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The Effect of Structural Changes on Magnetic Permeability of Amorphous Powder $Ni_{80}Co_{20}$

A. Maričić [1 *)], M. Spasojević [1], S. Arnaut [1], D. Minić [2], M.M. Ristić [3]
[1]Technical Faculty Čačak, Svetog Save 65, 32 000 Čačak, Serbia
[2]Faculty of Physical Chemistry, Studentski trg 16, 11 000 Belgrade, Serbia
[3] Serbian Academy of Sciences and Arts, Knez Mihailova 35, 11 000 Belgrade, Serbia

Abstract:

The structural changes of $Ni_{80}Co_{20}$ amorphous powder were tested during heating. The alloy was obtained by electrolysis from ammonia solution sulfate of cobalt and nickel on the titanium cathode. The differential scanning calorimetry (DSC) method was used to detect that the crystallization process of powder occured in two stages with crystallization peaks temperatures of the first stage at 690 K and of the second stage at 790 K. The effect of structural relaxation and crystallization of powder on magnetic properties was predicted by measurement of the relative magnetic permeability change in isothermal and nonisothermal conditions. On the basis of the time change of relative magnetic permeability at a defined temperature in the temperature range of the first and second crystallization peak on the thermogram, the kinetics of crystallization was defined. It was predicted, that in the initial time interval, in the range of the first crystallization peak, the rate of crystallization is determined by the rate of nucleation of the amorphous part of the powder. However, in the second time interval, the crystallization rate is determined by the rate of diffusion. In the range of the second peak, in the beginning the rate of crystal growth is determined by activation energy of the atom pass from smaller to bigger crystal grain. In second time interval, the rate of crystal grain growth is determined by the diffusion rate of atoms to the location of integration into bigger crystal grains. For all processes which determine the rate of crystallization in temperature ranges of both crystallization peaks, the Arrhenius temperature dependence of rate for those processes is obtained. The relative magnetic permeability of crystallized powder at 873 K, is smaller for about 30 % than the relative magnetic permeability of fresh powder at room temperature. However, structurally relaxed powder at 573 K has an about 22 % larger magnetic permeability than the same fresh powder at room temperature.

Keywords: *$Ni_{80}Co_{20}$, Amorphous powder, Magnetic permeability, Crystallization rate.*

1. Introduction

Amorphous and nano-structural metallic alloys are very important because of their specific properties, for fundamental research and also for application in electrical engineering, electronics and in the other industry branches. The powders of these alloys, thanks to their specific structure, have physical-chemical properties which are different from the properties

*) Corresponding author: marec@tfc.kg.ac.yu

of alloys with large crystal grains [1-15]. Application of nano-structural powders in modern technology is enabled by their specific properties. In the last twenty years, much research has been devoted to the determination of an optimized procedure for obtaining nano-structural powders with in advance determined stipulated properties. By adequate electro-chemical processes with applicable selected kinetic and operating parameters of electrolysis, it is possible to obtain powders of metallic alloys with stipulated physical-chemical characteristics [7, 8, 14-22]. The properties of electro-chemically obtained powders are often appreciably different than the properties of powder with the same chemical composition, obtained by another method. A study of structural changes of nano-structural alloys during heating enables definition of the conditions of thermal treatment for obtaining materials with specific properties [6-15, 22]. During heating dimensions of crystal grains, density of chaotically arranged dislocations, micro-strain, phase structure and the amount of the amorphous phase change in nano-structural materials [6-15, 22]. Those changes influence the electrical, magnetic, catalytic, corrosion and other properties. In this work the heating effect of electro-chemically obtained $Ni_{80}Co_{20}$ alloy from ammonia bath to the process of relaxation and crystallization is examined in order to determine how these processes affect the magnetic properties of these alloys.

Experimental

$Ni_{80}Co_{20}$ alloy powder was obtained by electro-deposition from an ammonia bath on the titanium cathode at a density of electric current j = 150 $mAcm^{-2}$ [23].

The crystal structure and grain size of the powders were determined by X-ray diffraction (XRD) annalysis. XRD measurements were performed on a Philips MRD diffractometer using Cu-K_α radiation.

