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To the Readers of METALLURGICAL TRANSACTIONS:

This first issue of METALLURGICAL TRANSACTIONS marks a major advance in the metallurgical profession. Two eminent publications, the TRANSACTIONS of The Metallurgical Society of AIME and the TRANSACTIONS QUARTERLY of the American Society for Metals, have now been merged into a single outstanding journal. Thus, our two sponsoring Societies are henceforth joined in the publication of high-standard papers dealing with a broad spectrum of metallurgically-oriented research and development. The Boards of TMS and ASM are confident that this new medium of informationdissemination in the field of metals science and engineering will serve its readers more effectively and comprehensively, as well as more economically, than would otherwise be possible.

In behalf of the metallurgical generations yet to come, and as Presidents of the respective Societies, we extend our collective appreciation to the committees and staff-members of both organizations whose vision and devoted efforts brought this new journal into existence.

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ADDRESS REPLY TO: THE METALLURGICAL SOCIETY, 345 EAST 47 STREET, NEW YORK, N.Y. 10017

Stress-Induced Pseudoelasticity in Ternary Cu-Zn Based Beta Prime Phase Alloys

HORACE POPS

"Pseudo" elastic strains up to approximately 15 pct have been observed in ternary Cu-Zn-Si and Cu-Zn-Sn β' phase alloys during loading in tension, compression, or bending. Metallographic observations and mechanical property studies have shown that the unusually large elastic strains occur by means of a stress-induced martensitic transformation. The relationship between spontaneous martensite and stress-induced martensite is discussed. The influence of temperature, composition, plastic flow stress, and elastic modulus upon the transformation induced pseudoelasticity are discussed.

IT is characteristic of the bcc β' phases that they transform martensitically during cooling and that this transformation is very sensitive to strain. In a few systems, such as Cu-Zn,¹ the martensite phase may form "thermoelastically", during which process the martensite plates grow as the temperature is lowered and shrink upon heating, with little or no temperature hysteresis. It is also common for a similar form of martensite to grow during application of stress and to shrink or disappear when the load is removed. The formation under stress of a reversible "elastic" martensite may result in rubber-like properties. This has been reported for In-Tl,² Au-Cd,³ and Cu-Al-Ni⁴ alloys. The term "super-elasticity" was used by Rachinger⁴ in 1960 to describe the large elastic strains (~4 pct) which occurred at room temperature during the stressing of single crystals of Cu-14.5 Al-3 Ni alloys. Superelastic behavior has also been observed in coarse grained polycrystalline Cu-Al-Ni alloys.⁵ Much attention has been focussed recently on the NiTi alloys⁶ (the Nitinols) which show a very unusual mechanical "shape-memory" effect as a result of a martensitic transformation.

Thermoelastic martensite was studied in previous investigations^{7,8} of ternary alloys based upon the Cu-Zn β' brass phase (Cu-Zn-Ni, Cu-Zn-Ag, Cu-Zn-Au, Cu-Zn-Cd, Cu-Zn-In, Cu-Zn-Ga, Cu-Zn-Si, Cu-Zn-Ge, Cu-Zn-Sn, and Cu-Zn-Sb). The data indicated that by a suitable choice of composition, these alloys might show rubber-like behavior under stress near room temperature. On the basis of preliminary experiments,⁹ the systems Cu-Zn-Si and Cu-Zn-Sn were selected for further detailed studies. The object of this research was to determine the extent of elasticity occurring as a result of the elastic martensite transformation. The role of applied stress, transformation temperature, and test temperature was also examined.

I) EXPERIMENTAL PROCEDURE

Alloys were chosen on the basis of their transformation temperatures, see Fig. 1, which depend upon chemical composition. M_s temperatures were se-

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lected in the range between room temperature and approximately -50° C. Six Cu-Zn-Si and Cu-Zn-Sn alloys, each corresponding to an electron-to-atom ratio of 1.395, were prepared and their nominal compositions are given in Table I. High-purity metals (>99.99 pct) were used, the alloys being melted and cast in sealed quartz tubes under a partial pressure of helium. Samples were homogenized in evacuated quartz capsules held for 24 hr at temperatures within the single β -phase field, ¹⁰ ~790° to 830°C. Since weight

Alloy	Cu	Zn	Si	Sn
A	62.5	36.5	1.0	
В	63.3	35.3	1.4	2
С	64.1	34.1	1.8	
D	62.9	35.9	-	1.2
E	63.8	34.55		1.6
F	64.9	32.9		2.2



