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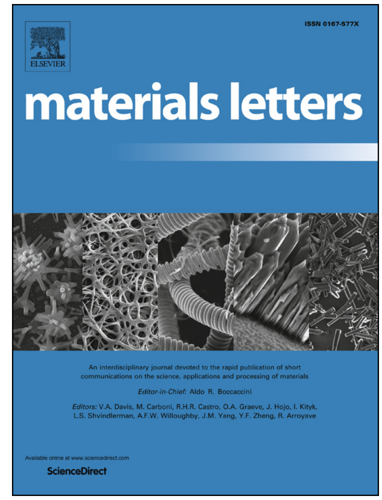

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Grain Ejection Associated with Thermocycling of Cu-Al-Ni Shape Memory Alloy

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Abstract

The phenomenon of metallic grain ejection is for the first time explained. An evaluation of ejected grains was performed in Cu-Al-Ni shape memory alloy load-free cycled up to 500 times within the temperature interval of reversible martensitic transformation (RMT). An intergranular fracture mechanism related to RMT is proposed to this phenomenon. By image processing, 8 grains/cm^2 after 500 cycles were measured. This is attributed to γ'_1 martensite transformation at the grain boundaries.

Keywords: Grain boundaries; shape memory materials; Cu-Al-Ni alloy; thermal cycling; martensitic transformation; grain ejection.

Introduction

Integrity is an important requirement for technological applications of shape memory alloys (SMAs). Baxevanis and Lagoudas [1] indicated that forthcoming commercial uses of SMAs in aerospace, automotive and energy conversion/storage systems are expected to drastically increase their fracture relevance. Actuators made of SMAs provide high energy density and can perform functions of several parts simultaneously [2,3]. These actuators might retain functionality despite the presence of a growing crack. Baxevanis and Lagoudas [1] also indicated that the fracture of SMAs under thermo-mechanical loading is, however, a complex phenomenon. It could involve reversible martensitic transformation (RMT). This reversible transformation was found to occur by means of the reaction in Cu-Al-Ni shape memory alloys [4,5]:

$$\beta_1 \leftrightarrow \beta'_1 + \gamma'_1 \tag{1}$$

Moreover, reorientation of martensite variant, transformation-induced plasticity and dislocation accumulation were also involved. Most reviewed experimental works on fracture of SMAs was related to NiTi alloys [6-11]. In the case of Cu-Al-Ni alloys, only works on single crystals [12-14], none on polycrystalline, were reviewed by Baxevanis and Lagoudas [1]. A possible reason is the fact that polycrystalline Cu-Al-Ni alloys are susceptible to intergranular fracture, which could impair their use in system subjected to thermo-mechanical or thermo-cycling conditions. Earlier works on Cu-Al-Ni intergranular fracture [15,16] postulated that RMT strain has a greater effect on grain boundary fracture than elastic strain. By contrast, Creuziger and Crone [17] presented results on Cu-13.95wt%Al-3.93wt%Ni implying that intergranular fracture is due to grain boundaries themselves and not an effect of the martensitic transformation. In a recent work [18], grain pullout from the metallographic surface of RMT thermal cycled

Cu-Al-Ni shape memory alloy, with composition similar to that of Creuziger and Crone [17], was for the first time surprisingly observed. This unexpected loss of pullout grains during cycling operation of a SMA is a practical problem related to its integrity, which deserves investigation. Even more recent investigations on polycrystalline Cu-Al-Ni shape memory alloys subjected to different heat treatments [19] and combined Cr addition with thermomechanical treatments [20] failed to report grain pullout in spite of intergranular fracture and microstructural embrittlement. Therefore, this phenomenon appears to be specifically related to a continuous cycling in the temperature interval of RMT.

In the present communication, surface grain pullout or ejection was quantitatively evaluated in the same previously investigated load-free thermocycled Cu-Al-Ni alloy [18]. A possible mechanism associated with martensitic transformation and intergranular fracture is proposed for this just recently discovered phenomenon.

Experimental

A polycrystalline Cu-13.7wt%-Al-4.0wt%Ni (Cu-13.7Al-4.0Ni) alloy was fabricated by plasma-melted followed by injection molding as described elsewhere [18]. A cast 27 x 25 x 5 mm alloy bar was heat-treated to stable ordered type DO_3 cubic β_1 at 850°C for 15 min and then water quenched to room temperature. Prismatic 5x5x5 mm specimens machined from the bar were subjected to load-free thermal cycling treatments (TCT) by heating up to 100°C, above austenite finishing (A_f) and cooling to -15°C, below martensite finishing (M_f). TCTs were conducted for 1, 100, 200, 300, 400 and 500 cycles. Phase identification during thermal cycling was performed by X-ray diffraction in a model XRD 7000 Shimadzu diffractometer operating with Cu-K_α

radiation in a 2θ interval from 25 to 100° and scanning steps of 0.03°. A sequence of reversible martensitic transformations $\beta_1 \leftrightarrow R \leftrightarrow \gamma_1' + \beta_1'$, in which the amount of phases is very sensitive to the number of cycles, was found and corroborated previous results [18].

