Production and Characterization Of Electroless Ni Coated Fe-Co Composite Using Powder Metallurgy In Microwave Furnace

Assoc. Prof. Dr. Ahmet YONETKEN (ayonetken@gmail.com)
Afyon Kocatepe University

Original Article

Keywords: Powder metallurgy, Microwave Sintering, Composite and Electroless Nickel Plating

Posted Date: December 20th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-1151074/v1

Production and Characterization Of Electroless Ni Coated Fe-Co Composite Using Powder Metallurgy In Microwave Furnace

Ahmet YONETKEN

Afyon Kocatepe University, Electrical Engineering Dept., Afyonkarahisar, Turkey
ayonetken@gmail.com, ORCID ID: http://orcid.org/0000-0003-1844-7233
Corresponding Author

Abstract

The specimens, magnetic properties materials, and microwave characteristics of Ni coated Fe and Co composites were researched by specimens produced by microwave furnace sintering at 1100°C temperature. A uniform nickel deposit on Fe-Co particles was coated previously to sintering by electroless coating deposition procedure. A composite consisting of quaternary additions, a metallic phase, Fe-Co inside of Ni matrix has been prepared under in a neutral atmosphere environment then microwave sintered. X-Ray Diffraction, SEM(Scanning-Electron-Microscope), Empedans Phase Analyzer were utilized to obtain structural data and to determine magnetic and electrical features such as dielectric and conductivity at the temperature range of 25-400C. The ferromagnetic resonance varied from 10 Hz to 1GHz and measurements were employed to characterize the features of the specimens. Empirical of findings obtained for the composition (Fe-%25Co)50Ni at 1100°C recommend that the best conductivity and hardness were obtained with 50Ni addition at a sintering temperature of 1100°C.

Keywords: Powder metallurgy, Microwave Sintering, Composite and Electroless Nickel Plating

1. Introduction

Nickel is a strategically important element used as an additive element in many commercial applications and in the military and aviation industries. And also It is used to give a green color in the glass industry. Nickel is above all an alloying element. Therefore, it has many uses as an alloy. These alloys are alloys made with copper, chromium, and aluminum etc. The magnetic characteristics of the Co-Fe-Ni triple alloys have been explored in the early 1950s [1] The properties of magnetic materials are known through research by Bozorth. Magnetic properties are recommended for many applications. An example is to save information to the head core in electronic storage unit [2-4]. It is obtain that. Iron-Nickel-Cobalt alloys with soft magnetic characteristics, radio frequency magnetron spray process was used. However, different alloy compositions have been used compared to electro-laminated films [5]. Low saturation magnetic field of Permaloy revealed the idea of searching for a material with better magnetic features. According to Permaloy, Fe-Co-Ni alloys have higher saturation magnetization properties [6]. The application of these materials on an industrial scale is limited by a absence of good information about the effect of the deposition condition on the microstructure, composition and magnetic features. In a ferrite we can observe the transition from ferromagnetic to paramagnetic state as temperature increases. In these materials, the activation energy was found to be lower in the ferro magnetic region than in the magnetic field [7-9]. The electrical properties of the ferrites are susceptible to the method of preparation, sintering temperature, time, heating and cooling rate [10-11]. Nickel-Cobalt alloys are nickel alloys commonly used for electroforming due to their magnetic features and high tensile strength [12]. It is used by integrating electrodeposition magnetic materials. For example, Micro Electro Mechanical Systems (MEMS), microactivators, sensors, micro motors and frictionless devices [13-20]. Electroless nickel coating has many advantages; the cost is low, easy to apply, a continuous and homogeneous coating formation occurs in powder particles [21]. It has been stated that microwave sintering consumes less energy in a short time together with the advantage of low temperature sintering [22-24]. The mechanical and magnetic properties of the electro-shaped nanocrystalline Fe-Ni thin film have been investigated and the Fe-Ni thin film has been commercially accepted with its grain size and thermal expansion coefficient and permeability properties [25–28]. Recently, there has been an increased interest in the use of magnetic materials due to their potential applications in ferrofluids, advanced magnetic materials, catalysts, optical and mechanical devices, high-intensity magnetic imaging, and medical diagnostics [29-35].

The aim of the study was to define the system and hyperfine interactions of selected Fe–Co–Ni alloys after Electrodeposited process as well as after heat treatment. X-ray diffraction (XRD) were used for the phase analysis. Moreover, the effective magnetic proporties and the Curie temperature of the alloys was defined from the temperature-dependent magnetize curve.

2. Experimental Method

The powder properties of Fe, Co, and Ni used in this work are as follows. It has a purity of 99.9% for Fe powders and a particle size of less than 40 µm. For Co powders it has a purity of up to 99.9% and a particle size of less than 140 µm. In the Ni deposition, Fe-Co powders were coated by using an electroless Ni coating method.

Ni coating was achieved by suspending the starting Fe-Co powders in a Ni containing solution ($NiCI_2.6H_2O$) at 90-95°C and by adding Hydrazine Hydrate ($N_2H_4.H_2O$) drop by drop and 30 vol.% Ammonia solution while keeping the pH at 8-10. With increasing temperature, As ammonia evaporation increases fort his reason, An ammonia dripping apparatus was used to keep the pH of the coating bath in the range of 8-10. By the way, the bath chemical was continuously mixed and the pH was constantly observed by utilized a Philips PW 9413 Ion-Meter. The reaction was allowed to continue until sufficient Ni was added for deposition coating all the Fe-Co powders, then, Ni coated Fe-Co powders were filtered out of the bath chemical by using a paper filter after washed several times with pure water and alcohol and then oven dried at 105°C. and then followed by sintering at 1100ºC for 30m using a microwave furnace. The purity of 99.9% for Co powders with a particle dimension lower than 150µm. The compositions of were figured out according to the variables in the formula Fex+Coy+Ni100-(x+y) (x+y= 40, 50, 66.66, 75 in at.%), that is, 50 at.% Fe, 25 at.% Co, Samples produced with powder metallurgy were prepared as 10g rectangular prism. The sample mixtures were shaped by uniaxial cold-hydraulic press using a high-strength steel mold. A compression of 400 bar was utilized to compress and shape the powder mixtures. Sintering time of 30 min at 1100 ° C in microwave oven was applied to the samples which were pressed cold chelated by using a neutral atmosphere. The specimens after sintering, the furnace was left to cool naturally. The hardness of the samples were compared with METTEST-HT hardness tester and Shimadzu AG-IS 100KN universal tensile equipment. Standard metallurgical sample preparation method was used to the samples. This was done to determine the microstructure in the sample. The contents of the coating bath are given in Table 1.

