IRRADIATION EFFECTS IN LOW T_C SUPERCONDUCTORS

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Abstract

The effects of neutron irradiation on the superconducting parameters T_c, B_{c2} and J_c of Nb_3Sn are reviewed in view of the determination of the radiation limit in the LHC upgrade magnets. The variation of J_c in binary as well as in Ti and Ta alloyed Nb_3Sn wires is presented. The coexisting defect mechanisms in irradiated Nb_3Sn type compounds are briefly presented and a model is discussed explaining the site exchange mechanism which leads to a decrease of atomic ordering after irradiation.

Based on calculations of F. Cerutti and coworkers (CERN), the neutron fluence at the inner winding of the quadrupole Q2a is estimated to values below 10^{18} neutrons /cm^2 for a life time of 10 years, which is within the safety margin with respect to the critical current density and B_{c2}.

INTRODUCTION

The construction of future colliders with higher energies implies the use of superconducting dipoles and quadrupoles with increasing magnetic fields. As an example, the envisaged magnetic field in the LHC upgrade project will be as high as 15 T. At the present state of knowledge, such dipoles have to be wound on the basis of round wires, which limits the possible choice of industrially available superconducting materials to Nb_3Sn, Nb_3Al (both crystallizing in the A15 type structure) and Bi-2212. However, Bi-2212 wires still exhibit weak mechanical properties, while Nb_3Al can be produced in kilometer lengths, but is not yet available in large quantities. The present paper will thus only consider the effect of high energy irradiation on the low T_c superconductor Nb_3Sn.

The question arises whether the intense radiation produced by the collisions in the high LHC upgrade energy collider has an influence on the magnetic fields produced by the superconducting dipoles and quadrupoles. Of particular interest for the present paper are the Phase I and Phase II upgrades, with a luminosity of 2.5×10^{34} cm^{-2}s^{-1} and 10^{35} cm^{-2}s^{-1}, i.e. 2.5 and 10 times larger than nominal LHC, respectively. This question is treated based on recent calculations of the maximum neutron fluence (also called radiation dose in a number of other publications) acting on a well localized magnet location (the quadrupole). These calculations have been recently performed by F. Cerutti et al. [1] at CERN.

The present paper is a review about the known neutron irradiation data on Nb_3Sn wires, based on neutron irradiation experiments performed before 1986. After this date, irradiation works have been performed on HTS superconductors and more recently, on MgB_2 [2]. The mechanism of irradiation effects in A15 type compounds will be discussed, and a model explaining the observed changes of the superconducting properties is presented. The variation of T_c, B_{c2} and the electrical resistivity ρ_o of Nb_3Sn wires with radiation will be discussed, as well as the effects on the critical current density, J_c, with and without additives.

These data show that the effect of neutron radiation fluence on the critical current density of Nb_3Sn wires during 10 years, the estimated lifetime of the LHC upgrades, will not affect the critical current density of the Nb_3Sn wires. This is also valid for the quadrupole magnets Q2a, where the highest radiation fluence is expected [1] (see also Fig. 1).

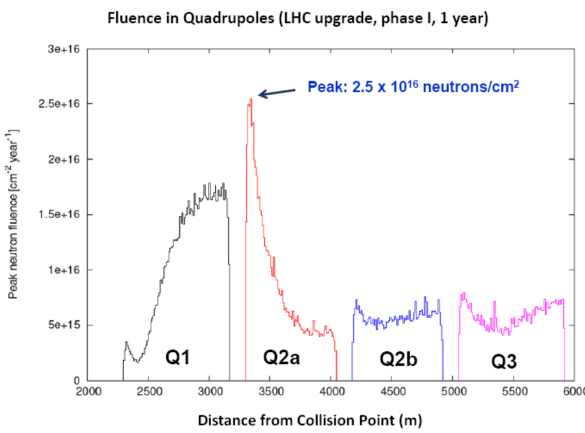

Figure 1: Calculated peak neutron fluence in phase I of LHC upgrade as a function of distance from the collision point, indicating a maximum for the inner winding of the quadrupole Q2a, as calculated by Cerutti et al. [1]).

THE NEUTRON RADIATION FLUENCE AT THE INNER WINDING OF LHC QUADRUPOLES

The collisions in a high energy collider produce a very

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intense radiation, its intensity increasing with energy. Neutron radiation has a strong influence on all the components of the collider, i.e. the superconductor, the stabilizer and the insulator. It is well-known that the superconducting properties of most superconductors depend on the radiation fluence, and the question arises whether significant radiation-induced effects (e.g. decreased values of T_c and of the critical field, increased resistivity, etc.) will lead to a reduced magnet performance within the time of operation, which may reach several years.

When calculating these effects, it is important to know the spectrum of the various radiations interacting with the magnets. At the collision site, a very strong radiation environment will be created. Estimates for the produced radiation at the collision site yield particle energies up to 10 GeV, the radiation being composed by charged pions, protons, neutrons, photons, electrons,…. [3]. It follows that adequate shielding is vital in order to reduce these radiations to an acceptable level. Taking into account the planned shielding conditions as well as the distance between the collision site and the magnets, the maximum radiation load acting on the magnets has been recently calculated by Cerutti et al. [1]. It was found that after interaction with the shielding material, the radiation spectrum reaching the closest magnets (the quadrupoles) will be composed by:

Photons	87 %
Neutrons	6 %
Electrons	3.5 %
Positrons	2.5 %
Pions (+/-)	0.4 %
Protons	0.15 %.

It has been shown that neutron radiation has a strong effect on the superconducting properties of Nb_3Sn, while the effect of photons, electrons and photons is negligible. Since the total amount of pions ad protons does not exceed 10% of the calculated intensity for neutrons, the present estimation will be done under the assumption that neutrons are the main source of damage to the superconductors. All other radiation sources will be neglected when considering the effect on the superconductor. Their effect on the insulator materials has to be analyzed separately and will not be treated in the present paper.

The calculations of Cerutti et al. [1] show that the neutron energy has a pronounced peak around 1 MeV. The neutron energies used in the previous works studying the effects of radiation damage on the superconducting properties of superconductors vary between 1 and 14 MeV (Sweedler et al. [4]) and 14 MeV (Weber et al. [5], Guinan et al. [6] and Weiss et al. [7]). A comparison between the various known data shows that within this energy range, one does not expect a strong difference between the effects of neutron radiation on the superconducting properties. As shown in Fig. 1, the highest fluence will be reached at the inner winding of the quadrupole Q2a, at a distance of 33 m from the collision point. The expected maximum radiation fluence was calculated to 2×10^{16} n/cm^2 per year, assuming an operation of 200 days a year. For a total operation time of 10 years, the maximum fluences at the inner winding of the quadrupole Q2a after the upgrades of Phase I and Phase II will thus be 2.5×10^{17} n/cm^2 and 10^{18} n/cm^2, respectively. In Section VI, the effect of these fluences on T_c and J_c will be analyzed for both, binary and alloyed Nb_3Sn wires. An explanation for the higher sensitivity to radiation damage for alloyed Nb_3Sn wires with respect to binary wires will be discussed.

THE EFFECTS OF NEUTRON IRRADIATION ON SUPERCONDUCTORS

Effect on T_c of various superconducting compounds

As mentioned above, the main damage at the magnet site in LHC upgrade phase I is expected to be caused by neutron irradiation. The overwhelming amount of neutron irradiation data of Low T_c (or LTS) superconductors has been performed prior to 1987, after which most work was performed on High T_c (or HTS) compounds. In Fig. 2, the variation of T_c vs. ϕt is shown for the most known Low T_c superconductors: Laves phases (HfV_2) [9], Chevrel phases ($PbMo_6S_8$) [10], MgB_2 [2]. T_c for NbTi [8] is not shown here, but is even less radiation sensitive than V_2Hf.

