



Title	Measuring Method of Non-Crystallinity for Amorphous Alloy by Ion Beam Bombardment
Author(s)	Takao, Yoshiyuki; Kanda, Yukinori; Kobayashi, Akira
Citation	Transactions of JWRI. 2012, 41(2), p. 27-31
Version Type	VoR
URL	https://doi.org/10.18910/24857
rights	
Note	

The University of Osaka Institutional Knowledge Archive : OUKA

<https://ir.library.osaka-u.ac.jp/>

The University of Osaka

Measuring Method of Non-Crystallinity for Amorphous Alloy by Ion Beam Bombardment[†]

TAKAO Yoshiyuki*, KANDA Yukinori**, KOBAYASHI Akira***

Abstract

Amorphous alloy is one of the newest materials, recently used in many industrial fields and it has many useful properties such as magnetic or mechanical. The properties will deteriorate when amorphous alloys are crystallized by unexpected heating or low temperature annealing for a long time. In order to evaluate the amorphous grade, we propose a new method based on investigating topographical changes of the amorphous surface by ion bombardment. When an amorphous alloy contains crystal grains, characteristic microfeatures such as cones and pyramids are produced and developed in proportion to the number of the grains on the surface by irradiation with ions. In the present work, the topographical changes of thin amorphous alloy wires ($Fe_{77.5}B_{7.5}Si_{15}$, 125 μm in diameter) annealed at several temperatures up to 650 °C about 10 hours for crystallization and then irradiated by 20 keV argon ions of 2×10^{23} ions/m² were studied using a scanning electron microscope. The relation between the topographical changes and the annealing conditions: the non-crystallinity of amorphous alloys is reported.

KEY WORDS: (Sputtering),(Ion Beam),(Amorphous Alloy),(Topographical Change),(Non-Crystallinity)

1. Introduction

Glass is well known as amorphous material and has been used for many years. Also, metals have been investigated regarding crystal structure and composition, and have been used in various industrial fields. In relatively late years, amorphous alloys were developed. Amorphous alloys are usually made of more than two kinds of metals together and various elemental compositions are selectable. The amorphous alloys therefore have the advantage of keeping useful properties of the constituent elements, because the elements exist as solid dispersions in the alloys homogeneously. Because of this property, amorphous alloys have more superior characteristics than polycrystalline metals, such as a superior magnetic characteristic which is useful to get a high-efficiency transformer and a strong magnetic head. However, these properties are spoiled when crystals are formed in the amorphous alloy. The crystallization process of the amorphous alloys has been studied systematically by Masumoto and et al.^{1,2)}, and it is reported that crystal grains grow in amorphous alloys in association with the condition of annealing temperature and time, and also grow for long time heating even if temperature is low.

Therefore, it is necessary for the product using amorphous alloy parts to know the degree of the

non-crystallization (non-crystallinity) for the parts, and their non-crystallinity strongly depends on their use history. So, it is important to develop a method to measure non-crystallinity of amorphous alloys.

There are two methods to measure non-crystallinity; one is the method using differential thermal analysis, the other is the method using transmission electron microscope (TEM). In the measurement method of amorphous non-crystallinity by the differential thermal analysis, there are problems that more than several tens of mg sample is needed to get enough calorific value for the analysis, and it is difficult to identify crystallized locations. In the method by TEM, there are problems that skill is required for the preparation of observation samples, and the observed range is quite small.

Sputtering is a phenomenon whereby the constituent particles near the surface of a solid are sputtered out when its surface is bombarded by energetic particles. The solid surface is eroded by the sputtering, and characteristic microfeatures such as cones, etch pits, and pyramids are produced and developed on the surface. Analytic studies about characteristic features produced by sputtering have been carried out by Stewart et al.³⁾, and their studies and analysis are based on the incident angle dependence of sputtering yield⁴⁻⁶⁾. The formation of characteristic microfeatures on the surface bombarded by

[†] Received on December 17, 2012

* Professor, Nishinippon Institute of Technology

** Emeritus Professor, Kyushu University

*** Associate Professor

Measuring Method of Non-Crystallinity for Amorphous Alloy by Ion Beam Bombardment

ions depends on the existence of a crystal grain, a grain boundary, a lattice defect, and an impurity, that causes the difference of a local sputtering yield on a solid surface.

