表1 メカニカル特性

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<td>メカニカル特性のAmorphous Metals</td>
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Mechanical Characteristics of Amorphous Metals*

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Synopsis

This paper reviews new informations obtained from the latest experiments concerned with the mechanical properties of amorphous metals. The selected subjects are of static and dynamic properties, plastic deformation and toughness, and several important effects on mechanical behaviors such as compositional, structural and environmental effects.

Amorphous metals are a new type material having consistently very high strengths together with a distinguished toughness and also they are classified as an elastic-perfectly plastic solid. In amorphous transition metal-metalloid alloys, the alloy system containing silicon and boron has high values of strengths and hardness, and a wide region of concentrations for becoming amorphous. An appreciable loss of ductility in some amorphous ferro-alloys takes place by aging at temperatures much lower than the crystallization temperature or by hydrogen embrittlement under a corrosive condition.

I. Introduction

Recently, many researchers are taking considerable interest in the mechanical properties of amorphous metallic materials. Because, their extremely disordered atomic arrangements must result in mechanical properties different from those of crystalline metals which inherently have a long range ordered structure. In earlier studies\(^{(1)}\)\(^{(2)}\) it has been found that they exhibit higher strengths and ductility in contrast to inorganic glasses which are generally extremely brittle. Since then various examinations of the mechanical behavior have been performed with a number of amorphous metals. While, at the end of 1973, Allied Chemical Corporation made public several kinds of amorphous iron-nickel base alloys produced in a semi-industrial scale with a trade name “Metglas”. Their success in production in a mass scale has made us believe that the practical use of these new materials is not far from realization. Under this circumstance it seems very significant to make a review of recent studies on the mechanical property of amorphous metals, I believe, one of the most important character.

Most of valuable results which have been obtained up to now are summarized

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* The 1673th report of the Research Institute for Iron, Steel and Other Metals.

in several review articles described by Gilman, Davis, Pampillo and ourselves. Accordingly, in this paper I will devote myself the discussion of our latest experimental results together with a brief comment on general characters. The selected topics are listed as follows: (1) static and dynamic properties, (2) plastic deformation and toughness and (3) several important effects on mechanical properties.

II. Static and dynamic properties

As is well known, amorphous metals have consistently very high values of static strength and hardness. For instance, the tensile properties of some typical amorphous metals are illustrated in Table 1, wherein upper column is the data of our experiments and the lower is the ones with Metglas. The highest nominal fracture strengths are about 350 kg/mm² for iron-base alloys, 300 kg/mm² for cobalt-base alloys and 270 kg/mm² for nickel-base alloys. These values are about the same or slightly higher than the highest value of conventional crystalline metals. In this table there are some discrepancies in the ratios of Young's modulus and hardness against the fracture strength. In the both cases our data are smaller by a few tens percent than the other values. It will be difficult to discuss on this discrepancy in detail, but this is probably due to some difference in the accuracy of measurements, in particular, of Young's modulus. Anyway, the ratio of Young's modulus to fracture strength is about 50 on the average, and the ratio of hardness

<table>
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<tr>
<th>Alloy system</th>
<th>Hardness (DPN)</th>
<th>Fracture strength (kg/mm²)</th>
<th>Young's modulus (kg/mm²)</th>
<th>E/σf</th>
<th>Hv/σf</th>
<th>Tc (°C)</th>
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<td>136</td>
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<tr>
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<td>420</td>
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<td>57</td>
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* Data from catalogs of Metglas by Allied Chemical Corporation.

to fracture strength is very close to 3.0, which is predicted from Hill's theory as pointed out by Davis.\(^8\) Also, the effect of the alloy composition on the strength will be discussed later.