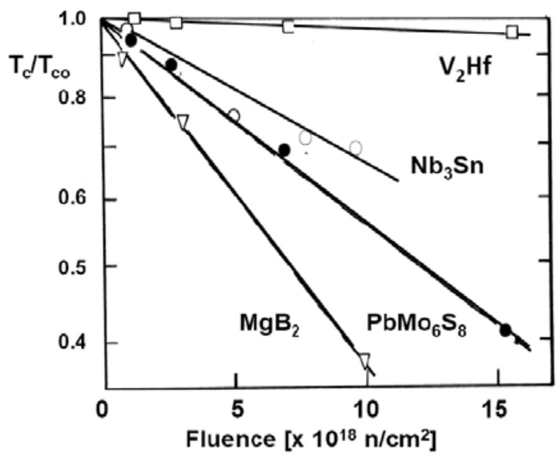

Figure 2: Variation of T_c vs. neutron fluence, ϕt, for various known low T_c superconductors. Laves phases (HfV_2) [9], Chevrel phases ($PbMo_6S_8$) [10], Nb_3Sn [4], MgB_2 [2].

Although NbTi and HfV_2 are little affected by neutron radiation, these materials must be excluded due to their too low H_{c2} values. Chevrel phases exhibit very high H_{c2}, but never reached the industrial level. In addition, they exhibit a relatively low T_c value (14 K in wires) and astronger radiation sensitivity with respect to Nb_3Sn. MgB_2 has also a stronger sensitivity to neutron irradiation than Nb_3Sn, but the value of B_{irr} is not high enough for magnets producing 15 T.

The data for the HTS compounds Bi-2212, Bi-2223 and Y-123 are not shown here, but it must be noted that even if they show a relatively strong decrease of T_c with neutron fluence, this would be of little importance for operation at 4.2 K. Indeed, T_c, J_c and B_{c2} would still be sufficiently high for this temperature. As mentioned above, mechanical stability for Bi-2212 and the tape geometry for Bi-2223 and Y-123 are at present important obstacles excluding their use in collider magnets.

Radiation effects on T_c of several A15 type compounds

It appears from Fig. 2 that Nb_3Sn is well suited for being used in LHC upgrade, no significant decrease of T_c being observed up to radiation fluences up to 10^{18} n/cm^2. The decrease of T_c in various A15 type compounds as a function of the irradiation fluence, ϕt, for neutrons with $E \leq 1$ MeV at irradiation temperatures $T_{irr} \leq 150$°C is represented in Fig. 3. The data on V_3Si, Nb_3Ge, Nb_3Al, Nb_3Pt, Nb_3Ga and Mo_3Os are extracted from Sweedler et al. [4,14], while V_3Ga was analyzed by Francavilla et al. [15] and $Mo_{.40}Tc_{.60}$ by Giorgi et al. [16]. It is remarkable that the variation of T_c with ϕt is very similar for V_3Si, Nb_3Ge, Nb_3Al, Nb_3Pt, Nb_3Ga and V_3Ga, in contrast to Mo_3Os and $Mo_{.40}Tc_{.60}$, where the decrease of T_c is considerably smaller.

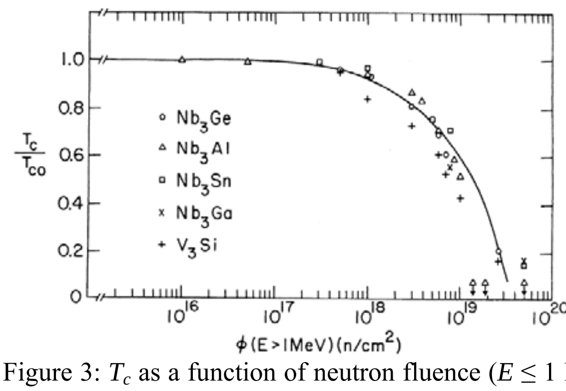

Figure 3: T_c as a function of neutron fluence ($E \leq 1$ MeV, $T_{irr} \leq 150$°C) for various A15 type compounds (all data from Sweedler et al. [4]).

From X ray diffractometry, Sweedler et al. [4,14] concluded that the decrease of T_c at doses up to 10^{19} n/cm^2 (where $T_c/T_{co} \sim 0.5$) is mainly caused by a decrease of the long-range atomic order parameter, S.

From X ray diffractometry, Sweedler et al. [4,14] established that the decrease of T_c at doses up to 10^{19} n/cm^2 (where $T_c/T_{co} \sim 0.5$) is mainly caused by a decrease of the long-range atomic order parameter. S. Pande [17] proposed another explanation for the observed correlation between radiation dose and T_c, based on TEM observations, revealing the presence of depleted zones in irradiated samples. As previously shown in an unpublished Internal Report (Flükiger [18]), calorimetric measurements constitute a definitive proof for the validity of the "disordering" model proposed by Sweedler et al. [4,14]. This will be discussed in detail in the following.

Irradiation temperature

At 1.8 K, the operation temperature of LHC and LHC upgrades, vacancies show any mobility, and site exchanges can only occur by the energy of transmitted by the multiple collisions. An increase of the irradiation temperature, T_{irr}, will lead to self-annealing effects. There is no complete series of measurements on irradiations at T < 10 K and T > 300K, and a comparison can only be made using the data of different authors, who worked at with neutrons of different energies, between 0.1 and 14 MeV. The conversion from 0.1 to 14 MeV is problematic, and several approaches to find appropriate conversion factors have given unsatisfactory results. For this reason, the nominal fluences will be used for the present comparison, well knowing that a certain error is introduced.

Söll et al. [19] performed neutron irradiations (E > 0.1 MeV) of Nb_3Sn diffusion wires at 4.6 K and moved the samples from the irradiation position to the test cryostat without warmup. The irradiation was performed up to a fluence of 3.9×10^{18} n/cm^2, after which a T_c reduction of 0.8 K was observed. After irradiation at >150 °C (in reality, the temperature before irradiation was 300 K) to 10^{18}/cm^2 (E>1 MeV), Sweedler et al. [4] reported a similar decrease in T_c, which indicates that changes in superconducting properties of Nb_3Sn after irradiation at > 150°C may be similar to effects of 4.6 K irradiation. This was later roughly confirmed by J_c measurements by Hahn et al. [20]. Söll et al. [21] extended their cryogenic irradiations to a fast neutron fluence of 10^{19} n/cm^2 (E>0.1 MeV) at $T_{irr} \sim 10$ K. A slight extrapolation of their results reveals that a fluence of ~ 1.2×10^{19} n/cm^2 would be required to reduce T_c to 13.5 K, the value obtained by Sweedler et al. [4] at >150 °C after a higher fluence of 5×10^{18} n/cm^2 (E>1 MeV). Again, this confirms that there is a small, but not negligible difference between the decrease of T_c after irradiation at ~ 10 K and at > 150 °C. It follows that there is only a weak recovery from 4.2 K to room temperature.

Meier-Hirmer and Küpfer [22] irradiated V_3Si single crystals with fast neutrons at 240 °C. As shown in Fig. 4, the decrease of T_c with increasing fluence ϕt is substantially reduced for T_{irr} = 240 °C [22] when compared with data taken at $T_{irr} \leq 150$°C by Sweedler et al. [23]. The observed lower decrease of T_c after heavy irradiation fluences with higher T_{irr} values has its origin in thermal recombination effects during irradiation. The data in Fig. 4 show that the activation of vacancies at between

$T_{irr} \leq 150$ and 240 °C has the effect of an annealing which increases the long-range order parameter of the crystal.