We pay therefore our attention to the sputtering property and suggest a method to evaluate amorphous non-crystallinity by the surface features of a irradiated amorphous alloy. The characteristic microfeatures are not produced on a amorphous alloy surface usually because sputtering yield is homogeneous in it. Since the characteristic microfeatures are produced on a surface of a crystallized amorphous alloy by ion bombardment, it is possible to evaluate its non-crystallinity by feature measurement of the surface. As this method only requires ion irradiation and surface observation by a scanning electron microscope (SEM), non-crystallinity of an amorphous alloy can be determined, releasing it from the problems that other methods have.

In this study, iron-based amorphous alloys were annealed at several temperatures up to 650°C about 10 hours for crystallization, and were irradiated by 20keV argon ions. The surface features of the irradiated alloys were observed with an SEM, and feature measurements from the SEM images were carried out. The relation between the topographical changes of the amorphous alloy surfaces and the annealing conditions, the non-crystallinity of amorphous alloys, is reported to suggest a new method of the non-crystallinity determination.

2. Experimental

The experiments have three consecutive stages of sample preparation, irradiation of the sample with electrically accelerated argon ions, and observation of the sample surfaces with a scanning electron microscope (SEM).

2.1 Sample preparation

The samples were prepared from thin wires of Fe-based amorphous alloy, which have an original diameter of 125 μ m, a length of 5mm and composition of Fe_{77.5}B_{7.5}Si₁₅. Tips of the samples were mechanically

Table 1 Annealing temperatures and times for amorphous samples.

Sample No.	Annealing temperature (°C)	Annealing time (hr.)
A	—	—
B	350	1
C	400	1
D	450	1
E	500	1
F	550	1
G	650	1
H	400	10

polished using silicon carbide and buffed with alumina powder of 0.3 and 0.05 μ m in a direction perpendicular to the wire axis. Samples were cleaned ultrasonically in acetone, were then annealed in a vacuum of 7x10⁻⁴Pa. The conditions of ample preparation are summarized in **Table 1**.

After annealing, a sample standing perpendicular to a foil of Fe-based amorphous alloy is set on a cylinder solid sample holder of brass (**Fig.1**). The foil of Fe-based amorphous alloy is to prevent the amorphous sample surface from contamination by sputtering of the brass sample holder. The sample together with a sample holder is put into a Faraday cup and is set in a target chamber so that an axis of the thin amorphous wire coincides with the axis of the Ar beam and is then irradiated with 20keV Ar ions.

2.2 Ion beam bombardment

Figure 1 shows the experimental setup for irradiation of amorphous samples. The apparatus is composed of a ion chamber, an analyzing magnet, a target chamber, and vacuum pumps. The ion chamber and the target chamber are individually evacuated with turbo molecular pumps, and their ultimate vacuums are 4x10⁻⁵Pa and 6x10⁻⁵Pa respectively. The ion chamber is composed of a plasma source of a hollow hot cathode type, an acceleration electrode, and an einzel lens. Ar ions produced in the plasma source are extracted and

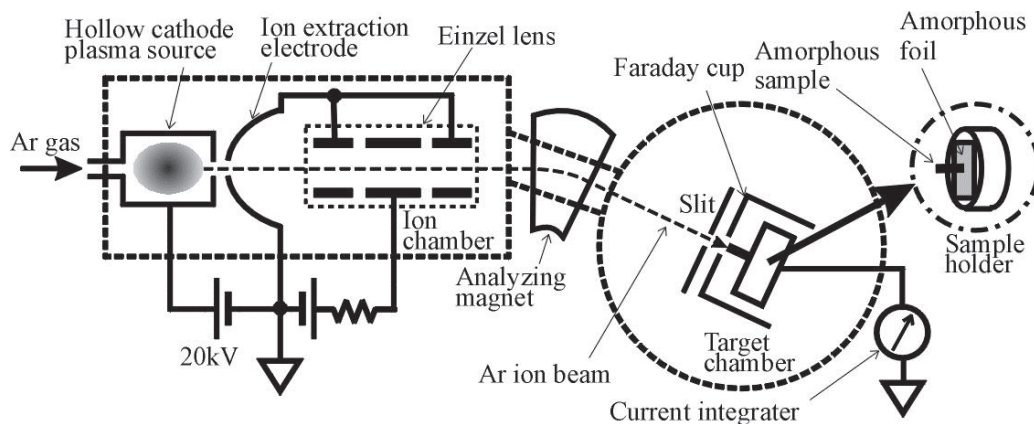


Fig.1 Schematic diagram of the experimental setup for irradiation of amorphous samples with Ar ions.

accelerated by electrical potential difference of 20kV between the plasma source and the acceleration electrode and are focused with the einzel lens. The Ar ion beam is deflected at an angle of 10 degrees in flight by a magnetic field in order to separate Ar ions from other ions, and is then introduced into the target chamber. A sample set in a Faraday cup is irradiated in normal incidence with Ar ions through an orifice of 2mm in diameter. The total charge of Ar ions is measured with a current integrator by the integration of the current from the sample and a Faraday cup surrounding the sample. The fluence of the bombarding 20keV Ar ions is 2×10^{23} ions/m² and the ion current is approximately 0.1 μ A. During bombardment, the pressure of the ion chamber and the target chamber raised to 4×10^{-3} Pa, 7×10^{-4} Pa respectively.