Optical microscopy (OM) analysis was carried out in an Olympus microscope only before quenching. The alloy bar was polished with emery paper and diamond paste to a mirror condition. The microstructure was observed without etching with polarized light. During all load-free thermal cycling treatments, the initially prepared metallographic surface was untouched and martensite relieves were preserved from one cycle to another. Average grain size and number of ejected grains were evaluated by means of a Scion™ image analyzer. Scanning electron microscopy (SEM) observation was conducted in a model SSX-550 Shimadzu equipment operating with secondary electrons at 15 kV.

Results and Discussion

Figure 1 shows by OM a typical microstructure of the as-quenched alloy surface before thermal cycling treatment (TCT). In this figure, a massive amount of martensite needles is observed in every grain and its extension is limited by the grain boundary. X-ray analysis (insert in Fig. 1) revealed that most of the martensite in the as-quenched structure is associated with γ'_1.

Insert Figure 1

Figure 2 displays, for another surface region, the sequence of images illustrating the continuous detachment of a surface grain (inside highlighted squares) until its final ejection after 500 TCT. In this figure, one should notice the high amount of γ_1'

martensite needles being developed in the neighbor grains. At every cycle, martensite needles are transformed during cooling to -15°C and suffer reversion on heating to 100°C. Their effect on grain boundaries is cumulative, while surface relieves are not necessarily increasing, as seen from Fig. 2a to Fig. 2b

Insert Figure 2

With greater magnification, it is also possible to see in Fig. 3 evidence of grain boundary rupture at 400 TCT. This effective beginning of intergranular rupture is clearly associated with the impingement of martensite needles from surrounding grains, mainly at the left side.

Insert Figure 3

The SEM micrograph in Fig. 4 reveals that the grain boundary being detached is covered with fine ripple marks. Although slip lines associated with dislocations activity should not be discarded, it is most probable that these grain boundary marks in a load-free thermal cycling are due to γ_1' martensite. In this regard, the Sakamoto and Shimizu [16] suggestion that incompatibility of deformation during RMT appears to be the reason for intergranular fracture in our polycrystalline Cu-13.7Al-4.0Ni alloy. Indeed, the image in Fig. 4 supports the suggestion of Sakamoto and Shimizu [16] rather than the Creuziger and Crone [17] proposition that intergranular fracture is not affected by RMT.

Insert Figure 4

As for the quantitative evaluation, a first ejected grain was observed only after 400 TCT. It was then estimated that two grains might have been ejected per cm^2 of the metallographic surface up to that number of cycles. After 500 TCT, a more precise evaluation of 8 grains/cm^2 could then be measured based on image processing. Owing to the effective intergranular embrittlement of polycrystalline Cu-Al-Ni shape memory alloys [16-20], eventually every grain will be affected. In principle, grains with a relatively larger ratio of bulk volume to grain boundary area, proportionally, would be weaker and prone to be ejected.

Conclusions

Quantitative results in association with both OM and SEM observations confirmed that grain ejection in a shape memory Cu-13.7wt%-Al-4.0wt%Ni alloy subjected to 500 load-free thermal cycles is an unquestionable and measurable phenomenon. The grain ejection was a consequence of the well known intergranular fracture of this type of alloy. The participation of γ_1' martensite transformation contributes to a complete separation of boundaries, which promotes the grain ejection from the metallographic surface.

Acknowledgements

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Captions of Figures

Figure 1. Optical microscopy of as-quenched, 25°C, Cu-13.7Al-4.0Ni alloy before thermal cycling treatment with corresponding XRD.

