Fifth International Conference on Cold Fusion
9-13 April, 1995 - Monte Carlo, Monaco
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Anomalous heat evolution of deuteron implanted Al on electron bombardment

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Abstract

Anomalous heat evolution was observed in deuteron implanted Al foils on 175 keV electron bombardment. Local regions with linear dimension of several 100nm showed simultaneous transformation from single crystalline to polycrystalline structure instantaneously on the electron bombardment, indicating the temperature rise up to more than melting point of Al from room temperature. The amount of energy evolved was more than 180MeV for each transformed region. The transformation was never observed in proton implanted Al foils. The heat evolution was presumed to be due to a nuclear reaction in D_2 molecular collections.

1. Introduction

In a previous paper [1], one of the authors (K.K.) reported an anomalous particle emission phenomenon from H_3^+ or D_3^+ implanted Al foil on 200 or 400keV electron bombardment. In the paper, the author presumed that fusion reactions to take place not only between deuterons but also between hydrogenes, which were embedded in the so called "Tunnel Structure" (T.S.), created in sub-surface layers of Al on the implantations. (The results will be supplemented by more detailed experiments and also by theoretical considerations in a recent paper [2].)One of the prominent features of this phenomenon was that it is not due to the energetic collisions between reacting particles. This was derived in the paper [1] from the calculation on the fusion reaction rate between knocked-on deuterons, produced by the electron bombardment, and embedded deuterons. In the present state of our knowledge, we cannot present any conclusive mechanism of the phenomenon, but the author presumes that β disintegration of proton on capturing secondary electron in highly ionized hydrogen or deuteron plasma of nearly solid state density may play a fundamental role in some aspect of the phenomenon.

In the present paper, a direct observation of this anomalous phenomenon via heat evolution leading to very local melting of deuterium implanted Al on 175 keV electron bombardment is reported. The direct observation was made by the transmission electron microscope (TEM), which at the same time served as an electron accelerator.

2. Experiment

We first implanted Al specimens with 25 keV D_2^+ ions. The specimens were prepared beforehand so as to enable TEM observations after the implantation. They were polished chemically from Al disk with 5^{mm} diameter and 0.1^{mm} thick using TENUPOLE chemical polishing machine. The purity of the Al was 99.99%, and the specimens were annealed at 400℃ for 3 hours before the polishing. After the polishing they have wedge shape with average thickness of more than 1 μ m over an area of about 1mm diameter and have a small hole of about 0.1mm diameter in the central part of each specimen.

The fluence of the implanted deuteron was chosen around 5×10^{17} D^+/cm^2, since below this amount of fluence only a bubble structure is formed in the sub-surface layer of Al foil, and above that so called "Tunnel Structure" (T.S.) is produced [3] .

The density of the implanted deuterium in the T.S., $1\times10^{22}D_2cm^{-3}$, was also estimated from the same kind of experiment measuring the implanted amount of hydrogen during and after the implantation at room temperature [2,3] . Again from these experiments, the implanted deuterium are presumed to situate at about 100nm depth from the implanted surface. As a consequence, more than ten times of the implanted depth remains unimplanted beneath the T.S..

3. Experimental results

Two micrographs of Fig.1 show the typical examples of TEM observations taken on the Al specimens implanted with hydrogen, (a), and with deuteron, (b), respectively. Regions of brighter contrast in both micrographs are the T.S. regions, where Al atoms are lacked and, instead, hydrogen or deuterium molecules are contained. In the micrograph (b), we can observe several areas, like the one indicated by an arrow, where something looks like microcrystallites are concentrated. These speckled areas appear instantaneously with the focusing of electron beam for the observation with a brighter contrast. They are not inherent to the implanted Al originally, but are attributable to the electron beam focusing effect. Before the focusing, when looking for the area to be observed with less focused beam, we can never observe such speckled areas. Further, these speckled areas can never be observable in hydrogen implanted Al under exactly the same experimental conditions with the deuteron implanted case. (So far we have done several ten times of the hydrogen implantation experiment.) This is readily seen in the micrograph (a) of Fig.1. To observe the speckled area, there is a region of optimum fluence of deuteron ions, which produce the microstructure like those shown in Fig.1. For lower fluence than that in the optimum region, we observe only bubble structures and never observe the speckled area, and for higher fluence, we observe, in addition to the T.S. structure, the bubble structure again but never observe the speckled area.