The stress-strain curves of these amorphous metals are quite characteristic in both tension and compression test. Figure 1 shows characteristic nominal load-extension curve\(^1\) for tension and stress-strain curve\(^9\) for compression of Pd-Si base alloys. It is noticeable that the specimens rupture abruptly after a permanent elongation of about 0.1 percent in their tension curves. In the compression curve the specimens deform with a jerky flow along localized shear bands above room temperature after sharp yielding. This characteristic stress-strain curve with a jerky flow can not be observed below room temperature. According to Pampillo and Chen\(^9\) the jerky flow is caused by a partial relaxation of the atomic rearrangements, such as regeneration of the destroyed short range order, produced by plastic deformation. They believe that there is a critical temperature, where partial relaxation of rearrangements may occur quickly as the rearrangements take place, and below this temperature the atomic mobility is too small and plastic deformation becomes stable.

Recently several studies on the fatigue properties of amorphous metals were carried out by tension-tension mode. Figure 2 is the experimental results of Davis\(^10\) and of ours\(^11,12\) on the stress-lifetime behavior of Pd-Si base alloys.

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Fig. 1. Load-extension curves of Pd\(_{90}\text{Si}_{10}\) alloy by tension (left) and stress-strain curves of Pd\(_{77.5}\text{Cu}_{4}\text{Si}_{18.5}\) alloy by compression (right).

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\(^8\) L.A. Davis, Scripta Met., 9 (1975), 431.


\(^12\) T. Ogura, K. Fukushima and T. Masumoto, Scripta Met., 9 (1975), 979.
under various conditions of constant mean stress or constant minimum stress. Here, \( \sigma_m \) is the mean stress acting on the sample, \( \sigma_y \) is the yield strength, and \( \sigma_s \) is the average applied stress. As a result, it was pointed out that for large values of the ratio \( S \) amorphous metals exhibit a lifetime about three orders of magnitude greater than steels with comparable fracture strength, and that the long lifetime is reached at stress ratio of about 0.07 in Ni-Fe base alloys and about 0.15 in Pd base alloys.\(^{(10)}\) From our experiments\(^{(11,12)}\) the dynamic properties of amorphous metals are summarized as follows: The fatigue strength is a high level and a sharp knee is observed on the S-N diagram. Here, the fatigue fracture results from the nucleation of a crack, which grows gradually accompanied by striation markings on the fracture surface. Also, from the study of the propagation of fatigue cracks, it is concluded that the rate of crack propagation can be expressed as a power function of \( K_1 \) shown by equation (1). Here, \( n \) is approximately 4. It is also concluded that the size of plastic zone \( Z_p \) is related to \( K_1 \) as equation (2), where \( m \) is about 2. These values are in good agreement with those predicted theoretically.

\[
\frac{dl}{dn} = CK_1^n \\
Z_p = AK_1^m
\]

III. Plastic deformation and toughness

1. Mode of deformation

At temperatures well below the glass transition temperature \( T_g \) amorphous
metals are ductile on the microscopic scale, while they are brittle macroscopically. Namely, slip deformations occur in a localized shear band in the cases of uniaxial tension and compression. On the other hand, at temperatures around $T_g$ plastic deformation occurs homogeneously by viscous-like flows. As an example the temperature dependences of fracture strength and elongation for Pd-Si alloy are shown in Fig. 3. As temperature increases above $150^\circ C$, the strength decreases rapidly and the elongation increases. One of the important aspects of amorphous metals is that in several experiments\(^{(13,14)}\) a strain hardening is negligibly small even though it is either softening or hardening. Since ideal plastic materials should become unstable in tension and begin to neck as soon as yielding takes place, this fact implies that amorphous metals can be considered as elastic-perfectly plastic solids.

Fig. 3. Temperature dependence of fracture of maximum stress (*) and elongation of amorphous (AM), metastable (MS-I, MS-II) and stable (S) phases in Pd$_{80}$Si$_{20}$ alloy.

