# Characterization of Ag-Ni nanocomposites in amorphous SiO<sub>2</sub> glass synthesized by silver and nickel dual ions implantation

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### 1 Introduction

It is known that metal nanoparticles (NPs) exhibit some specific properties, which are not appeared in bulk materials, such as catalytic activities, magnetic properties, electric conductivity and light absorption. Their unique properties come from the size, shape, structure, alloying state and so on. Therefore, many researchers trying to synthesize various kind of nanoparticles. Most metal nanoparticles are fabricated by chemical reduction method. This method has some advantages that is mono dispersive stable nanoparticle in large quantities. On the contrary, a peculiar state of nanoparticles such as non-equilibrium phase alloy ones are not possible to synthesize by the method of ion irradiation into solid. So far, we have been trying to synthesize new alloy nanoparticles in solid by metal ions irradiation. The ion irradiation method is a method that can be injected with highly controlled amount and ion species. In addition, it is reported that some metals nanoparticles formed during ion implantation without annealing. Therefore, we have challenging to synthesize alloy nanoparticle, which is not solid solution mixed at room temperature in phase diagram, by dual ions implantation. Also, the structure and light absorption of nanoparticles fabricated by this method were estimated by X ray diffraction and UV-vis spectroscope.

Previously, we have successfully synthesized pure Ag nanoparticles with control the size from 5 to 20 nm diameter in amorphous SiO<sub>2</sub> glass by Ag ion implantation. Also, the shape of Ag nanoparticle successfully modified from the spherical to elliptic shape by swift heavy ion irradiation [1,2]. In this study, we have tried to synthesize Ni-Ag alloy nanoparticles. In this alloy system, nickel and silver atoms are not dissolve in wide chemical composition. In this study, we been trying to synthesize Ni-Ag nanoparticles into SiO<sub>2</sub> glass by changing order of irradiation ions. After irradiation, all samples were characterized the light absorption, local structure, local atomic bonding and electronic state.

## 2 Experimental

Transparent amorphous silica glass  $(SiO_2)$  plates  $(5 \times 5 \times 1 \text{ mm}^3)$  were prepared as an implantation target material. The targets were irradiated with 380 keV Ag ions and 200 keV Ni ions at room temperature by using ion implanter at Takasaki Advanced Accelerator Research Institute, Institutes for Quantum and Radiological Science and Technology (QST-Takasaki). Dual ions implantation has done by changing order. Figure 1 shows a schematic image of Ag and Ni dual ions implantation into SiO<sub>2</sub> glasses. Ag ion implantation into SiO<sub>2</sub> followed by Ni ion implantation, and the other is vice versa. The implantation fluence of Ag ions was from  $5 \times 10^{15}$  to  $7 \times 10^{16}$  /cm<sup>2</sup> and that of Ni ions was from  $1 \times 10^{15}$  to  $1 \times 10^{17}$  /cm<sup>2</sup>. After each ion implantation, the optical absorption spectra were measured by Ultraviolet visible absorption spectroscopy and characterized nanostructure by using the positron annihilation coincidence Doppler broadening (CDB) measurement, Grazing Incidence X-ray Diffraction (GIXD), and X-ray photoelectronic spectroscopy (XPS) at BL27A, KEK photon factory. We define the first Ni ion implantation followed by Ag ion implantation as NA, and reverse order implantation as AN.



Fig.1 Schematic illustration of Ag implantation for SiO<sub>2</sub> glass followed by Ni ion implantation for same target.

#### 3 Results and Discussion

Figure 2 shows that photographs of SiO<sub>2</sub> glasses with Ag and Ni ions implantation. It is seen that the color difference between these four kinds of samples Ag, Ni, NA and AN implantation. These differences measured by UV-vis absorption spectroscopy showing that the intensity and position of absorption peak originated from surface plasmon resonance (SPR) of metal nanoparticles formed in SiO<sub>2</sub> clearly different as shown in figure 3. SPR peak of firstly Ag ion irradiated sample is broadened than that of revers ordered irradiated sample. In general, SPR



Fig.2 Photographs of (a)Ag, (b)Ni, (c)Ag <- Ni, (d)Ni <- Ag ions implantated SiO<sub>2</sub> glasses.



Fig.3 UV-vis spectra of (a) AN: Ag <- Ni and (b) NA: Ni <-Ag ions implanted SiO<sub>2</sub> glasses.



Fig.4 GIXD spectra of Ni and Ag ions implanted  $SiO_2$  glasses. (a) AN and (b) NA

absorption profile depends on a kind of chemical element of nanoparticle and its shape. Therefore, this result showing that the structure and/or electronic state of formed nanoparticles by Ni and Ag ions implantation with different irradiation order NA and AN is not same.

Figure 4 shows the focused GIXD patterns for Ni and Ag implanted SiO<sub>2</sub>. As seen in this figure, diffraction peaks around 44 degrees, which shows Ni (111) or Ag (200) plane, appeared a single peak for NA, but split it for AN. Also, both peaks dose not match for that of pure Ag and Ni. This result reveals that the formed nano composites with changing implantation order may have lattice distortion and their structure is not same, even if the same concentration (irradiation fluence) of ions are irradiated. Therefore, the irradiation sequence of multi-ions can be considered as the control parameter of nanocomposite formation in solids. These results well agree with the result of XPS measurement. Figures 5 and 6 show Ni  $2p_{3/2}$ ,  $2p_{1/2}$ and Ag  $3d_{5/2}$ ,  $3d_{3/2}$  core level XPS spectra of AN and NA irradiated SiO<sub>2</sub>, respectively. No peak shifts are observed in these figures, but the peak intensities, which indicate the amount of metal bonds, are reversed between AN and NA. Therefore, the irradiation order of Ni and Ag ions is considered to be a control parameter for nanocomposite formation.



Fig.5 Ni- $2p_{3/2}$  and  $2p_{1/2}$  core level XPS spectra for SiO<sub>2</sub> glasses with Ag and Ni ions irradiation (AN).



Fig.6 Ag- $3d_{5/2}$ ,  $3d_{3/2}$  core level XPS spectra for SiO<sub>2</sub> glasses with Ni and Ag ions irradiation (NA).

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#### References

[1] T.Yamada, K.Fukuda, S.Semboshi, Y.Saitoh, H.Amekura, A.Iwase and F.Hori, Nanotechnology **31**, No.45 (6) 455706 (2020).

[2] T. Yamada, A. Takano, K. Sugita, A. Iwase, M. Maekawa, A. Kawasuso and F. Hori, Trans. Mat. Res. Soc. Japan **45**, [4] pp.127-130 (2020).

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