For crystallographic investigation of the speckled areas, selected area electron diffraction was tried on about 500nm area surrounding the speckled area. Those inserted in Fig.1 show the diffraction patterns taken on the areas seen in the micrographs. One should pay attention that (b-1) in Fig.1 (b), which was taken at the speckled area, clearly demonstrates the polycrystalline pattern with co-central spotty circular rings. On the other hand, the diffraction patterns from the normal areas, (a-1) and (b-2), show only usual Bragg spots showing single crystalline Al. So far we have observed five polycrystalline rings from several speckled areas of different specimens. The lattice constants corresponding to the rings are tabulated in Table 1, together with the lattice constants and planes inhirent to Al.

To confirm the correspondence between the polycrystalline rings and the speckled area, dark field images were taken using several spots on the rings. Fig.3 (a) shows the bright field image of the speckled area with the diffraction rings, and (b) shows the dark field image of the same area taken with several diffraction spots on the rings. They show evidently that the polycrystalline spots originate from several parts of the speckled area, which appear brighter in (b).

The appearance of the speckled areas on the electron beam focusing is so rapid that we could not follow the detailed process of the appearance during the observation. However, after the appearance the images change rather gradually as seen in Fig.3. In this figure, (a) was taken in roughly 10 seconds or so after the appearance, and (b) and (c) were taken sequentially in less than 60 seconds after (a). These micrographs show rather gradual polycrystallization, judging from the growth of small crystallites as seen in (b) and (c), and the gradual change of equal thickness fringes as shown by an arrow. The change of the equal thickness fringes is presumed due to the thermal conditions around the region. The image did not change anymore after (c).

These observations indicate a rapid melting and gradual crystallization, which is rather conceivable if we assume that the reactions in D_2 collections occurred suddenly and was maintained for a short time, evolving the large amount of heat.

In addition to the above observations, we tried stereographic observations of the speckled region with change of the tilt angle of about 6 to 8 degrees to indentify the depth of the layer in which the polycrystals are laying. As a result, we found that they are contained within the surface layer of the specimen with thickness of about 100nm above the T.S..

The experimental results described above clearly show that the speckled areas appeared on the electron bombardment is due to the local transformation from single crystalline to polycrystalline Al.

As far as the present authors are aware, the transformation from single crystalline to polycrystalline structure of Al metal with purity of 99.99% or above can never be achieved without melting and subsequent rapid solidification. One might inquire that the melting may induce evaporation of Al in high vacuum of electron microscope. However, the evaporation of Al on the melting does not take place usually due to the firm protection of the melt surface by the oxide film even after the chemical etching.

Now we evaluate the amount of the heat evolution necessary for the observed local melting. We know already that the implanted deuterium molecules aggergate at the depth of around 100nm from a surface on the implantation of 25 keV D_2^+, referring to the previous experiment with 25 keV H_2^+ [3] . From our observations so far undertaken, the extent of the transformed region occupies, on the average, 1×10^{-9}cm^2 of the surface. Taking the cover thickness of Al on the T.S. as 100nm, weight of Al contained in the polycrystalline transformation becomes 2.7×10^{-14}g. The heat necessary to raise the temperature of this amount of Al from 300K to melting point 933K is $q = 633 \times 0.25 \times 2.4 \times 10^{-14} = 4.3 \times 10^{-12}$ cal, where the specific heat of Al in this temperature range is 0.25 cal/g · K. The latent heat of melting of Al is 2.58×10^3 cal/mol [4] , which gives the total latent heat for the melting of the above amount of Al $L = 2.58 \times 10^3 \times 2.7 \times 10^{-14}/27 = 2.58 \times 10^{-12}$ cal. Therefore, the whole heat necessary for the melting becomes, averaging on the several transformed regions, $Q = q + L = 6.9 \times 10^{-12}$ cal = 180 MeV for each transformed region.