Then, as temperature increases toward the glass temperature, the mode of deformation apparently changes from the inhomogeneous type to more homogeneous type around a certain temperature. This critical temperature seems to be fairly lower than the glass temperature depending on the strain rate. Figure 4 shows the temperature- and strain rate-dependence of the strength and mode of deformation.\(^{(15)}\)

The chained line indicates an approximate boundary between inhomogeneous and homogeneous mode of deformation. In the two regions there are distinct differences in the strain rate dependence of strength as well as the mode of deformation. In the homogeneous region the strength increases as the strain rate increases, while in the inhomogeneous region its dependence is small or opposite. The temperature dependence of strength also changes at this boundary. The strength is less sensitive to temperature in the inhomogeneous region, whereas a strong temperature dependence exists in the homogeneous region. These facts imply that the flow can be controlled by an athermal mechanism at lower temperatures and by a thermally activated one at higher temperatures. Such a difference in the mechanism may be supported by several experimental results on the structural change during plastic deformation. Inhomogeneous deformation causes disordering in the amorphous state,\(^{(16)}\)\(^{(16, 17)}\) while homogeneous deformation promotes ordering of atoms.\(^{(15)}\)

Figure 5 is a schematic presentation of the temperature dependence of the stress in homogeneous and inhomogeneous states constructed from the previous arguments for the plastic deformation of amorphous metals.\(^{(18)}\) Here, the stress is a value necessary to induce continuous deformation at a constant strain rate, and


\(^{(17)}\) H.S. Chen, Scripta Met., 9 (1975), 411.

Fig. 5. Schematic diagram showing the variation of the mode of deformation with temperature. The vertical axis denotes the stress which is necessary to deform amorphous metals at a constant rate. If the testing temperature is higher than $T_p$, homogeneous deformation takes place because the stress is smaller than that for inhomogeneous deformation.

Fig. 6. Temperature dependence of hardness of amorphous Pd$_{80}$Si$_{20}$ alloy. $T_g$ is the glass transition temperature and $T_c$ the crystallization temperature.

the critical temperature $T_p$ is defined at the point where the stresses of two modes coincide with each other. We have found recently the similar behavior in the high temperature hardness.\(^{(19)}\) Figure 6 shows the hardness of Pd-Si alloy plotted against temperature. The two turning points observed clearly at 320°C and 380°C well coincide with $T_g$ and $T_c$, respectively, while the one at 190°C seems to correspond to $T_p$. As seen in the figure, below this temperature indentation with

\(^{(19)}\) A. Inoue and T. Masumoto, Scripta Met., to be published.
microhardness tester produces deformation markings of a coronet, and above this temperature no marking is found under the similar conditions.

This critical temperature resembles to the so-called equicohesion temperature which has been proposed in the phenomenological study of the fracture of crystalline metals. At present, to obtain better understanding of this temperature, comparison of the resistance against inhomogeneous and homogeneous deformations is important and its variation with the strain rate should be clarified extensively.

2. Inhomogeneous deformation

On the flow character of inhomogeneous deformation quantitative data are hardly obtained because of its occurrence within a limited, localized shear band except for data obtained by compression test\(^{(9)}\) which is usually carried out under complex conditions. In general, the apparent yield and fracture stresses are higher than those of the major elements composing amorphous metals in the crystalline state. Based on the dislocation model the theoretical strength of amorphous metals is estimated as 0.03 to 0.05 of the Young's modulus. However, because of experimental difficulties, there is no exact value of the critical resolved shear stress of amorphous metals, which can be compared with the theoretical value.

In a tensile experiment using a conventional tensile testing machine, amorphous metals rupture in a brittle manner at temperatures below \(T_g\). Because of this fact it is difficult to observe the generation of slip lines and the behavior of the displacement along these lines. Recently, we developed a new type of tensile testing machine for the purpose of studying the detailed process of slip deformation

![Graph](image)

Fig. 7. Load-elongation curve of amorphous \(\text{Pd}_{60}\text{Si}_{20}\) alloy obtained under extension speed of about \(1 \times 10^{-4}\)/sec at 20°C by using a new developed hard type tensile testing machine.
and the critical resolved shear stress of amorphous metals. Figure 7 shows a load-elongation curve of amorphous Pd$_{80}$Si$_{20}$ alloy at 20°C. This sample yields abruptly at the time when the slip takes place and the slip proceeds in an intermittent manner along a particular slip band. This fact shows that the slip takes place when the shear stress acting on this slip plane exceeds a certain critical value of the applied stress. In this experiment, the critical resolved shear stress of Pd$_{80}$Si$_{20}$ alloy was estimated to be about 95 kg/mm$^2$. Moreover, by estimating the relationship between slip and applied stress it was clarified that the strain hardening is inoperative.

