Textures and Microstructures, 1988, Vols. 8 & 9, pp. 415–426
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Printed in the United Kingdom

Texture and Microstructure of Meltspun Shape Memory Alloys

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(Received July 27, 1987; in final form August 24, 1987)

Dedicated to the memory of Professor Günter Wassermann

Shape memory effects are based on the shear stress sensitivity of the orientation of lattice variant and invariant shear which is associated with reversible martensitic phase transformation. The effect of texture and microstructure of the high temperature phase was investigated with copper based shape memory alloys. Rapid solidification can produce either a columnar grain structure with a pronounced ⟨100⟩ fibre texture, or an equiaxal structure with a grain orientation close to random. It could be shown that the columnar and textured grain structure produces the maximum shape memory effect in β-Cu alloys if the ribbon shaped specimen is exposed to tensile stress parallel to the length of the ribbons. These effects are explained by the influence of primary texture of the high temperature phase on the texture which originates in the martensitic low temperature phase if the transformation occurs under an external load.

KEY WORDS: Shape memory effect, Cu-based alloys, meltspinning, rapid quenching, pole figures, anistropy.

INTRODUCTION

The maximum strain which can be obtained in shape memory alloys either for one-way, two-way effects or pseudoelasticity is limited by

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the crystallography of lattice variant shear (Bain strain) of the particular crystallographic system (Wayman, 1964; Otsuka and Shimizu, 1986; Hornbogen and Wassermann, 1956; Wassermann, 1936). This shear may vary between more than 20 degrees for the b.c.c.→f.c.c. or f.c.c.→b.c.c. or f.c.c.→h.c.p. transformations and a few degrees for transformations which imply only slight tetragonal or rhombohedral distortions. Most materials are polycrystals, and this shear occurs inside of grains. The bulk shape change is affected by the orientations of the individual grains and, in addition, by the constraint which each grain experiences by grains in its environment.

Consequently, the maximum transformation shear can only be realized in single crystals with optimal orientation with respect to the external load, which transforms the high temperature phase into a martensitic single crystal. The behavior of polycrystalline materials will be modified by two parameters: The orientation distribution, i.e. the texture of the polycrystal and the microstructure, predominantly the grain size and shape.

The subject of the present investigation was to find a microstructure which leads to maximum bulk shear in Cu-based shape memory alloys. For this purpose experiments were conducted to create different textures and grain structures. Two methods are established for the production of semifinished products or parts of SM-alloys: Conventional casting, plastic deformation and heat treatments or powder metallurgical methods (Elst *et al.*, 1986; Duerig *et al.*, 1982). They both possess merits and disadvantages. In deformed alloys recrystallization may provide a chance to produce recrystallization textures and defined grain sizes. The powder metallurgical method will predominantly produce a textureless structure and a small grain size which is related to the initial powder size.

In our work, production of the bulk material is attempted by direct solidification from liquid. For this purpose the meltspinning method was applied (Stoobs and Wood, 1979; Eucken, 1987). It is known that solidification textures can originate by columnar growth of crystallization fronts. It is intended to vary the solidification conditions so that a wide range of microstructures can be obtained and to compare their properties as solidified with those which are known from materials produced by conventional methods.

MATERIALS AND EXPERIMENTAL METHODS

The chemical composition of the alloys used for this investigation is given in Table 1. All of these alloys were induction melted and subsequently meltspun on a copper wheel. The cooling conditions could be modified by choosing different circumferential wheel velocities which, in turn, produce ribbons of variable thickness (5 to 100 μm) and cooling conditions. The microstructure of these ribbons were investigated by light and scanning electron microscopy in order to determine structural homogeneity, grain size and grain shapes.

Subsequently, tensile tests were conducted in the temperature range in which shape memory effects occur in order to determine the extent of the shape changes associated with the tranformation. The tensile test specimens were produced by spark erosion cutting from the ribbon. The shape and width is given in Figure 1. The thickness of the specimen was the thickness of the ribbon. A special holder with soft iron grips was used for tensile testing. The strain was measured by an extensometer clipped between the specimen grips.