Here, we would like to mention that the above estimation of the energy evolution could be smaller than that of whole energy evolved, since we have totally neglected the heat conduction though Al specimen. As mentioned before, the bottom side of the specimen below the T.S. is far thicker than the top side. So, a large amount of heat evolved in the D_2 collections in T.S. is presumed to flow out of the specimen though the thick bottom part to the specimen holder. Therefore, the heat responsible for the melting of the top area of the specimen must be a part of the whole heat evolution.

4. Discussions

One of the authors (K.K.) has published a short paper [1] describing the particle emission from implanted Al on the electron bombardment. The present experimental results have close similarities with this particle emission experiment in two aspects. First of all, it is requisite to focus the electron beam to observe both the polycrystalline transformation and the particle emission. Secondly, the optimum implantation fluence around $5 \times 10^{17} D^+/cm^2$ is common in both experiments. Neither lower nor higher fluence does produce the heat evolution and the particle emission as well. we have to keep the fluence within roughly ± 10% around the above value. However, the difference of the two kinds of experiment is that in the present experiment we could never observe the heat evolution in hydrogen implanted case, and, on the other hand, in the particle emission experiment, we could observed the particle emission in both hydrogen and deuterium implanted cases. We presume that though the reactions in deuterium are accompanied with the heat evolution, the reactions in hydrogens, on the other hand, could be such as not accompaning heat evolution. The two reactions are not necessarily the same reactions at all.

Here, we like to add that the primary electrons with energy around 200 keV has little interactions with the embedded deuterium, and hydrogen as well, in the present experimental conditions. However, internal secondary electrons produced in Al due to the primary electron bombardment have strong inelastic interactions with the deuterium leading to the ionization of them. In justification of this model, this interaction is capable of explaning the dependence of the phenomenon on both the electron beam focusing and the microstructure in the subsurface layer which are described above. These points will be discussed fully in [2] .

In a subsequent paper [5] , we will discuss more fully on possible heating mechanisms based on solid state properities of Al, such as the insulating effect of the T.S. on the electron bombardment, difference of implantation depth of hydrogen and deuterium, and the decrease of melting point of the thin Al surface layer. However, we can show experimentally and theoretically that these mechanisms do not contribute to the observed melting. Further, we can show that the mean kinetic energy of the embedded deuterous in the T.S. possibly acquired by some mechanism, whatever it may be chemical or nuclear nature, must be more than 30eV to melt the surface layer.

These considerations strongly suggest the presence of some kind of nuclear reactions in the present situation.

Reference

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[2] K. Kamada, M. Nakajima, M. Ogawa, T. Goto and H. Kakihana; Proc. 7th Int. Conf. on Emerging Nuclear Energy System(ICENES'93), p168 Sept.1993.
[3] K. Kamada, A. Sagara, H. Kinoshita and H. Takahashi; Rad. Effects 103 119 (1987): K. Kamada ; J. Nucl. Mater. 169 141 (1989).
[4] from American Institute of Physics Handbook, 3rd edition, McGraw Hill, New York, 1972.
[5] submitting.