Table 1. The Nickel bath and ratios

	(Fe-%20Co)%60Ni	(Fe-%25Co)%50Ni	(Fe-%33Co)%33Ni	(Fe-%37,5Co)%25Ni
Iron(Fe)	8g	10g	14,06g	15g
Cobalt (Co)	4g	5g	7,03g	7,5g
Nickel Chloride ($NiCl_2.6H_2O$)	72g	60g	39,6g	30g
Hydrazine Hydrate ($N_2H_4.H_2O$)	20%	20%	20%	20%
Distile Water	80%	80%	80%	80%
Temperature (°C)	90-95°C	90-95°C	90-95°C	90-95°C
pH Value	10	10	10	10

Shimadzu XRD-6000 model that X-Ray Diffraction Analyzer was run with CuK alpha radiation at 2 degrees per-minute as scan value. LEO 1430 VP model Electron Microscope with Oxford EDX analyzer was utulized for sample microstructural properties and element composition analysis. In the impedance phase analyzer, 1 Hz-frequency frequency was selected. Nova Control Alpha Impedance Phase Analyzer (AEPA) was used in the temperature range 0-500 C. 1.5KW impedance analyzer produced by Hundsangen / Germany was used to measure electrical properties. The shaped (Fe + Co + Ni) samples were calculated using the volumetric variation of the composite samples after sintering (d = (m)mass / (V) volume) (Fig. 1). The capacity of composite specimens were compared with Archimedes' principle. Unless otherwise indicated, The percentages and ratios used in the article are given as weight percent for the samples.

2. Results

2.1. Characterization of specimens

In this paper, powdered (pressed) samples were prepared using powder metallurgy method and sintered in microwave sintering furnace at argon atmosphere at 1100ºC. The samples are prepared for density as physical properties, hardness and compression strength values from mechanical properties were examined. Electrical properties were investigated by SEM, X-ray analysis in metallographic examinations. The density-composition change curve of the samples sliced in the microwave is given in Figure 1. Density values for different compositions were determined. The highest density of composition was compared with 4,5gr/cm^3 in the (Fe-%37,5Co) %25Ni composition.

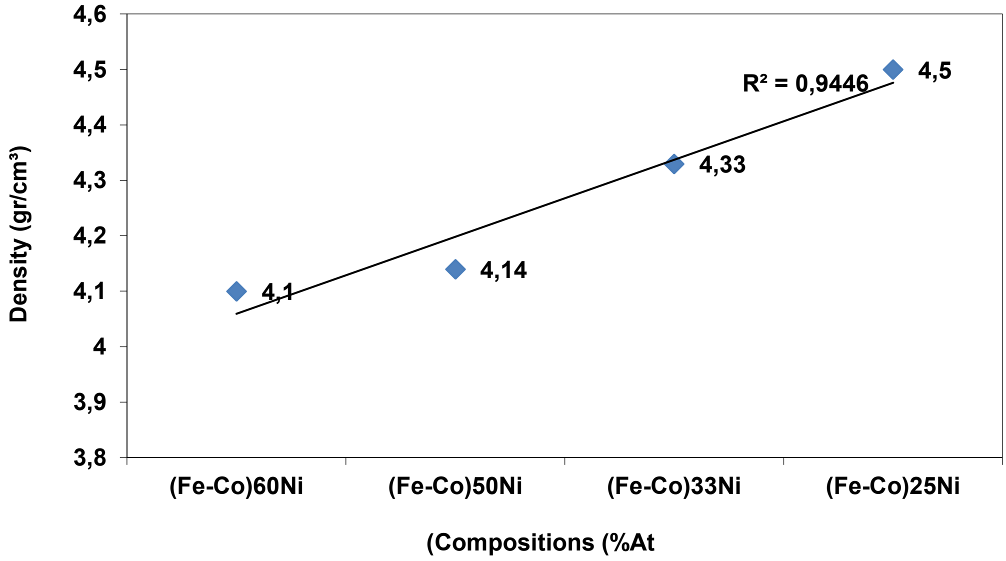

Fig.1. Density - Hardness - Compositions curve in specimens

The hardness-composition variation graph of samples sintered in the microwave sintering furnace is shown in Fig. 2. So that, As a result, the highest hardness value (Fe-37.5%Co)%25Ni of composite specimens fabricated by electroless Ni coating the way was measured as 78.18HB at (Fe- 37.5%Co)%25Ni composition.