The X-ray measurements of textures were carried out on a fully automatic texture goniometer (Hirsch *et al.*, 1986) using Cu K_α radiation. {110} and {200} pole figures were measured from both sides of each ribbon: The bottom surface which was in contact with the cooling wheel and the top surface. (For small ribbons several pieces were put together in order to obtain a sufficiently large area

Table 1 Composition, phase and grain structure of the alloys and rapidly quenched specimen

Alloy number	Alloy composition (in wt %)	M_s K (°C)	Phase at ambient temperature	Grain structure of investigated specimens
1	Cu 24.35% Sn	213 (−60)	austenite	columnar
2	Cu 24.3% Sn	<293 (<20)	austenite	equiaxed
3	Cu 11.7% Al 3.5% Ni	>293 (>20)	martensite	columnar some equiaxed
4	Cu 12.8% Al 3.3% Ni	353 (≈80)	martensite	columnar some equiaxed

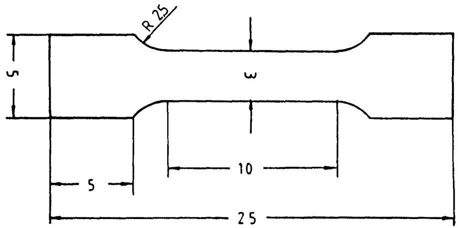

Figure 1 Shape of tensile test specimen (thickness: 5 to 150 μm = ribbon thickness).

of $14 \times 24\,\mathrm{mm}^2$.) Prior to the pole figure measurement the bragg angles θ_i were determined individually for each material by a $\theta/2$ scan. Only the reflection mode was applied with a maximum tilting angle $\alpha = 85°$. The corrections for background intensity and defocussing effects were taken from off-Bragg angle measurements (separately measured for each sample) and from the evaluation of the α dependent intensities of a reference sample (with random texture).

EXPERIMENTAL RESULTS

Light microscopy showed that principally two different types of homogeneous microstructure were obtained: equiaxed and columnar grains (Figure 2).

A special method to reveal the grain structure is mercury embrittlement and subsequent analysis of the fracture surface by scanning electron microscopy (Figure 3).

All alloys are obtained as homogeneous ordered b.c.c. phases, the structure of which has been described in earlier papers (Eucken and Hornbogen, 1985).

The textures of the two materials investigated in high temperature ordered bcc austenite phase are shown in Figures 4 and 5 in form of a) {110} and b) {200} pole figures, each measured from the upper side (top side: *t*) and lower side (bottom side: *b*, i.e. the one which was in contact with the copper wheel), respectively.

Figure 4 shows the polefigures typical for the columnar microstructures of alloy 1. For this ribbon (Figures 4a–d) a rather distinct texture is found with the character of a fibre texture, especially

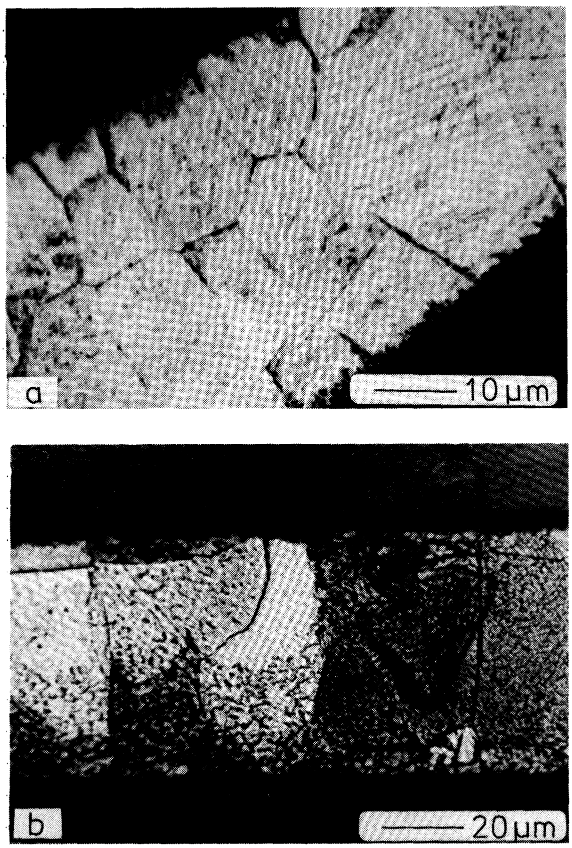

Figure 2 Light micrographs of cross sections: a) equiaxed grain structure, occurs at slower cooling rates, b) columnar grain structure, occurs at high cooling rates.

pronounced for the upper side ("$1-t$", Figure 4a) where an intensity maximum of 2.6 times random occurs in the {100} and 5.8 times random in the {200} (Figure 4c) pole figure. The high intensity of the latter is due to the fact that the fibre axis is a ⟨200⟩ axis, tilted ~12° from the sheet plane normal (centre of the pole figure) towards the negative reference direction RD, which indicates the rotation direction of the copper wheel. The texture of the lower side "$1-b$" (Figures 4b and d) is similar to "$1-t$", but also shows characteristic differences. It is less pronounced (2.1 and