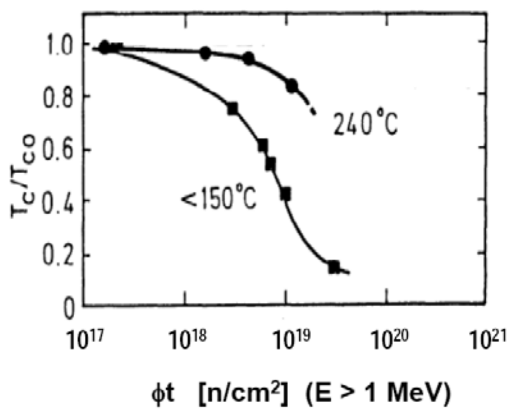

Figure 4: T_c of V_3Si as a function of fast neutron irradiation fluence ϕt for various irradiation temperatures. $T_{irr} \leq 150$°C (Sweedler et al. [23], $T_{irr} = 240$°C (Küpfer et al. [22]).

COEXISTING DEFECT MECHANISMS IN IRRADIATED A15 TYPE COMPOUNDS

From the wealth of published data, it follows that the decrease of T_c in irradiated A15 type compounds has been attributed to three competing defect mechanisms. These mechanisms are analyzed in the following.

Homogeneous Defects (decrease of long-range atomic ordering)

The "defects" are here lattice sites occupied by the wrong atoms (in analogy to the quenched state), also called "antisite defects". A decrease of the long-range order parameter S (defined by van Reuth et al. [24] in irradiated A15 compounds was first observed by Sweedler and Cox [23] in Nb_3Al. It was later confirmed by Moehlecke et al. [25] for Nb_3Pt and by Cox and Tarvin [26] for V_3Si, while Schneider et al. [27] reported a decrease of S in Nb_3Al after irradiation with 700 keV N^{2+} ions. It is important to note that the attribution of the observed effects on the superconducting properties of A15 type compounds after irradiation to a change of atomic ordering implies the assumption of *homogeneous* disordering over the sample volume. The decrease of T_c is explained by changes of the electronic density of states at the Fermi level.

Inhomogeneous defects (disordered microregions or "depleted zones")

The defects are represented by the depleted zones of 4 nm diameter, generally observed by means of TEM microscopy in solids after high energy irradiation [17]. The depleted zones can be either highly disordered or amorphous, and were also called "disordered microregions" by Pande [17, 24]. The presence of inhomogeneities was also observed by Nikulin et al. [29], by means of small angle neutron scattering. The inhomogeneous defect mechanism [17] assumes that the matrix enclosing the depleted zones is essentially unaffected by the radiation, in contrast to the antisite defect mechanism (disordering) [14,18], which is based on a homogeneous decrease of the atomic order parameter over the whole sample volume.

As pointed out by Pande [17], the weakening of A15 superstructure lines after irradiation, (which is interpreted as a decrease of S [14,18], could also arise from an increasing volume fraction of disordered microregions (also called depleted zones), concentrated in regions of ~ 5 nm diameter. The inhomogeneous defect mechanism explains an overall degradation of the super-conducting properties by the proximity effect [17] between the ordered, high T_c matrix and the disordered low T_c micro-regions of a size comparable to the coherence length, ξ_o ~ 4 nm.

Static Displacement of the Atoms from their Equilibrium Positions

This kind of defect was first observed by Meyer [30] and Testardi et al. [31] after heavy irradiation of V_3Si single crystals by means of the channeling technique. The existence of all three defect types α, β and γ is based on sound experimental data. It is also ascertained that all three types of defects occur simultaneously during irradiation. In the past, each of these defects has been independently made responsible for the decrease in T_c. Depending on the total radiation fluence, each one of these defect types will be dominant over the concurrents. As will be shown in the following, antisite defects (or decreasing atomic ordering) are mainly responsible for the observed decrease of T_c for radiation doses up to ~ 10^{19} n/cm^2, while inhomogeneous defects and static displacement are dominant for higher doses, where the value of T_c is too low for being of interest for the present considerations. The effects of high energy radiation on A15 type compounds are complex and are schematically represented in Fig. 5.

Volume change after irradiation

The number of vacancies in unirradiated A15 type compounds at 300K is generally below 2 x 10^{-3}, the experimental error limit. This is in agreement with other dense structures, for which vacancy concentrations of the same order of magnitude have been reported. The only case where a measurable amount of vacancies could be produced in A15 crystals was reported by Cox and Tarvin [26], who found that the density of a V_3Si single crystal after a fast neutron irradiation dose of 22.2×10^{18} n/cm^2 decreased by -0.3%. This observation allows the estimation whether a lattice expansion of Nb_3Sn filaments after irradiation has to be taken into account for LHC upgrade II.

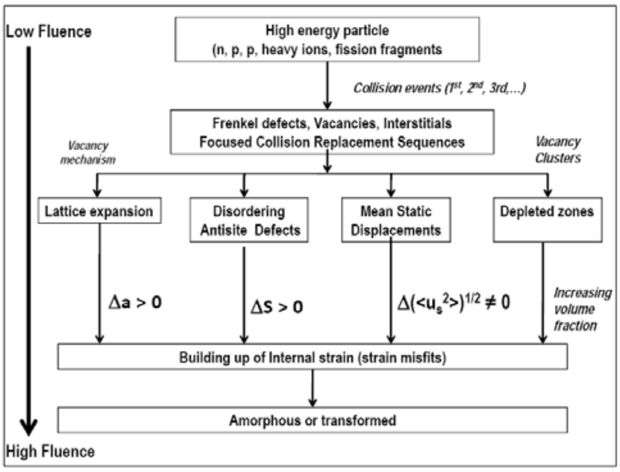

Figure 5: Schematic representation of the radiation induced effects in A15 type compounds, with increasing fluence (R. Flükiger [18]).

Assuming a total fluence of 10^{18} n/cm^2, on the quadrupole Q2a after 10 years of operation, the decrease of density inside the Nb_3Sn filaments is estimated to -0.03%. This would correspond to a volume expansion of + 0.03%, and thus of a linear expansion of ~ 0.01%, thus creating an additional stress acting on the filaments. A comparison with the effect of uniaxial stress or compressive transverse stress on J_c of Nb_3Sn wires shows that an increase of stress by 0.01 % as a consequence of irradiation would cause a small, but non negligible effect on J_c. Additional precise irradiation measurements should be performed on multifilamentary Nb_3Sn wires samples, not only as single wires but in the same configuration as in the cable, comprising all reinforcing and insulating elements. This rough estimation shows that that the additional stress effect due to radiation damage on J_c after 10 years of operation of LHC upgrade phase II may be below 5%.

CORRELATION BETWEEN ANTISITE DEFECTS AND T_C AFTER NEUTRON IRRADIATION

Several arguments can be invoked for proving the validity of antisite disorder as the dominant mechanism influencing the variation of T_c vs. ϕt for radiation doses up to ~ 10^{19} n/cm^2:

- Homogeneous distribution of the antisite defects over the whole sample after irradiation, detected by specific heat measurements;
- Comparison between the variation of T_c vs. the order parameter S for A15 type compounds after irradiation and after fast quenching from high temperatures (in both cases, S is measured by diffraction methods);
- Irradiation with high energy electrons and neutrons.

Homogeneous distribution

The question about a *homogeneous* distribution of the damage can be answered on the basis of the known low temperature specific heat measurements before and after irradiation for the A15 type compounds Nb_3Sn [32], Nb_3Al [33] and V_3Si[34].