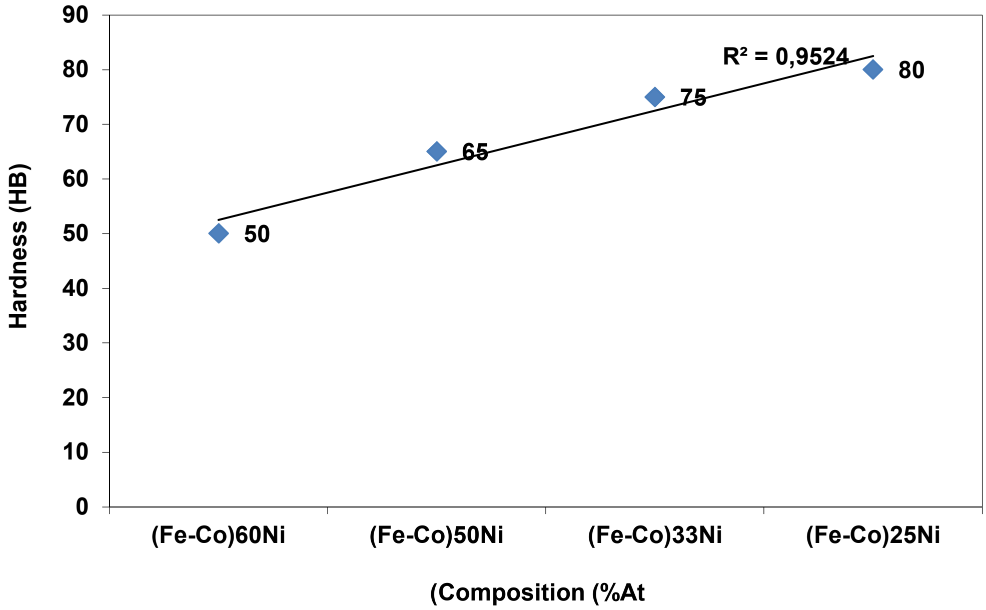

Fig.2. Hardness - Compositions curve in specimens

Electrical conductivity has already been referred to in discussing the bonding of atoms. It is largely determined by a lot of electrons in the conduction teritory, and by the number of of the holes in the valans band. Average distance traveled by electrons freely in metals will affect the value of electrical conductivity and its temperature coefficient. Resistance and conductivity were applied to determine the electrical properties of the materials produced. The properties of resistance and conductivity are inversely opposite to each other in materials. The resistance of a material describes how much difficulty it has passed to the passage of electrical current. Conductivity is generally represented by the Greek character sigma (σ) and compared with S m-1. Resistance (ρ) with resistance (R), cross section (S) and length (L) related formula: ρ = R.S / L Individual resistance values of components, Fe, Co and Ni in %60Ni coated Fe-Co Composite are given as 8.85x10-8Ωm, 6.24x10-8Ωm and 6.84x10-8Ωm at 20 °C, respectively. The total resistance of composite consisting of Fe, Co and Ni can be theoretically calculated with respect to their atomic fraction in the mixture as (0.27x8.85 + 0.13x6.24+0.6 x 6.84)10-8Ωm which is equal to 7.29 x10-8Ωm. Results showed that conductivity decreases with increasing frequency of the currect and it becomes completely non conductive at the frequency of 1MHz. Within 1KHz - 10KHz range, the change in resistance varies slightly and at 100KHz, the dependency of frequency on the temperature change becomes ineffective (Fig. 3).

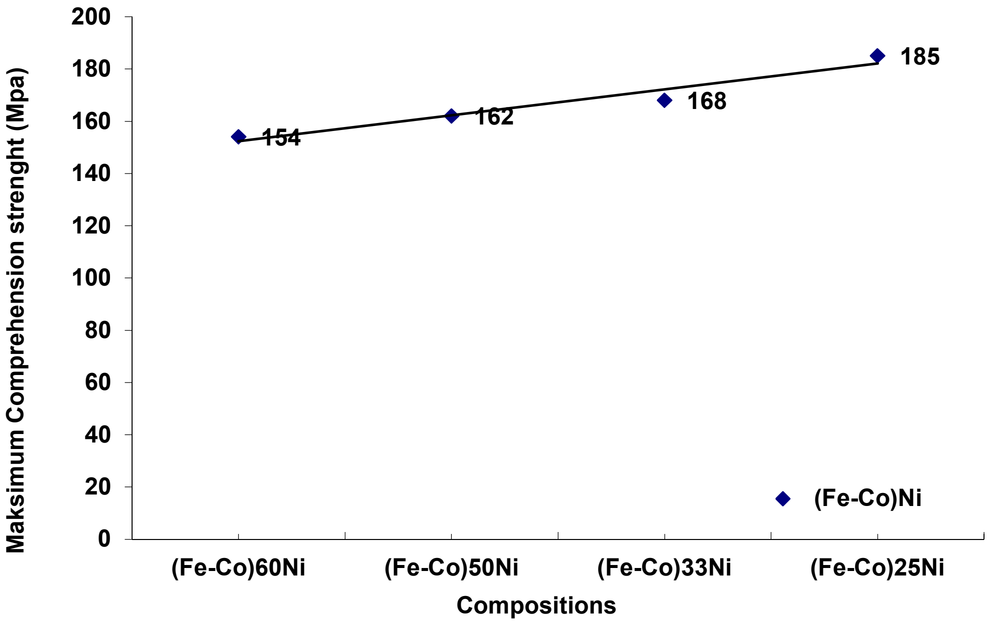

Fig.3. Comprehension Strenght - Compositions temperature curve in specimens

Electrical conductivity has already been referred to in discussing the bonding of atoms. It is largely determined by a lot of electrons in the conduction teritory, and by the number of of the holes in the valans band. The average path followed by electrons freely in metals will affect the value of electrical conductivity and its temperature coefficient. Resistance and conductivity were applied to determine the electrical properties of the materials produced. The properties of resistance and conductivity are inversely opposite to each other in materials. The resistance of a material describes how much difficulty it has passed to the passage of electrical current. Conductivity is generally represented by the Greek character sigma (σ) and compared with S m-1. Resistance (ρ) with resistance (R), cross section (S) and length (L) related formula: ρ = R.S / L Individual resistance values of components, Fe, Co and Ni in %60Ni coated Fe-Co Composite are given as $8.85x10^{-8}\Omega m$, $6.24x10^{-8}\Omega m$ and $6.84x10^{-8}\Omega m$ at 20 ºC, respectively. The total resistance of composite consisting of Fe, Co and Ni can be theoretically calculated with respect to their atomic fraction in the mixture as $(0.27x8.85 + 0.13x6.24+0.6 x 6.84)10^{-8}\Omega m$ which is equal to $7.29 x10^{-8}\Omega m$. Results showed that conductivity decreases with increasing frequency of the currect and it becomes completely non conductive at the frequency of 1MHz. Within 1KHz - 10KHz range, the change in resistance varies slightly and at 100KHz, the dependency of frequency on the temperature change becomes ineffective (Fig. 4).

