THE SAXS STUDY OF MICROSTRUCTURE OF SOME AMORPHOUS AND CRYSTALLINE ALLOYS

P. Dubček, M. Stubičar, J. Lukatela and B. Leontić
Department of Physics, Faculty of Sciences and Institute of Physics of the University,
41001 Zagreb, P.O. Box 304
and
A. Janosi
Institut für physikalische Chemie der Universitat Graz,
A–8010, Graz, Heinrichstrasse 28

INTRODUCTION

Small-angle X-ray scattering (SAXS) is a very promising method for investigating the size (or shape) distribution function of submicroscopical inhomogeneities usually present in different Crystalline or amorphous alloy system (1). This method is particularly suitable for systems where the electron density difference between the inhomogeneities and the matrix appears to be rather high, e.g. between GP zones and the matrix in Al-Ag crystalline oversaturated solid solution. This method is also applicable if one expects changes in compositional or topological short range order.

This communication deals with SAXS investigation of changes in microstructure observed in hydrogen doped Zr_2Ni metallic glass and Cu_2Ti metallic glass isochronaly annealed at various temperatures up to 773 K. In addition, the results obtained for metallic glasses are compared to those obtained for samples quenched conventionally (from solid state – CQ samples) and ultrarapidly (from the liquid state – URQ samples) built of Al-16 wt. % Ag crystalline alloy.

EXPERIMENTAL

The amorphous samples were prepared by melt spinning (2) from master alloys of predetermined composition and the crystalline ones by means of the mill device (3). The SAXS spectra were registered using the standard Kratky type X-ray camera and a scintilation counter connected to a pulse height analyser. The entrance and the counter

slit widths were 100 μm and 250 μm respectively. Ni-filtered Cu radiation was used in all our SAXS experiments. The experimental SAXS curves were corrected for the influence of collimation by Vonk's method (4). Isothermal or isochronal heat treatment was performed using oil bath at temperatures bellow 500 K, and the treatment was performed in vacuum of the order of 10^{-4} Pa at higher temperautres.

RESULTS AND DISCUSSION

Fig. 1 shows typical SAXS spectrum changes observed for Cu_2Ti amorphous samples during half an hour annealing in 50 K steps from the room temperature up to the temperature indicated in the figure. The curves show that, except at very small angles, amorphous Cu_2Ti alloy has negligible SAXS intensity. However, the annealing at higher temperatures causes additional changes in the structure of the sample, which alter local electron density and thus SAXS intensity is enhanced. This is a consequence of the increasing difference of the electron density caused by development of at least two different phases.

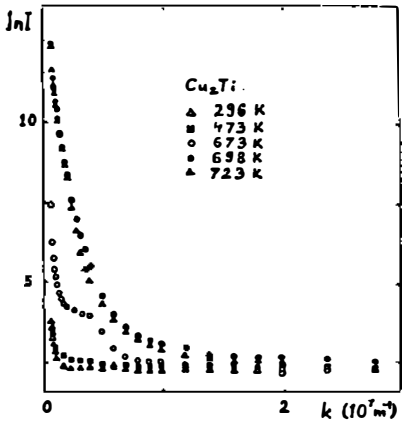

Fig. 1.

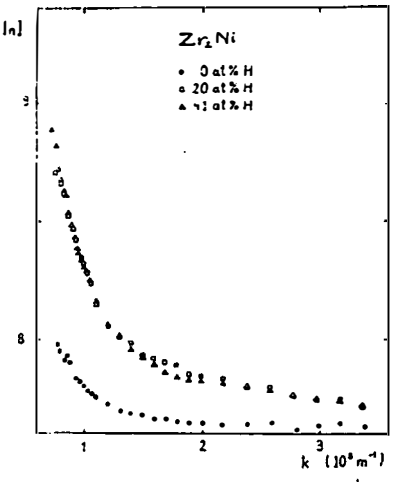

Fig. 2.

The SAXS results shown in Fig. 2 belong to Zr_2Ni system doped with hydrogen up to various atomic percentages. A similar explanation might be offered here, i.e. the observed changes could be explained by the fact that the introduced hydrogen is unevenly distributed throughout the sample. Hydrogen atoms cling preferably to Zr atoms (5), and, in addition, they could fill existing voids in the glass. Thus agglomerates, variously sized and rich in hydrogen, could be formed in the specimen. Moreover, the average size of the agglomerates increases with the hydrogen concentration.

