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無容器浮遊凝固した SiO_2-FeO 非晶質の強磁性

Magnetic properties of glass FeO-SiO_2 by containerless levitation method

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1. Introduction

Iron containing silicates are interested in the fields of earth science, material science and steel process engineering. In materials science, magnetism by iron ions in silicates is interested, therefore the binary system of Fe_3O_4-SiO_2 is investigated from the magnetics viewpoints [1)]. However, the binary system of FeO-SiO_2 is not much investigated because magnetic properties are not expected from its equilibrium phase diagram [2)]. Recently, the magnetism of FeO-CaO-SiO_2 system glass was reported [3)]. The magnetism would be Fe^{3+} ion segregation in the glass metrics. Since the FeO-CaO-SiO_2 glass was easy to make glass, the initial glass samples were made by quenching of high temperature liquids using cooling plate. Then by annealing the glass samples with several conditions, magnetic properties were investigated. From this investigation, we try to make a FeO-SiO_2 glass to expect the magnetism as same as FeO-CaO-SiO_2 system. However, FeO-SiO_2 glass has not been made by the same method of previous research. We adopted the containerless method by using aerodynamic levitation to make glass of FeO-SiO_2 system in FeO rich compositions. The glass samples made under some conditions of cooling rate and the atmospheric gas showed the magnetism. In this report, we show the details of sample preparation of FeO-SiO_2 glass and the results of magnetic properties measurements.

2. Experimental Procedure

2.1. Sample preparation

A master samples of FeO:SiO_2=40:60mol% were prepared from reagent grade, SiO_2, FeO powders (KOUJYUNDO Ltd.). Powders of SiO_2 and FeO were dried at elevated temperatures to 300℃ with 14 h in order to evaporate H_2O contained in raw powders. A weighed mixture of the oxides was placed in a Cu cooled crucible and sintered at 1000℃ in the dried air by CO_2 laser irradiations to make a shape of sample to levitate by the aerodynamic levitation (**Figure 1**). The sintered sample with a diameter of about 2mm was levitated using the aerodynamic nozzle to achieve containerless melting and solidification. Levitation conditions were in dried air and sample melting was achieved by the CO_2 laser irradiations. Highest temperature for melting samples and cooling rate to solidification were variously changed. The sample temperature was monitored by

single color pyrometer with emissivity of one because the emissivity value of FeO-SiO_2 melt has not been obtained and reported still yet. **Figure 2** shows example of temperature profile during melting and solidification. From the result, we did not find the recalescence phenomena of crystallization from the undercooled melt with releasing laten heat of solidification. Therefore, the samples were formed glass with amorphous structure.

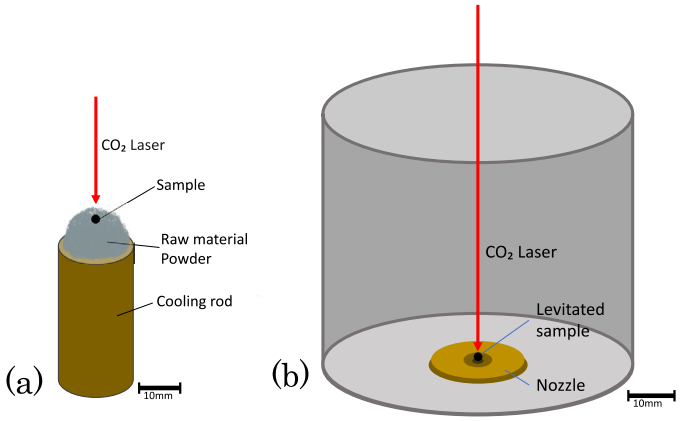

Figure 1. Schematic figure of FeO-SiO_2 sample preparation: (a) laser sintering method for making initial samples and (b) aerodynamic levitation method for glass samples.

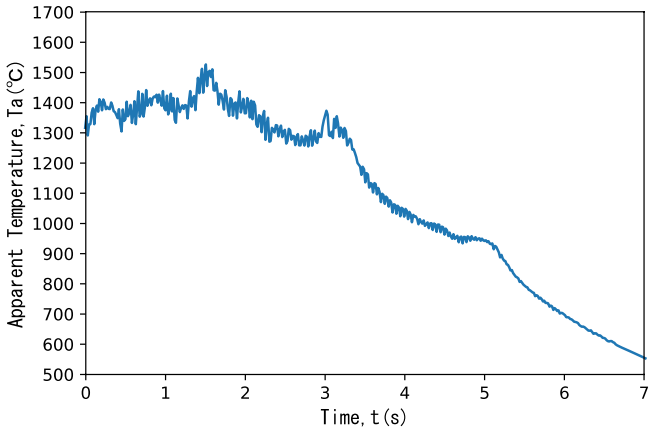

Figure 2. Temperature profiles of cooling of aerodynamic levitated FeO:SiO_2 = 40:60 samples.

2.2. Measurement

Glass samples obtained by above procedures were measured the magnetic and structural properties. Magnetization measurements were carried out by the vibrating sample magnetometer (VSM) at room temperature. The solidified samples were observed by the scanning electron microscope (SEM) for observation of segregations inside samples and measured by x-ray diffraction (XRD) method for structure properties.