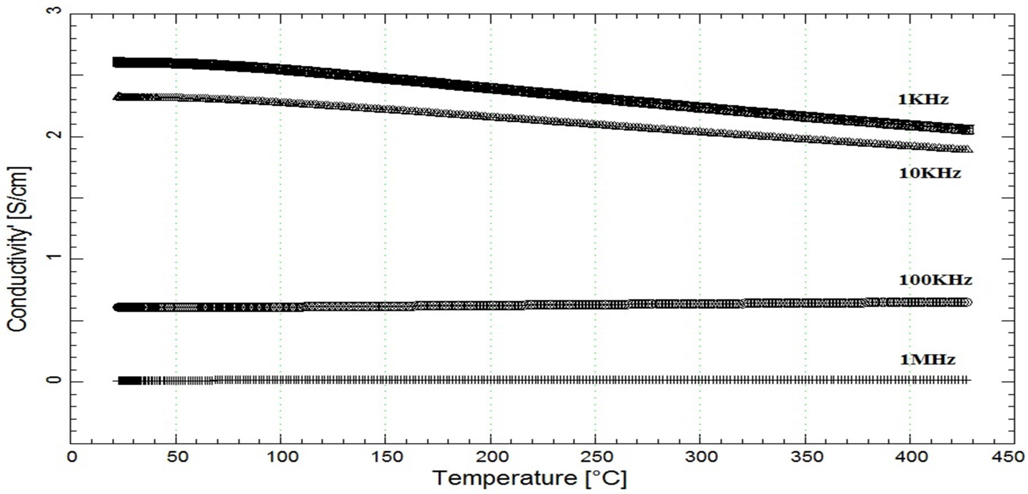

Fig. 4. AEPA graphs of The compositions-dependent change in conductivity -received %60Ni Coated Fe-%20Co Composite,

The theoretical total resistance of %50Ni Coated Fe-Co Composite was calculated as $(0.33x8.85+0.17x6.24+ 0.5x\ 6.84)10^{-8}\Omega m=6.86x10^{-8}\Omega m$ with the same method as given above. The change in composition eventually changed the total resistance from 2.6 S/cm in %60Ni coated Fe-Co to 2.8 S/cm in %50Ni coated Fe-Co composite compared with 1 KHz. In the range of 1-10KHz the conductivity decreases with increasing operating temperature. At 100KHz, the effect of temperature on the conductivity appears to be very small (Fig. 5).

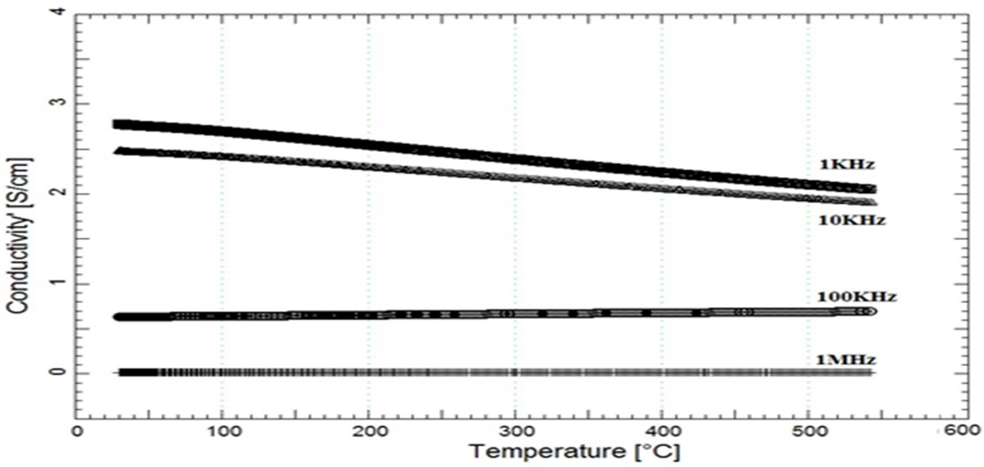

Fig. 5. AEPA graphs of The compositions -dependent change in conductivity -received %50Ni Coated Fe-%25Co Composite,

The theoretical resistivity of 33%Ni coated Fe-Co-Ni composites is given as (0.44x8.85+0.23x6.24+ 0.33x 6.84)$10^{-8}\Omega m$ = 7.57x$10^{-8}\Omega m$ and the resistance was experimentally measured to be 2.1 S/cm at 1KHz. Within 130-160ºC the conductivity fluctuates dramatically to increase but above 160C the conductivity decreases. As expected from other experiments that the conductivity becomes very low at 1MHz. The conductivity decreases with increasing temperature within 1KHz-10KHz, however, at 100KHz the effect is not considerable (Fig. 6).

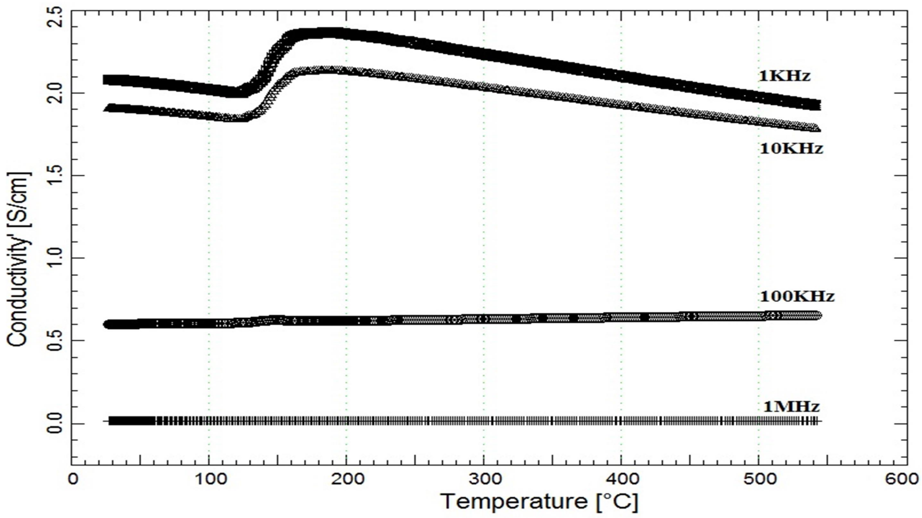

Fig. 6. AEPA graphs of The compositions-dependent change in conductivity -received %33Ni Coated Fe-%33Co Composite,

The effect of lower Ni addition on the total resistance was also studied with 25%Ni. The total theoretical resistnace was calculated as (0.5x8.85+0.25x6.24+ 0.25x 6.84)$10^{-8}\Omega m$=7.69 x$10^{-8}\Omega m$ but experimentally found to be 2.2 S/cm at 1KHz. At frequencies of 1 and 10 KHz and within the range of 40-500ºC, a distinctive fluctuation in the conductivity values was observed. At 100KHz the temperature effect was not important and it became nonconductive at 1MHz (Fig. 7).