2.3 Observation of sample surface by a scanning electron microscope

The observation was carried out in the direction perpendicular to the irradiated sample surface (parallel to an axis of the Ar ion beam) using an SEM at scanning electron voltage of 15kV and magnification up to 1500 times.

3. Results and Discussion

A series of SEM micrographs is given in **Figure 2 and 3** showing topographical changes of the Fe-amorphous alloy wires irradiated with Ar ions.

3.1 Topographical change of sample surface by ion bombardment

3.1.1 Unannealed amorphous sample after irradiation

Figure 2 shows the irradiated feature of an unannealed sample (as-given) of Sample A. Figure 2 (a) is a whole image of Sample A. All samples were irradiated in the direction indicated by arrows in Fig.2 (a). The surfaces of the sample A shown in Fig.2 (b), (c) were etched uniformly and has no microfeatures. Other samples were also irradiated by Ar ions and were observed by an SEM in the same direction for Sample A.

3.1.2 Irradiation features of sample surfaces annealed for one hour

Samples B~G (listed in Table 1) are annealed

samples for one hour at several temperatures. Figure 3(a)~(e) show the surface change dependence on the annealing temperature after irradiation. Micro features of craters and etch pits have increased in proportion to the annealing temperature. At the annealed samples above 450°C, the micro-features overlap and form into net-like structure. The mesh of the net-like structure becomes of higher density at the higher annealing temperature. The topographical changes are caused by the difference of the sputtering yield between crystalline and amorphous regions. The sputtering yield of the amorphous region is smaller than that of the crystalline region. As the higher density of crystal grains are formed at the higher temperature, the mesh of the net-like structure becomes smaller.

3.1.3 Irradiation features of sample surfaces annealed for ten hours

Sample H is an annealed sample for ten hours. Figure 3(f) shows the surface feature of the annealed sample after irradiation. On comparison between the surface in Fig.3(f) and that in Fig.3(b), microfeatures of the surface in Fig.3(f) are more complicated than that in Fig.3(b). On comparison between the surface in Fig.3(f) and that in Fig.3(d)(e), mesh of the surface in Fig.3(f) is wider than those in Fig.3(d)(e). This is caused by the difference of crystallization process in those amorphous samples. The number of small size crystal grains increases at the high temperature for a short time. On the other hand, larger crystal grains grow at low temperature for a long time annealing. The feature of the irradiated surfaces depends on their crystallizing process.

3.2 Comparison of irradiated sample surface by image data processing

Feature measurement by image data processing was carried out to define the topographical changes of irradiated sample surfaces. Average of roughness height was used as a representative value to define the characteristic of the surface feature. The averages of roughness height for the samples were calculated from SEM grayscale images and the results are shown in **Fig.4**. The averages of roughness height increase in proportion to annealing temperature, and these values are associated