The measurements of Cort et al. [33] for Nb_3Al are represented in Fig. 6, showing the specific heat jump at T_c after irradiation at 150°C at a fluence of 13×10^{18} n/cm^2. The transition width was found to increase from 0.8 to 1.3 K, for a significant decrease of T_c from 18 to 10K. The compound V_3Si was studied by calorimetry by Viswanathan et al. [34] after neutron irradiation at 200°C at a fluence of 22.2×10^{18} n/cm^2. A decrease of T_c from 17 to 7.5 K was found, the width ΔT_c increasing slightly from 0.5 to 1.2 K. The specific heat measurements of Karkin et al. [32] on Nb_3Sn before and after neutron irradiation are shown in Fig. 7. These authors studied the effect of neutron irradiation at 80°C in Nb_3Sn. At a dose of 10^{19}n/cm^2 they found a T_c value of 12.5 K, while the transition width ΔT_c measured by specific heat increased from 0.8 to 2.5 K.

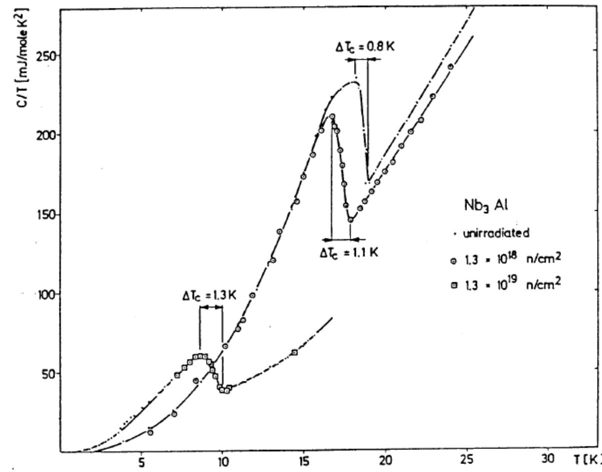

Figure 6: Specific heat of Nb_3Al before irradiation and after neutron doses of 1.3×10^{18} and 1.3×10^{19} n/cm^2, respectively. The linear representation C/T vs. T has been chosen in order to visualize the moderate increase in ΔT_c with dose (after Cort et al. [33]).

Table 1. Specific heat data of the compounds Nb_3Sn, V_3Si and Nb_3Al after neutron irradiation. Indicated are the T_c values before and after irradiation as well as the transition width ΔT_c.

Compound	Mass (g)	T_{co} (K)	T_{irr}(°C)	Fluence	T_c (K)	T_c/T_{co}	ΔT_c	
Nb_3Sn	11.0	17.9	80	10 x 10^{19}	12.5	0.69	2.5	[28]
Nb_3Al	0.2	18.7	150	13 x 10^{19}	9.6	0.51	0.5	[29]
V_3Si	1.5	17.1	200	22 x 10^{19}	7.5	0.44	0.7	[30]

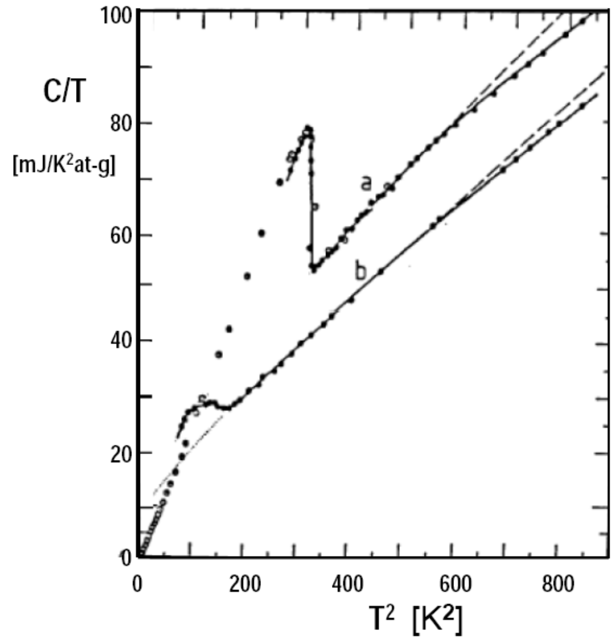

Figure 7: Specific heat of Nb_3Sn, a) in the unirradiated state and b) after a dose of 10^{19} n/cm^2 according to Karkin et al. [32]. For unirradiated Nb_3Sn at lower temperatures, data of Junod et al. [35] have been superposed.

When comparing the transition width of superconducting compounds after irradiation, one has to take into account the nuclear heat recovery, which depends on the size of the sample. Viswanathan et al. [34] pointed out that V_3Si single crystals with masses of ≥ 1 g exhibited additional heating during irradiation, leading in one case even to a visible darkening of the surface, i.e. to estimated temperatures T_{irr} of 300°C and above. This is due to increasing difficulties in transferring the nuclear heat to the sample environment (Argon gas) with increasing volume to surface The masses of V_3Si and Nb_3Al were 1.5 and ~1 g, respectively, in contrast to Nb_3Sn, which had 11 g [32]. The latter had a T_c value of 12.5 K, which is markedly higher than 9.9 K, the value estimated from the behavior of T_c vs. ϕt for small samples in Fig. 3.

The corresponding values for Nb_3Al and V_3Si are 9.6 and 7.6 K, which is in good agreement with the values reported in Table 1. It can thus be estimated that the increase of T_c due to nuclear heat recovery at ϕt=10^{19} n/cm^2 for this "heavy" Nb_3Sn samples is ~ 1.5 K. It follows that for the 3 systems Nb_3Sn, Nb_3Al and V_3Si, heavy neutron irradiations up to fluences of 2×10^{19}n/cm^2 only causes a very small enhancement of the transition width, ΔT_c. It is interesting that for a neutron fluence reducing T_c to about 50% of the initial value, the transition width ΔT_c is only enhanced by ≤ 1K.

In the specific heat curves, there is no trace of the original superconducting transition temperature, T_{co}, in any of the 3 systems Nb_3Sn, Nb_3Al and V_3Si after irradiation. This implies that the amount of depleted zones (with considerably lower T_c) is very small and cannot account for the observed decrease of the superstructure lines. After having shown that the damage is homogeneous, this is a strong argument in favour of the antisite defect mechanism as the dominant one up to 10^{19} n/cm^2.

Comparison between irradiated and quenched A15 type compounds

Quench induced disordering constitutes a particularly simple case, where the only possible defects are the antisite defects (up to a few %) and quenched-in vacancies (≤ 0.02% at 300K). No direct comparison between neutron irradiation and quenching has been performed on Nb_3Sn and the data reported here concern the system Nb_3Pt, another A15 type compound.

Fig. 8 shows the variation of T_c for Nb_3Pt vs. the long-range order parameter, S, after fast quenching from temperatures up to 1900 °C by Flükiger et al. [36] and after from neutron irradiation (E>1 MeV) by Moehlecke et al. [37].

It follows from Fig. 8 that the variation of T_c vs. S is the same, regardless whether the compound was fast quenched or irradiated. This constitutes an additional argument in favour of variations of the degree of atomic for the observed decrease of T_c after irradiation up to fluences of 10^{19} n/cm^2.

Irradiation with High Energy Electrons and Neutrons

According to Seeger [38], the complex situation in an irradiated crystal can be described as follows. The high energy particle transfers a kinetic energy, E_T, to an atom of the target, the primary knock-on atom. At sufficiently high values of E_T, this atom will be removed from its equilibrium lattice site: a Frenkel defect is formed. Due to inelastic collisions with electrons, the energy of the primary knock-on atom towards the end of its path falls to values of the order of E, the displacement energy (~ 25 eV), and almost every collided atom is displaced [39].

