LIQUID QUENCHED METALLIC METASTABLE ALLOYS*

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Introduction

Metastable phases in physical metallurgy are the rule rather than the exception. These phases are generally obtained by quenching a solid alloy from high temperature and tempering, or aging it at a lower temperature in order to obtain the desired physical properties. Quenching alloys from the liquid state did not receive much attention from the metallurgist until approximately ten years ago when new techniques became available to reach high enough a rate of cooling to affect the normal process of nucleation and growth during the transition from liquid to solid. Obviously, the well known techniques of cooling a solid with a fluid, either a liquid or a gas moving at high speed, are not applicable to liquid alloys, and the only other method of heat removal is by conduction to a solid surface. The two methods now widely used to achieve this purpose are the "gun" technique in which a liquid globule is projected at high speed on a copper substrate, and the "piston and anvil" technique in which a liquid globule is squeezed between two copper lined flat surfaces (Duwez et al. 1963, Pietrokowsky 1963). The rate of cooling obtained by these techniques is not yet very accurately known, but the orders of magnitude of the average rate of cooling are 10^5 to 10^6 °C/sec for the piston and anvil technique and as high as 10^6 to 10^8 °C/sec for the gun technique. The first metastable phases obtained by rapid cooling from the melt were a complete series of solid solutions in Ag-Cu alloys (Duwez et al. 1960) which under equilibrium conditions are two-phase eutectic-type alloys. These results were predictable since in this system a solid solution would be expected from the Hume-Rothery rules governing the formation of solid solutions. The second type of metastable phase discovered soon after the Ag-Cu alloys was a new crystalline hexagonal closed packed phase in Ag-Ge alloys (Duwez et al. 1960), which does not exist under equilibrium conditions. The third and probably last type of metastable alloy obtained by quenching from the liquid state was an amorphous phase in the Au-Si system (Klement et al. 1960) which represents the ultimate in quenching rate, since a liquid-like atomic arrangement could be obtained in the solid state. Many papers presented at this conference deal with the first two types of metastable alloys and I will limit my discussion to amorphous metallic alloys.

Amorphous Metallic Phases

Soon after the first amorphous phase obtained by liquid quenching was reported in binary Au-Si alloys, it was pointed out that the composition of this phase lies close to equilibrium eutectic and that this eutectic occurs at a very low temperature compared with the melting point of gold (Cohen et al. 1961). This simple observation served as a guide to discover other binary alloys susceptible to lead to an amorphous phase after quenching. The Pd-Si binary alloys, with a eutectic at about 16 at.% Si and a eutectic temperature of about 800^{o}C, compared with a melting temperature of 1550^{o}C for Pd, turned out to be the easiest to quench by both the gun and the piston and anvil technique and have been the subject of numerous investigations (Duwez et al. 1965). The Pd-Si amorphous alloys are quite stable, with a rapid crystallization temperature of about 420^{o}C. Kinetics data indicate that no crystallization can be detected after more than 6 months at 200^{o}C (Duwez 1967).

Many binary allow systems involving a transition metal and phosphorus have low eutectics comparable to that in Pd-Si alloys. Quenching from the liquid state, however, generally results in an amorphous matrix in which microcrystals of either the transition metal or of a phosphide or both are present in small quantities. Binary alloys belonging to this class are Fe-P, Ni-P, Pd-P and Pt-P. It was found, further, that pseudo-binary alloys containing two of the above mentioned compositions can be quenched into an entirely amorphous structure. Studies have been reported on the ternary system Pd-Ni-P, Pd-Fe-P (Maitrepierre 1969) and Pt-Ni-P (Sinha 1970). Another ternary alloy of interest is FePC, which is amorphous around the atomic composition Fe_{75}-P_{15}-C_{10} and is strongly ferromagnetic (Duwez et al. 1967, Lin 1969).