Fig. 1—Transformation temperature (M_g) of ternary alloys based upon the Cu-Zn β' phase. e/a = 1.395.

losses were negligible, the compositions after melting and homogenization were assumed to be the same as the nominal compositions. In order to prevent precipitation of the fcc α phase, the alloys were quenched from the homogenizing temperature into an iced brine solution. Metallographic examination was carried out on samples electropolished in a 10 pct solution of KCN in water using alternating current. The samples were completely β' prior to the mechanical deformation studies and exhibited a coarse grain size (~1 to 5 mm). An Instron machine was used for tensile tests, threepoint bending, and compressive loading. Testing was done at temperatures between the M_s of the sample and approximately 100°C. A liquid (in most cases water) was used to maintain a constant specimen temperature for the tension and compression tests. The progress of the transformation occurring during loading of a bend sample was observed using a Reichert Metallograph and a specially constructed deformation jig. Several single crystals were grown by the Bridgman technique and were used for compression studies.

II) RESULTS

A) Effect of Applied Uniaxial Stress. The tensile curve of a typical Cu-Zn based ternary alloy is shown in Fig. 2. Upon initial loading, stress increases linearly with strain. With further loading, the stressstrain relationship is very much different from that of conventional polycrystalline materials. When the load is increased beyond what appears to be the proportional limit, the curve becomes nearly horizontal and the strain increases approximately 10 pct in this plateau region. At higher loads, the stress again varies linearly with strain, and the point of deviation from linearity has been termed the plastic yield point. The total strain measured up to this point appears to indicate a seemingly elastic nature because the specimen acquires little or no permanent deformation (set) after unloading. Apparent elastic strains as high as 15 pct have been measured in coarse grain polycrystalline specimens. In several specimens of alloy F a reduction in the cross section was detected visually at high strains, but disappeared when the load was removed.

Serrations were generally visible in the plateau re-



Fig. 2–Typical stress-strain curve for a silicon-bearing alloy deformed in tension at room temperature. Total elastic strain is ~ 15 pct.

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gion of the tensile curve, as may be seen in Fig. 3. Upon unloading, the stress drops rapidly at first, and then becomes nearly horizontal, producing a characteristic hysteresis loop. As in the case of loading, serrations are visible in the horizontal portion of the unloading curve. The shape of the curve in this region was quite irregular and differed from sample to sample. Although a small permanent set usually remained in the test sample after unloading, large elastic strains were reproduced upon reloading.

The point of deviation from linearity in the tensile curves prior to the plateau region occurred at different values of stress depending upon the alloy composition. Assuming that a strain induced martensitic transformation occurs at the point of deviation, the stress required to induce this transformation ought to be dependent upon the strength or stability of the matrix and, therefore, the temperature at which the sample is deformed. Accordingly, polycrystalline tensile specimens of several ternary Cu-Zn-Si and Cu-Zn-Sn alloys were deformed elastically over the temperature range between the M_s of the alloy and 100°C. Fig. 4(a) shows the stress-strain curves at temperatures between 27° and 78°C for a silicon-bearing alloy. The stress at departure from linearity on loading is plotted as a function of test temperature in Fig. 4(b). At the lower test temperatures, very little stress is required to induce the transformation. As the test temperature is increased, stress also increases. The plot of loading-stress vs temperature results in a straight line which extrapolates to the M_s temperature at zero stress. Data obtained from the unloading curves also give a straight line plot, but in this case extrapolate



Fig. 3—Portion of tensile curve for a ternary Cu-Zn-Sn alloy deformed at room temperature showing characteristic hysteresis loop after unloading. Serrations are associated with the formation of elastic martensite.

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to the initial transformation temperature during heating (As) at zero stress. Similar behavior was observed for all ternary alloys containing silicon; the tensile stress required to initiate a martensitic transformation became zero at the M_s temperature.

Tensile stress-strain curves for the Cu-Zn-Sn alloys were similar to those described for the Cu-Zn-Si alloys, as shown in Fig. 5(a). The stress upon loading becomes zero near the M_s temperature and increases linearly with increasing test temperature. Some of the tensile curves contain two peak maxima, each resembling a plastic yield point (see curve for 57° C); however, little or no permanent set was observed after unloading. Since the samples had very large grains, it is assumed that these elastic instability points are associated with the onset of a martensitic transformation in different grains. It should be noted that the applied tensile stress produces



plastic deformation at 81°C, but no permanent set occurs in samples tested below 78°C. Therefore, for this particular specimen, pseudoelasticity occurs in the temperature range between -6°C (the M_s) and less than 81°C.