Crystallization of the powder was investigated by the differential scanning calorimetry (DSC) method. Thermograms were obtained on a Shimadzu instrument at a heating rate of 20 $Kmin^{-1}$ under the flow of pure nitrogen.

Measurements of relative magnetic permeability were performed using a modified Maxwell method, based on the action of an inhomogeneous field on the magnetic sample. The magnetic force measurements were performed with a sensitivity of 10^{-6} N in an argon atmosphere.

Results and Discussion

$Ni_{80}Co_{20}$ alloy powder was obtained by electrolysis from an ammonia solution of sulfate nickel and cobalt on the titanium cathode. XRD-analysis confirmed that the powder is composed of a phase of solid solution nickel and cobalt with nano-crystals of medium value with dimensions from 12 nm with a surface centered cubic lattice (FCC) and from an amorphous phase positioned between the crystal grains. The Rietveld method [24] was used to determine that 75 % fresh powder has an amorphous structure, and that 25 % has a crystal structure.

A DSC thermogram of fresh powder of alloy $Ni_{80}Co_{20}$ is shown on fig. 1. The obtained thermogram shows that the relevant structural changes in the powder occur in the temperature interval from 623 K until 943 K. It is possible to notice two expressed crystallization peaks with maximums on 690 K and 790 K. The enthalpy of the first crystallization peak was $\Delta H_1 = 66{,}7 \cdot 10^3$ J kg^{-1}, while the enthalpy of the second crystallization peak was $\Delta H_2 = 56{,}0 \cdot 10^3$ J kg^{-1}.

The X-ray diffractogram obtained after heating $Ni_{80}Co_{20}$ powder at 723 K shows that during heating crystallization of the amorphous part of the powder and growth of crystal

grains of the FCC phase on account of amorphous phase take place. These changes dictate the appearance of the first crystallization peak on the thermogram.

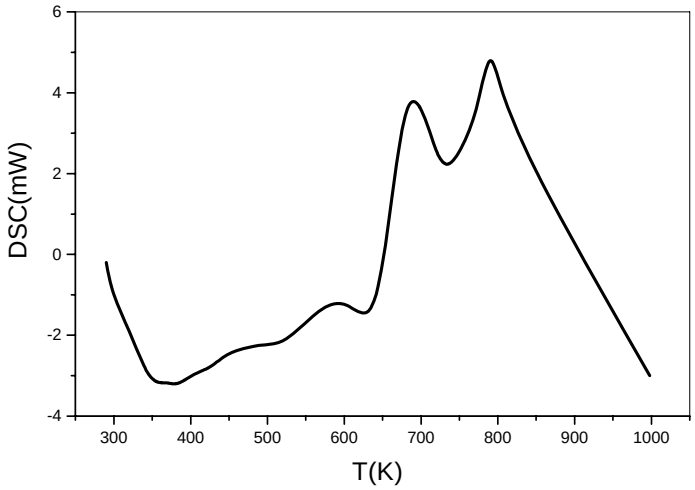

Fig. 1. DSC thermogram of $Ni_{80}Co_{20}$ powder alloy obtained at j = 150 mAcm^{-2}. The heating rate was 20 Kmin^{-1}.

The X-ray diffractogram obtained after heating of the powder at 923 K shows, that during heating relevant growth of crystal grains occured. The second crystallization peak on the thermogram is the consequence of the growth of bigger crystal grains on account of the smaller ones.

Structural changes of the electro-chemically obtained powder during heating are followed by measuring of relative magnetic permeability change. Fig. 2 shows the magnetic permeability changes during heating of fresh powder until 573 K (curve a).

The curve a shows that magnetic permeability increases and reach a maximum at 493 K during powder heating. After heating the powder was cooling to room temperature.

