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Arch. Metall. Mater. **65** (2020), 4, 1277-1280

DOI: 10.24425/amm.2020.133684

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WAVE ABSORPTION PROPERTY OF HIGH ENTROPY ALLOY FABRICATED BY SOL-GEL PROCESS

In this study, the magnetic properties and wave absorption characteristics of high entropy alloys are investigated. The high entropy alloys with FeNiMnCoCu, FeNiMnZnCo, and FeNiZnCoCu compositions were synthesized by the sol-gel method. After the sol-gel process, the annealing process and hydrogen reduction process was performed. FeNiMnCoCu and FeNiZnCoCu were revealed soft magnetic property. The saturation magnetization was 12 emu/g and 36 emu/g, respectively. And The coercive force was –45 Oe and –34 Oe, respectively. The high entropy alloy with these compositions was revealed wave absorption property at above 10 gigahertz frequency region. And it has shown the trend that wave absorption frequency has decreased with the sample thickness increasing.

Keywords: High entropy alloy, Wave absorption property

1. Introduction

High entropy alloys(HEA) can be defined in two ways. The first definition is based on composition, and the second definition is based on configuration entropy. The definition by composition is a single-phase crystalline alloy composed of 5~35 at% of five or more atoms. And the definition by configuration entropy is defined as an alloy whose configuration entropy (ΔS_{conf}) is greater than 1.6R (R is gas constant), regardless of the number of phases at room temperature [1]. In general, multi-element alloys had characterized in that when the number and amount of alloying elements increase, intermetallic compounds are formed and the mechanical properties of the material become weak [2]. However, since high entropy alloys cannot be identified as major atoms, high entropy alloys form high configuration entropy and thus do not form intermetallic compounds, and do not become weak. The lattice structure of HEA has BCC, FCC and mixed phases thereof. Such high entropy alloys are known to increase in strength even when lattice deformation occurs due to the mixing of elements with similar atomic radii [3]. Since HEA exhibits excellent physical properties, researches to develop and utilize them is continuously conducted. However, we have confirmed that HEA had properties as wave absorbers. A wave absorber shall exhibit at least one of the following three loss mechanism: magnetic loss, conduction loss, and dielectric loss. The most commonly used loss as a wave absorber is known as magnetic loss. We will check the magnetic properties of HEA, measure the permittivity and permeability, and confirm the propagation and absorption properties of HEA. As a result, it is intended to study the wave absorber that has excellent physical properties of HEA.

2. Experimental

In this study, HEA(High Entropy Alloy) of 5 atoms system was synthesized by sol-gel process. The compositions of HEA were FeNiMnCoCu (FNMCC), FeNiMnCoZn (FNMCZ), FeNiZnCoCu (FNZCC). Each HEA is designed with 5 elements of 20 wt%. The starting materials used in the sol-gel process were $Fe(NO_3)_3 \cdot 9H_2O$(98%), $Ni(CH_3COO)_2 \cdot 4H_2O$(99%), $Zn(CH_3COO)_2 \cdot 2H_2O$(99%), $Cu(CH_3COO)_2 \cdot H_2O$(99%), $Co(CH_3COO)_2$(99%), $Mn(NO_3)_2 \cdot 4H_2O$(98%). The starting materials were mixed in the reaction bath by stoichiometry. And the same moles of citric acid($C_6H_8O_7$) and ethylene glycol($C_2H_6O_2$) as starting materials were added to the mixture. D·I water was added to dissolve the mixture. And ammonia solution (NH_4OH) was added until pH6 of the mixture solution. The reflux device was installed on the upper reaction bath, and 5°C cooling water was circulated in this. Ammonia solution was added to the mixture until the hydrogen ion concentration change to pH6. The temperature of the mixture solution was controlled by an outer

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thermocouple. The mixture had been heated to 800°C, stirred as 400 rpm, and kept 12 hours for make the sol. The sol was heated and stirred to 100°C without the reflux device on a hotplate for water evaporation to make the gel. And the gel was annealed at 900°C for powder synthesis. The heating rate was 5°C/min and kept 3 hours. Then, the powders had undergone a hydrogen reduction process. The hydrogen reduction process had processed during 6hours at 900°C. Made like this high entropy alloys were analyzed by XRD, TEM, VSM and network analyzer to confirm the lattice structure, magnetic properties, and wave absorption property.