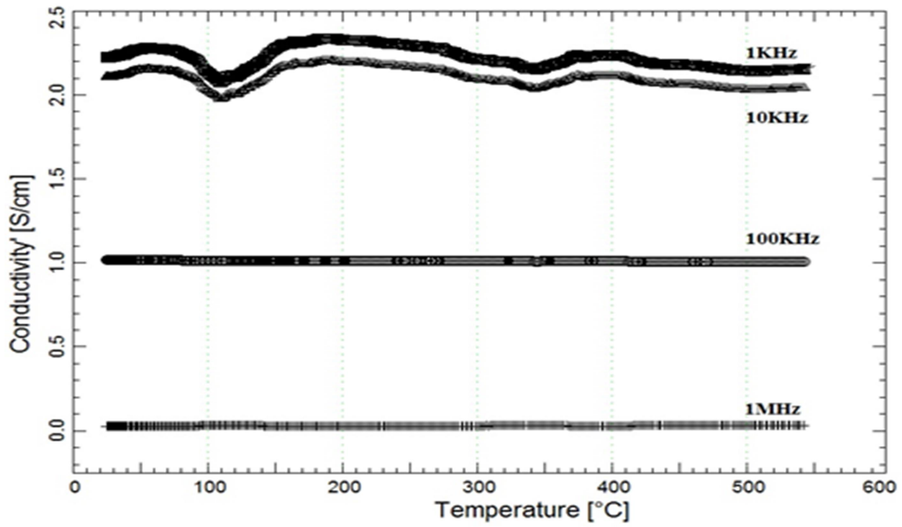

Fig. 7. AEPA graphs of The compositions-dependent change in conductivity -received %25Ni Coated Fe-Co Composite,

Metallographic analysis

Fe-Co powders are sintered by uniaxial press after sintering. In the SEM image grain growth was observed. It is seen that the boundaries between the particles become apparent. Intercalarily, There are pores in the grain boundaries that are homogeneously distributed in places (Fig. 8).

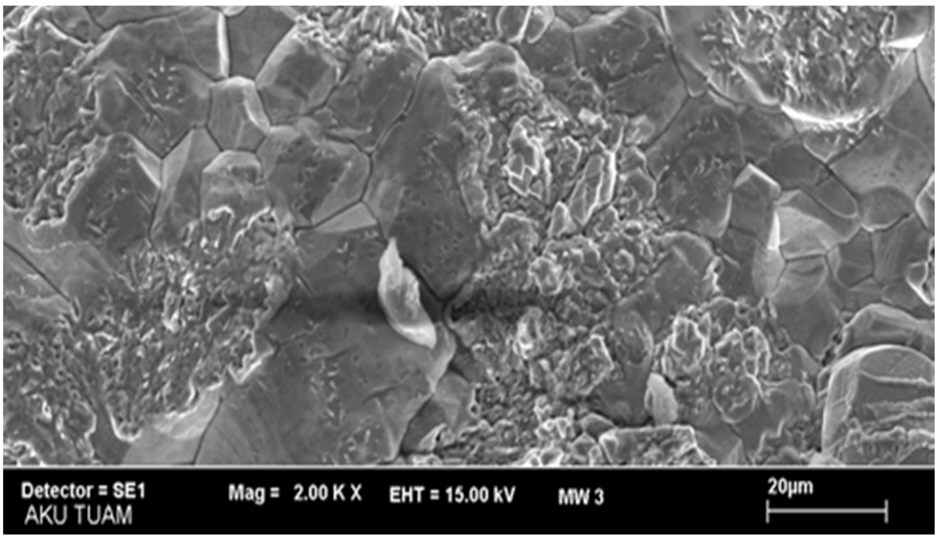

Fig. 8. SEM micrographs of %25Ni coated Fe-%37,5Co, Mag.2kX

Scanning Electron Microscopy (SEM) and XRD analyzes were performed in order to reveal the effect of Electroless Nickel coating on the particles and to identify the sample formed. Fig. 9, Microwave sintering at 1100°C for different compositions produced. In the samples, the non-flowing Ni coating formed the Ni layer with low pore density. It has shown a higher green product mechanical property prior to sintering process (Fig. 9).