3. Result and discussion

Figure 3 shows the sample shape of FeO:SiO_2=40:60 mol% by optical observations. The shape is spherical and has not faced planed, therefore from the figure we identified that the sample formed glass. XRD results

were show no peaks corresponds crystalline phase (Fe_2SiO_4 and SiO_2(tridymite)) from expectation from the phase diagram for FeO:SiO_2=40:60 mol% compositions. **Figure 4** shows the results of the magnetization measurements by VSM. From the results, we found the small hysteresis loops, and the other samples made by different conditions of cooling rate and atmosphere showed the small magnetization of saturations with the large hysteresis loops. **Figure5** is observation results by SEM the inside of samples shown in **Figure 4**. In **Figure 5**, some segregations are appeared in the main matrix phase. From compositional analysis by the energy dispersive x-ray analysis (EDX), these segregation parts were not corresponded to FeO and Fe_2SiO_4. If the segregation parts are caused the magnetism, the expect compositions are Fe_3O_4. However, the parts compositions were not corresponded Fe_3O_4. Therefore, it is not understood the magnetism of the samples of FeO:SiO_2=40:60 mol% from these results, we tried to analyze the magnetization curve by the singularity value decomposition (SVD) method proposed by Sasayama et al.[4] in to the initial part of magnetization curves shown in **Figure 4.** From the SVD results, we find that the small particles with a diameter of 10nm caused by the magnetization curve origin. The size of particle inside the main matrix phase cannot observe by SEM. From the size of magnetic particle, we attribute the Fe nanoclusters appeared in main matrix phase for the magnetism origin of the present glass samples. Therefore, for the future we need to attribute the magnetism origin by more microscopic measurement method electron spin resonance, transmission electron microscope and magnetic scattering using synchrotron radiation.

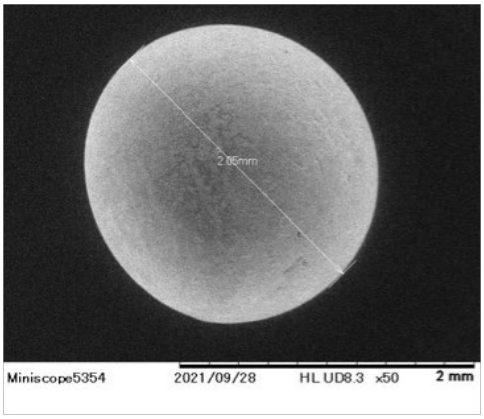

Figure3. SEM images of as solidified samples of FeO:SiO_2=40:60.

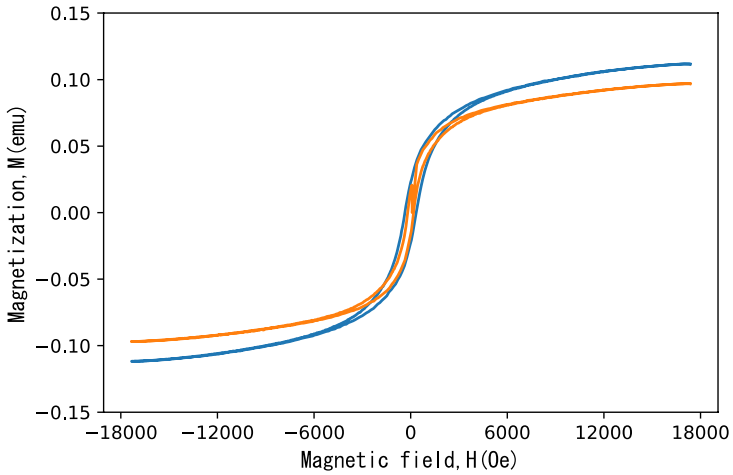

Figure 4. Magnetization of FeO:SiO_2=40:60 glass samples by VSM at room temperature.

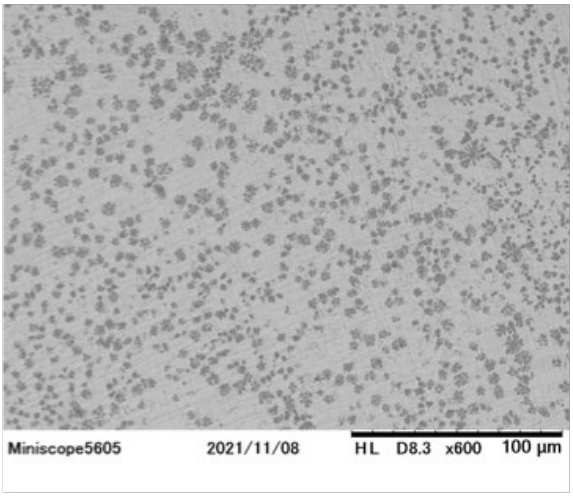

Figure 5. SEM images of inside of specimens shown in Figure 3 for EDX analysis.

4. Conclusion

We succeed to make glass sample of $FeO:SiO_2$=40:60 mol% by the containerless method using aerodynamic levitation with suitable colling and atmospheric conditions. The sample was observed the magnetization with hysteresis loops, but we cannot observe magnetism phase in the sample by SEM and EDX analysis. Therefore, From the present research could not be clarified the origin of magnetization of $FeO:SiO_2$=40:60 mol% glass samples, but from the magnetization curve analysis we estimated that Fe nanoclusters in the main matrix phase segregating are origin the magnetism.

References

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