3. Homogeneous deformation

The flow character at temperatures above $T_p$ is more quantitatively investigated by creep testing.$^{(13),(21-24)}$ Creep deformation of amorphous metals occurs homogeneously at temperatures above $T_p$, and especially near $T_g$. It is known that a transient creep is induced and that the deformation is of viscoelastic origin.$^{(23,24)}$ There is a distinct difference from the creep curve of crystalline metals which exhibits also viscoelastic flow (diffusion creep) at just below the melting point. By contrast with case of diffusion creep, the creep rate of amorphous metals decreases as the creep strain increases. This phenomenon shows that there should be some strengthening effect, such as an increase in creep activation energy, an increase in internal stress and a decrease in uncertain elements which participate in the deformation. The structural change to a more stable state is also considered to be responsible for this effect of the testing temperature is much higher than $T_p$. It seems that we need to investigate in more details these factors mentioned. Furthermore, diffusion measurements in amorphous metals would be desirable for drawing more definite conclusion from the creep experiments.

4. Ductility and toughness

As mentioned above, amorphous metals are brittle in the tensile experiment at room temperature. On the other hand, large strains are found to exist in compression and bending tests. Also, amorphous metals can be deformed severely by cold-rolling without cracking. Therefore, they can be classified as ductile glass in contrast to oxide glass.

On this view we measured the mode III fracture toughness of three alloys using a tear test of a trouser-leg type.$^{(25)}$ Davis$^{(10)}$ also examined the thickness dependence of fracture toughness to Ni-Fe base alloys in single edge notch and

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center cracked panel configurations. In Fig. 8 the fracture toughness $K_c$ measured by Davis is plotted against the sample thickness for Fe-Ni base alloys, together with our data. It is found that the fracture toughness is about 30 kg/mm$^{3/2}$, which is close to the values of high strength steels if we can assume a higher yield strength and a low modulus for amorphous metals.\(^{(16)}\)

**IV. Several important effects on mechanical properties**

1. Compositional effect

In amorphous metals which contain, almost invariably, metalloid atoms as one of the constituents, the strength is greatly affected by the compositions of the alloys. In a recent study\(^{(26)}\), the compositional effects on the hardness were examined using amorphous iron-base alloys with various alloying elements. Figure 9 shows the hardness plotted against the concentration of alloying elements in Fe-P-C alloy, where iron atoms are replaced by other transition element atoms. It is clearly seen that hardness increases by addition of Ti, V, Cr and Mn, while it decreases by addition of Co, Ni and Cu, whose atomic numbers are larger than Fe's. Thus, alloying elements from Ti to Cu show the group number effect of the periodic table. This effect can be seen more clearly in Fig. 10, where the hardness is plotted against the concentration of average outer electron $s$ and $d$ of metallic

Fig. 9. Microhardness plotted against the concentration of alloying elements (X) in amorphous (Fe-X)$_{80}$P$_{13}$C$_{7}$ alloys.

Fig. 10. Microhardness plotted against the average outer electron (s, d) concentration of metallic atoms (e/a) for amorphous (Fe-X)$_{80}$P$_{13}$C$_{7}$ alloys.

atoms. The data show that there is a correlation between hardness and average concentration of the outer s, d electrons. After some arguments we have proposed that the major role of the outer electrons of transition element in amorphous metals is not to increase the cohesive energy of solid like in the case of crystalline solids, but to weaken the bond strength associated with metalloid atoms which seem to play the dominant role in determining the flow property of amorphous metals. The same effect\(^{(27)}\) is also seen in Fig. 11, which shows the change of hardness in

Fig. 11. Microhardness of amorphous \((X, Y)_{30}\text{Si}_{10}\text{B}_{12}\) alloys with \((X, Y) = \text{Fe, Co, Ni}\).

Fig. 12. Relationship between the microhardness and the total atomic percent of metalloids for Fe-base, Co-base and Ni-base amorphous alloys with combinations of metalloids (C, P, B, Si).