Figure captions

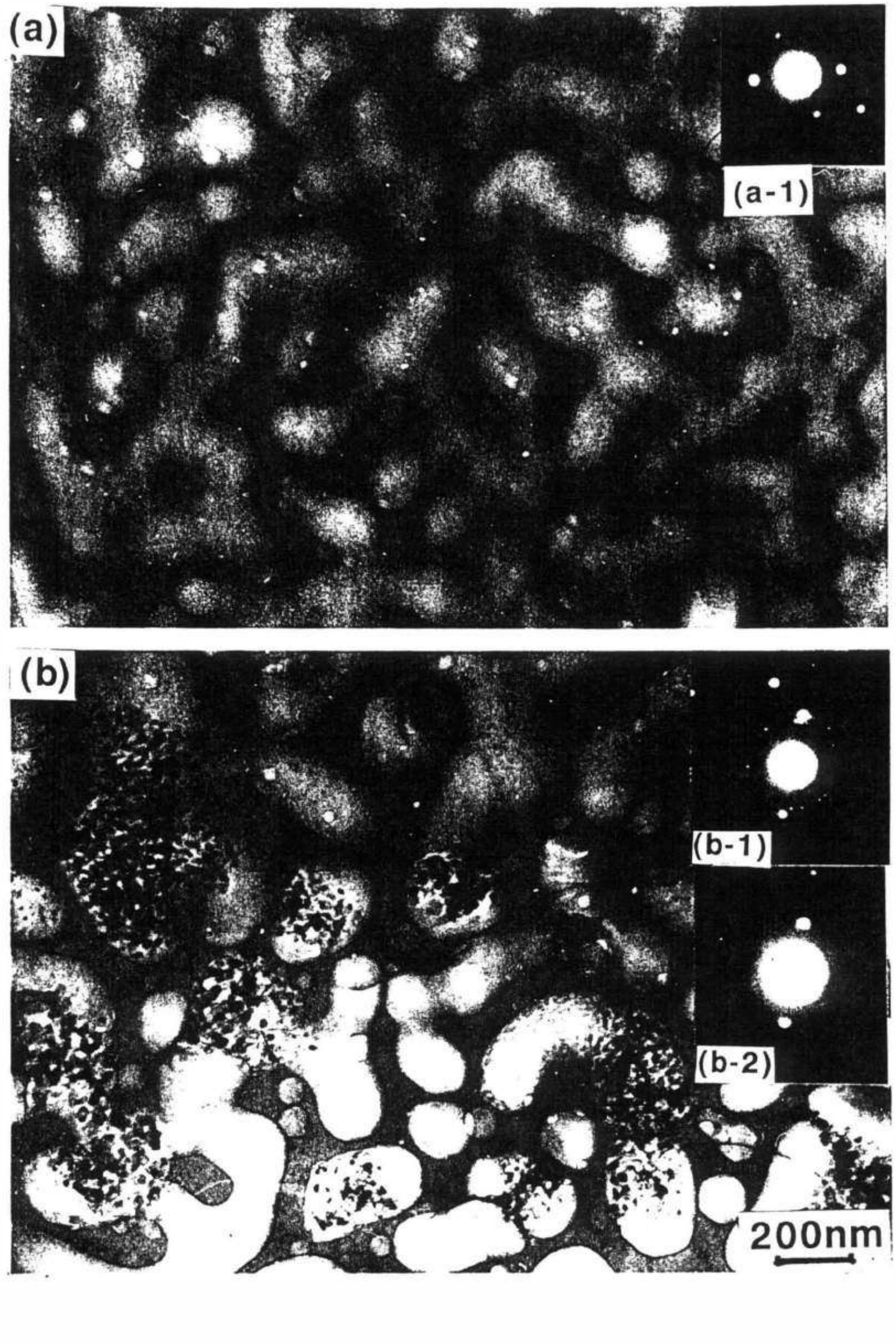
Fig.1 Transmission electron micrographs of hydrogen, (a), and deuterium, (b), implanted Al. An example of the polycrystallized area on the electron bombardment is shown by an arrow in (b). Selected area diffraction patterns taken on normal T.S. area, (a-1) and (b-2), and that taken on the polycrystallized area, (b-1), are inserted.

Fig.2 Bright field, (a), and dark field, (b), images taken on the polycrystallized area in a deuterium implanted Al. The dark field image was taken with several diffraction spots on the circular rings of the inserted selcted area diffraction pattern.