3. Results and discussion

In this study, We had synthesized HEAs, that the composition of FNMCC, FNMCZ, FNZCC (F:Fe, N:Ni, M:Mn, C:Co, C:Cu, Z:Zn) by sol-gel process and hydrogen reduction process. Each element had the same molar ratio of 20 wt%. XRD analysis was performed for the lattice structure analysis of the synthesized HEA.

XRD patterns of FNMCC, FNMCZ, FNZCC were displayed in Fig. 1. Three patterns in Fig. 1 is revealed the peak of broad width. It means that synthesized powder is HEA, as the characteristic of HEA's XRD pattern. The black line is the pattern of FNMCC, the red line is FNMCZ, and the blue is FNZCC. The FNMCC and FNMCZ have revealed the same degree of deflection peaks, and it was confirmed phase of FCC and BCC. And The pattern of FNZCC is confirmed that is a pattern of BCC.

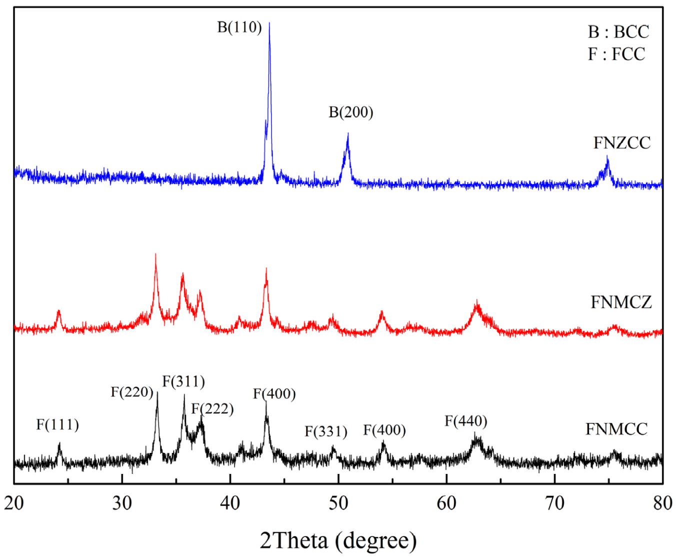


Fig. 1. XRD patterns of FeNiMnCoCu, FeNiMnCoZn, FeNiZnCoCu

In order to confirm the magnetic property of High entropy alloys(HEA), VSM analysis was processed. And the results of VSM analysis were displayed in Fig. 2 as M-H hysteresis loop. FNMCC and FNZCC revealed the saturation magnetization of 12 emu/g and 36 emu/g, respectively, and it's the coercive force was –45 Oe and –35 Oe, respectively. Because of the HEA has lattice structure of random arranged atom, it seemed it has a paramagnetic property. Because the arrangement of atoms is not constant, it seems that the saturation magnetization and coercive force had a low value. And FNMCZ revealed non-magnetic property as saturation magnetization was 0.3 emu/g, coercive force was –11 Oe.

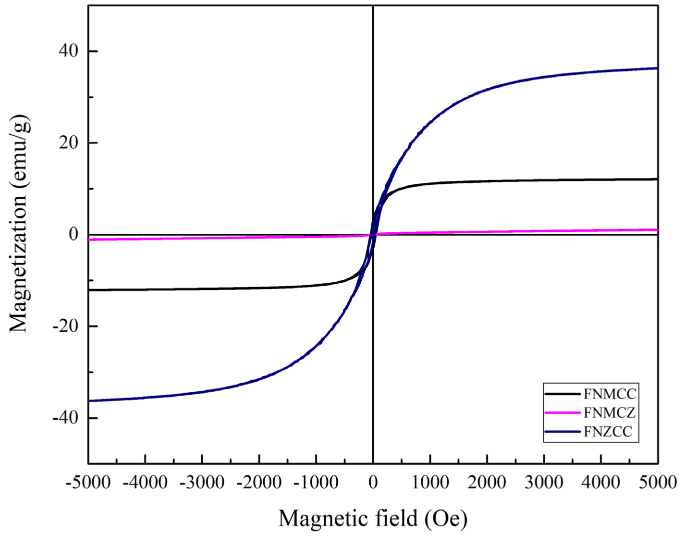


Fig. 2. M-H hysteresis loop of FeNiMnCoCu, FeNiMnCoZn, FeNiZnCoCu, annealing temperature at 900°C for 3 hours