Binary systems consisting of Fe, Co and Ni with a constant concentration of Si and B. In each case the hardness changes monotonically. Also, its value is high in the cases of Fe and Co, but decreases appreciably by addition of Ni element.

In Fig. 12, the relationship between the hardness and the total atomic percent of metalloids is shown for Fe-base, Co-base and Ni-base alloys with various combinations of metalloids such as C, P, B and Si. From this figure ones are able

to find the region of metalloid concentrations where the alloys become amorphous. A Si-B system has a high value of hardness and a wide region of concentrations for becoming amorphous. After making a statistical analysis we have found that the effect of metalloids on hardness increases in the order of P, C, Si and B in Fe-base alloys, and as far as the hardness is concerned the B-base system and B-Si system are the most prominent alloy system. In the figure the hardness of Metglas is also plotted, which seems to be consistent with our results.

In amorphous metals it is more advantageous to have metals which show not only a high value of hardness but a high crystallization temperature, which can be considered as a measure of structural stability. A correlation between these two quantities is shown in Fig. 13 and also Si-B system is shown to have the best quality in this respect. This is why our recent studies of amorphous metals are concentrated intensively on this system.

![Graph showing microhardness vs. crystallization temperature for different metalloid combinations](image)

Fig. 13. Correlation between the microhardness and the crystallization temperature for Fe-base, Co-base and Ni-base amorphous alloys with combinations of metalloids (C, P, B, Si).

2. Structural effect

Since the amorphous phase is thermodynamically unstable, the transformation to the crystalline phase occurs during cooling from the molten state with a relatively slow rate, or heating and aging at elevated temperatures. In general, the ductility of amorphous metals disappears gradually as crystallization proceeds. Recently it has been found that an appreciable loss of ductility in iron-base alloys is induced by aging even at temperatures appreciably lower than the crystalliza-
tion temperature (determined by means of X-ray and electron analyses). Figure 14 shows the relation between crystallization and fracture strain estimated by a bending test. $E_f$ equals to unity means that no fracture occurs even after perfect bending. As seen in the figure embrittlement takes place within a very short time at low temperatures prior to crystallization. Figure 15 also show the

![Graph](image-url)

**Fig. 14.** Temperature and time dependences of the fracture strain ($E_f$) and crystallization diagram of amorphous Fe$_{80}$P$_{13}$C$_7$ alloy.

![Graph](image-url)

**Fig. 15.** Changes in the fracture strain by aging at 300°C for Pd-base, Fe-base, Ni-base and Co-base amorphous alloys.

change in fracture strain by aging at 300°C of Pd, Fe, Ni, Co-base alloys.\(^{(31)}\) In iron-base alloys the fracture strain decreases by aging at 300°C, and also the Fe-P-C alloys are more brittle than the Fe-Si-B alloys. In contrast other alloys stay as ductile. Therefore, it seems that embrittlement after aging at low temperatures is not a general character of amorphous alloys, but only characteristic to the amorphous ferro-alloys.

The development of embrittlement by aging at the incipient state of crystallization may be caused by a change in the bonding nature between the constituent atoms such as an increase of the covalent bonds due to ordering of atoms in the short range. In addition there are other empirical evidences which are appeared to be related to the structure dependence of flow. For example amorphous metals such as Co-P and Ni-P synthesized by electro- or chemical deposition are entirely brittle in spite of the fact that the same alloys produced from the liquid state are ductile. As mentioned above, the ductility of some of amorphous metals is affected sensitively by a slight change in the structure which is not easily detected by conventional methods used for structural analyses. Accordingly, better experimental procedures should be needed to provide more detailed information about the atomic configuration in short range and the bonding nature of atoms in relation to the amorphous structure and flow mechanism.

