Mat. tech. San Francisco, 80

STUDY AND CHARACTERIZATION OF THE ELECTRICAL PROPERTIES OF THE FeNi (50-50 at.%) ALLOYS DURING FAST NEUTRON IRRADIATION

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Ni -Cr?

ABSTRACT

By means of electrical resistivity the order-disorder transformation was studied during fast neutron irradiations on FeNi (50-50 at.%) and NiCr (80-20 wt.%) alloys. The results obtained with FeNi are in agreement with the literature. The critical temperature of the order-disorder transformation obtained was Tc = (319 ± 9) °C and the activation energy Ea = (0.49 ± 0.05) eV. As for NiCr (80-20 wt.%) alloy the main results obtained was the following: a) Tc = (536 ± 4) °C with Ea = (1.26 ± 0.14) eV. Also in this case, the results are in agreement with the literature, but they are obtained during irradiation in spite of those obtained without irradiation for the same material, b) electrical properties characterization with technological applications.

I. INTRODUCTION

The present paper aims at a contribution to the materials science and technology using and developing methods of selection and quality control of nuclear materials, mainly those related with the nuclear technology. For this, resistivity method was used in order to obtain the characterization of the electrical properties as well as radiation damage studies in conditions that simulate those of power reactors. Other methods were also used in the Radiation Damage Dept. of IPEN as it can be seen in the followings references [(1), (2)].

The order-disorder transformation was considered as one process of the structural changes. This is a physical process during which a metallic binary alloy goes from an ordered state to a disordered state or vice versa, the order being marked by all atoms distributed in a regular and periodic arrangement[(3)]. The long range order (LRO) parameter is represented by η and the short range order (SRO) by ζ. In the case of complete disorder $\eta = 0$ and $\zeta = 0$, while for complete order one has $\eta = 1$ and $\zeta = 1$. Consequently; $0 \leq \eta \leq 1$ and $0 \leq \zeta \leq 1$.

II. SAMPLES

The alloys studied were FeNi (50-50 at.%) and Ni Cr (80-20 wt.%). The FeNi (50-50 at.%) alloy was studied in continuation of Marchand's work [(4)]. The samples of these materials came from Johnson-Mathey and they were annealed during 1 h in Helium atmosphere at 1,100 °C followed by quenching. The agreement between the results of this work and those of Marchand [(4)] is excellent.

The samples of NiCr (80-20 wt.%) alloy produced by Villares (Brasil) were wiredrawn in order to reduce the 3mm wire diameter to 1mm square section wire which corresponds to 85% reduction in cross section[(5), (6)]. This proceedure was made to obtain the electrical properties characterizations according to the standard of ASTM B 70-56 and B 63-49. After the wiredrawing, the samples were annealed at 900°C in Argon atmosphere and then, quenched with the purpose to give the samples the same normalized initial state, characterized by a high vacancies concentration and a disordered state. With this heat treatment the tensions created during the wiredrawing are also annealed out. The figure 1 illustrates a phase diagram of this alloy which was suggested by Hansen[(7)].

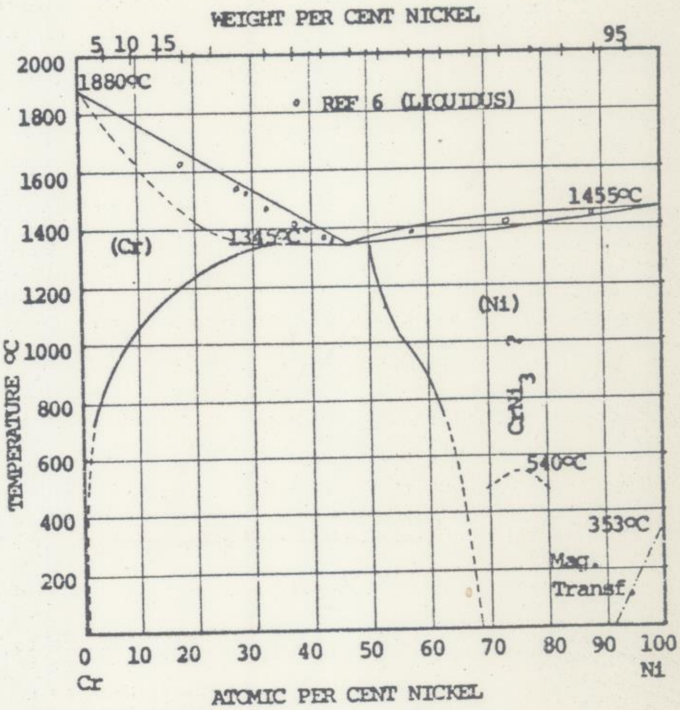

Fig. 1 - Phase diagram of CrNi.