The real and image of permittivity and permeability were measured using a network analyzer to verify the radio wave absorption property of the high entropy alloy. The results were calculated to confirm the reflection loss using equation 1 and 2.

$$\Gamma = \frac{Z_L - Z_0}{Z_L + Z_0} \tag{1}$$

$$\text{Reflection loss} = -20\log|\Gamma|^2 \tag{2}$$

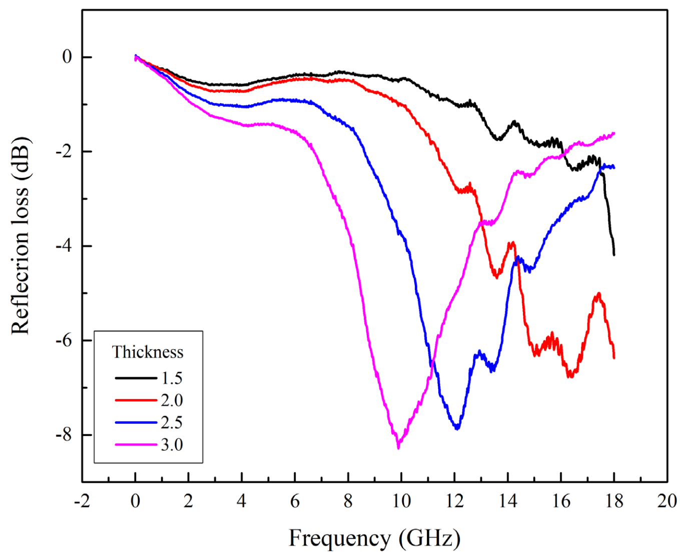


Fig. 3. Reflection loss of FeNiMnCoCu according to thickness with paraffin wax binder

The results of the calculated reflection loss of each composition were displayed in Figures 3, 4 and 5. Both FNMCC and FNZCC composition showed the change of reflection loss characteristics with thickness. As the thickness of the sample increases from 1.5 mm to 3 mm, the frequency of reflection loss decreased. It can be inferred that the sample of FNMCC 1.5 mm will appear above 18 GHz. It can be seen that as the thickness of the sample increases, the frequency at which reflection loss occurs decreases to 10 GHz. Also, this can be seen in the FNZCC. It can be inferred that the 1.5 mm and 2.0 mm samples will exhibit reflection loss frequencies above 18 GHz. The 2.5 mm and 3.0 mm samples showed reflection loss frequencies at 13.6 GHz and 11.9 GHz, respectively.

Reflection loss showed that increased with sample thickness increasing. FNMCC showed a reflection loss of –7.2 dB at 2.5 mm and –8.3 dB at 3.0 mm. FNZCC showed a reflection loss of –9.8 dB at a 2.5 mm sample and –10.8 dB at a 3 mm sample. This shows that FNMCC and FNZCC exhibit wave absorption characteristics by the same loss mechanism.

In order to confirm the loss mechanism of FNZCC and FNMCC, permittivity and permeability were displayed in Fig. 6, 7. Comparing the tangent loss of permeability and permittivity with the reflection loss frequency, it can be seen that the reflection loss frequency was not matched with tangent loss. Thus, it can confirm that FNMCC and FNZCC have a conduction loss, not magnetic loss and dielectric loss.

Since the waves be absorbed and converted into heat, the conduction loss is closely related to the resistance. Therefore, the reflection loss of copper contained FNMCC and FNZCC is higher than that of FNMCZ, and it can be said that the change

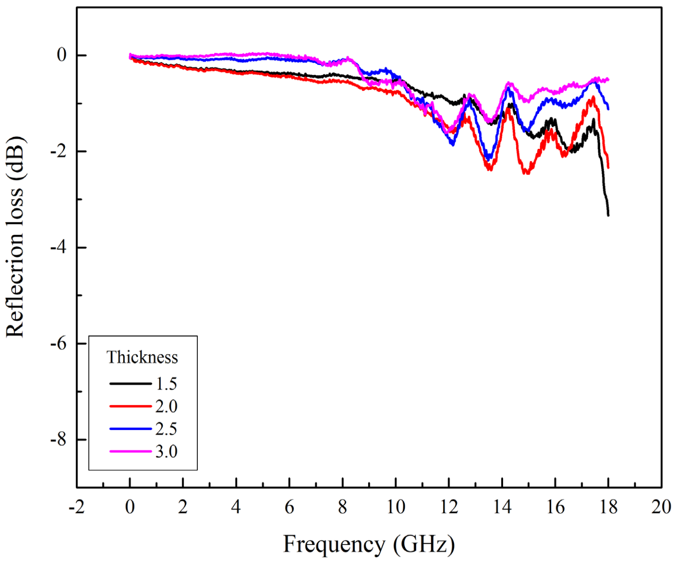


Fig. 4. Reflection loss of FeNiMnCoZn according to thickness with paraffin wax binder

