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THERMOMAGNETIC ANALYSIS AND MARTENSITIC PHASE TRASITIONS IN Cu-Mn-Al HEUSLER ALLOY

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Abstract. In the Cu-Mn-Al Heusler alloy ternary system the martensitic transformation may proceed in solid solutions having concentration nonuniformities related to small coherent particles forming during the degradation of the high temperature phase and having no spontaneous martensitic transformation on cooling. These particles may have a size well below the critical size of martensite nucleation. In the present work we study the temperature dependence of the structure and magnetic properties of Cu-Mn-Al in the temperature interval including the phase transition. Forward (cooling) and reverse (heating) structural transformations were studied directly by optical microscopy and illustrated by movies showing the motion and transformation pathways of multiple martensite interfaces. Simultaneous temperature measurements of the AC magnetic susceptibility gave a possibility to estimate the volume changes of high- and low-temperature phases during the transformation necessary for modelling the behaviour of this type of alloys.

1. INTRODUCTION

The development of new functional materials changing their shape and size under the action of external fields (electric, magnetic, thermal, *etc*.) is an important issue of current science and technology [1,2]. A special class of functional materials is metal alloys possessing a property of spontaneous shape recovery – shape memory effect. The magnitude of this effect may be as high as 30% [2]. The reversibility of such large non-elastic deformations is conditioned by the unusual structural rearrangement of the material – martensite reactions, elastic twinning, dislocation motion. If, in addition, the material is ordered ferromagnetically (e.g., ferromagnetic Heusler alloys [3]), new possibilities for the magnetic control of the shape and size of the samples are opened.

In the ternary system of Heusler alloys Cu_2MnAl it is possible to observe martensite transformation in solid solutions containing concentration nonuniformities [4-6], such as small inclusions precipitating during the decomposition of the high-temperature β_1-phase of Cu–Mn–Al, coherently coupled with the matrix without the spontaneous martensitic tansformation during cooling [4-6]. The size of these precipitates may be well below the critical size for martensite nucleation. The problem of the mechanism of martensite transformation in such type of alloys considered as a sequence of the formation of new phase nuclei, their growth and enlargement of orientational variants is of great interest both from academic and technological point of view [7,8]. Unfortunately, there is a lack of experimental information on this problem; in particular, little is known about the processes of structural realignment of Cu-Mn-Al alloys in the region of temperatures including phase transition points. In this view in the present work we performed a study of

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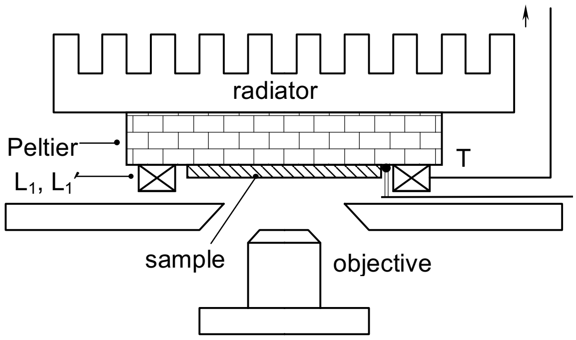

Fig. 1. Scheme of the AC initial susceptibility temperature dependence measurement simultaneously with microscopic observation.

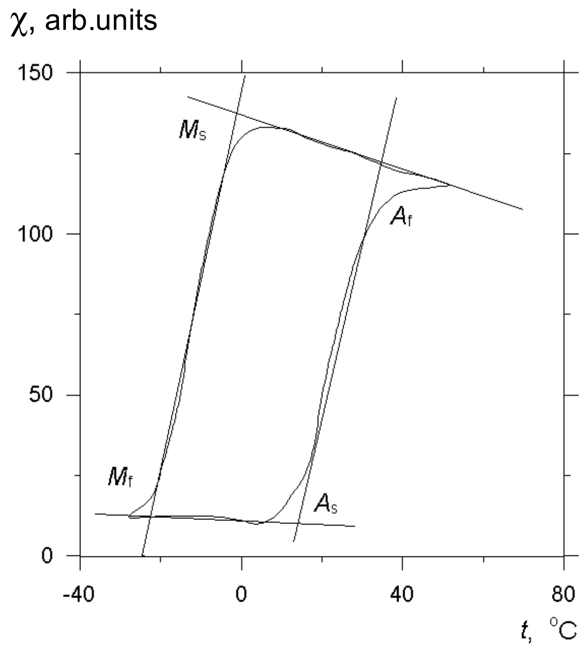

Fig. 2. Temperature dependence of the initial magnetic susceptibility for the Cu_2MnAl sample aged 6 hours at 500K.

the martensite transformation under changing temperature by direct optical microscopy observations supplemented by thermomagnetic analysis.