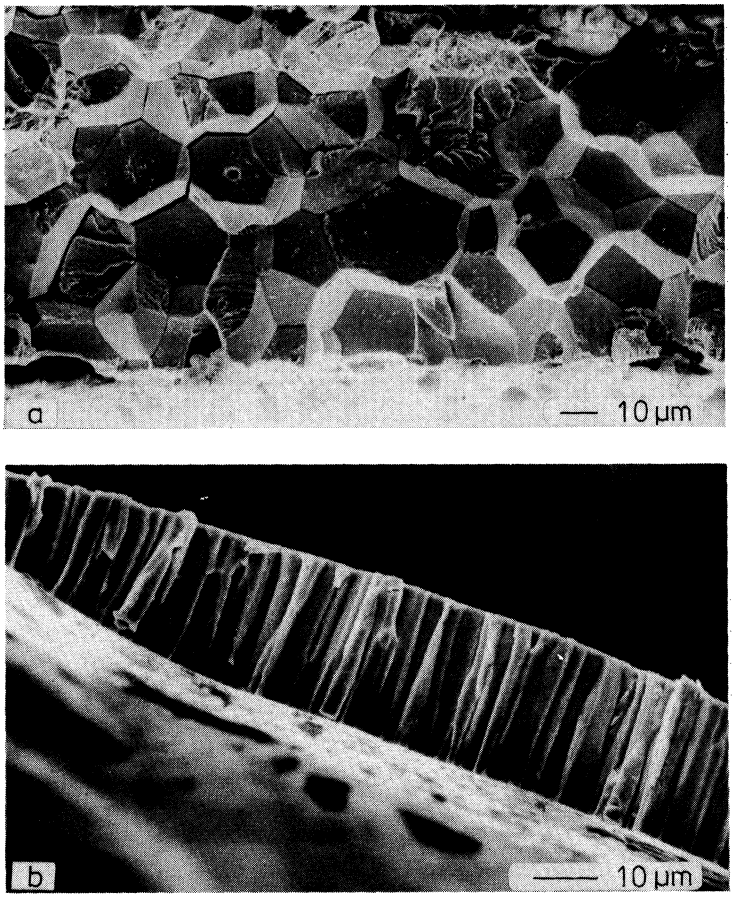

Figure 3 Mercury embrittlement makes the grain structure visible in SEM: a) equiaxed grains, b) columnar grain structure.

$4.9 \times$ R) and it appears to be more symmetric than $1-t$ showing no shift of the {200} fibre axis towards RD. (Both textures show a certain scattering along the equator line, to the left and to the right, i.e. by rotation around RD. This, however, should not be overestimated since it was rather difficult to adjust the small ribbon in an exact horizontal position to the measured surface, which might cause such an effect. (The adjustment parallel to RD was much better!)

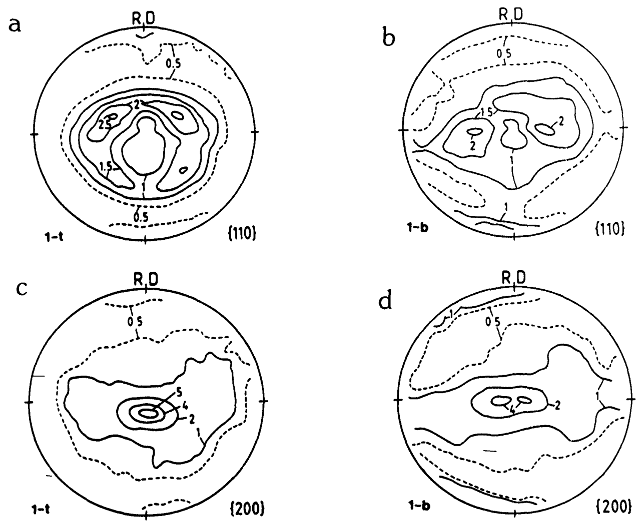

Figure 4 Polefigures of an alloy 1 ribbon with columnar grain structure: a) {110} pole figure of top surface, b) {110} of bottom (contact) surface, c) {200} of top surface, d) {200} of bottom surface.