Large elastic strains were also produced in the ternary beta brass type alloys by compressive loading. A representative stress-strain curve for a single crystal of Cu-Zn-Si is shown in Fig. 6. This curve indicates typical features of transformation induced pseudoelasticity in that it exhibits an initial region during which stress is proportional to strain, a plateau region where the "apparent" elastic modulus*

*The slope of the stress-strain curve does not give the true elastic modulus of the matrix, but indicates the occurrence of a phase transformation. The strains are not elastic in a true sense, rather the martensitic process results in "pseudo" elastic strains which resemble elasticity in that the strain is reversible.

decreases to zero or becomes negative, a hysteresis loop, and large elastic strains. The procedure of loading and unloading a polycrystalline compression sample at one temperature and repeating the cycle at different test temperatures produced transformation stress vs test temperature results shown in Fig. 7.





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Fig. 5-Variation of pseudoelasticity with test temperature for Cu-Zn-Sn alloy F deformed in tension. (a) Stress-strain curves showing onset of transformation and plastic deformation. (b) Stress vs temperature for loading and unloading.

Test Temperature, °C

(b)

20 30 40 50 60 70 80

0L 20

10

MsJLAs



Fig. 6–Stress-strain curve at $27^\circ \rm C$ for a single crystal of Cu-Zn-Si alloy C deformed in compression.



Fig. 7—Transformation stress vs temperature for alloy C deformed by compressive loading.

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As in the case of tensile loading, the stress corresponding to the departure from linearity in the stressstrain curve increased linearly with increasing test temperature. Similar trends were also observed when the stress obtained from the unloading curve was plotted against the test temperature.

B) Martensitic Transformation Under Bending Stress. A typical load vs deflection curve for a Cu-Zn-Si sample deformed at room temperature is shown in Fig. 8. On bending the specimen, large deflections are produced and the apparent elastic modulus approaches zero. Upon unloading, the specimen snaps back to its original shape, characteristic of "rubberlike" behavior. A hysteresis loop was observed in all specimens exhibiting this type of behavior. Metallographic examination during loading revealed that plates (traces are formed in the plane of polish by a section through the martensite. The traces will be referred hereafter as plates, or platelets.) of a martensitic phase form and grow side-by-side into the β' phase matrix under the application of stress. Fig. 9 shows a plot of length of plate vs deflection for a Cu-Zn-Sn sample deformed elastically at room temperature. A small increase in deflection (proportional to the applied strain) produces a small increase in



Fig. 8-Typical load vs deflection curve for pseudoelastic alloys deformed in three point bending, alloy B.



Fig. 9–Growth of elastic martensite with bending stress in alloy $\mbox{A}.$

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length. The arrest in growth occurred when martensite impinged upon grain boundaries of the β' phase.

The first platelets that were observed to grow were those already present, and most likely were produced by initial mechanical polishing. A number of new plates subsequently nucleated at the compression side of the sample and grew into the matrix with increasing stress. Their appearance corresponded to a point of departure from linearity in the load vs deflection curve. Transformation generally occurred at the compressive side with individual grains responding differently to the applied stress. This suggests that the formation of elastic martensite is dependent on the orientation of the grains. Martensite plates stopped growing at α precipitates or at the original β' phase grain boundaries, as shown in Fig. 10. They decreased in length upon unloading and usually disappeared when the load was removed. The plates which formed last upon loading disappeared first upon unloading but were stable to a lower stress level on unloading.

When the sample was reloaded, martensite formed at identical locations in the structure. A simple bending experiment was conducted to show the effects of stress (tensile vs compressive) upon the transformation behavior, see Fig. 11. The photomicrographs shown in Fig. 11(a) was obtained from the tensile side of a bend sample. Several different variants of the martensite habit plane are visible in each grain. Most of the plates disappeared when the load was removed, Fig. 11(b). When the area shown in Fig. 11(a) was reloaded under compressive stress, the strain induced martensite formed on different habit planes, see Fig.



Fig. 10-Martensite plates formed by elastic deformation in alloy A. Magnification 80 times.