2. EXPERIMENTAL

Initial materials for the alloy preparation were copper (99.97%), electrolytic manganese (99.7%) and aluminium (99.99%). Melting was performed by induction heating in pure argon atmosphere in alumina crucibles. After homogenization and before aging the samples were water quenched from 1123K. Metallographic sections were prepared by mechanical fine grinding followed by electrolytic polishing. The microstructure was observed with the aid of metallographic optical microscope equipped by a cooling/heating stage working in the temperature range from +120 to –40 °C. Both heating and cooling was accomplished with a thermoelectric Peltier cooler (Electrosolutions, USA) by changing the current direction through the Peltier battery. Hermetic chamber with drying agent was used to prevent hoar-frosting of the sample at low temperatures.

It was shown by Kokorin *et al.* [4-6] that ferromagnetic particles coherently coupled with the metal matrix may be characterized by magnetic measurements. In this way it becomes possible to determine the start and finish temperatures of direct and reverse martensitic transformations by purely magnetic means. In the present work for this purpose we used the temperature measurement of the initial alternating current (AC) magnetic susceptibility. A differential scheme with two measuring coils and an exciting solenoid was built into the microscope cooling/heating stage so that it was possible to perform the magnetic measurements simultaneously with microscopic observations of the surface structure of the same sample (Fig. 1). The amplitude of the exciting AC field was of the order of 10 microtesla at a frequency of 1200 Hz. Lock-in amplifier connected via ADC to the computer with digital filtering was used to measure the amplitude and the phase of the signal from the sample. Calibrated platinum resistor (Honeywell HEL 775 series) was employed for the temperature measurement.

The microstructure was observed with the aid of differential polarized light microscope. In this method, two images of the same field of view are obtained for different (symmetrical) angular settings of the microscope analyzer resulting in an inversion of the optical contrast. Digital subtracting of the images results in the compensation of non-informative background so that the contrast of the resulting image may be greatly enhanced.

3. RESULTS AND DISCUSSION

Fig. 2 shows the temperature dependence of a Cu_2MnAl sample aged for 6 hours at 500K after

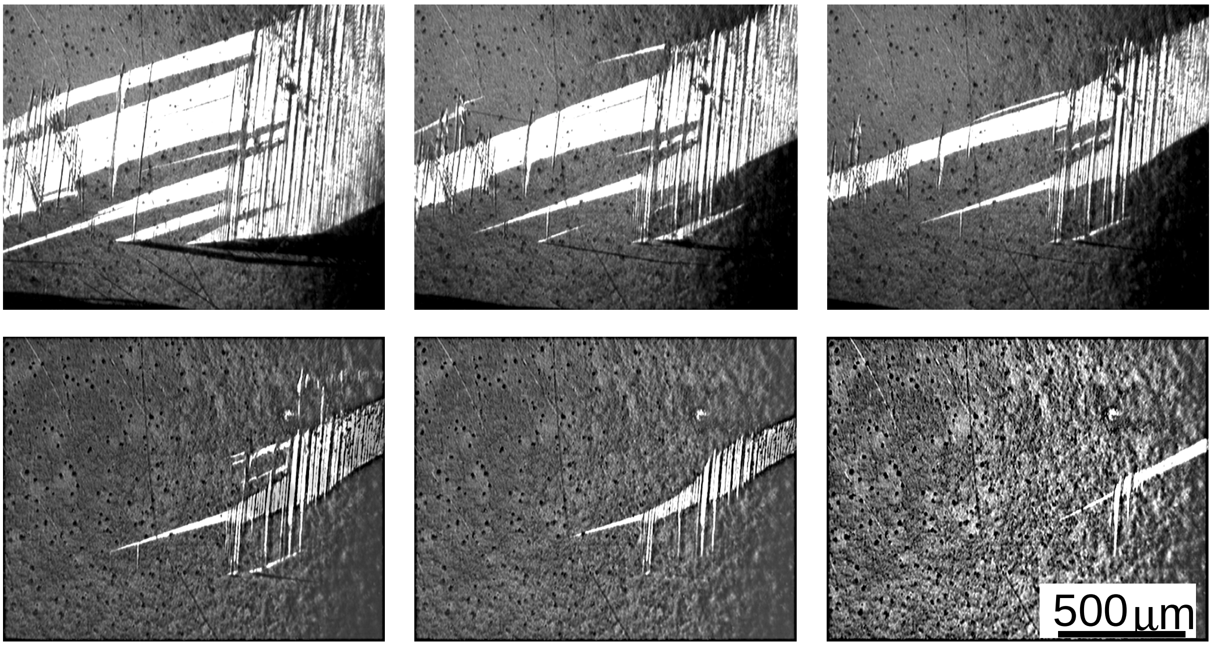

Fig. 3. Fragments of a movie demonstrating the decrease of the martensite fraction with heating of the sample from 23.5 °C (upper left frame) to 27 °C (lower right frame).