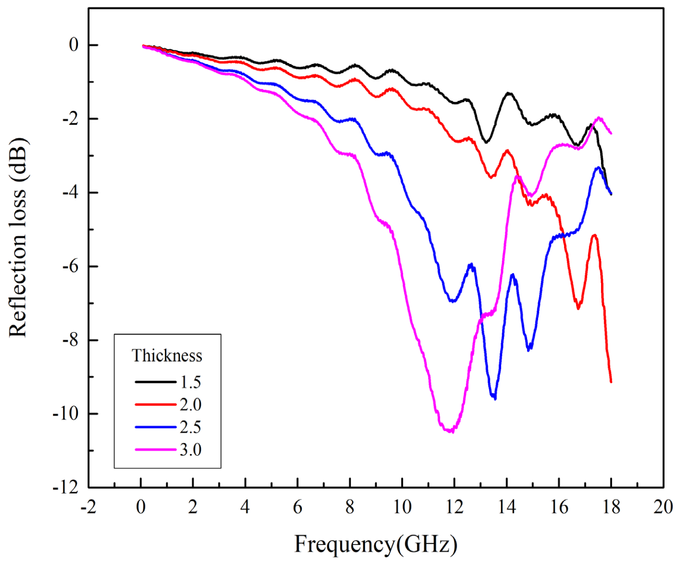
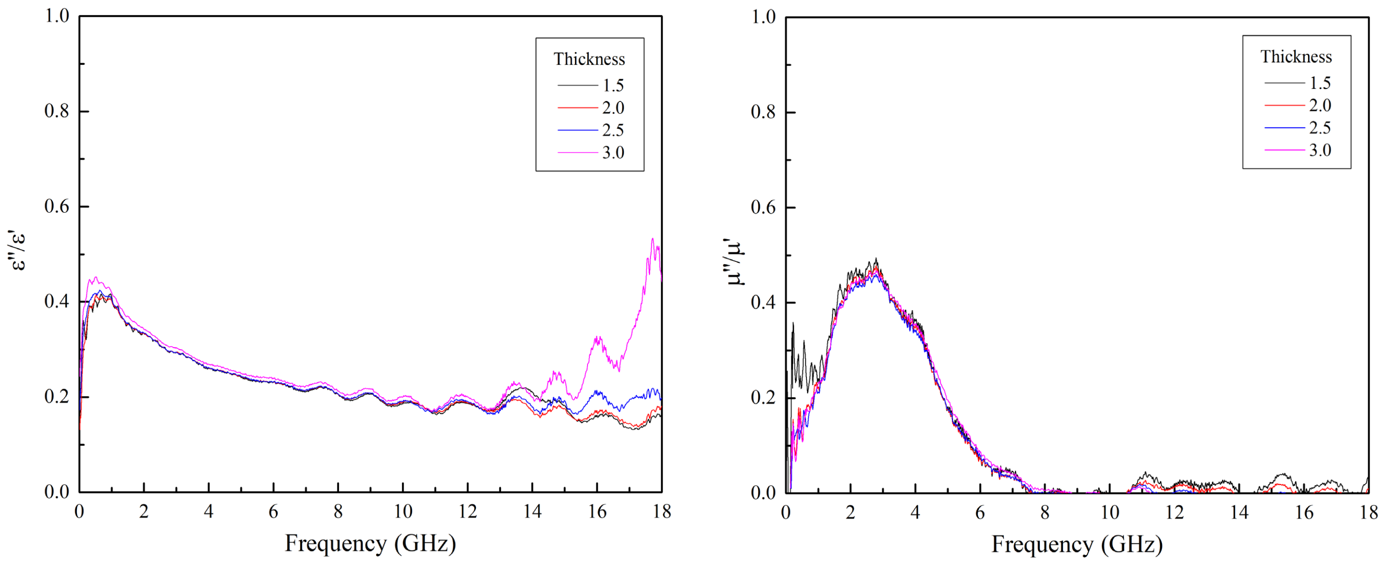