In the range 70-80 at.% of Ni and close to 540°C, there is a transformation, supposed to be an order-disorder transition[(8), (9)].

The main aim of this work is to define this transition and to measure the temperature in which it occurs. Table 1 gives a nominal composition of NiCr (80 - 20 wt.%) alloy.

TABLE 1

ELEMENT	wt.%
Ni	79.500
Cr	18.800
C	0.140
Si	1.020
Mn	0.030
P	0.012
S	0.005
Ti	0.260
N	156 ppm
Al	0.060

III. EXPERIMENTAL PROCEDURES.

Electrical resistivity (ρ) measurements were made with a DANA - 5800A digital multimeter which incorporates an "Ohms Converter" module. The temperature (T) was controlled by RT-3000, SETARAM electronic temperature controller which permits annealings with ± 1 °C precision. The measurements were made during irradiation (in situ) using an appropriate irradiation device with controlled temperature and atmosphere. Many of the measurements were made also without irradiation.

IV. RESULTS AND DISCUSSION.

The electrical properties characterization is shown in the figure 2, which refers to sample 1. With this sample, three linear annealings was made: curve a, first annealing - from temperature 20°C at 900°C; curve b, second annealing - from temperature 20°C at 750°C ; and curve c, third annealing from temperature 20°C at 750°C. In this case the measurements were made without irradiation. Also without irradiation, the figure 3 shows a number of linear annealings increasing and decreasing temperature for sample 2 without normalizing heat treatment. One observes an anomalous behavior of ρ X T, because, as it is kown, the ordered state shows a ρ smaller than that in the disordered one [4]. It must be taken into account that the sample was reduced 85% in cross section, what is a considerable cold work. In this hardened state a first linear annealing was made at a rate of ≈ 3°C/min for both increasing and decreasing temperature. The result was the following: the resistivity increases with T (temperature) to approximately 510°C and thereafter it decreases to T ≈ 750°C, increasing again after this temperature; during the annealing with decreasing T, the value of ρ at room temperature became higher. In this aspect the anomalous behavior appears. Initially hardened sample is in the disordered state. After the first annealing, it reaches some extent of order degree and; it was expected that the value of ρ to be lower than that in the disordered state, but it became 14% larger, at room temperature. Further annealings with increasing and decreasing of T leaded the values of ρ to a saturation in which the values of electrical resistivity coincide.

This phenomenon was observed by Yano[8] who by means of resistivity measurements supposed to refer to an order-disorder transformation at approximately 540°C based in Ni_3Cr (77.2 wt.%Ni) composition. This behaviour of ρ with T, observed in this work was also observed by Taylor[9]. This author indicates the critical temperature as Tc = (544 ± 4) °C which was determined by specific heat measurements.

Knowing that neutron irradiation enhances the ordering processes in an alloy [10], a plot was made (fig.4) that compares the linear annealings made with 3 samples annealed at 900°C and quenched at room temperature (disordered). The behaviour of this curves expresses clearly the agreement with the arguments above. The curve a of sample 3 represents the increase of ρ with T, without irradiation; the curve b of sample 4, during irradiation shows the increase of ρ with T by values a little larger than of curve a and the curve c of sample 5, after 6 hours of irradiation (3h at 590°C, 2h at 550°C, 1h at 520°C) shows ρ increasing with T by values still larger than those of curve b. Thus it can be concluded that when alloy Ni_3Cr has a greater degree of order its electrical resistivity increases.

In considering what was said above, it is possible to formulate an hypothesis regarding the isothermal annealings during neutron irradiation: one should expect the occurrence of an increase in ρ, during isothermal annealings "in situ". The analysis of isothermal annealings made during neutron irradiation will be con-