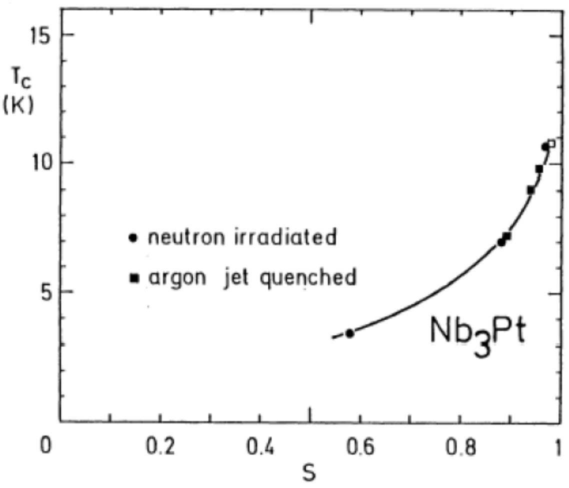

Figure 8; T_c vs. long-range order parameter S (determined by X ray diffraction) for Nb_3Pt after fast quenching from different temperatures [36] and after neutron irradiation [37].

As a result of this displacement cascade, regions with high concentrations of vacancies are formed, called "depleted zones" by Seeger [38], which are surrounded by a zone enriched with interstitial atoms. In A15 type compounds, the existence of depleted zones has first been reported by Pande [17,24], who called them "disordered microregions". In reality, the structure of this region can be either amorphous, of the A15 type (but strongly disordered), or of another structure type, as for example bcc (A2 type) in Nb_3Al [40] or in $Nb_{\sim 3}Si$ [41]. The term "disordered zone" seems thus to be more appropriated and will be adopted in the present work.

A further strong argument in favour of ordering effects was given by Ghosh et al. [42] and Rullier-Albenque et al. [43]. These authors used the fact that electrons with energies of the order of 1 MeV do not produce the displacement cascade (or disordered zones) described above, in contrast to irradiation with fast neutrons or high energy ions. Due to their low mass, electrons of 1 MeV create only isolated Frenkel defects (Schulson [44]). Rullier-Albenque et al. [43] have irradiated Nb_3Ge films (with T_{co} = 19.5 K) with 1 MeV neutrons and with 2.5 MeV electrons, respectively, while Ghosh et al. [42] performed an analogous experiments on Nb_3Sn films, which were irradiated with 2 MeV He ions and with 2 MeV electrons. Both investigators came to the same conclusion, i.e. a strong decrease of T_c is observed, regardless of the nature of the projectile. In particular, it was found that the ratio $\Delta T_c/\Delta\rho_{irr}$ (where ρ_{irr} is the increase of residual resistivity after irradiation) for Nb_3Ge does not differ between fast neutron and high energy electron irradiation. For $\Delta\rho_{irr}$ values between 0.1 and 10 μΩ·cm, the ratio $\Delta T_c/\Delta\rho_{irr}$ remained almost unchanged, close to 0.1 K [43]. Although these experiments do not give a direct evidence that the order parameter is the same in both cases, they lead to an important conclusion: the presence of "disordered zones" is not necessary to cause a decrease of T_c. This result is analogous to that of fast quenching experiments, and furnishes an additional proof for atomic ordering effects as the dominant effect causing the decrease of T_c after neutron irradiation.

THE SITE EXCHANGE MECHANISM IN IRRADIATED A15 TYPE COMPOUNDS

Vacancy diffusion: the "virtual" lattice site (or split vacancy)

In the tightly packed A15 structure, each atom is closely surrounded by its neighbors. The atoms would need to be considerably compressed before any two could squeeze past one another and interchange positions as required for ordering changes. Among different possible diffusion mechanisms, vacancy diffusion is the most probable one. It is based on the fact that at thermal equilibrium each solid at a temperature above zero contains a certain number of vacant lattice sites. An atom will now jump into a neighbouring vacancy, thus creating a new vacant site. Atoms and vacancies undergo a series of position exchanges, in order that a very small number of vacant lattice sites is sufficient to induce a substantial diffusion. Most studies dealing with diffusion have been undertaken on bcc or fcc structures. In the A15 structure, however, the situation is more complex, the *6c* and *2a* sites being not equivalent from the point of view of electronic bonding. Indeed, interactions beween atoms on the *6c* and on the *2a* sites are characterized by metallic bonding, white the intrachain bonding between two A nearest neighbors is of the covalent type, as was shown by Staudenmann [45], who established electron density maps of V_3Si. This covalent bond is correlated with the very short AA distances on the chains of A15 type compounds, which are noticeably shorter than the sum of the atomic radii of the A element. This configuration leads to highly nonspherical shapes for the A atoms. The region of covalent bonding between two A atoms on *6c* sites will be called "overlapping region" in the following. This region corresponds to a high electron density in V-Si [45].

The question arises whether this overlapping (or covalent bonding) between two A neighbors on the chain sites still resides if one of them is next to a *6c* vacancy. Welch et al. [46,47] have shown by means of pair potential calculation that such an individual vacancy of an A atom on a 6c site is unstable. They found that the state of lower energy corresponds to a configuration where one of the two A atoms adjacent to the single *6c* vacancy is shifted towards a new site which is equidistant from the next two A neighbors (see Fig. 9). This particular type of "split vacancy" [46,47], located in the overlapping region corresponds to a nonequilibrium state and will thus in the following be called "virtual" lattice site [48].

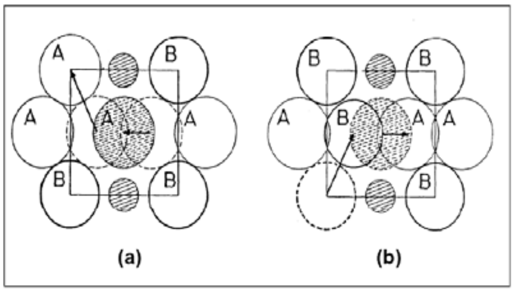

Figure 9: A ←→B exchanges by vacancy diffusion in A15 type compounds A_3B. a) Jump of an A atom into a B vacancy, followed by the occupation of the "virtual" site by the neighbour A atom, b) Jump of a B atom into an equilibrium A site (two-step processes). Small circles: overlapping region between two A atoms or region of covalent bonding (Flükiger [18,48]).

It thus appears that the vacancy diffusion mechanism in the A15 type structure comprises two steps, in contrast to the one-step mechanism acting in simple bcc or fee structures. Since the calculations of Welch et al. [46,47] show that the mobility of *6c* vacancies is quite high, the author has used the “virtual site “ concept as a basis for a mechanism of homogeneous disordering in irradiated A15 tape compounds [18,48].

Disordering by Focused Replacement Collision Sequences

In spite of the fact that the observations in Section V strongly support a homogeneous decrease of the long-range order parameter over the whole sample volume in irradiated A15 type compounds, a question arises: How is it possible that a homogeneous decrease of S implying site exchanges over several lattice spacings can occur during irradiations at temperatures $T \leq 100°C$, where little thermal diffusion takes place?

Under these conditions, disordering in irradiated A15 type compounds can be caused either by a) local cascade replacements or b) focused replacement collision sequences, a superposition of both being even more probable.

Local Cascade Replacements

Interaction between high energy incident particles and the lattice atoms produces a cascade, i.e. the atoms are displaced to produce Frenkel defects in a three-dimensional zone, the disordered (or depleted) zone [38,39]. Simultaneously, an even larger number of atoms change their mutual positions (i.e. they are replaced), thus producing locally a decrease of the degree of ordering. In principle, it could be argued that the atomic replacements produced in the cascade would be sufficient to produce disorder in the whole A15 lattice.

However, this argument cannot be generalized, since electron irradiation of A15 type compounds causes the same effect on the superconducting transition temperature, T_c, and the electrical resistivity [43] but produces only isolated Frenkel defects rather than depleted zones (or cascades). Thus, an additional mechanism must be effective in causing a homogeneous decrease of the order parameter. Such a mechanism, requiring atomic transport over several interatomic distances (the condition for a collective phenomenon) is constituted by the focused replacement collision sequences.