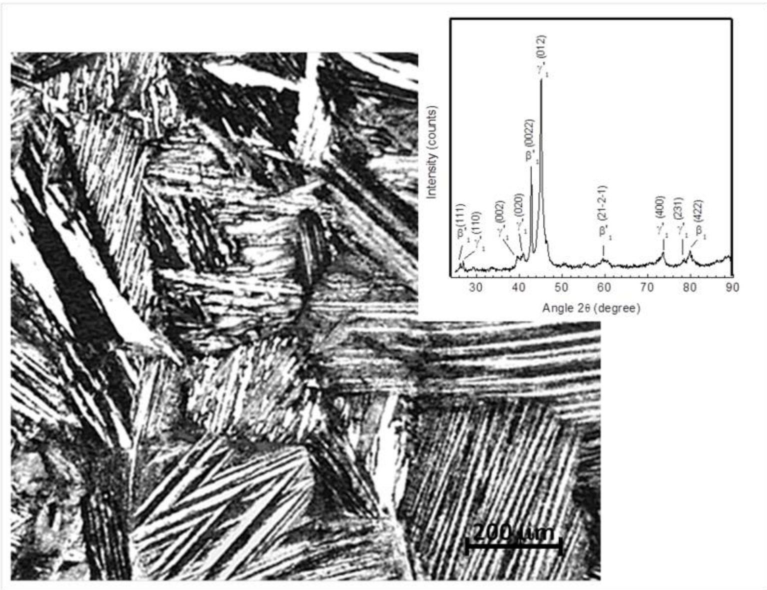


Figure 2. Optical microscopy of metallographic surface of load-free thermal cycled Cu-13.7Al-4.0Ni SMA after (a) 100 cycles, -15°C: grain inside highlighted square is being detached; (b) 300 cycles, -15°C: gain inside highlighted square is still detaching; and (c) 500 cycles, -15°C: empty space inside highlighted square left from ejected grain.

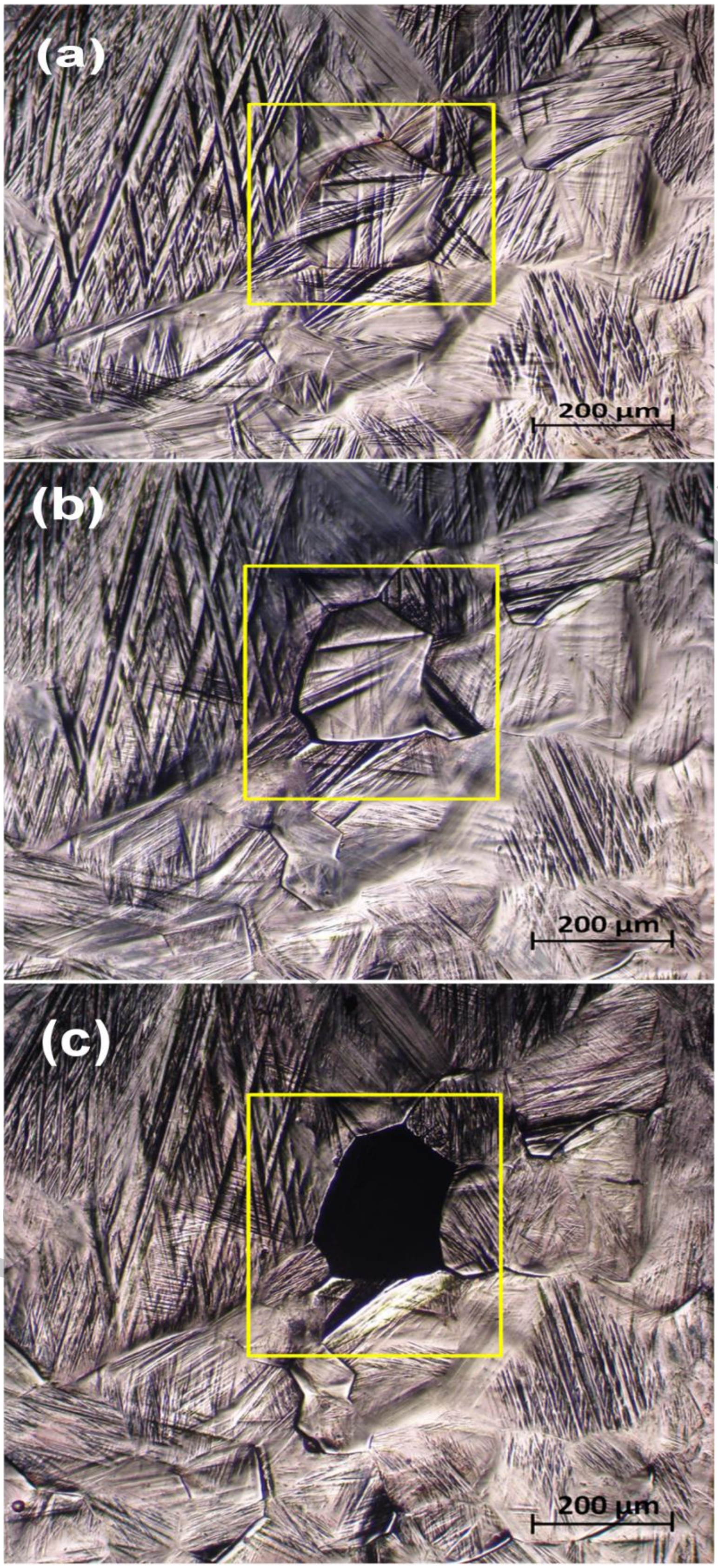

(a)
200 µm
(b)
200 µm
(c)
200 µm

Figure 3. Optical microscopy with higher magnification of the same grain followed in Fig. 2 after 400 cycles, -15°C.

