovery, and material remains reliable for extended periods.

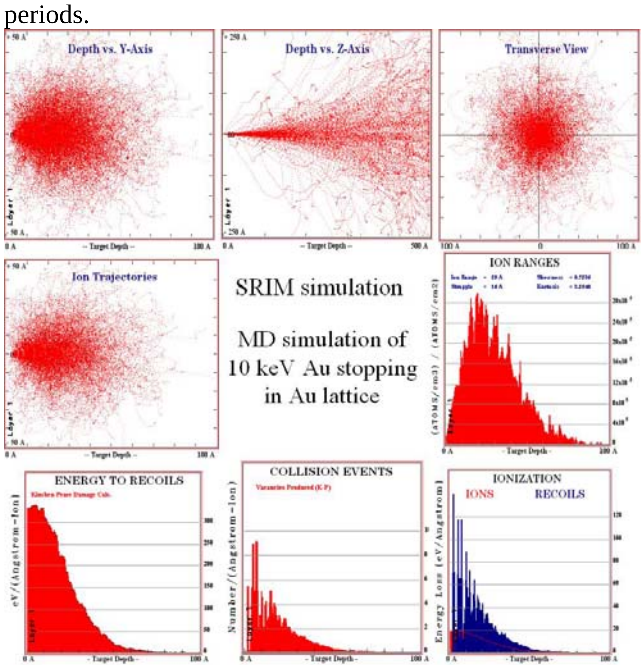

Fig. 1. Gold lattice damage inflicted by a recoiled Au atom

The figure above shows the simulations made by Monte Carlo Codes (MC) SRIM for a gold foil, where a 10 keV gold atom recoil was induced by a few hundred keV neutron scattering [1].

The entire slide presents the case of a 10 keV gold atom recoiled in the gold lattice or mono-crystal. The upper-left image shows the behavior of the lattice, while the rest of the charts represents the recoiled atom path inside the lattice and its associated parameters, how far it goes, how it deposits its energy, etc [10].

3.1 New cladding materials

A main problem in the actual fuel cladding system is given by the radiation damage. Experiments elaborated in 1960s showed the rate of failure of the fuel pellets versus surface temperature and burnup shown in Fig. 2.

To this data, we have added the fit line, of the failures that shows that at 1960s fuel 1% of burnup is equivalent to about 400 C more on the surface.

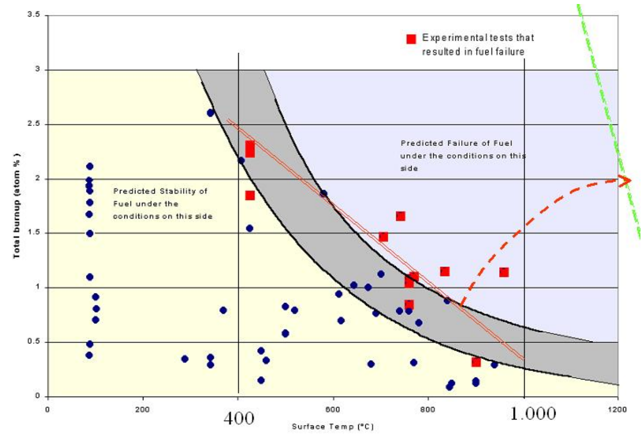

Fig. 2. Rate of failures dependence on burnup and operating temperature.

Our goal is to displace this red double line shown in Fig.2 representing the fuel failures versus burnup and temperature towards higher grounds, represented by the green dashed curve.

We have to consider that there is a damage induced by the fission products in the fuel and a radiation damage induced in the cladding, due to dislocations, that makes the cladding swallow and getting some embitterment.

4 SELF-RECOVERING MATERIALS

What matters is not how the material behaves during 100 ps when the spike energy sharing takes place, but what happens after that, and if the material maintains its original shape and properties, as Fig. 3 shows.