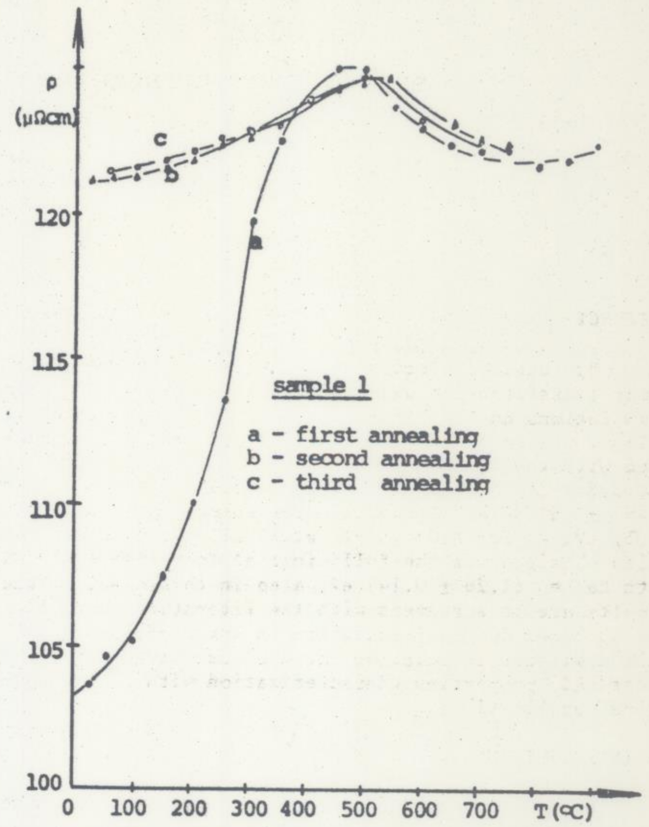

Fig. 2 - NiCr(80-20 wt.%) alloy. Electrical Properties Characterization, without irradiation. Linear annealings.

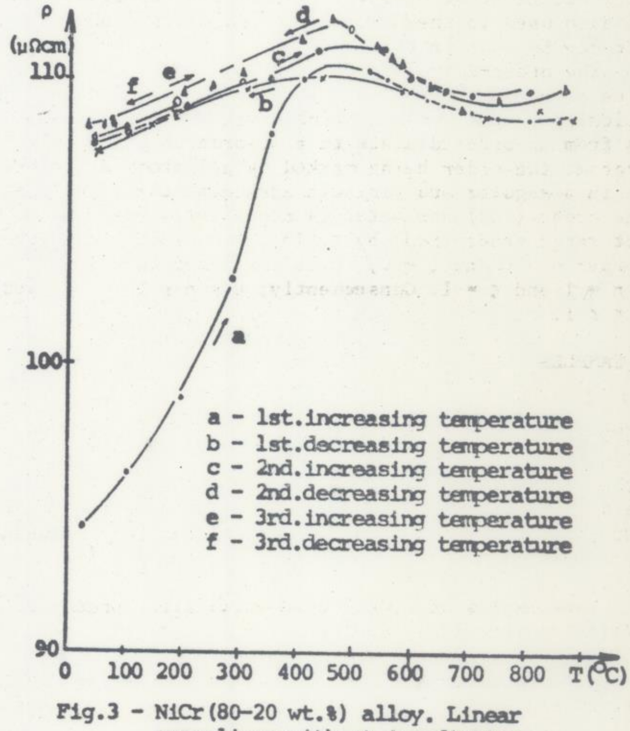

Fig.3 - NiCr(80-20 wt.%) alloy. Linear annealings without irradiation.

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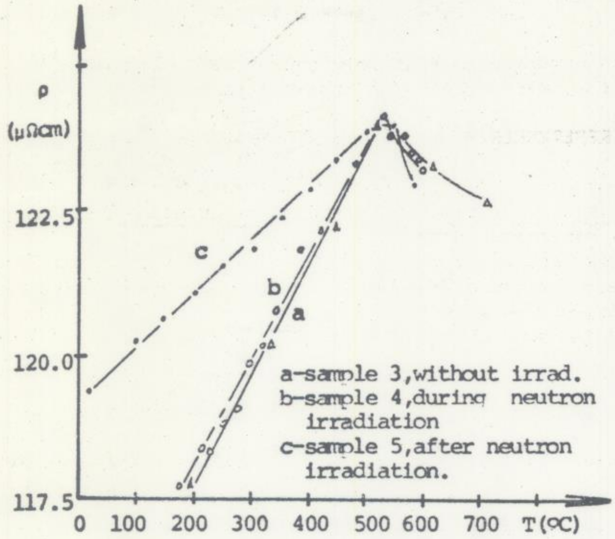

Fig.4 - NiCr(80-20 wt.%) alloy. Linear annealings.