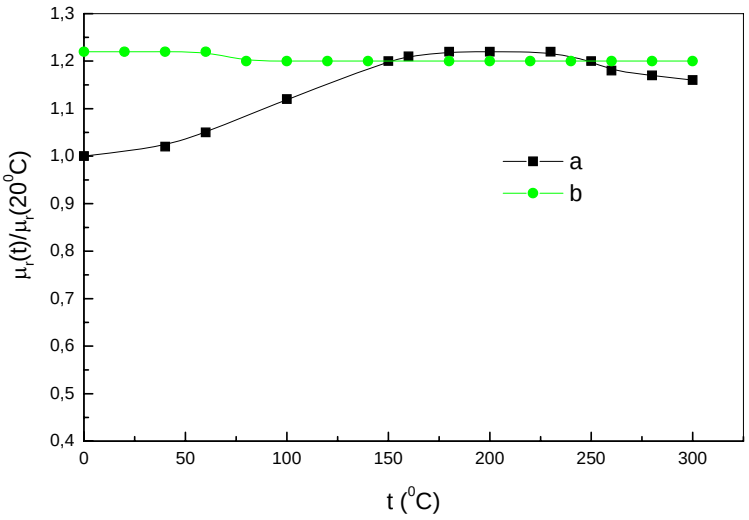

Fig. 2. Temperature dependence of relative magnetic permeability of $Ni_{80}Co_{20}$ powder: *a* – first heating and *b* – second heating. The heating rate was 20 Kmin^{-1}.

During cooling, the magnetic permeability did not change. This indicates that during structural relaxation in powder, irreversible structural changes occured. After structural relaxation, the cooled powder has an about 22 % higher magnetic permeability than the same

fresh powder. After that, the powder was heated for a second time, at 573 K. During the second heating, the magnetic permeability did not change relevantly (fig. 2, curve b). This indicates that during the second heating, relevant structural changes in the powder did not occur.

During structural relaxation, the thermal energy and external magnetic field with an intensity of 8000 Am^{-1}, caused mobility of magnetic domains walls. By organising the structure on short distances, the atoms, in line besides oriented magnetic domains, cross at a slightly lower potential level and their magnetic moment tends to be in the direction of magnetic moments of atoms in the nearby oriented domain. This leads to an increase of magnetic domains. Thus the termal energy and relatively weak external magnetic field during structural relaxation, causes extracting oriented magnetic domains, which also increase the magnetisation and magnetic permeability.

Curve (a) on fig. 3, shows the temperature dependence of the relative magnetic permeability change during heating fresh powder $Ni_{80}Co_{20}$ until 873 K. The Curie temperature of electro-chemically obtained powder is 873 K. The relative magnetic permeability of fresh powder, decreases appreciably with temperature increase in the temperature interval from 573 K until 873 K. This decrease is caused by: a) dissipation of orientation of magnetic domains by the activity of thermal energy and b) structural changes in the powder. Decreasing of magnetic permeability of fresh powder (curve a), in the temperature interval from 543 K until 640 K, is caused just by thermal energy. In the interval from 640 K until 763 K, magnetic permeability decreases because of dissipation orientations of domains by thermal energy and by crystallization of the amorphous part of the powder. The intense reduction of magnetic permeability from 763 K until 873 K is caused by dissipation orientations of domains and creation of larger crystalline grains.

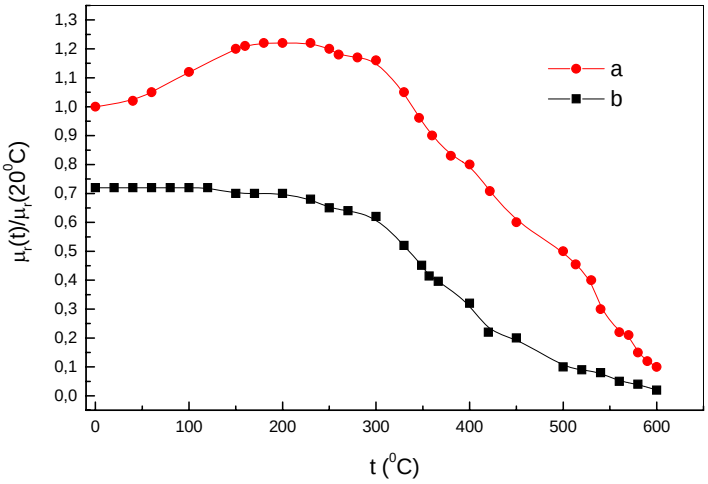

Fig. 3. Relative magnetic permeability temperature dependence of powder $Ni_{80}Co_{20}$: *a* – first heating and *b* – second heating. The heating rate was 20 $Kmin^{-1}$.