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Fig. 11-Martensite formation under elastic bending stress in alloy F. (a) Tension side of the sample. (b) Load removed. Note lack of martensite plates. (c) Reloaded in compression. Note martensite plates lying on different planes from those shown in (a).

(c)

(a)

(b)

11(c). While loaded elastically, the specimen was cooled below the M_s until spontaneous martensite formed. It was observed that many of the existing strain-induced martensite plates decreased in length and disappeared, while other plates grew in length. Also, some of the thermoelastic martensite phase formed along different planes (possibly different variants of the same plane) in the matrix.

Elastic martensite very often has a different morphology from martensite formed by plastic deformation. Fig. 12(a) shows a region near the fracture surface of a permanently deformed tensile sample. The



Fig. 12—Martensite formation in alloy E deformed plastically in tension. (a) Martensite formation resulting from heavy deformation. Magnification 77 times. (b) Interaction between martensite platelets. Higher magnification.

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strain-induced martensitic phase is wavy when deformation is relatively heavy and broader than the elastic martensite. Intersection between different platelets is common, and often results in a microstructure similar to that shown in Fig. 12(b); this has a more regular pattern, and is similar to the thermoelastic martensite.

C) Elastic Moduli. A dynamical torsion pendulum method was used to determine the modulus of rigidity, G, in ten different ternary systems based on the Cu-Zn system. Details of the sample preparation and apparatus are described elsewhere.¹¹ Room temperature data were obtained for polycrystalline alloys. The spontaneous transformation temperature, which was varied by choosing different chemical compositions, was in the range between -18° and -169°C. Alloy compositions, transformation temperatures, and G values are given in Table II. It may be seen that the shear moduli of the ternary alloys are approximately the same as the binary Cu-Zn β' phase alloys, *i.e.*, $\sim 2 \times 10^{11}$ dynes per sq cm. G does not appear to be related to the martensitic transformation temperature, Ms, in contrast to some other alloy systems.12

III) DISCUSSION

Pseudoelastic strains as large as 15 pct have been observed in single crystals and polycrystalline samples of Cu-Zn-Si and Cu-Zn-Sn beta phase alloys during loading in tension, compression, or bending. This phenomenon may be described as a stress induced pseudoelasticity. For simplicity, the term STRIPE will be adopted in the following discussion. STRIPE differs from normal elastic behavior in that stress does not vary linearly with strain. In addition, the stress-strain curve during loading is not the same as the curve during unloading. The appearance of a hysteresis loop and the small amount of strain remaining in the sample after unloading suggest that the martensitic transformation is not completely reversible on unloading. Interaction between martensite plates and either prior β' phase grain boundaries or α precipitates, and possible interactions between plates on different habit

Alloy No.	At. pct. Cu	At. pct. Zn	At. pct. 3rd Element	M _s Temper- ature, °C	G × 10 ⁻¹¹ dynes per sq cm
G	60.5	39.5		- 126	2.00
H-1	60.15	39.6	0.25 Ni	-128	1.64
H-2	59.45	39.8	0.75 Ni	-136	1.84
H-3	57.7	40.3	2.0 Ni	- 169	2.55
I-1	60.25	39.5	0.25 Ag	- 139	1.84
I-2	59.75	39.5	0.75 Ag	-153	1.81
J-1	60.25	39.5	0.25 Au	-126	1.73
J-2	59.75	39.5	0.75 Au	- 132	1.71
K	60.5	39.25	0.25 Cd	- 149	1.41
L-1	60.65	39.2	0.15 Ga	- 135	1.80
L-2	61.25	38.0	0.75 Ga	- 99	2.88
L-3	62.0	36.5	1.5 Ga	- 84	1.93
M	61.25	38.0	0.75 In	- 140	1.95
N	61.0	38.75	0.25 Ge	- 107	1.60
0-1	61.0	38.75	0.25 Sn	-124	1.57
0-2	62.0	37.25	0.75 Sn	- 99	2.0
0-3	63.5	35.0	1.5 Sn	- 44	2.0
P-1	61.0	38.5	0.25 Si	-142	1.73
P-2	62.0	37.25	0.75 Si	- 96	2.05
P-3	63.5	35.0	1.5 Si	- 18	2.91
Q	62.75	36.5	0.75 Sb	- 37	2.14

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Table II. Shear Moduli of Ternary Cu-Zn Based β' Phase Alloys

plane (variants) within a single grain may produce localized plastic deformation. Plastic deformation is irreversible, and hence would result in permanent set after unloading.