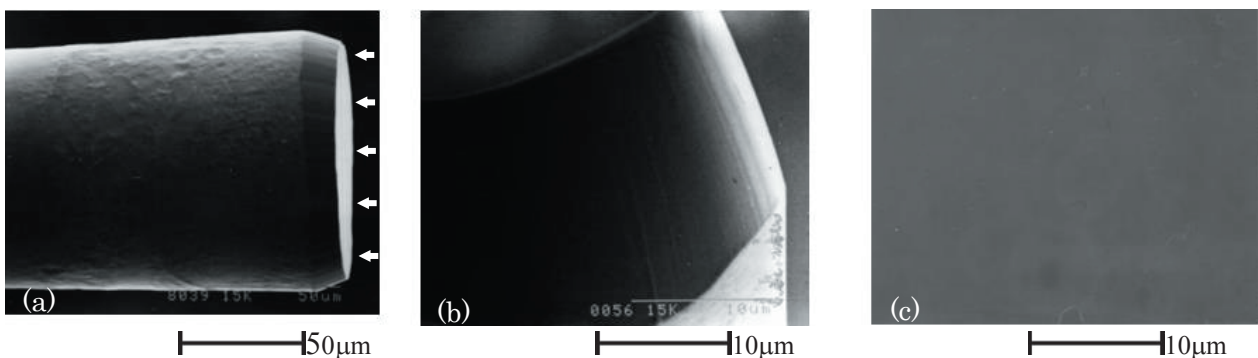


Fig.2 Topographical change of Sample A (as given amorphous sample of $\text{Fe}_{77.5}\text{B}_{7.5}\text{Si}_{15}$) surface irradiated by 20keV argon ions of 2×10^{23} ions/m². (a) whole image, samples are irradiated in the direction indicated by arrows; (b) edge of the irradiated sample A surface; (c) sample A surface observed in the direction parallel to an axis of the Ar ion beam.

Measuring Method of Non-Crystallinity for Amorphous Alloy by Ion Beam Bombardment

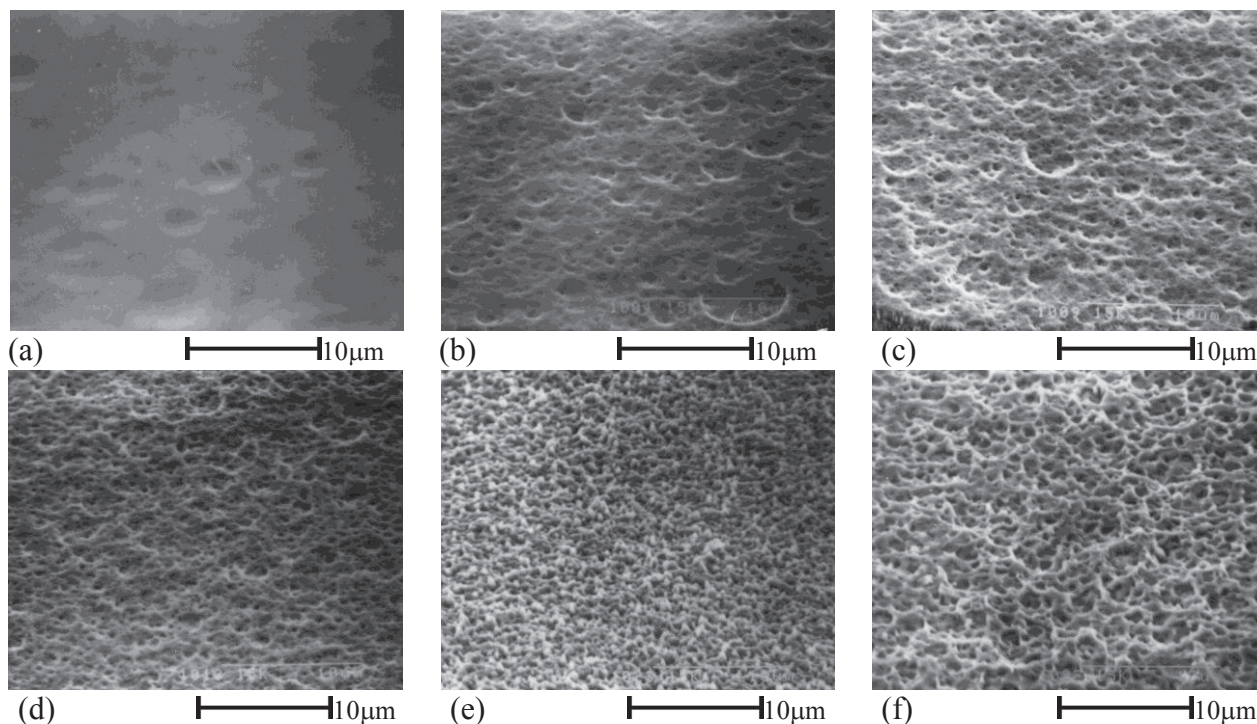


Fig.3 Photographs of topographical changes of crystallized amorphous alloy surfaces irradiated by 20keV argon ions of 2×10^{23} ions/m². The amorphous samples were annealed to crystallize for an hour at several temperatures: (a) 350°C (sample B); (b) 400°C (sample C); (c) 450°C (sample D); (d) 500°C (sample E); (e) 650°C (sample G). (f) The amorphous sample was annealed for ten hours at 400°C (sample H).

with the SEM images (Fig.3(a)~(e)) observed results described above. In particular, many cones are shown in Fig. 3(e), and it is thought that the amorphous sample is considerably crystallized and then the average of roughness height increased. In addition, the average of roughness height for sample H becomes larger conspicuously than that for sample C and this indicates that the surface features of irradiated samples are closely related to the crystallization process.