3. Environmental effect

As the final subject the environmental effect on the mechanical effect on the mechanical properties of amorphous iron-base alloys is discussed. We reported before that the iron-base amorphous alloys containing a small amount of chromium exhibit remarkably high resistance to chemical corrosion, particularly to pitting corrosion.\(^{(32,33)}\) However, it was observed recently that some iron-base alloys suffer quite easily environmental effects such as hydrogen embrittlement (HE)\(^{(34,35)}\) and stress corrosion cracking (SCC),\(^{(31,85)}\) and their plasticity is almost lost by these effects. The main effects observed in several our experiments are summarized as follows:

1) Under cathodic polarization hydrogen embrittlement occurs in the most of solutions.
2) Highly corrosion resistant alloys are susceptible to hydrogen embrittlement at

corrosion potential and under anodic polarization only when tensile stress is applied in strongly acidic solutions with a certain amount of chloride ions.

3) Stress corrosion cracking, which is a synthetic effect of anodic dissolution and stress, may not take place. Embrittlement during traction in corrodenets are attributable to hydrogen embrittlement.

4) Fe, Ni and Co base alloys containing a large amount of metalloids with a strong affinity for hydrogen are more susceptible to hydrogen embrittlement than ordinary steels due to cooperative action of stress and corrosion.

5) When a stress is applied to low corrosion resistant alloys, hydrogen embrittlement may occur even under atmospheric conditions perhaps due to corrosion by water vapor.

These effects will be explained by indicating several examples. Figure 16 shows the stress-strain curves in different environments of amorphous Fe-Cr-Ni-P-C alloy having high corrosion resistance. The fracture stress is about 270 kg/mm² in air. In strongly acidic solutions with a certain amount of chloride ions, the fracture stress decreases remarkably. Under cathodic polarization where the reduction of hydrogen ions takes place, the fracture stress decreases to about 100 kg/mm² due to hydrogen embrittlement as shown by the curve 4. Even in the case that hydrogen reduction does not occur, a remarkable decrease of fracture stress takes place as seen by the curves 2 and 3. From examinations in more details we found that it is probably caused by hydrogen embrittlement due to local cell corrosion. Some nature of hydrogen in amorphous alloys was studied. According to our experimental results an absorbable amount of hydrogen is higher than that in crystalline metals and degassing of hydrogen seems to occur.

![Fig. 16. Stress-strain curves of amorphous FeCr₇₄Ni₃₅P₁₄C₁ alloy in air and H₂SO₄+NaCl solutions at strain rate of 4.2×10⁻⁶ sec⁻¹.](image-url)
gradually in air even at room temperature. Also the fracture stress is recovered completely after several hours. Furthermore, it was found by Nagumo and others\(^{(34)}\) that diffusion constant in amorphous alloys is much smaller by about \(10^{-4}\) than that in iron or steels, and is rather close to that in covalent crystals. They estimated the activation energy for diffusion of hydrogen is about 2000 cal/mol. Such absorptive power and diffusibility of hydrogen atoms in amorphous structure are attributed to relatively high susceptibility to hydrogen embrittlement of amorphous alloys. Recently we observed that a delayed fracture occurs in Fe-P-C alloys exposed under stresses above the critical level in air at room temperature.\(^{(36)}\) The result is shown in Fig. 17. Such a drastic decrease in plastic flow seems to be mainly due to a harmful effect of hydrogen atoms diffused into inhomogeneous shear slip bands. However, further investigation seems to be needed in order to understand more definite roles of hydrogen atoms on the bonding nature of atoms.

![Fig. 17. Delayed fracture of amorphous Fe\(_{40}\)P\(_{13}\)C\(_7\) alloy under various stresses at room temperature in air.](image)

V. Final remarks

In the present paper an outline of the mechanical properties of amorphous metals was presented by showing a number of examples from our studies. All informations explained so far are very macroscopic ones, because we do not have any definite model for the short range structure which gives a significant influence on the intrinsic nature of amorphous metals. Besides the mechanical properties it is desirable to know other physical properties, which are playing significant role in the conventional solid state physics. Examples are the measurement of Gruneissen constant, validity of Wiedemann-Frantz law and study of vibrational modes in amorphous materials. Also, the statistical mechanical treatment which has just been applied to spin glasses should also be applied to the present problems to clarify the thermal and mechanical properties.

\(^{(36)}\) T. Masumoto, H.M. Kimura and S. Arakawa, to be published.