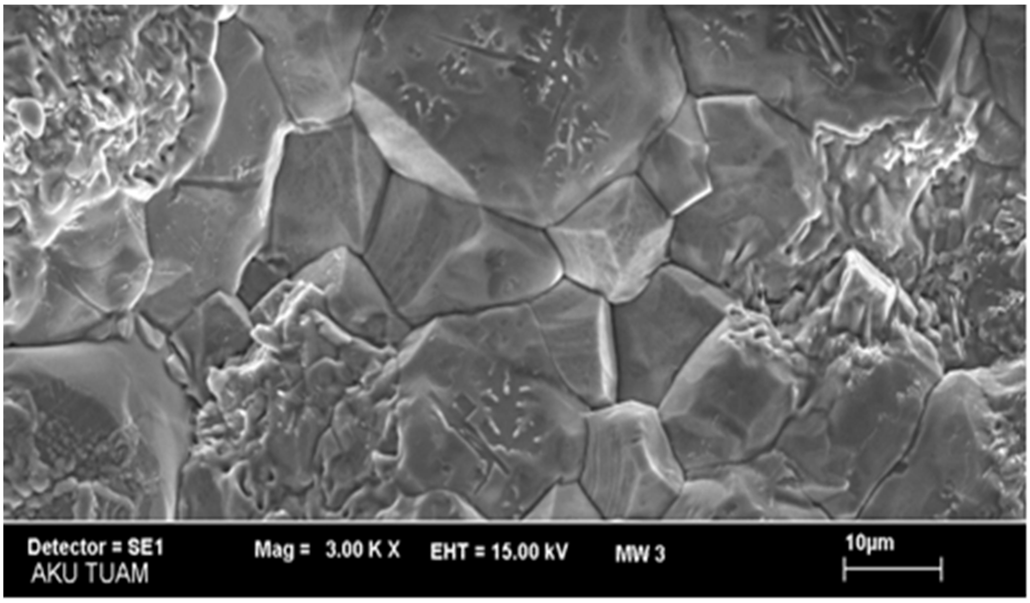

Fig. 9. SEM micrographs of %25Ni coated Fe-%37,5Co, Mag.3kX

It is given by linear EDX analysis on the sample of its (Fe-%37,5Co)%25Ni composition. As a result of the analysis, the peaks of Ni, Co and Fe elements were found. In addition to these elements, the oxygen peak was also found. The lack of high purity of argon gas during sintering is effective in seeing oxygen. As a result of the analysis, the elements show a homogeneous distribution on the linear line. (Figure 10).

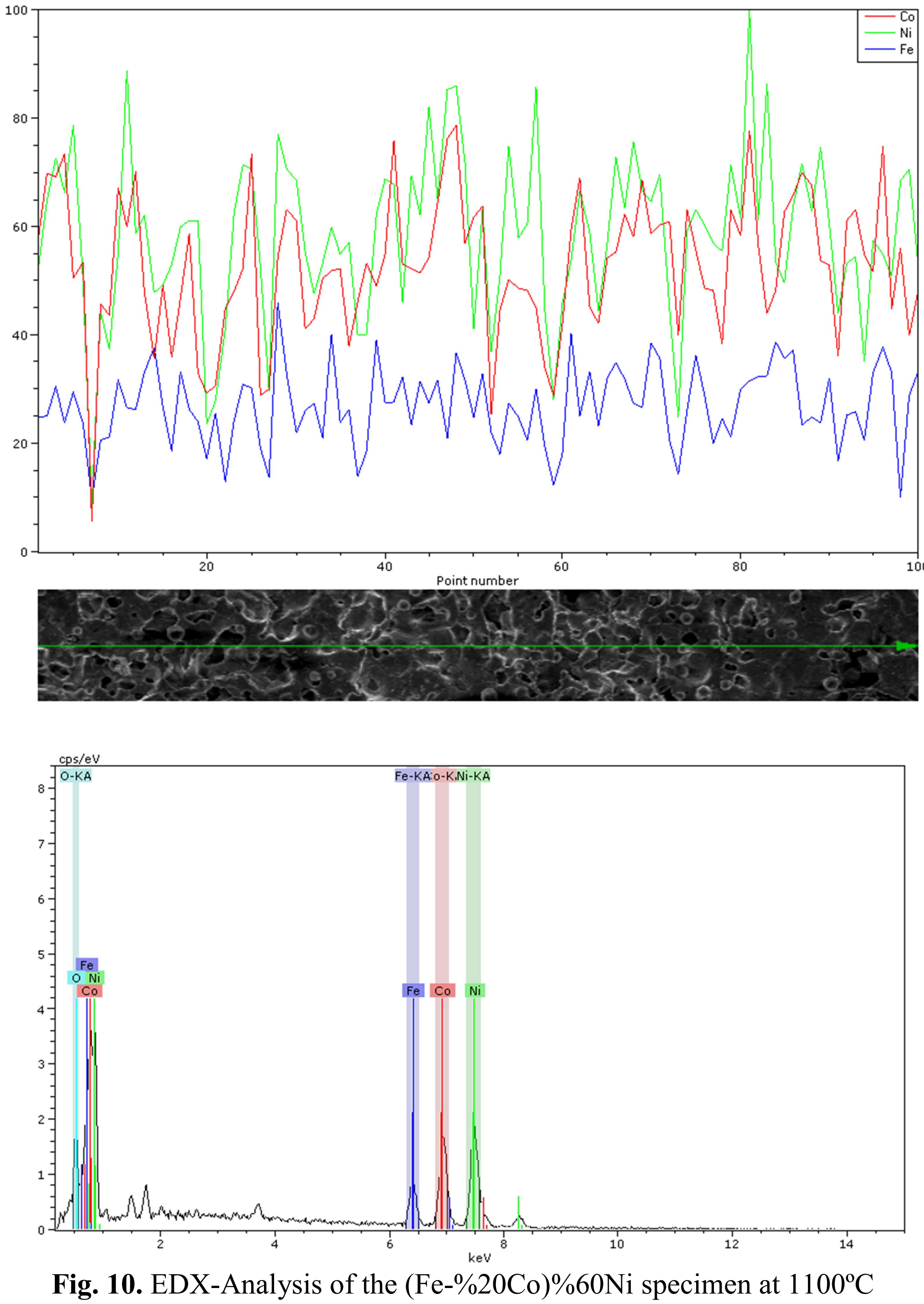

Fig. 10. EDX-Analysis of the (Fe-%20Co)%60Ni specimen at 1100ºC

In Figure 11, Ni, CoFe, $FeNi_3$ and FeNi phases climaxs can be observed in the XRD analysis from (Fe-%20Co)%60Ni composite specimens sintered at microwave oven at 1100°C. The further addition of Ni particles into the matrix appears to have increased the amount of $FeNi_3$ phase as seen in Fig. 10 and Fig.11. The intensity of peaks of FeNi in 25%Ni added specimens have more intermetallics phase compared to 60% Ni added specimens. This caused a decrease in the number of intermetallics phase, forming mostly $FeNi_3$ which has more Ni atoms since Ni is more available for the formation of higher Ni compounds. The increase of Ni also caused the appearance of pure Ni peaks. The Fe and Co,Fe phase was also affected by the addition of Ni.