Focused Replacement Collision Sequences.

The so-called focusing replacement collision sequences, introduced by Seeger [38,39] represent the transport of matter and energy in irradiated crystals along dense crystallographic directions. Regardless of the crystal structure, an interstitial atom produced by irradiation processes is transported several interatomic distances away from its associated Frenkel vacancy, along these "focalizing" crystallographic directions. Focused replacement collision sequences can occur at the external boundaries or depleted zones with sizes of 4 to 5 nm for neutron irradiation [17,24] and up to 15 nm for irradiation with ^{32}S ions [50]. At these boundaries, highly disordered regions are adjacent to the matrix, and a sufficient but still small number of virtual sites is thus occupied, a necessary condition for the occurrence of focused replacement collision sequences (it may be recalled that this number is still very small, < 0.1 %). As irradiation further proceeds, the number of occupied virtual sites will increase, thus leading to an increase of site exchange processes. At the same time, thermal reordering will occur, as follows from the recombination theory of Liou and Wilkes [51].

It can be immediately seen that in the unirradiated state the replacement collision sequences in A15 type compounds occurring in the <100> and <111> directions, i.e. along the chains and the diagonal of the cube, are not effective in producing A ←→ B site exchanges. The only focusing direction where collision sequences could in principle produce A ←→ B site exchanges is the <102> direction. However, the atomic sequence in the <102> direction is oABAoABAo, the space between two ABA sequences coinciding with the region of overlap of two A atoms on the chain being perpendicular to the {100} plane. It can be easily shown that this configuration excludes extended site exchanges. Indeed, the potential encountered by A or B atoms on their way along the <102> direction, calculated using a Born-Mayer interatomic potential [52]shows that $V_{A\rightarrow B}$ and $V_{B\rightarrow A}$ are of the same order of magnitude, ~ 10 eV, while $V_{A\rightarrow A}$ reaches 52 eV.

The situation in irradiated crystals is, however, quite different from that encountered prior to irradiation. With the occupation of the virtual sites (a very small number of occupied non-equi1ibrium sites represented in Fig. 9 is sufficient), the potential $V_{A\rightarrow A}$ is considerably lowered. The new sequence of atoms in the focalizing <102> direction around the occupied virtual site is now oABAAABAoABAo or oABABABA0ABAo, depending on the occupation of the virtual site by an A or a B atom, respectively (see Fig. 10).

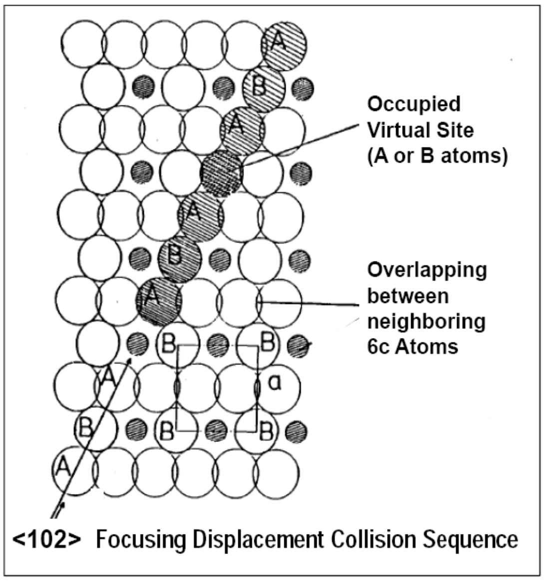

Figure 10: Focusing replacement collision sequences in A15 type compounds. Representation of the {100} plane of the A15 lattice (the radii are scaled for Nb_3Al). The small circles correspond to the overlapping region between two A atoms belonging to the chains perpendicular to the {100} plane. The occupation of the virtual site by an A or a B atom leads to the sequences oABAAABAo or oABABABAo, respectively instead of oABAoABAo as in the unirradiated case, thus enabling A←→B site exchanges in the <102> direction, a is the lattice parameter (Flükiger [18,48]).

As mentioned above, a very small amount of vacancies (or occupied virtual sites) is sufficient for the occurrence of the present mechanism. It should be recalled that for example, density measurements have revealed ~ 0.3 % of lattice vacancies in neutron irradiated V_3Si after a fluence of 22.2×10^{18} n/cm^2, corresponding to a decrease of T_c to a value less than one half of the initial T_c value. Due to the very large number of replacement collisions over the whole crystal volume during the whole irradiation time, the virtual sites will be many times occupied and abandoned again, alternatively by A or B atoms, respectively. The latter constitute a "bridge" between neighbouring ABA sequences, thus allowing A ←→ B exchanges over several interatomic distances. This is a necessary condition for a homogeneous decrease of the degree of ordering over the whole crystal after irradiation at low temperatures.

A very important conclusion can be drawn from a consideration of the close neighborhood of an occupied "virtual" lattice site. Its nearest neighbors, 6 A and 2 B atoms, are in close contact with the interstitial, which is either an A or a B atom. In the {100} plane, this leads to a hexagonal arrangement, as shown in Ref. 18. If the interstitial atom is of the A type, the occupation of the virtual site leads to 4 additional AA contacts, with AA distances of 5/16a = 0.559 a, thus 6% more than the AA distances in the chains, a/2. In addition, there are 2 AB contacts with interatomic distances of a/2 compared to 9/16a in the unirradiated A15 lattice, .i.e. 6% shorter.

This situation can be compared to that in quench disordered A15 type compounds, where the AB distances are the same, i.e. a/2. The case of a B element occupying the virtual site with 4 AB and 2 BB close contacts is more interesting. The AB distances are now 5/16a, i.e. they are still the same as in the unirradiated A15 structure, but the BB distances are now 36% shorter (a/2 with respect to 3a/2). This would mean that in each case, BB distances shorter than the sum of two B radii would be encountered. Such an "overlapping" between B elements is only possible in the nonequilibrium situation caused by low temperature irradiations or in a dynamic situation during site exchanges at high temperature. It is particularly interesting to consider the case of nontransition B elements, where such close BB contacts are expected to cause strong electrostatic repulsive forces. The occurrence of such BBB sequences furnishes the key for understanding the causes of the lattice expansion and the static displacements observed in irradiated A15 type compounds (see Fig. 7).

From these remarks, it can be recognized that the diffusion in the A15 type structure is not a single vacancy diffusion as suggested by Sweedler et al. [23] nor it corresponds to a so-called interstitialcy diffusion mechanism, where the atom diffuses from a normal site to an interstitial site. In the case of the A15 structure, the situation is complicated by the fact that a jump into the virtual site (which is an interstitial site) requires simultaneously a rearrangement of the other atoms.

NEUTRON IRRADIATION AND ITS EFFECTS ON J_C AND B_{C2} OF MULTI-FILAMENTARY NB_3SN WIRES

There are only two investigations on alloyed Nb_3Sn wires, by Weber et al. [5] (E>0.1 MeV) and by Weiss et al. [7] (E=14 MeV). The latter compared the behavior of binary and alloyed Nb_3Sn wires at T_{irr} = 300K and performed J_c vs. B measurements at higher fields (between 14 and 20 T).

Variation of T_c

As shown in Fig. 11, the variation of T_c vs. ϕt at low fluences is very similar for binary and alloyed wires: at 10^{18} n/cm^2 the T_c reduction is close to 4% for fluences up to ~ 20% at 2×10^{18} n/cm^2 [7]. At the highest fluences in Ref. 7, i.e. 4×10^{18} n/cm^2, ΔT varies between – 2.3 and – 2.6 K for the binary wires. The variation ΔT for alloyed wires is slightly higher: - 2.9 K for Ti additions and – 3.3 K for Ta additions. The slightly larger decrease for Ta additions may be caused by the considerably larger amount of Ta (3.5 at.%) in the filaments with respect to Ti (~ 1 at.%). As recently shown, the fact that Ta and Ti occupy different lattice sites prior to irradiation (Ta on Nb sites, Ti on Sn sites) may give an explanation for the observed difference [53]. As will be shown in the following, a considerably larger difference is observed when comparing the variation of J_c with fluence.