The figures 5 and 6 show the annealings made during irradiation with the samples 6,7,8,9 and 10. The curves in the figure 5 refer to the first annealing made with each sample, i.e., each annealing corresponds to one sample, every sample having the same initial state (disordered). The curves in the figure 6 were obtained with the sample 9. One can observe in these figures that isothermal annealings up to T ≈ 540 ºC show a decrease of the resistivity in the first part of the annealing, increasing again afterwards. Since in this case the annealing was made with disordered samples and, with initial vacancies concentration, it is supposed that in the first instants of the annealing a high quantity of vacancies is annealed out by migration to the fixed sinks and by vacancy-interstitial recombination, which causes a decreases in the resistivity. Also, in these initial instants of the annealing occurs a migration of vacancies, consequently there will be a formation of SRO (short range order), although with $0 < \zeta << 1$, but which nevertheless contributes to the increase of ρ and appears during the annealing processes, and is due to the remaining vacancies migration that facilitate the ordering processes by means of exchange of position between the atoms [11], [12]. On the other hand, isothermal annealings with T below 540ºC show that the electrical resistivity increases during the annealings. Here also, is present an annealing out of vacancies by migration to fixed sinks and the vacancy-interstitial recombination, but the main contribution to the resistivity comes from vacancies migration due to the exchange of the atomic sites which, at T < 540ºC, occupy the sites in the lattice favorable to the formation of LRO and SRO, both in considerable degree, which can be expressed as $0 << \eta < 1$ and $0 << \zeta < 1$, respectively.

The figure 7, represents the data of figures 5 and 6 normalized. In this figure one can observe more clearly the change in the behaviour of ρ where curves <u>a</u>, <u>b</u>, <u>c</u> and <u>d</u>, refer to T < 540ºC and <u>e</u>, <u>f</u>, <u>g</u> and <u>h</u>, refer to T > 540ºC.

V. DETERMINATION OF Tc AND Ea.

Within limits mentioned above and based on plots of the figure 5, it is possible to settle the critical temperature of order-disorder transition of the NiCr (80-20 wt.%) alloy between 540 and 532ºC;

$$T_c = (536 \pm 4)\ ºC,$$

which is comparable with that obtained by Taylor[9] by means of specific heat, and without irradiation;

$$T_c = (544 \pm 4)\ ºC.$$

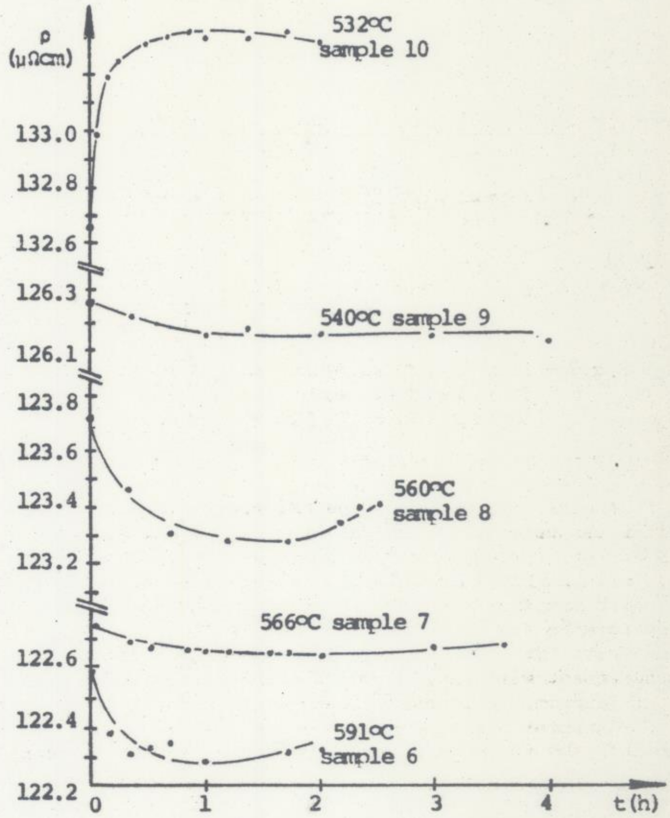

Fig.5 - NiCr(80-20 wt.%) alloy. Isothermal annealings during neutron irradiation. All samples have the same initial heat treatment.