The changes in SAXS spectrum observed for convetionally and ultrarapidly quenched Al–16 wt. % Ag samples after annealing at 393 K are shown in Fig. 3. The diameter distribution function of the existing inhomogeneities is shown in insert. It was calculated using Vonk's program and experimental SAXS data obtained for a liquid quenched sample aged over ten years at 300 K.

It is evident from Figs. 1 and 2 that the observed effects in SAXS spectra are caused by changes in the samples microstructure during the treatment. Moreover, evaluation of all the scattering curves, e.g. using Guiner approximation, indicates that submicroscopical inhomogeneities existing in samples grow in size during the treatment. Growing of inhomogeneities is evident (see Fig. 1) if heat treatment is carried out near or above the crystallization temperature where electron density difference between possibly coexisting phases occurs more rapidly. Although one can conclude about the degree of change in microstructure on the basis of the observed SAXS spectra, it is not possible to make reliable predictions on changes of other properties. This is because nonlinear correlation might exist sometimes. Our SAXS data indicate that there are weak changes in SAXS spectrum well bellow the crystallization temperature in spite of the fact that a maximum in isochronal microhardness curve is observed for Cu_2Ti system, or an increase in microhardness for about 30 pct. was registered in the case of Zr_2Ni system when it was doped with hydrogen (7).

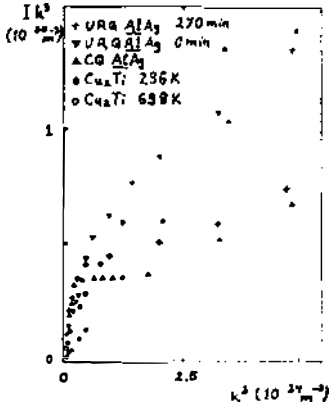

Fig. 3.

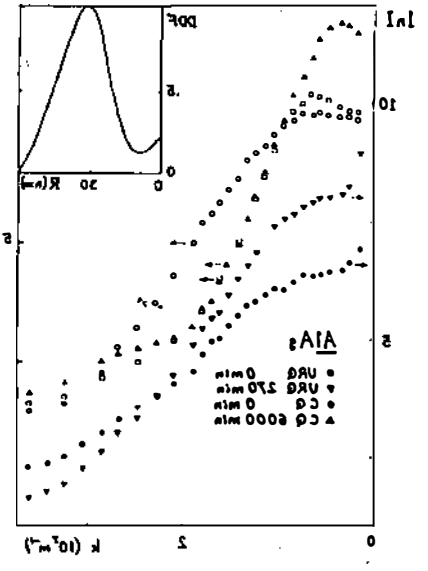

Fig. 4

Additional analysis of SAXS curves, e.g. using Porod plot (see Fig. 4) indicates that both for the crystalline Al—Ag and for the studied glassy systems Porod law is not obeyed. This could be explained by assuming that a diffuse boundary exists between inhomogeneities and the matrix phases, or some additonal scattering (e.g. Laue monotonic) might mask this behaviour (8). Additonal SAXS investigations have to be done in order to decide which of the offered explanations is correct.

CONCLUSION

Valuable informations have been obtained on the changes of microstructure of Zr_2Ni and Cu_2Ti glassy systems caused by heat treatment or by hydrogen doping.

REFERENCES

1. A. Guinier and G. Fournet: Small-angle Scattering of X–Rays, Wiley and Sons, New York, 1955.
2. H.H. Liebermann and C.D. Graham Jr., IEEE Trans. Magnetics, 12 (1976) 921.
3. E. Babić, E. Girt, R. Krsnik and B. Leontić, J. Phys. E, 4(1970)1014
4. C.G. Vonk, J. Appl. Cryst., 5(1975)340.
5. K. Tanaka, N. Hamasaka, M. Yasuda and Y. Fukai, Solid State Commun, 30(1979)173
6. B. Gržeta, M. Stubičar, N. Cowlam and R. Trojko, to appear in Phil. Mag.
7. B. Leontić, J. Lukatela and M. Stubičar, J. Non-Crystal. Sol. 54(1983)81.
8. a) V. Gerold in: Small-Angle X-Ray Scattering, Ed. by H. Brumberger, Gordon and Breach, New York, 1976.
   b) A. Naudon et J. Caisso, J. Appl. Cryst. 7(1974)25.