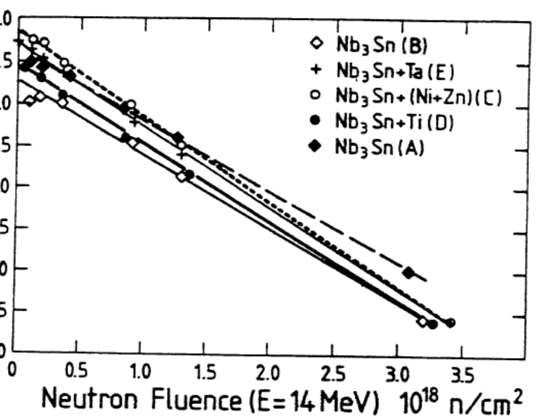

Figure 11: T_c vs. fluence ϕt for binary and alloyed Nb_3Sn multifilamentary wires after irradiation with 14.8 MeV neutrons at 300 K. Wire A: binary, 10'000 filaments, wires B, C, D and E: 19 filaments (Weiss et al. [7]).

Binary multifilamentary Nb_3Sn wires

In view of the use of multifilamentary alloyed Nb_3Sn wires in the LHC upgrade collider, it is interesting to know the effects of the expected neutron radiation on J_c at 15 T at fluences up to 10^{19}n/cm^2. As mentioned in Section V, fast neutron irradiation of Nb_3Sn causes a decrease of the long-range order parameter, S, which causes the decrease of T_c.

A second consequence of the decrease of S is the enhancement of the electrical resistivity ρ_o, which is correlated to the enhancement of the upper critical field, B_{c2}. At low fluences, this leads to a situation where T_c decreases, but B_{c2} increases. This means that at a given magnetic field, the initial values of J_c also increase: all known investigations report a maximum of J_c. At higher fluences, the decrease of T_c becomes the dominant effect, and B_{c2} decreases again.

An example for the binary multifilamentary wire [7] is shown in Fig. 12. The maximum of J_c/J_{co} and B_{c2} is reached at 0.8×10^{18} n/cm^2. The increase of the ratio J_c/J_{co} at the maximum fluence is strongest for the highest fields and reaches ~ 2.1 at 15 T and ~ 3.5 at 20 T.

As mentioned before, a certain difference is observed between the fluence values given in different papers. This is illustrated by a comparison with a publication of Weber et al. [5], where the same maximum of J_c/J_{co} ~ 2 was reached after neutron irradiation at 300 K for a fluence of 1.1×10^{19} n/cm^2 (E>0.1 MeV). It follows that the conversion factor between 14 MeV and > 0.1 MeV is close to 1.4. The maximum of B_{c2}* was reached at the same ratio J_c/J_{co}, the initial value of 21.4 T being enhanced to ~ 22.5 T [7].

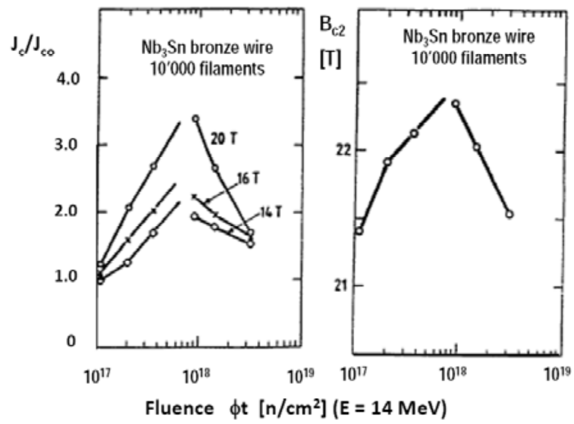

Figure 12: J_c/J_{co} and B_{c2}* of a 10000 filament binary Bronze Route Nb_3Sn wire after neutron irradiation at 300K (E=14 MeV). B_{c2}* is the critical field at 4.2 K determined by the Kramer extrapolation (Weiss et al. [7]).

Alloyed multifilamentary Nb_3Sn wires

The trend towards a stronger degradation for neutron irradiated wires observed for T_c vs. ϕt (see Fig. 13) is even accentuated when comparing the behavior of the normalized critical current density, J_c/J_{co}, and the upper critical field, B_{c2}, with fluence. The results for the Ti alloyed wire with the composition $(Nb\text{-}1.6wt.\%Ti)_3Sn$ are represented in Fig. 13. The most evident differences between the irradiated binary wires in Fig. 12 and the Ti and Ta alloyed wires are [7]:

a) The fluence ϕt_m at which T_c and B_{c2}* reach their maximum is 0.18×10^{18} n/cm^2 for the Ti alloyed wire (Fig. 11), i.e. it is four times smaller than for binary Nb_3Sn wires, where ϕt_m is 0.8×10^{18} n/cm^2. For the Ta alloyed wire, ϕt_m is very close to that of the Ti added wire: it is slightly below 0.2×10^{18} n/cm^2, as shown in Fig. 12. In their paper, Weiss et al. [7] also mention another additive, Ni+ Zn, which leads to a smaller enhancement of J_c, the amount of substituted elements in the filaments being smaller. For this rather unusual additive, the value of ϕt_m

lies at ~ 0.25×10^{18} n/cm^2, i.e. close to the values for Ti and Ta alloyed wires. It can be concluded that irradiation of alloyed Nb_3Sn wires with 14 MeV neutrons at 300 K has a very similar effect on the maximum value of J_c/J_{co}.

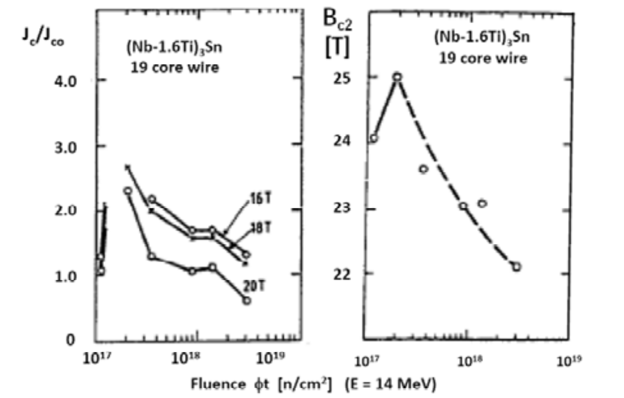

Figure 13: J_c/J_{co} and B_{c2}* of a 19 core (Nb-1.6wt.%Ti)$_3$Sn produced by the Bronze Route after neutron irradiation at 300 K (Weiss et al. [7]).