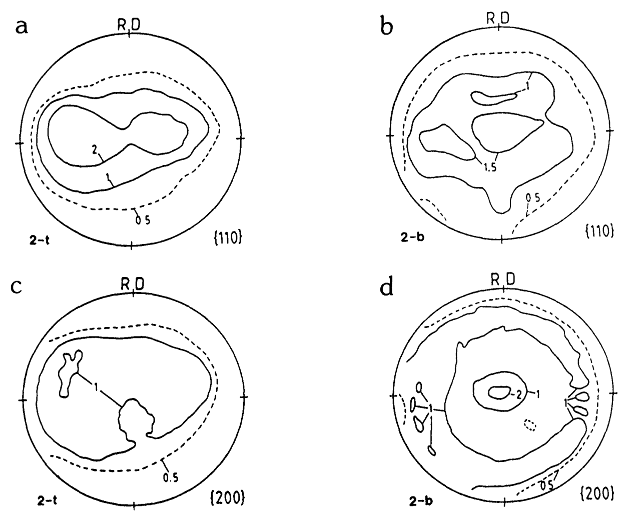

Figure 5 Polefigures of an alloy 2 ribbon with equiaxed grain structure, a) {110}, top surface, b) {110}, bottom surface, c) {200}, top surface, d) {200}, bottom surface.

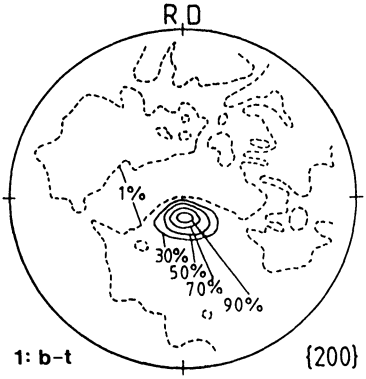

Figure 6 {200} difference pole figure of alloy 1 ribbon (columnar grain structure).

For a better estimation of the differences between sample "$1-t$" and "$1-b$" the difference between the two pole figures were calculated and plotted in relative (%) intensity of the maximum difference in Figure 6. It clearly shows that the main difference between the two sides is the strength and the position of the $\langle 200\rangle$ fibre texture. The tilted $\langle 200\rangle$ fibre axis dominates the {200} difference pole figure. Large areas in the difference pole figures are $\leq 1\%$ (dotted areas in Figure 6).

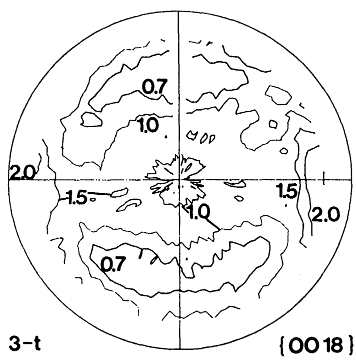

Figure 7 {0018} polefigure of alloy ribbon (with columnar austentite grains) after stress free martensite transformation.

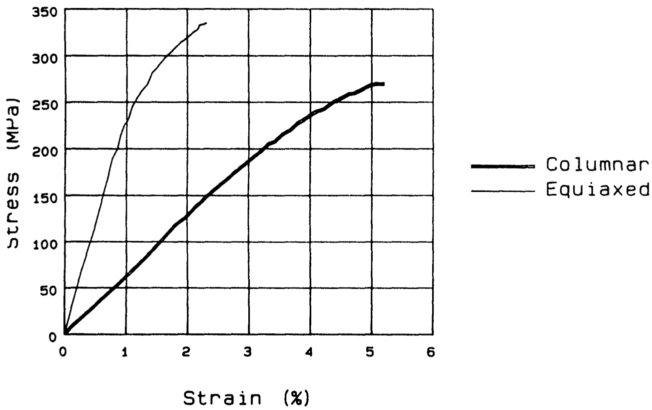

Figure 8 Typical results of tensile tests: Ribbons with fibre textured columnar grain structure exhibit a larger amount of recoverable strain at a lower stress level (a)) compared to texture free equiaxed grain ribbons (b)) in copper based shape memory alloys (alloy 4).

The textures of alloy 2 ribbon with an equiaxed grain structure (Figure 5a–d) are much weaker with maximum intensities of $1.6 \times R$ in both {110} pole figures and 2.2 or 2.8 in the {200} pole figure. Thus here the textures appear to be closer to random and no distinct axis occurs in the {200} pole figure.

Figure 7 shows a pole figure of the upper surface of the columnar microstructure after stress-free martensitic transformation. It is quite evident that the high degree of randomness has been introduced by various shear systems of the martensitic transformation.

