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Review of Irradiation Effects on Ferrites: Results in the World from 1970 to 1995

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Abstract. In this paper, we present the main irradiation effects observed on ferrites in the world between 1970 and 1995. The crystallographic and magnetic changes induced by irradiation are shown for various kinds of particles : fast neutrons, low energy ions and high energy heavy ions and for different ferrite structures : spinels, garnets and hexaferrites. The observed effects depend both on the mechanism of particle crystal interaction and on the structure of the ferrite. From crystalline point of view, several effects can be induced : increase of lattice parameter, change of site and valence for cations and anions, amorphisation, creation of tracks showing either elongated extended defects or continuous amorphous cylinders. From magnetic point of view, the following effects can be induced : - the magnetization and the Curie temperature can either increase, or decrease, or be constant, - the anisotropy can change and even cancel. These effects are reproducible and stable at 300K. Recently, it was found that Fe_3O_4 is the most irradiation resistant ferrite.

1. INTRODUCTION

Many experimental results have been published on the new properties of spinels and garnets ferrimagnetic oxides after irradiation by various kinds of particles - electrons, γ -photons, fast-neutrons, protons, low-energy ions and high-energy heavy ions. These investigations are of interest in several fields, e.g., in the nuclear industry to know the behaviour of ferrites near a reactor, in space radiation to know the behavior of devices using ferrites, in bubble garnets whose properties are improved by low energy ion bombardment. During the irradiation of a crystal, the mechanisms of particle-crystal interactions inducing crystallographic changes are mainly of two kinds : the nuclear elastic collisions and the electronic inelastic collisions. These mechanisms produce displacements of electrons and ions in the crystal.

2. FAST-NEUTRON AND LOW-ENERGY-ION IRRADIATIONS

In the two cases of fast-neutron and low-energy-ion irradiations, the particle-crystal interaction is governed by the same mechanism of elastic nuclear collisions.

2.1 Spinel and hexagonal ferrites irradiated by fast-neutrons

The typical irradiation conditions by fast neutrons in nuclear reactor were following : neutron energy $E_n > 0.1$ MeV ; neutron flux density $0.8\text{--}1.5 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$; temperature approximately 350 K.

We first show the effects of fast neutron irradiation on saturation magnetization n_B , for different ferrites : on figure 1, for spinels (Mn-Zn ferrite I1I, Ni-Zn ferrite I1I, NiFe_2O_4 I2I) and on figure 2, for $\text{BaFe}_{12}\text{O}_{19}$ I3I. In the case of spinel structure

Figure 1: magnetization n_B vs neutron dose (spinels)

Figure 2: (1) magnetization n_B , (2) n_B canting angle ($\text{BaFe}_{12}\text{O}_{19}$)

(NiZn, MnZn and NiFe₂O₄), the changes are interpreted by a statistical redistribution of cations over unequivalent sites : thus, nickel ions can move from octahedral to tetrahedral sites and cause an increase in n_B . In the case of hexagonal structure (BaFe₁₂O₁₉), a decrease in n_B and a non collinear magnetic order are observed. These changes are interpreted as arising from the creation of Fe³⁺ cation vacancies in the "trigonal" 2b sites that weaken the exchange interaction between the spinel blocks.

Now, let us consider the effects of fast neutron irradiation on Curie temperature T_c 141. Depending on composition, T_c can either decrease, or increase, or be constant. We show in figure 3 one typical example of the change in T_c of NiZn ferrites : in that case, T_c increases with the dose 141. This behaviour is well interpreted by the redistribution of cations : the transfer of magnetic ions from octahedral to tetrahedral sites can cause an increase in number of exchange linkages and explain the increase in T_c . This effect has recently been summerized in ref. 151.

Figure 3: initial permeability vs temperature (NiZn spinel)

Figure 4: initial permeability vs neutron dose, at 150 K (spinels)

More, the effects of fast neutron irradiation on magnetocrystalline anisotropy K_1 have been also observed 161. So, the permeability, which is proportional to $1/K_1$, shows a peak versus neutron dose (figure 4). The peak is clearly due to a cancellation of K_1 (which is the sum of various single-ion contributions) : the irradiation induces displacements of Ni²⁺ in tetrahedral sites, giving a positive contribution to K_1 161.

Finally, we also mention the effects of fast neutron irradiation on magnetite 1141. Recently, it has been found that magnetite Fe₃O₄ is very resistant to the fast neutron irradiation : for 2×10^{20} n/cm², very few change is observed on the properties. This can be related to the simultaneous existence of empty sites and mixed-valence iron ions in the magnetite structure, which makes the rearrangement of ions after irradiation easier, while this hardly influences the magnetic parameters 1141.

2.2 Garnet ferrites irradiated by low energy ions and by fast neutrons

In 1971, it was discovered that the properties of bubble ferrimagnetic garnets could be improved by low energy ion implantation 171. Since then, the magnetic and crystallographic properties of ion-implanted garnets have been studied 181. In figure 5, we show an example of variations of lattice volume and magnetization of garnets induced by low energy ion Ne⁺ and H₂⁺, from ref.191, and we compare these variations to the case of fast neutron irradiation, from ref. 1101 and 1111. For both ion-implanted and fast-neutron irradiated garnets, a similar behavior is observed : there are an increase in the lattice volume and a decrease in magnetization, and the curves are of the same shape.