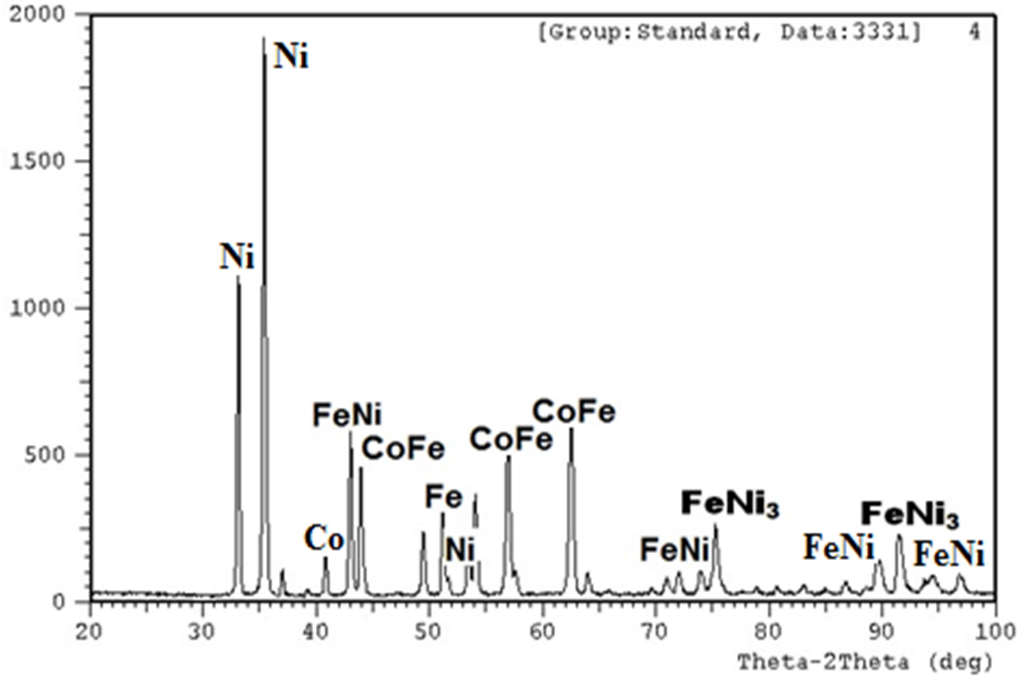

Fig. 11. X-Ray Diffraction of the (Fe-%37,5Co)%25Ni specimen at 1100ºC

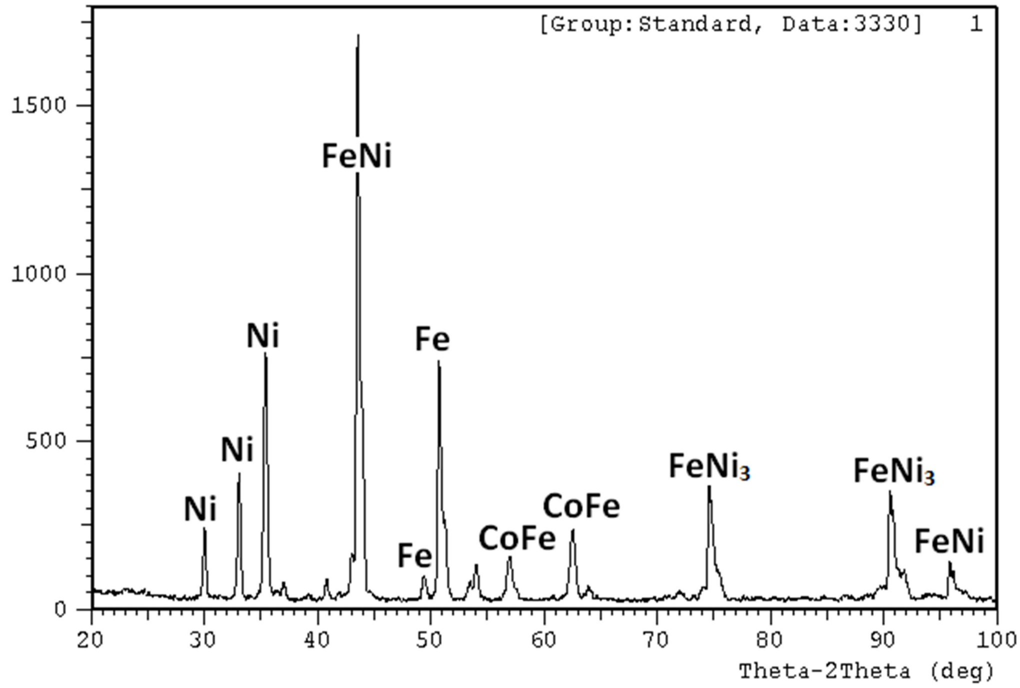

Fig. 12. X-Ray Diffraction of the (Fe-%20Co)%60Ni specimen at 1100ºC

4. Conclusion

Electroless Nickel coating process is a method that has many advantages in improving the mechanical properties of materials. It helps us to save energy by providing the advantage of syntax especially at low temperatures. It also shows resistance against oxidation in manufactured materials. It provides the advantage of sintering as well as easy shaping of powder materials. There is a direct relationship between theoretical and experimental resistance that has been obtained from Impedance Phase Analyzer. A resistance measurement could be made upto 100KHz above which the resistance was unreadable. Upto 500°C, the conductivity decreased linearly in 60% and 50%Ni additions but in 33% and 25%Ni additions there appears to be fluctions in the resistance values due probably to decreased conductivity with lower percentage of connecting Ni coated particles.

Acknowledgements

This study was supported by Afyon Kocatepe University. We would like to thank to TUAM for their support.

Declaration

Raw data were generated at Afyon Kocatepe University Technology Research and Application Center (TUAM). Derived data supporting the findings of this study are available from the corresponding author Ahmet YONETKEN on request.

Author Contributions

Conceptualization, A.Y.; methodology, A.Y.; software, A.Y.; validation, A.Y; formal analysis, A.Y.; investigation, A.Y.; resources, A.Y.; data curation, A.Y; writing—original draft preparation, A.Y.; writing—review and editing, A.Y.; visualization, A.Y.; funding acquisition, A.Y; All authors have read and agreed to the published version of the manuscript.