Typical results of tensile tests are shown as Figure 8: the textured material deforms to a much higher degree as compared to the equiaxed random structure. In addition the stress to induce the transformation strain is higher for the random orientation.

DISCUSSION

An understanding of the two types of microstructure can be based on different types of nucleation of crystals. The columnar grains nucleate at the interface between liquid and the wheel and grow through to the upper surface. The sharpening of the texture can be explained by growth selectivity of the [100] orientation (Wassermann and Grewen, 1962). The deviation of the fibre axis at the upper surface can be understood by the fact that the wheel is moving while the crystal is growing. Equiaxed grains should originate most likely by individual probably heterogeneous nucleation of grains in the interior of the melt. Some authors imply a secondary recrystallization process which occurs at intermediate temperatures during cooling (Caesar and Köster, 1987).

The effect of texture on the bulk transformation induced strain is shown in Figure 8. The columnar grain structure grows about perpendicular to the length of the ribbon which is exposed to a tensile stress. In loading direction [100] and [110] orientations are dominating because of the texture. It is well known that martensitic shear occurs in a (110) $[1\bar{1}0]$ shear system. Cube texture or to a somewhat lesser degree cube fibre texture provides the optimum orientation for maximum shear stress in this particular shear system. In addition the columnar structure creates favourable conditions for minimum constraint of the interfaces. Therefore values for the bulk shape change can be obtained which approach the theoretical value (De Vos *et al.*, 1979) which would be obtainable only by single crystals of optimum orientation. On the other hand the equiaxed microstructure on average crystallizes with a less favourable orientation. A smaller shear stress is acting in the direction of the shear transformation tensor. A high constraint exists, as given by the fact that most of the grains are surrounded by other grains which impede their individual shape change.

The role of texture in shape memory alloys has been summarized in Table 2: the intial orientation distribution can be favourable or random with respect to phase transformation. There are 12 equivalent $\{110\}\langle 1\bar{1}0\rangle$ shear systems for the transformation into martensite. Consequently a further randomization will occur in either the original random or the textured material if the transformation takes place without external shear stress.

If the transformation occurs under stress final saturation conditions will be defined by the fact that only the one, the most favourable, shear system will be left to transform the material. In this case the material containing favourable texture will be trans-

Table 2 Role of texture in shape memory alloys (β: high temperature "austenite" phase; α_M: low temperature martensite phase; ε: recoverable strain = transformation strain; γ: shear angle; σ: external stress)

Austenite β grain structure	Poly crystal β random (equiaxed grain)	β textured (columnar grain)	Single crystal
transformation to martensite α_M without external stress: $\sigma = 0$	α_M random $\gamma = 0 \Rightarrow \varepsilon = 0$	α_M random $\gamma = 0 \Rightarrow \varepsilon = 0$	α_M random $\gamma = 0 \Rightarrow \varepsilon = 0$
transformation to martensite α_M with external stress: $\bar{\sigma} \geq \bar{\sigma}_{sat}$	α_M (?) $0 < \gamma_\beta < \gamma_{\beta\alpha}$	α_M textured $\gamma_\beta \Rightarrow \gamma_{\beta\alpha}$	α_M single crystal $\gamma = \gamma_{\beta\alpha}$ $\varepsilon = \max$
	$\Rightarrow \varepsilon \approx \max$		

forming into a pronounced new texture which can be directly derived from the crystallography of lattice variant shear. Each grain of the originally random material will also transform into martensite of unique orientation, which however will inherit the scatter of the original β grains. As a consequence in average these grains will also contribute less shape change by shear in direction of the external tensile load. The texture obtained by columnar growth has proved to be an orientation favourable to stress affected phase transformation. There is a possibility that less favourable orientations exist. In such a case the shape change could be less even than that which is obtained by the textureless random material.

In the case that all twelve $\{110\}\langle 110\rangle$ orientations occur in equal frequencies the bulk shape change should be 0. In saturation only the most favourable one will occur. The shape change approaches the theoretical limit given by the crystallographic condition. The SM-shear angle γ is $0 \leq \gamma \leq \gamma_{\alpha\beta}$.

Our work has shown that the actual bulk deformation $\varepsilon = 2\gamma$ can be affected by three factors:

1. the texture of the β phase,
2. the grain size of the material and
3. (as it is well known) the amount of external stress.

The optimum shape memory shear is represented by coarse grain material with optimum texture. It has to be exposed to the critical stress which provides the shear stress sufficient to complete selectivity of the shear systems.

Acknowledgement

The support of the German Science Foundation (DFG) is gratefully acknowledged.

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