Curve (b) on fig. 3 shows the relative magnetic permeability change of the same sample during second heating. During second heating up to 873 K no structural changes occured. Thus the relative magnetic permeability did not change in the temperature interval from 293 K until 573 K. Decreasing of magnetic permeability of the crystal sample with increasing of temperature in the temperature interval from 573 K until 873 K was caused by the change of individual magnetic domain direction under the influence of thermal energy. Fig. 3 shows that the magnetic permeability of the crystalline sample is about 30 % less than the magnetic permeability of the same fresh sample. The magnetic permeability of crystallized powder is 50 % less than the magnetic permeability of powder annealed up to 573 K (fig. 2). The crystalline powder of electro-chemically obtained alloy $Ni_{80}Co_{20}$, has lower

magnetic permeability than the annealed powder, as the orientation of individual domains is more compilaced in the crystalline powder and the motion of oriented domain walls is reduced.

The kinetics of powder crystallization is determined by measuring of time changes of the relative magnetic permeability, at the defined temperatures, in temperature ranges of the first and the second expressed crystallization peak on the thermogram.

Fig. 4 shows that in the initial time interval, a linear dependence exists of the logarithm of relative magnetic permeability $\ln(\mu(\tau)/\mu(\tau_0))$ on time (τ).

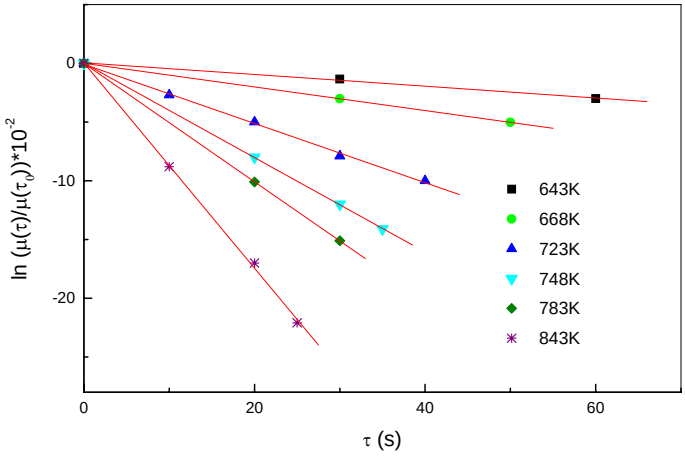

Fig. 4. Dependence of the logarithm of relative magnetic permeability $\ln(\mu(\tau)/\mu(\tau_0))$ as a function of the time duration of heating (τ),at the temperatures: ▪ 643 K ; ◦ 688 K, ▲723 K ;▼ 748 K , ♦ 783 K, ∗ 843 K .

The obtained dependences, shown on fig. 4, show that, in the first time interval in the temperature range of both expressed crystallization peaks on the thermogram, crystallization is an activated reaction of the first order. This indicates that in the temperature range of the first crystallization peak, the rate of crystallization in the first time interval is determined by the rate of nucleation, but in the temperature range of the second crystallization peak, the rate of crystallization is determined by the activation energy of transfer of atoms from smaller crystal grains to larger crystal grains.

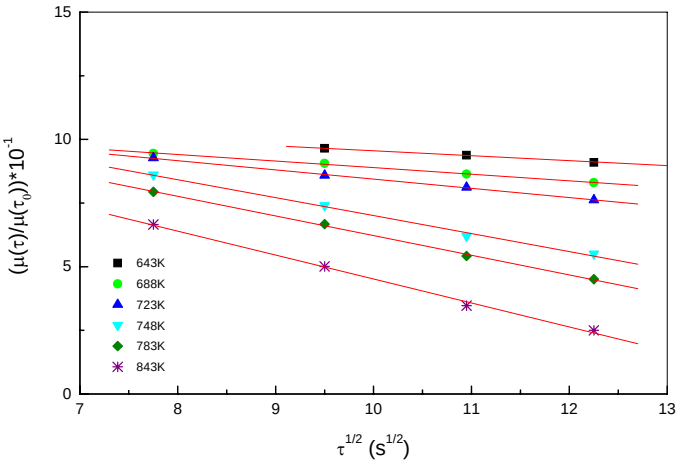

Fig. 5. Dependence of relative magnetic permeability $\mu(\tau)/\mu(\tau_0)$ on $\tau^{1/2}$ at temperatures: ▪ 643 K ; ◦ 688 K, ▲723 K ;▼ 748 K , ♦ 783 K, ∗ 843 K