At temperatures above the M_s (but below the T_0 temperature, *i.e.*, the temperature at which the chemical free energies of the parent and product phases are equal), the β' phase is thermodynamically unstable and may transform martensitically when sufficient stress is applied to overcome nonchemical free energies. In an early study of the martensitic reaction in steels, Scheil¹³ postulated that an (austenitic) lattice becomes both mechanically and thermodynamically unstable at the M_s temperature and that a critical resolved (elastic) shear stress is required to promote the martensitic transformation. Mechanical instability at the M_s implies that the elastic modulus is zero. From the present shear modulus experiments, G was approximately 2×10^{11} dynes per sq cm for all of the ternary Cu-Zn based β' phase alloys, irrespective of their M_s temperature. If the shear modulus is to become zero at the M_s , one would expect some reduction in the G value as M_s is approached. Since this has not been observed, it is concluded that the β' phase does not become mechanically unstable.

Internal stresses produced by the change in volume accompanying the transformation may be added to the external applied stress. The total stress must be less than the elastic limit of the β' phase if the resulting martensite phase is to be in equilibrium with the matrix. If this condition is satisfied, it is expected that growth of plates would be controlled by the rate of change of applied stress. This has been verified by metallographic examination of alloys during bending, see Fig. 9; the martensite phase grew slowly in the lengthwise direction with increasing applied stress and disappeared on the removal of the stress. The effect of increasing or decreasing the applied load is in this case equivalent to cooling or heating the specimen through the M_s temperature. Therefore, it should be possible to obtain STRIPE in other systems containing thermoelastic martensite. In the binary Cu-Zn β -phase alloys, stress induced martensite forms under tensile¹ or compressive15 loading, and for small loads disappears when the load is removed. In spite of this mechanical reversibility, the total elastic strain achievable under conditions of reversibility is less than $-\frac{1}{2}$ pct. Adding only a small amount of a third element such as silicon or tin increases the reversible elastic strains up to ~15 pct. A likely explanation for this rather remarkable increase is that the third elements increase the amount of thermoelastic martensite,⁸ and expand the range of stress in which the elastic martensite occurs.

Plastic deformation cannot occur until the sum of the applied and the internal stresses exceeds the elastic limit of the β' matrix. Whereas the stress required to produce plastic flow generally decreases with increasing temperature, the stress required to induce the elastic martensitic transformation increases with increasing temperature, see Figs. 4 and 7. Consequently, elastic martensite will occur more easily at lower temperatures. The variation of the stress to form elastic martensite and the plastic flow stress with temperature are illustrated schematically in Fig.

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13. The curves intersect at a temperature which will be called the "critical" temperature, i.e., that temperature where the plastic flow stress is the same as the stress required to form elastic martensite. Plastic flow (such as slip, twinning, or nonreversible strain induced martensite) occurs at lower stress levels above this temperature. The shaded region in Fig. 13 illustrates the ranges of temperature and stress in which pseudoelasticity is expected; the lower limit in temperature is the M_s , and the upper limit is the critical temperature. Increasing the strength of the matrix should raise the plastic flow stress curve and increase the critical temperature. Consequently, the temperature range for STRIPE will be extended. Additions of various third elements to Cu-Zn alloys will undoubtedly increase the strength of the β' phase by solid solution strengthening. A likely consequence of this strengthening is an increase in elastic behavior. Increasing the M_s temperature will shift the transformation stress curve toward high temperatures and therefore decrease the extent of pseudoelasticity.

The most characteristic features of pseudoelasticity may be summarized as follows:

a) Very large "pseudo"elastic strains are produced as a result of a stress induced reversible martensitic transformation; the phenomenon may be called <u>stress</u> induced pseudoelasticity (STRIPE).

b) The stress induced martensite phase is elastically balanced within the matrix, so that it disappears upon unloading.

c) Applied strains plus internal strains surrounding the martensite phase must be less than the plastic flow stress of the matrix.

d) Stress required to promote a martensitic transformation in β -brass type alloys increases with increasing test temperature.

e) Pseudoelasticity occurs only in the temperature range between the M_s and a (higher) critical temperature (defined as that temperature where the plastic flow stress of the matrix is equal to the stress required to form elastic martensite).

f) STRIPE is more likely to occur at low temperatures and in samples containing a large amount of thermoelastic martensite.





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