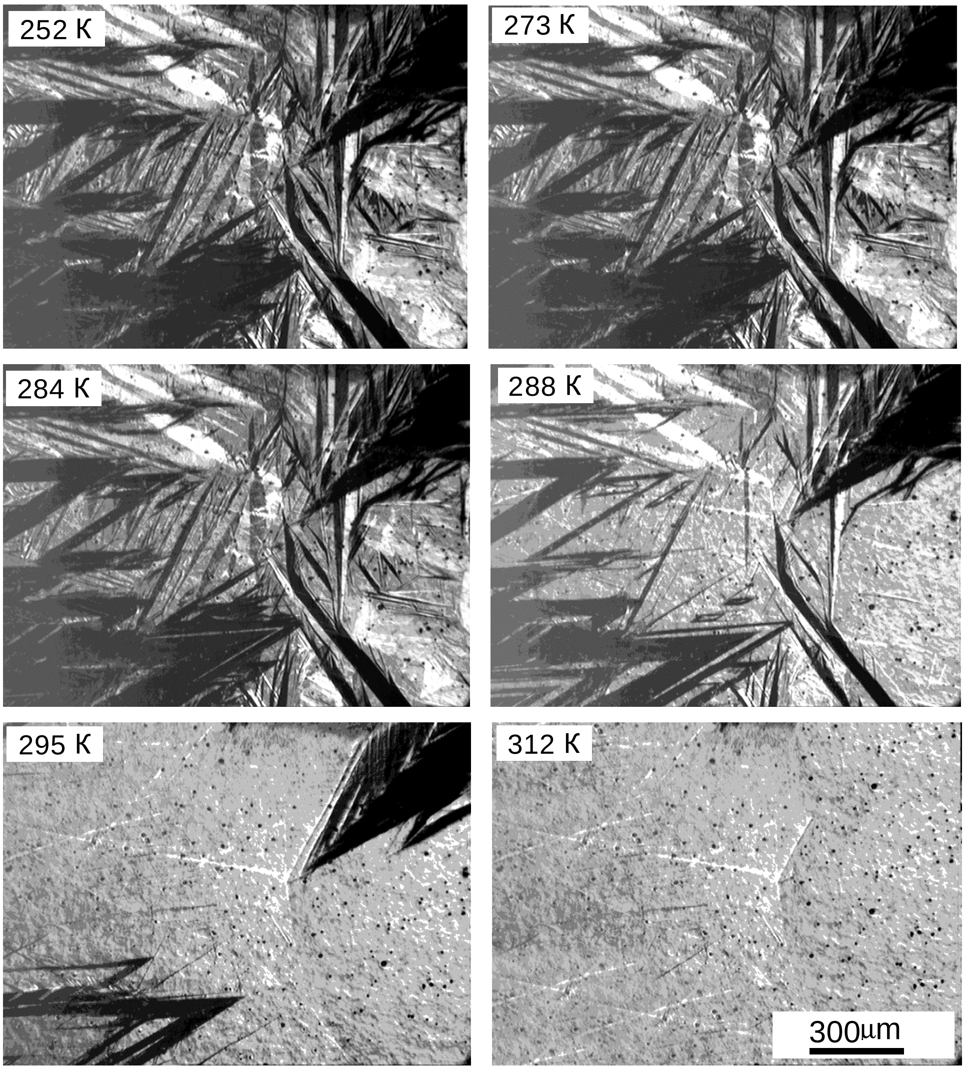

Fig. 4. Change of the microstructure of Cu–Mn–Al sample during heating from 252K (100% martensite phase) to 312K (100% austenite).