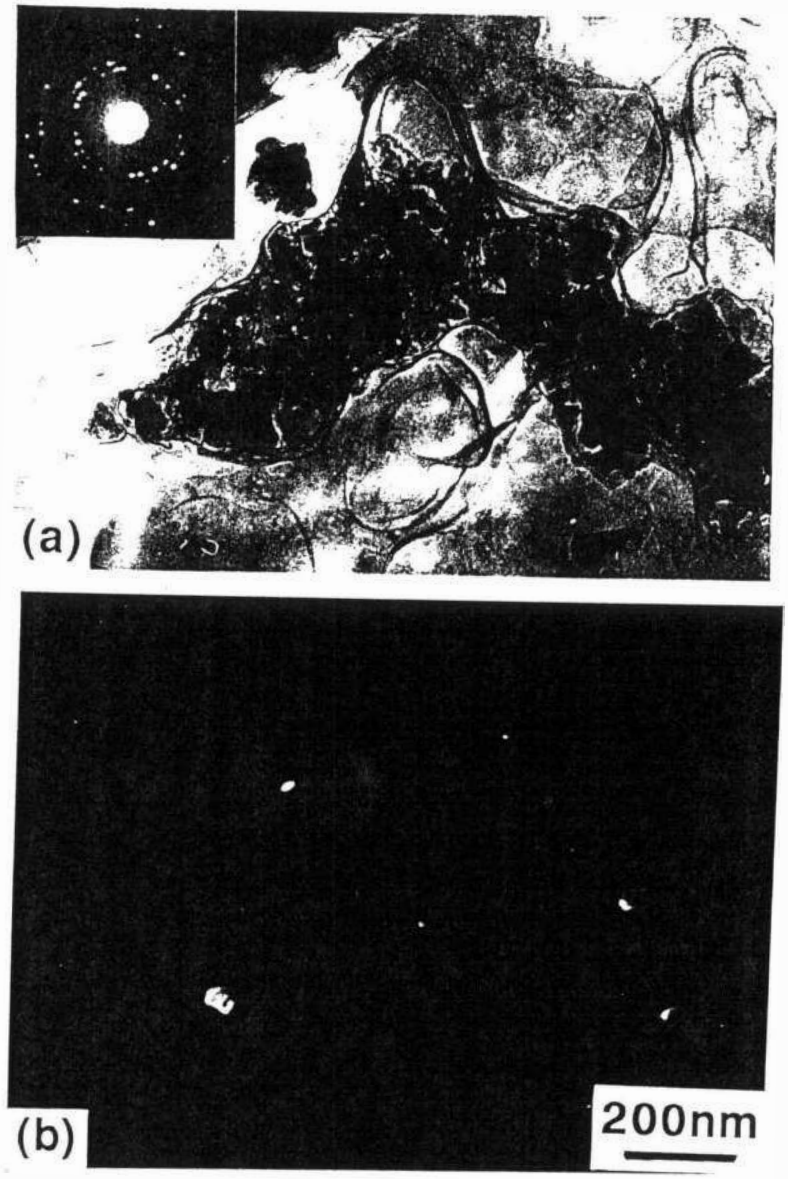
Fig.3 Sequential TEM micrographs of the same area of a derterium implanted Al, which were taken in less than 60 seconds after the appearance of the speckled area. (a) was taken in less than 10 second, then (b) and (c) were taken in less than 60 seconds sequentially after (a).

able 1 Lattice constants of the polycrystalline aggregates determined y electron diffraction with the camera length L=570mm and the wavelength f electron $\lambda=2.99\times10^{-3}$nm. Lattice constants and corresponding lattice lanes of Al are shown together for reference purpose.