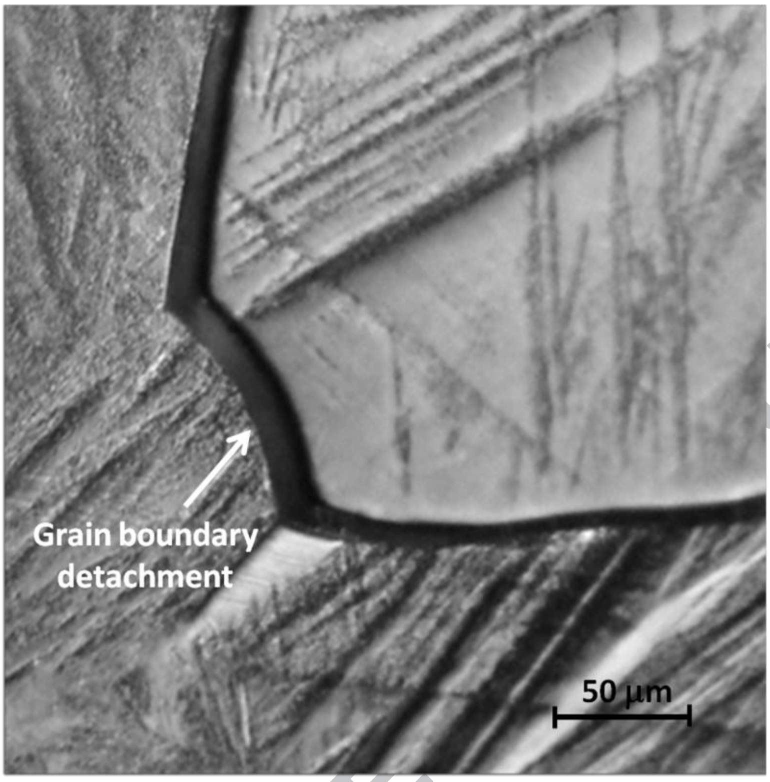
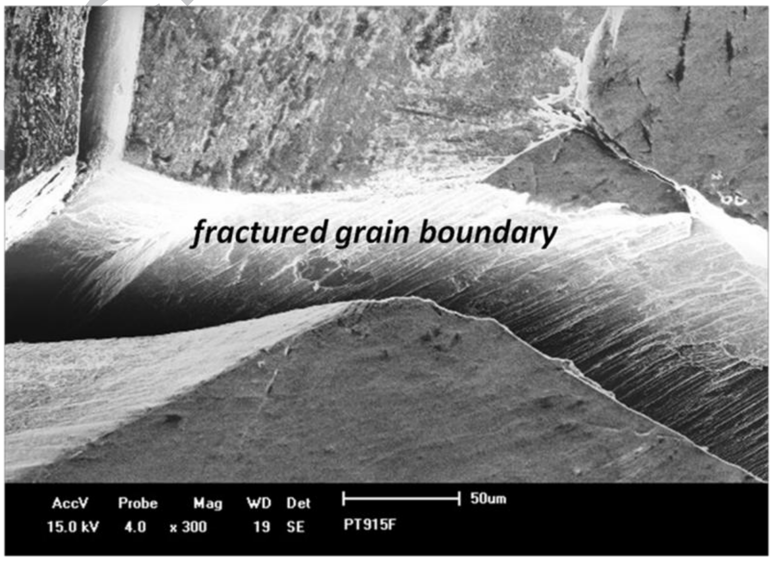


Figure 4. SEM of grain boundary intergranular fracture causing detachment before grain ejection.



HIGHLIGHTS

- A recently discovered phenomenon of grain ejection is for the first time explained.
- In CuAlNi alloy, the first ejected grain was observed after 400 load-free thermocycles.
- After 500 thermocycles, an average of 8 grain was ejected per square centimeter.
- Surface grain ejection was a consequence of intergranular fracture.
- Martensite transformation related to thermocycles causes grain boundary separation.