Figure 5 shows the annealing temperature-time dependence of phase changes for the amorphous alloy and was referred from Ref. 1. The solid lines in the figure

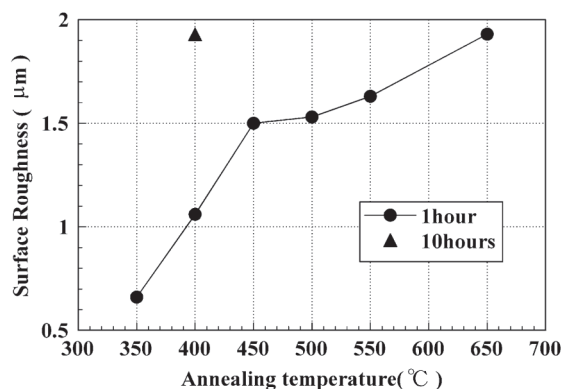


Fig.4 The feature extraction of surface roughness from SEM pictures of sample B~H.

indicate boundary conditions of phase changes for the amorphous alloy of $\text{Fe}_{78}\text{B}_{10}\text{Si}_{12}$ ¹⁾. Although the composition of the amorphous alloy between this study and Ref.1 are slightly different, it is thought that the samples in this study are distributed from amorphous phase to a final stable phase through two metastable nonequilibrium phases of MS-I and MS-II¹⁾. Since the phase change from amorphous to MS-I occurs at 420°C at the annealing time of one hour in Fig.5, it is thought that the tendency of the averages of roughness height curve significantly changes at around 450°C in Fig.4. On the other hand, Sputtering yields differ very slightly

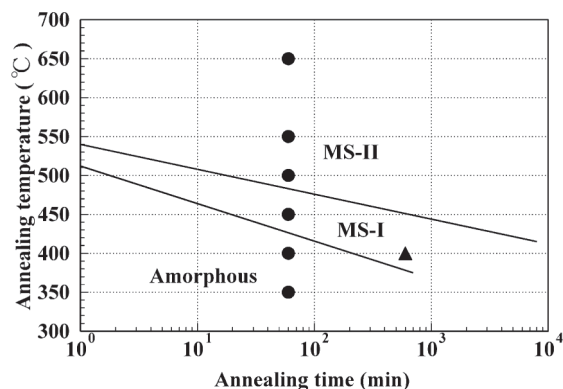


Fig.5 The annealing temperature-time dependence of phase change for the amorphous alloy. The solid lines are referred from Ref. 1.

between MS-I phase and MS-II, because the curve of the averages of roughness height from Sample D to G increases slowly.

4. Conclusion

Fe-based amorphous alloy samples ($\text{Fe}_{77.5}\text{B}_{7.5}\text{Si}_{15}$) were annealed at several experimental conditions and irradiated by 20keV Ar ions. Topographical changes of the samples were investigated and the obtained results are as follows.

- (1) The features of the amorphous alloy surfaces irradiated by Ar ions depend on the crystallization processes.
- (2) By using the sputtering method, the determination of amorphous grade in a small area of amorphous solids are possible.
- (3) There is a significant difference of the sputtering yield between crystalline and amorphous region.
- (4) The difference of the feature between the annealing

sample for a short time at a high temperature and that for a long time at a low temperature is caused by the difference in growth process of the crystal grain.

References

- 1) T. Masumoto, A. Inoue and H. Kimura, Crystallization Process of Iron-, Nickel-, and Cobalt-Based Amorphous Alloys Containing Silicon and Boron, J. Japan Inst. Metals, 41 (1977) 730-737 (in Japanese).
- 2) T. Masumoto, K. Suzuki, et al., Amorphous kinzoku no kiso, Ohmsha (1982) (in Japanese).
- 3) A.D.G. Stewart and M.W. Thompson, Microtopography of Surfaces Eroded by Ion-Bombardment, J. Master. Sci., 4 (1964) 56-60.
- 4) H. Fetz, Z.Phys., 119 (1942) 590.
- 5) V.A. Molchanov and V.G. Telkovskii, Sov. Phys. Doklady. (1961) 6.
- 6) P. Sigmund, Theory of Sputtering. I. Sputtering Yield of Amorphous and Polycrystalline Targets, Phys.Rev., 184 (1969) 383-416.