Figure 5: changes in volume, magnetization, at 300K vs dose

Figure 6: magnetization variations vs volume variations

In figure 6, we have replotted the magnetization variations as a function of the lattice volume variations for Ne⁺ ions, H₂⁺ ions and fast neutrons 1121. A striking concordance is observed in that a single law is obtained, which is characteristic of the garnet ionic structure after irradiation, irrespective of the nature and quantity of irradiating particles. The continuous line is the linear law corresponding to the model based on the transformation of Fe³⁺ into Fe²⁺ proposed in ref. 1131 to interpret the

properties of fast neutron irradiated $\text{Y}_3\text{Fe}_5\text{O}_{12}$. In order to confirm the existence of Fe^{2+} , M.O. Kerr experiments should be interesting. In conclusion, the analogy between the effects of low energy ion and fast neutron irradiation suggests that the induced ionic structure transformation is the same. Furthermore, note that a partial amorphization is also observed in fast neutron irradiated garnets.

3. HIGH ENERGY HEAVY ION IRRADIATIONS

With swift ($E_{\text{ion}} > 500$ MeV) heavy ions the ion-matter interaction is governed by inelastic collisions with the target electrons resulting in a collective excitation of the electron gas. Thus, the main parameters to consider during these high energy irradiations are the fluence, the electronic stopping power $(dE/dx)_e$ and the ion velocities. At these high energies, the ratio $(dE/dx)_e / (dE/dx)_n$ is larger than 1500 in most of the materials such that the irradiation fluence can be much smaller at high than at low energy, giving rise to individual defects in the wake of the ions.

3.1 $\text{Y}_3\text{Fe}_5\text{O}_{12}$ garnet irradiated by swift heavy ions 115-171

During the last decade, the garnet structure has been extensively studied in the field of irradiation effects induced by swift heavy ions, due to the existence of new heavy ion accelerators such as the GANIL in Caen (France). The heavy ions which have been used so far, are : Ar, Kr, Xe, Pb and U 118-231.

From crystalline point of view, let us consider the damage induced along the ion path. High Resolution Electron Microscopy (HRTEM) is a privileged tool to investigate the defects morphologies. As shown on figure 7 in the case of Xenon ions, swift heavy ions leave in their wakes cylinders of amorphous matter called tracks extending along the ion range (a few hundreds of nm). Depending on the incident ion masses and energies (velocities), and on the structure and physical properties of the irradiated compounds, the radius of the track as well as the homogeneity of the damaged core zone can change considerably. For instance, for the garnet YIG, a correlation (fig. 8) can be found between the variations of the effective track radii R_e , deduced from Mössbauer and magnetic measurements, versus $(dE/dx)_e$ and ion velocities, and the morphologies of the tracks observed by HRTEM. At very high fluences and low ion velocities where high $(dE/dx)_e$ can be obtained, a recrystallisation process has been observed in amorphized garnet films which induces the formation of nanograins (10 nm).

Figure 7 : latent tracks observed by HRTEM

Figure 8 : effective track radii and damage morphologies

From magnetic point of view, the heavy ion irradiation induces a specific anisotropy of the magnetic properties [24]. In most materials, the irradiation induces a specific volume increase of the matter. This increase may in turn generate radial stresses in the bulk surrounding the track core. In the case of weak magnetocrystalline anisotropy K_1 , the stress can generate changes in the orientation of the magnetization M_s through the magnetostriction coefficient λ_s . A complete rotation of M_s , that turns parallel to the ion path, has been observed in Mössbauer spectroscopy by vanishing of intermediate lines, both in the garnet YIG (fig. 9a) and in the spinel NiFe_2O_4 (fig. 9b), at low fluences, before the track overlapping.

3.2 Appearance of magnetization in the spinel ZnFe_2O_4 1251

Swift heavy ion irradiation (Kr, Xe) of ZnFe_2O_4 ceramics induce the creation of a strong ferrimagnetic behaviour as shown by Mössbauer spectroscopy (fig. 9c) and magnetization measurements whereas the initial material exhibits only an antiferromagnetic behaviour below $T_N = 10$ K. It is worth noting that the created magnetization is also oriented along the ion path direction (fig. 9c). This result can be understood only on the basis of cationic displacements on the tetrahedral and octahedral sites of the spinel structure induced by amorphisation-recrystallisation processes. Other spinels (NiFe_2O_4 , MgFe_2O_4), although already ferrimagnetic at 300K, exhibit also increases of the magnetization both after swift heavy ion irradiation as well as after neutron irradiation.

3.3 Trapping of magnetization in the spinel magnetite Fe_3O_4 1141 1261

The damage induced by swift heavy ion irradiation strongly depends on the material type, for the same (dE/dx) and ion velocities, some materials like MgO and Al_2O_3 show no damage, whereas garnets or quartz can be totally amorphized.

Figure 9 : Mössbauer spectra, at 300K, of polycrystals of : (a) $\text{Y}_3\text{Fe}_5\text{O}_{12}$; (b) NiFe_2O_4 ; (c) ZnFe_2O_4 .

Magnetite is a very resistant material since, after a high energy lead irradiation, only some spherical extended defects, surrounded by the dark contrast characteristic of a stressed domain, can be observed along the ion path. The core of these tracks remains crystallised with a structure close to the spinel one. In the stressed bulk material surrounding the damaged zones, the Ms orientation must depend on the sign of λ_s . Indeed, for magnetite, the sign of λ_s is reversed with respect to the one of the garnet, such that Ms should rotate perpendicularly to the ion path. This result has been observed on Mössbauer spectra [141] [261], but only for weak Pb ions fluences (0.8×10^{12} and 1.2×10^{12} Pb ions/cm²). Thus, the magnetic moments are trapped in the stressed domains and do not participate any longer to the resulting magnetization of the samples which decreases. Upon increasing the fluence, the extended defects start to overlap, removing the stressed domains and resulting in a relaxation of the trapped moments which induce an increase of the total magnetization [141] [261].

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