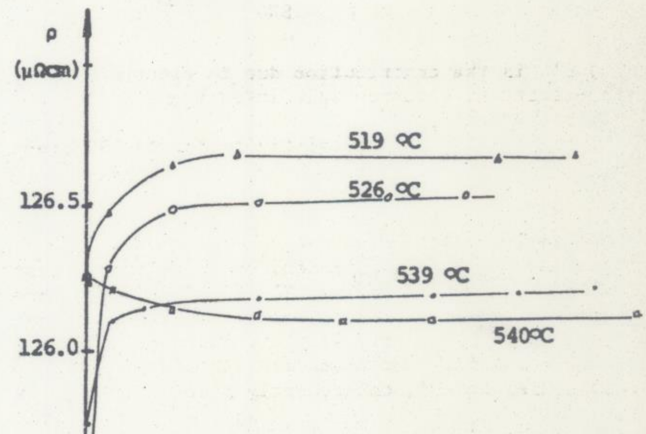

Fig.6 - NiCr(80-20 wt.%) alloy. Isothermal annealings during neutron irradiation. Sample 9.

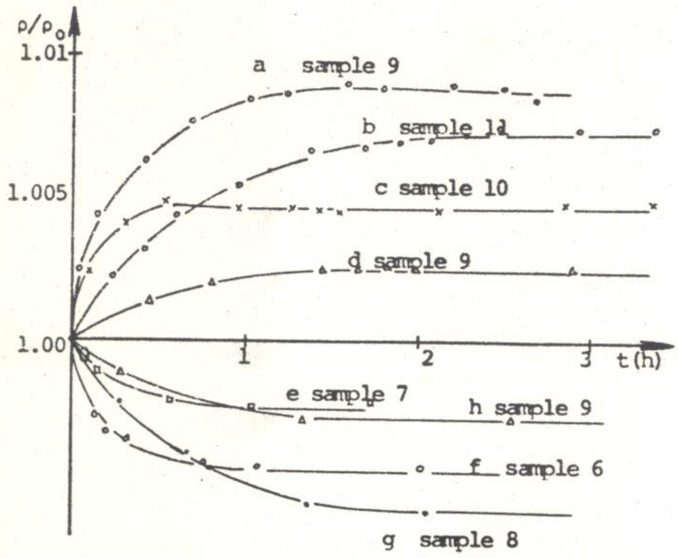

Fig.7 - NiCr (80-20 wt.%).Normalized plots of figs.5 and 6. a,b,c,d - T 540 °C and e,f,g,h - T 540°C.

Another evidence of the existence of this transition can be observed in the figures 2,3 and 4. The samples of NiCr(80-20 wt.%) alloy, consisted mainly of 79.5 wt.% Ni and 18.8 wt.% Cr, in agreement with table 1, which admits the existence of Ni_3Cr phase of fcc structure in the range from 70 to 80 wt.% Ni, in agreement with the phase diagram of the figure 1 and also in agreement with Taylor and Floyd [13]. Based on phase diagram, it is possible to suppose the inexistence of diferent phases over and under T = 540°C. This permits the following interpretation of the linear annealings of figures 2,3 and 4: i) temperatures between room temperatures and T = 535 °C (Fig.4).In The beginning of the annealing the sample was disordered.In this case the electrical resistivity can be writen as:

$$\rho = \rho' + \rho_{SRO} + \rho_{LRO} ,$$

where:

a) ρ' is the contribution due to electron-fonon, electron-electron, electron-spin interation, plus the residual resistivity,

b) ρ_{LRO} electrical resistivity due to LRO processes,

c) ρ_{SRO} electrical resistivity due to SRO processes.

One admits ρ' as being ever increasing with temperature, thus ($\rho_{SRO} + \rho_{LRO}$) contributs to the increase of ρ, because at temperature T < 535 °C, there are long range and short range ordering processes, ii) at temperatures T > 535°C (fig.4). In this case, the LRO disappears completely and there are only SRO processes present and, consequently ρ decreases, in other words, $\rho_{LRO} = 0$ and ρ_{SRO} contributes to the ordering.

Thus it is possible to determine from figure 4, that the critical order-disorder temperature is,

$$Tc = (535 \pm 5)\ °C.$$

The activation energy Ea for the diffusion of vacancies in the annealings processes was determined by means of the curves of figure 5 with four temperatures: T = 591, 566, 560 and 540°C. The value obtained was:

$$Ea = (1.36 \pm 0.14)\ eV,$$

which agrees with that obtained by Feder and Cahn[14] for the alloy Fe_3Al with the same structure as Ni_3Cr.

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