Fig. 5. Reflection loss of FeNiZnCoCu according to thickness with paraffin wax binder



a) Tangent loss (ε''/ε') b) Tangent loss (μ''/μ')

Fig. 6. Loss tangent of a) permittivity and b) permeability (image/real), FeNiMnCoCu according to thickness with paraffin wax binder

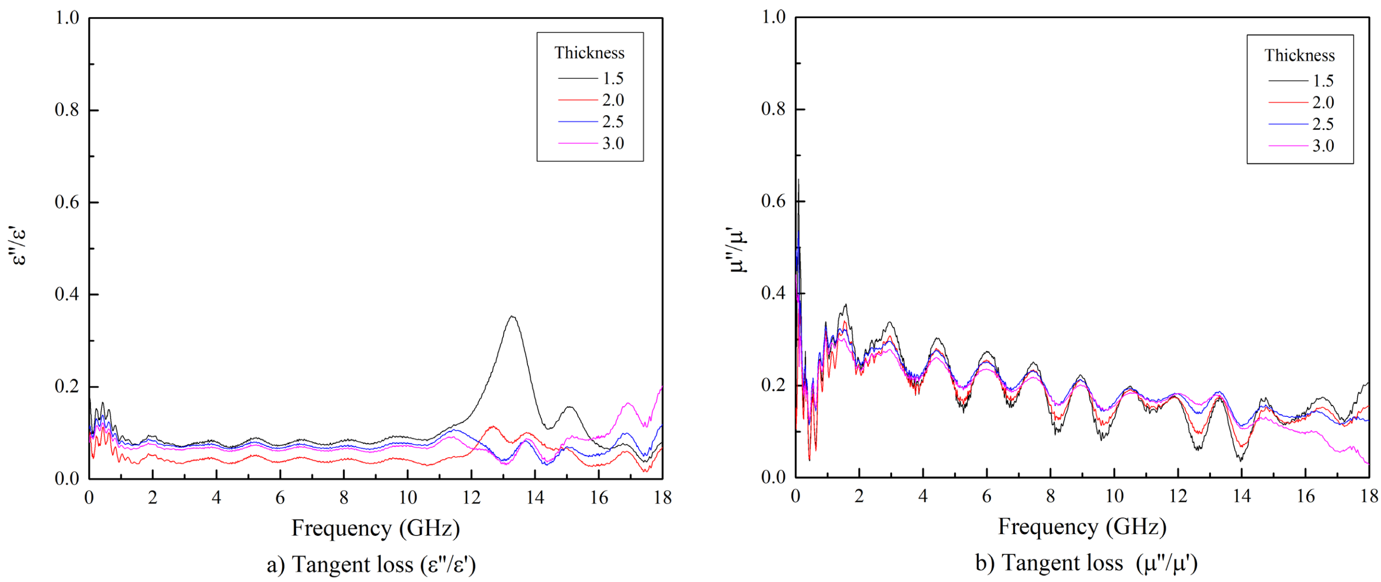


Fig. 7. Loss tangent of a) permittivity and b) permeability (image/real), FeNiZnCoCu according to thickness with paraffin wax binder

of frequency according to thickness is related to this. The high entropy alloy synthesized in this experiment can be used as an x band (8~12 GHz) as the wave absorber due to conduction loss.

4. Conclusions

In order to confirm the application of wave absorbers utilizing magnetic properties of high entropy alloys, powders of 5-atom system high-entropy alloys, FNMCC, FNZCC, and FNZCC, were synthesized by the sol-gel process. Segregation occurred during the annealing process. In this study, FNMCC and FNZCC showed soft magnetic properties as the saturation magnetization were 12 emu / g and 36 emu / g, and the coercivity was –45 Oe and –34 Oe. As a result of measuring the reflection loss, the FNMCC and FNZCC showed the appropriate frequency wave absorption characteristics as X-band wave absorbers. It is confirmed to be caused by conductive loss. Changing the composition and thickness of the high entropy alloy can change the frequency of a wave absorber.

Acknowledgments

This work was supported by Incheon National University Research Grant in 2018.

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