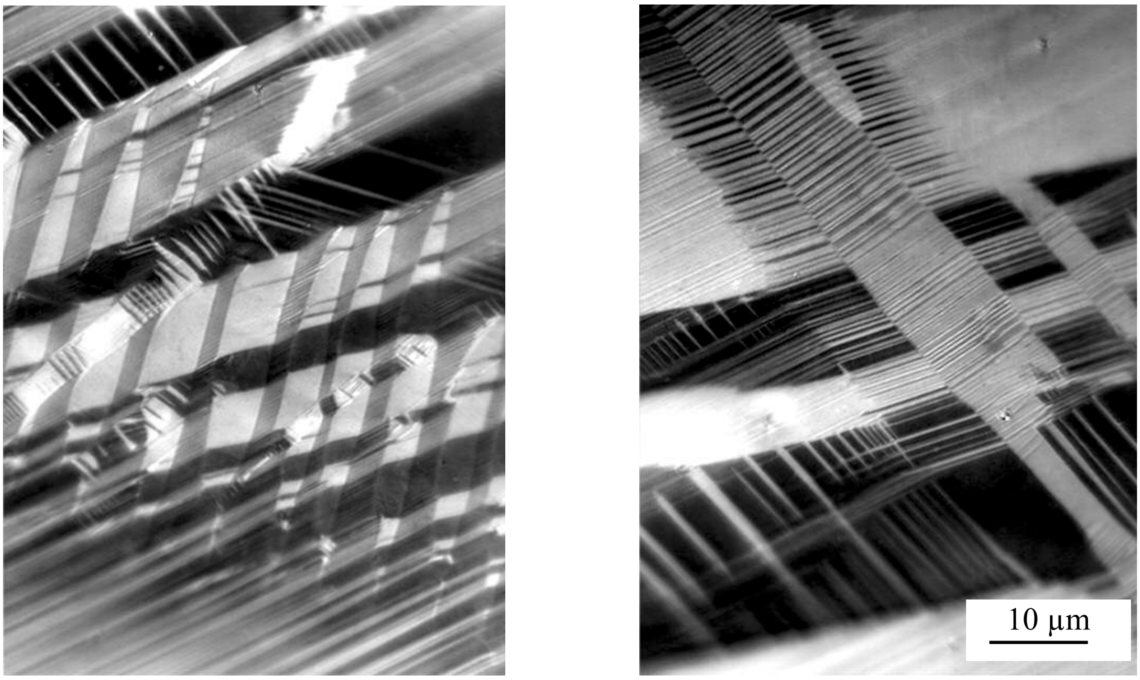

Fig. 5. Complex martensite structures with various types of inhibition, fragmentation, absorption, and intersection of twins.

quenching in water from 1123K. It is seen that magnetic susceptibility χ increases slightly with cooling from +80 to 0 °C, afterwards χ decreases sharply. Parallel observation of the microstructure shows that this region of sharp decrease of the susceptibility corresponds to spontaneous arising and avalanche-like development of the martensite twin structure. At a temperature of -20 °C the martensite structure occupies the whole volume of the sample and the process of phase transformation is finished. On continuing of the thermal cycle (heating from -20 to +80 °C) the reverse process of transition to the high-temperature austenite phase is finished at about +40 °C. For this state polarized light observation shows only the coarse austenite grain structure of the crystal having no noticeable internal fine structure.

Observations of the structure show that there are some specific features of the metallographic sections of Cu-Mn-Al samples under study. Flat polished surfaces prepared on the samples being in the austenite state (at the temperatures above the martensite – austenite (M-A) transformation) acquire a microscopic relief corresponding to the martensite twin structure arising after the A-M transformation. Repeated heating results in a restoration of the flat surface of the austenite phase. If the polishing is performed in the martensite state the relief appears in the austenite state. However, in this case the flat surface is not restored on cooling. The mechanism of this unusual behaviour is not clearly understood.

Fig. 3 shows the fragments of a movie illustrating the changes of the microstructure with heating from 23.5 to 27 °C. Estimation of the relative volume of the martensite phase from the obtained images gives a value of 30% for 23.5 °C. This figure is in good accordance with the estimate obtained from the temperature dependence of initial susceptibility shown in Fig. 2 based on a reasonable assumption of linear relation between the relative normalized value of χ and the martensite fraction.

The microstructures in Fig. 4 illustrate the structural phase transition from the state of 100% martensite at 252K to the state of 100% austenite at 312K. Comparison of the thermomagnetic analysis data (Fig. 2) with the microstructures presented in Figs. 3 and 4 gives an evidence of good correspondence between the estimates of the martensite and austenite start and finish temperatures and the relative volumes of these phases.

It is worthwhile to mention the wide variety of martensite microstructures forming during the reactions of accomodation due to the tendency of minimizing the elastic stresses [7–9]. Fig. 5 shows some complex martensite structures corresponding to different types of intersections and twin full and partial secondary interlayers, cross-shaped intersections, interlayer inhibition, interlayer absorption, fragmentation, and polysynthetic structures of submicron width (Fig. 5).

4. CONCLUSIONS

The processes of formation and realignment of the low-temperature phase of aged Cu-Mn-Al alloys were observed directly by the differential polarization optical microscopy during the structural austenite – martensite phase transition. Direct estimates of the relative volume of the phases by the results of optical microscopy observation are consistent with those inferred from the temperature dependence of the initial magnetic susceptibility. The variety of martensite structures of Cu-Mn-Al forming during the phase transitions are similar to the polysynthetic structures observed in other shape memory materials, so they may be adequately described by the previously developed computer simulation methods [7,8].

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