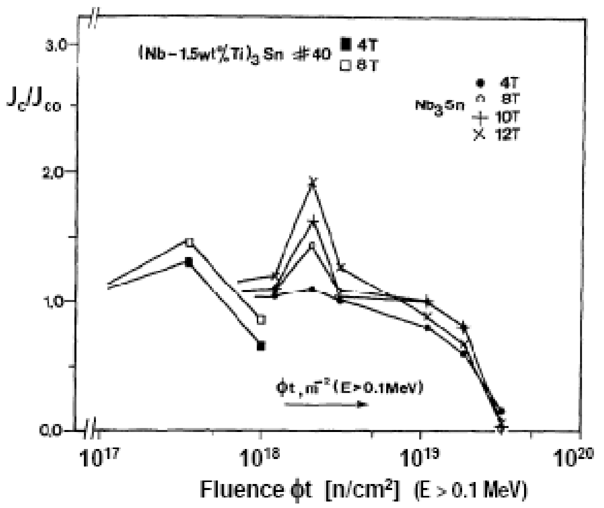

Figure 14: J_c/J_{co} and B_{c2}* of a 19 core (Nb-1.6wt.%Ti)$_3$Sn produced by the Bronze Route after neutron irradiation at 300 K (Weber et al. [5]).

b) The enhancement of J_c/J_{co} at ϕt_m for the same value of $h^*=H/H_{c2}^*$ is smaller for alloyed than for binary wires. At $h^*=0.7$, J_c/J_{co} for the Ti and Ta alloyed wires is ~3 and 1.2, respectively, which is considerably smaller than for binary Nb_3Sn (~ 4.5 for the binary 19 core wire and 2.2 for the binary 10'000 filament wire) (see Figs. 12).

c) In view of the application in LHC upgrade, it is interesting to know at what fluence J_c/J_{co} starts to be lower than 1 under an applied field of 15 T. It is remarkable that for both binary wires, this is the case for fluences close to 1 x 10^{19} n/cm^2, which is considerably higher than for alloyed wires. For the additives Ti and Ni+Zn, the corresponding fluences are estimated to 5 and 4 x 10^{18} n/cm^2. A much stronger effect was observed for the Ta alloyed wire, where J_c/J_{co} = 1 is already reached at 0.3 x 10^{18} n/cm^2! This tendency is confirmed by neutron

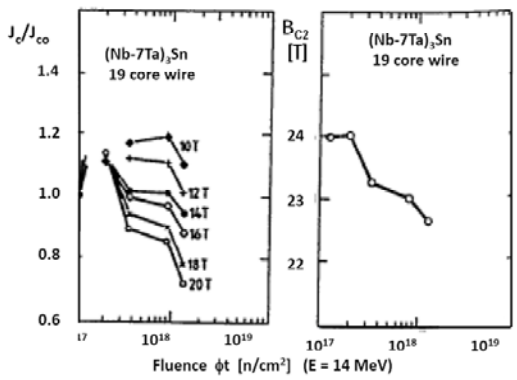

Figure 15: J_c/J_{co} and B_{c2}* of a 19 core (Nb-7wt.%Ta)$_3$Sn produced by the Bronze Route after neutron irradiation at 300 K (Weiss et al. [7]).

irradiation measurements of Weber et al. [5], who reported measurements up to 12 T and found J_c/J_{co} = 1 at 7×10^{18} n/cm^2 for binary Nb_3Sn wires, but 0.8×10^{18} n/cm^2 for (Nb-1.5wt%Ti)$_3$Sn wires.

It follows that alloyed wires are much more sensitive to radiation than binary wires. This can be explained by the disordering effect of the various additives. Indeed, it was found in unirradiated samples that the maximum of J_c is reached when the initial residual resistivity ρ_o of the ordered binary Nb_3Sn system is enhanced to values of 30-35 μΩ·cm by the substitution of the additives into the A15 lattice [53]. In binary wires, the necessary fluence to reach this optimum value of ρ_o (and thus B_{c2}) is considerably higher than in alloyed wires. Before irradiation, the ρ_o value of alloyed wires is already close to the optimum value of 30-35 μΩ·cm. Irradiation will thus only lead to a small enhancement of B_{c2}, followed by a decrease with higher fluences.

CONDITIONS FOR NB3SN WIRES IN THELHC UPGRADE

Radiation damage on the superconducting properties

As shown in Fig. 1 [1], the highest fluence in LHC upgrade will occur at the Quadrupoles Q2a, the second highest fluence being expected for the quadrupole Q1. For a total operation time of 10 years, calculating 200 operation days a year, the total fluence at the inner winding of the quadrupole Q2a after the upgrades of Phase I and Phase II will be 0.25×10^{18} n/cm^2 and 10^{18}

n/cm^2, respectively. For the quadrupole Q2 (external winding), the corresponding fluences are 0.17×10^{18} n/cm^2 and 0.68×10^{18} n/cm^2, the total fluence for the quadrupoles Q2b and Q3 being considerably smaller. No problem is expected for the dipoles. Since the highest radiation damage will be expected on the inner winding of the quadrupole Q2a, the present estimation will be limited to this location.

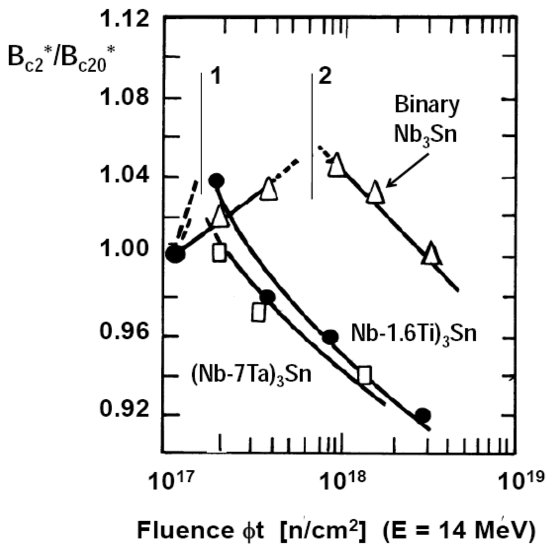

Figure 16: Variation of the upper critical field of binary and alloyed Nb_3Sn wires (additives: Ti and Ta), after Weiss et al. [7].

Radiation induced change of Cu resistivity and thermal stability

A point to be discussed when analyzing the properties of Nb_3Sn wires in view of operation in a LHC upgrade collider at ≥ 15 T is the effect of neutron radiation on the thermal stability. Little is known about the change of the electrical resistivity of pure Cu submitted to neutron irradiation. Fabritsiev et al. [54] studied the effects of neutron irradiation on the electrical resistivity of precipitation hardened (PH) and dispersion strengthened (DS) copper alloys in the fast neutron reactor BOR-60 with doses of $8\text{-}16\times10^{21}$ n/cm^2 and in the mixed spectrum neutron reactor SM-2 with doses of $3.7\text{-}5.5\times10^{21}$ n/cm^2. The experimental data on the change $\Delta\rho$ in electrical resistivity of DS-type copper alloys irradiated in the BOR-60 reactor show that irradiation to 7-10 dpa at T=340-450 °C causes a drop in electrical conductivity by not more than 20%. The obtained results show that in mixed-spectrum reactors the rate of $\Delta\rho$ normalized to the dpa is about 20 times as high as in fast neutron reactors. The conclusion is made that the calculations performed for ITER must take into account the presence of appreciable fluxes of thermal neutrons in certain components of the reactor. The latter will play a decisive role in the drop in thermal conductivity of copper alloys in these components. This problematics may be of smaller importance in LHC upgrade, the neutron spectrum being narrower. Nevertheless, this question must be studied in detail if the thermal stability of the magnets has to be guaranteed for the whole lifetime of approximately 10 years.

CONCLUSIONS

An overlook of the effects of neutron irradiation on the superconducting parameters T_c, B_{c2} and J_c of Nb_3Sn wires as reported in the literature has been given in view of the determination of the radiation limit in the LHC upgrade magnets. The coexisting defect mechanisms in irradiated Nb_3Sn type compounds are briefly presented and a model is discussed explaining the site exchange mechanism which leads to a decrease of atomic ordering after irradiation.

The variation of J_c in binary as well as in Ti and Ta alloyed Nb_3Sn wires is presented, showing a higher sensitivity to neutron radiation for the latter. Based on calculations of F. Cerutti and coworkers (CERN), the neutron fluence at the inner winding of the quadrupole Q2a is estimated to values below 10^{18} neutrons /cm^2 for a life time of 10 years, which is within the safety margin with respect to the critical current density and B_{c2}.

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