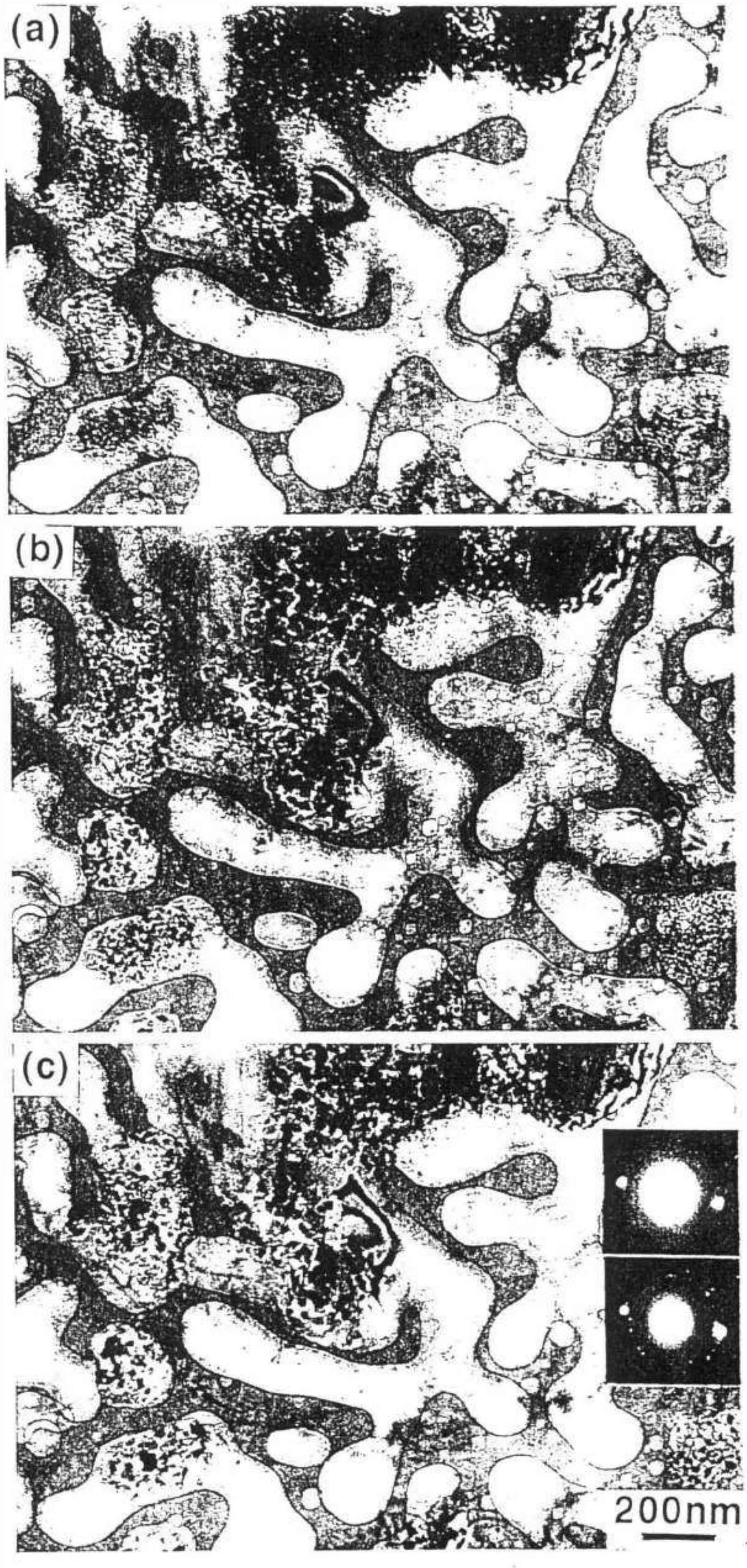
ing	d_{obs}(A)	d_{obs}(A)	d_{obs}(A)	d_{obs}(A)	d_{obs}(A)(hkl)
1	2.4024	2.3976	2.3549	2.3385	2.338(111)
2	2.0366	2.0609	2.0287	-	2.024(200)
3	1.4570	1.4511	1.4505	-	1.431(220)
4	1.2473	1.2398	1.2213	-	1.221(331)
5	0.9266	-	-	-	0.9289(331)



Kamada Fig. 1



Kamada Fig. 2



Kamada Fig. 3