In the second time interval (fig. 5), there is a linear dependence of the relative magnetic permeability $\ln \mu(\tau)/\mu(\tau_0)$ on $\tau^{1/2}$. This indicates that in the second time interval in both temperature ranges, the rate of crystallization is determined by the rate of diffusion atoms until the place of integration in crystal grains.

On the base of diagrams shown on fig. 4. and 5, a linear dependence of the rate crystallization logarithm on $\frac{1}{T}$ is established. This indicates that there is an Arrhenius dependence of the nucleation rate, rate of transition of atoms from smaller to larger crystalline grains and diffusion rate on temperature.

Conclusion

By DSC it is demonstrated that electro-chemically obtained $Ni_{80}Co_{20}$ powder, made from 75 % of amorphous phase and 25 % of FCC phase of solid solution nickel and cobalt with nanocrystal average dimension from 12 nm, crystallizes in two temperature ranges with crystallization peak temperatures at 690 K and at 790 K.

Measurements of the magnetic permeability relative change during time at a constant temperature in the temperature range of the first crystallization peak, show that in the initial time interval, the rate of crystallization is determined by the rate of nucleation. In the second time interval, the rate of crystallization is determined by the rate of diffusion of nickel and cobalt atoms.

In the temperature range of the second crystallization peak, in initial time, the rate of growth of crystal grains is determined by the activation energy of transition atoms from smaller to larger crystal grains. In the second time interval, the rate of growth of crystal grains is determined by the diffusion rate of atoms to the place of integration into larger crystal grains.

By thermomagnetic measurements it was shown that the relaxed powder at 573 K has about 22 % larger magnetic permeability than the same fresh powder at room temperature. However, powder $Ni_{80}Co_{20}$, crystallized at the temperature of 893 K, has about 50 % lower magnetic permeability than structural relaxed powder at room temperature.

The research results have shown that the electro-chemical process may produce nano-structural powders whose functional properties can be changed through the simultaneous effect of thermal energy and an external magnetic field.

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Садржај: *Испитане су структурне промене аморфног праха легуре $Ni_{80}Co_{20}$ током загревања. Прах је добијен електролизом из амонијачног раствора сулфата кобалта и никла на катоди од титана. Диференцијалном сканирајућом калориметријом (DSC) утврђено је да се кристализација праха одвија у две темепературске области са температурама егзотермна максимума 690 K и 790 K. Ефекат структурне релаксације и кристализације праха на магнетна својства установљен је мерењем промене релативне магнетне пермеабилности у изотермским и неизотермским условима. На основу временске промене релативне магнетне пермеабилности на одређеној темепратури у температурској области првог и другог егзотермна максимума на термограму, одређена је кинетика кристлизације. Установљено је да у почетном временском интервалу, у области првог егзотермна максимума, брзину кристализације одређује бризна нуклеације аморфног дела праха. Међутим у другом временском интервалу брзину кристализације одређује брзина дифузије. У области другог егзотермног максимума у почетку брзину раста кристалних зрна одређује енергија активације преласка атома са мањих на већа кристална зрна. У другом временском интервалу брзина раста кристалних зрна одређена је брзином дифузије атома до места уградње у већа кристална зрна. За све процесе који детерминишу брзину кристализације у температурским областима оба егзотермна максимума на термограму добијена је Аррхениус-ова зависност брзине тих процеса од температуре. Релативна магнетна пермеабилност искристалисаног праха на 873 K је на собној темепратури за око 30 % мања од релативне магнетне пермеабилности свежег праха. Међутим прах структурно релаксиран на 573 K има на собној температури за око 22 % већу магнетну пермеабилоност од свежег праха.*

Кључне речи: *$Ni_{80}Co_{20}$, аморфни прах, магнетна пермеабилност, брзина кристализације.*