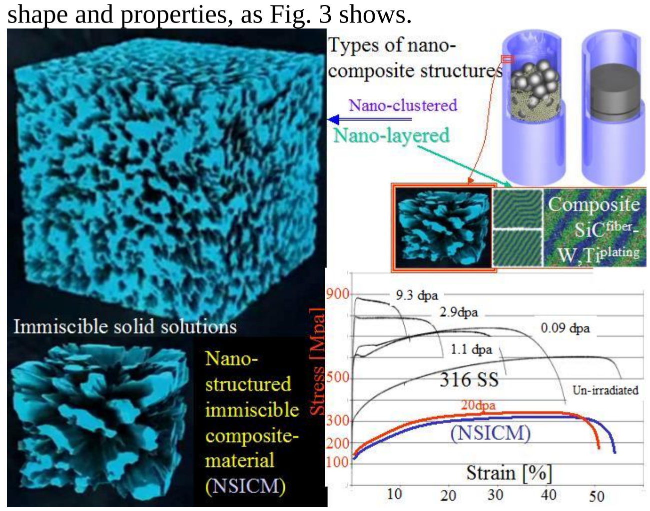

Fig. 3: Radiation robust self-repairing nano-structured materials.

In the right-low corner is presented a chart showing the variation of stress versus strain as function of radiation dose in [dpa] (displacements per atom) for the most used since 1960s stainless steel – the 316, and for the new material, generically called nano-structured immiscible composite material (NSICM). Therefore, using the previous knowledge, we, now answer the question about how to create a material that to be immune to radiation damage.

Such a structure has to lock the position and recover into a self-similar structure after the damage.

Even it is not having as high mechanical performances as the SS316 its radiation damage resilience makes it a favorite for high burnup applications, and long life nuclear structures.

There are two types of structures that may exhibit this property. One is made on a plurality of layers being a composite material that transforms into a 3D double interlaced mesh being stabilized by the immiscibility of the phases creating a system of solid solutions. This kind of material is an invar under radiation, and once it reached the equilibrium it remains there maintaining the same properties over a wide dose range or equivalent fuel burnup.

A LANL scientist of Chinese origin, Xian-Ming Bai shows the ability of a material to resist radiation damage is determined by how well the microstructure can remove vacancies and interstitial defects in equal numbers . I agree with him when he says that: ”However, the exact processes by which this happens are poorly understood, and the search for promising materials has been largely heuristic”. Bai describe a mechanism in copper that suggests that nano-materials in general might have exceptional radiation resistance, being exactly what I stated since 2004 in various publications. He’s interpretation is supported by some previous experimental findings and suggests new directions for trial materials.

In fact, in nano-materials, that are subject to Bragg peak the interface is all along the peak, and the crystalline boundary, more, if the size of the spike is comparable with the grain size, the crystallite may split into smaller crystals, in the process called amorphization . If this process takes place in refractory or semiconductor materials, the diffusion speed is low and the lattice properties alteration occurs, due to absorbed dose, which generated displacements.

5 CONCLUSIONS

Nuclear safety and nuclear reactor’s performances are improved by radiation robust nano-structures.

This material represents a breakthrough, increasing the fuel safety and making structural elements from materials that have the parameters immune to radiation damage.

The structures are mechanically stable with dose taking the radiation damage and recover to similar structures in the same locations using nano-cluster special properties, due to dynamically distributed active interfaces.

The structures may have low neutron interaction cross-section and be chemically inert versus cooling and drain fluids.

- reduces irradiated structure and cladding waste mass
- does not react with coolants catalysts like Zirc Alloys.
- allows ultra high burnup without recladding

These materials have also several issues:

- Are difficult to manufacture,
- are requiring advanced nano-materials knowledge
- may be insensitive to a reduced type of radiations, or exhibit radiation energy dependence for properties alteration effect.

This material represents a breakthrough, increasing the fuel safety and making structural elements from materials that have the parameters immune to radiation damage. Some other neutron economy optimizations may also contribute to long-life of the nuclear structures.

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When specking about advanced radiation robust materials, we have also to use common sense, and to appreciate that for the safety reasons, even if the material can withstand to an increase of absorbed dose by two orders of magnitude, we may not use that structure providing an non economical ultra high burnup, but an assembly of methods, achieving the same goal with minimum cost.

There are many structures able to exhibit high radiation resilience [10] and a dedicated research will be the most appropriate in order to collect the already available knowledge unitarily and apply in the technological developments.

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