Funding

This research was funded by A. Yönetken

Institutional Review Board Statement

Not applicable.

Informed Consent Statement

Not applicable.

Data Availability Statement

The processed data necessary to reproduce these findings are available upon request with permission.

Conflicts of Interest

The authors declare no conflict of interest.

References

[1] R.M.Bozorth,Ferromagnetism,vol.V,D.VanNostrandCompany,NewYork, 1951 160.

[2] T. Osaka, Electrochim. Acta 45 (2000) 3311.

[3] X. Liu, G. Zangari, L. Shen, J. Appl. Phys. 87 (2000) 5410.

[4] S.U. Jen, H.P. Chiang, C.M. Chung, M.N. Kao, J. Magn. Magn. Mater. 236 (2001) 312.

[5] Y.M. Kim, D. Choi, S.R. Kim, K.H. Kim, J. Kim, S.H. Han, H.J. Kim, J. Magn. Magn. Mater. 226–230 (2001) 1507.

[6] T. Osaka, M. Takai, K. Hayashi, K. Ohashi, M. Saito, K. Yamada, Nature 392 (1998) 796.

[7] Suckov, A.E., Fiziko Metallov I Mettalovedene7 1959 p.p 317.

[8] Ghani, A.A. and Mirijasov, N.Z Sov.Phys.Sol.State (USA) 13 1972 p.p2627

[9] Reslescu, M.N., C.R.Hebd.Sean.Acad.Sci.263 1969 p.p 136

[10] Kulkarani V R, Todhar M M and Vaingankar A S 1986 Indian J.Pure. Appl. Phys. 24, p.p 294

[11] Rezlesus N and Rezlesus E Phys. Status. Soldi 23 1974 p.p 575 doi:10.1002/pssa.2210230229

[12] J. Hormes, J. Gottert, K. Lian, Y.andL. Jian, Nucl. Instr. Meth. Phys. Res. Sect. B-Beam Interact. Mater. Atom 2003, 199 , 332.

[13] J.W. Choi, T.M. Liakopoulos, C.H. Ahn, Biosens. Bioelectron. 16 (6) (2001) 409.

[14] Y.W. Yi, C. Liu, IEEE J. Microelectromech. Syst. 8 (1) (1999) 10–17.

[15] J.W. Judy, R.S. Muller, Sens. Actuators (Phys. A) A53 (1996) 392.

[16] J.W. Judy, R.S. Muller, IEEE J. Microelectromech. Syst. 6 (1997) 249.

[17] C.H. Ahn, M.G. Allen, IEEE Trans. Ind. Electron. 45 (1998) 866.

[18] T.M. Liakopoulos, M. Xu, C.H. Ahn, in: Proceedings of the Technical Digest Solid-State Sensor and Actuator Workshop, vol. 19, Hilton Head Island, SC, USA, 1998.

[19] T.S. Chin, J. Magn. Magn. Mater. 209 (2000) 75.

[20] J.W. Dini, Electrodeposition, The Material Science of Coatings and Substrates, Noyes Publications, 1992, p. 292.

[21] Yönetken, A. Fabrication of Electroless Ni Plated Fe–Al_2O_3 Ceramic–Metal Matrix Composites, *Trans Indian Inst* Met 2015. **68**(5), 675-681. https://doi.org/10.1007/s12666-014-0497-1

[22] Yönetken, A , Erol, A . Sintering and Characterization of SiC Reinforced Ni Powders in Microwave Furnace. International Journal of Engineering Research and Development, 2020, **12** (1) , 83-89 . DOI: 10.29137/umagd.474003

[23] Ban, Z.G., Shaw, L. L., 2002. Synthesis and processing of nanostructured WC-Co materials, Journal Of Materials Science, (37), 3397 – 3403

[24] Upadhyaya G.S., Bhaumik S.K.,1988. Sintering of submicron WC-10wt.%Co Hard Metals Containing Nickel And Iron Material Science And Engineering-A, Volume:249, 105–106, 1988.

[25] Nagayama, T., Yamamoto T., Nakamura T., 2016 Thermal expansions and mechanical properties of electro-deposited Fe–Ni alloys in the invar composition range. lectrochim. Acta 205, 178–187

[26] Y. Liu, L. Liu, Z. Wu, J. Li, B. Shen, W. Hu, 2010, Grain growth and grain size efects on the thermal expansion properties of an electro-deposited Fe–Ni invar alloy. Scr. Mater. 63, 359–362

[27] Peng, Y., Zhu, Z., Chen, J., Ren, J., Han T., 2014, Research on pulse electro-deposition of Fe–Ni alloy. AIP Adv. 4, 31301–31305.

[28] Matsui I., Mori H., Kawakatsu T., Takigawa Y. Uesugi T., Higashi K., 2015, Mechanical behavior of electrodeposited bulk nanocrystalline Fe–Ni alloys. Mater. Res. 18, 95–100

[29] S. Sarkar, S. Pande, S. Jana, A.K. Sinha, M. Pradhan, M. Basu, S. Saha, S.M. Yusuf, T., 2009, pal, J. Phys. Chem. C 113 6022–6032.

[30] C. Xu, S. Sun, Dalton Trans., doi:10.1039/B900272N.

[31] O.B. Miguel, M.P. Morales, P. Tartaj, J.R. Cabello, P. Bonville, M. Santos, X. Zhao, S.V. Verdaguer,2005, Biomaterials 26 5695–5703.

[32] T. Jafari, A. Simchi, N. Khakpash, J. 2010, Colloid Interface Sci. 345, 64–71.

[33] K.S. Suslick, M. Fang, T. Hyeon,1996, J. Am. Chem. Soc. 118, 11960–11961.

[34] C.W. Kim, H.G. Cha, Y.H. Kim, A.P. Jadhav, E.S. Ji, D.I. Kang, Y.S. Kang, J. 2009, Phys. Chem. C 113, 5081–5086.

[35] Alagiri M., Muthamizhchelvan C., Ponnusamy S., 2011, Structural and magnetic properties of iron, cobalt and nickel nanoparticles, Synthetic Metals 161, 1776–1780, doi:10.1016/j.synthmet.2011.05.030