The number of amorphous alloys obtained sofar by liquid quenching is not large enough to formulate definite rules governing the occurance of amorphous phases. Three observations can be expressed as follows: 1/ An amorphous phase is likely to be found near a eutectic composition providing the eutectic is of the "deep" type, meaning that the eutectic temperature is low with respect to the melting point of the predominant metallic constituant and the liquidus curve drops sharply with the addition of the alloying element. This condition may be called "The deep eutectic condition". 2/ Ternary or quaternary alloys are in general easier to quench into an amorphous state than binary alloys, providing the first condition is fulfilled for the two binary alloys involved. This seems to imply that in an alloy containing a large number of different atoms it is easier to prevent the normal process of crystallization. This condition may be called "The confusion principle". 3/ The amorphous alloys obtained by liquid quenching reported in the literature sofar always contain a transition metal as a major constituant (from about 70 to 80 at.%) and a non-metal such as B, C, Si, Ge, As, or P. It seems that as a condition for glass formation from the liquid state, a certain amount of covalent bonding is required. This could be called the "Covalent bonding condition". It is interesting to note that the "deep eutectic condition" mentioned above is generally found in binary alloy systems involving a transition metal and a non-metal of valence 3 or more. Hence, conditions 1 and 3 may eventually be reduced to one more fundamental condition related to the electronic configuration of the constituants of the alloys.

Properties of Amorphous Metallic Phases

Amorphous metallic alloys are likely to have unusual physical properties because of their unusual structure. The electrical resistivity of an amorphous Pd_{80}-Si_{20} alloy has a very small temperature coefficient between 1.5°K and 200°C (about $17^{-5}/^{\circ}C$). Such a singular behavior for a metallic alloy is attributed to the fact that most of the scattering is due to the structural atomic disorder and the contribution of the thermal scattering is very small. When a transition metal such as chromium is substituted for Pd in the alloy, a minimum in the electrical resistivity curve is observed (Tsuei et al. 1969). The temperature at which the minimum occurs increases with increasing concentration of Cr and is about 200°C for 7 at.% Cr which is about the maximum concentration above which the amorphous state cannot be obtained. Between the minimum and absolute zero, the resistivity increases steadily with decreasing temperature, and this alloy constitutes a very sensitive resistance thermometer for low temperatures. In addition it has been found that the resistivity of this alloy is not affected by very heavy doses of radiation (Lesueur 1968). The probable reason for this insensitivity to radiation is the fact that radiation cannot introduce more defects than those already present in the amorphous alloy. The existence of a minimum in the electrical resistivity-temperature curve of amorphous alloys containing Cr is related to the so-called Kondo effect previously observed in crystalline alloys. One major difference, however, is that the Kondo effect in crystalline alloys is generally found in alloys containing a rather low percentage of magnetic impurity, and the temperature at which the minimum occurs is very low. In amorphous alloys the magnetic impurity content may be as high as 10 at.%. Previous theories based on the hypothesis of a dilute impurity alloy are not applicable, and this problem is an important one in theoretical solid state physics. Several amorphous alloys have been found to be ferromagnetic. In particular, the Fe_{75}-P_{15}-C_{10} alloy has a saturation magnetization of about 7000 G, a cohersive force of 3 Oe and a Curie temperature of 320^{o}C (Duwez et al. 1967). Although the existence of ferromagnetism in a non-crystalline solid has been predicted by Gubanov (Soviet Phys, Solid State 2, 468, 1960) no such ferromagnets were found until recently. Many questions are still unanswered on the exact nature of ferromagnetism in amorphous solids. It is not yet clearly established whether or not domain boundaries exist in amorphous ferromagnets, and if so, what is their origin, their width and their mobility. These new alloys present a challenge to experimental and theoretical physicists. The study of the mechanical properties of metastable phases has not yet received much attention, probably because of the difficulties associated with the small size of the available specimens. Judging from the relatively large number of papers dealing with this subject presented at this conference, it is anticipated that an increasing and continuing effort will be made to overcome this apparent lack of interest in the subject.

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