STUDIES OF THE Cu–Mn SYSTEM

E.-M. Nicht, G. Brauer

Positron Group of TU Dresden at Research Centre Rossendorf Inc.
P.O.Box 510119, 01314 Dresden, Germany

Zs. Kajcsos and B. Molnar

Central Research Institute for Physics, Hungarian Academy of Sciences
P.O.Box 49, 1525 Budapest, Hungary

The superstructures Cu_3Mn and Cu_5Mn have been characterized for the first time by X-ray and differential scanning calorimetry measurements. In addition, positron lifetime measurements are presented in order to show the defect structure of these alloys. It turned out that the annealing behaviour is equal for the two different alloys and the two different deforming processes. An annealing temperature of 300°C is not sufficient to obtain a defect free state.

PACS numbers: 78.70.Bj

1. Introduction

The changes in mechanical properties of reactor pressure vessel (RPV) steels during their irradiation in a nuclear power plant are known as neutron embrittlement. The observed changes are thought to be correlated with the formation of precipitates. In RPV steels type 15kh2MFA and 15kh2NMFA, respectively, carbides are assumed to be such precipitates [1], whereas in RPV steels type A533B the formation of CuMn precipitates is suggested. Based on calculations, given in Refs. [2, 3], the precipitates are thought to be Cu_3Mn and Cu_5Mn, respectively.

Several investigations have already been done to enlighten the properties of the system Cu–Mn [4–6]. In the Cu–Mn phase diagram the existence of two regions with well-developed short-range orders is assumed [7] (Fig. 1). These Cu–γ–Mn short-range-order regions are interpreted as the superstructures Cu_3Mn and Cu_5Mn, respectively.

The goal of our studies is to enlighten the properties of Cu_3Mn and Cu_5Mn and to solve questions concerning defect structure, ordering effects and thermal stability of these alloys. By means of X-ray investigations the lattice parameters are to be determined. Differential scanning calorimetry (DSC) measurements are used to show ordering effects. Finally, the defect structure and annealing behaviour of these alloys are firstly enlightened by positron lifetime measurements and first results shall be presented.
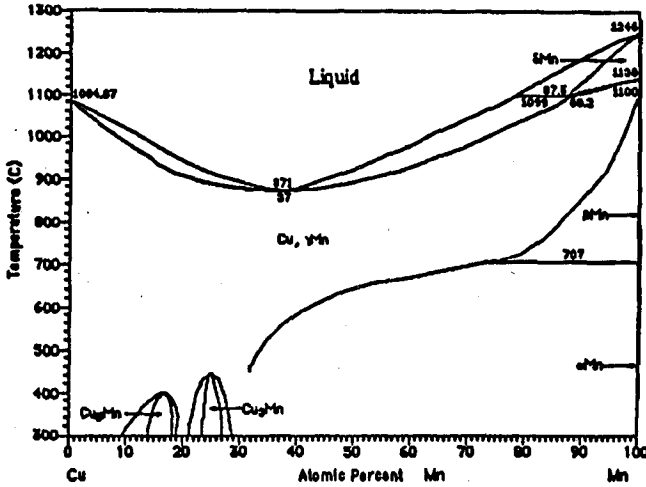
Fig. 1.   Cu–Mn phase diagram.

2. Experimental

To study the above-mentioned system, ingots of Cu–16at% Mn and Cu–25at% Mn were alloyed. These ingots were deformed by hammering and rolling, respectively. The different ways of deformation were chosen to investigate whether different deformation processes cause different defects and therefore different results of the lifetime measurements. The samples were prepared by separating of 1 mm thick discs from the hammered rods and by manufacturing of 8 × 8 mm pieces from the rolled sheets (thickness 1 mm).

To reach the equilibrium and defect free state according to Ref. [4], a set of samples was annealed at 580°C/24 h and aged at 300°C under vacuum for a long time (4 d and 60 d, respectively). To study the changes of the defect structure in more detail, isochronal annealing (20 min) up to 620°C has been performed using another set of samples.

X-ray measurements were used to determine the lattice parameters in the cast, the deformed as well as the aged state. DSC measurements should demonstrate the formation of the superstructures Cu_3Mn and Cu_5Mn by pointing out small transformation heats resulting from solid state reactions. In the transition range heat is released and a peak is indicated. Our measurements were carried out with a device from NETZSCH. The heating rate amounts to 2 K/min.

The positron lifetime measurements were performed at the conventional device at MLU Halle with a time resolution of 278 ps (FWHM). The source supporting foils consisted of Al with a thickness of 2 μm. The source correction was calculated to 11.9% using a formula given in the literature [8]. The source correction terms were estimated to have a lifetime (intensity) of 165 ps (88%) and 465 ps (12%) and are due to annihilations in the source supporting Al-foil and at the ^{22}NaCl/Al interface and the activity itself, respectively. All measurements were

carried out at room temperature (20°C). The evaluations of the time spectra were performed by an updated version of the well-known POSITRONFIT EXTENDED program package [9].

3. Results and discussion

The results of the X-ray measurements show that the treatment did not influence the values of the lattice parameters, but the chemical composition of the studied alloys. The measured lattice parameters are shown in Table.

TABLE

Lattice parameters of CuMn (radiation Cu).

	Cu_3Mn	Cu_5Mn
$a = b$ (A)	2.61	2.60
c (A)	3.69	3.67

The interpretation of the DSC measurements show that the transition from the ordered γ-range to the CuMn-phase takes place at nearly 450°C for Cu_3Mn and about 400°C for Cu_5Mn.
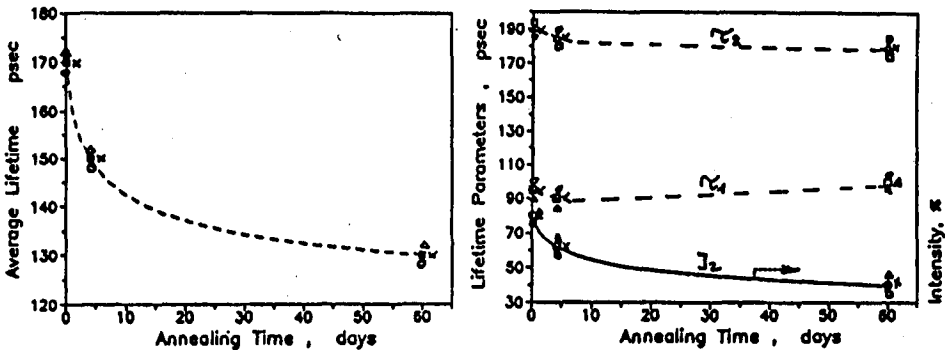

Fig. 2. Average positron lifetime $\bar{\tau}$ and lifetime parameters τ_1, τ_2 and I_2 as a function of annealing time (annealing temperature 300°C), in Cu_3Mn — $\triangle$ cold rolled, o cold hammered, in Cu_5Mn — $\square$ cold rolled, $\times$ cold hammered.

In Fig. 2 the development of the average positron lifetime τ and the lifetime parameters τ_1, τ_2 and I_2 of all Cu–Mn samples are presented as a function of the annealing time at 300°C.

The possible decomposition of the measured lifetime spectra into two components points out that positrons can either be trapped by lattice defects (τ_2, I_2) or annihilate as free positrons (τ_1, I_1). Using the one-defect trapping model the bulk lifetime can be calculated as

$$\lambda_b = 1/\tau_b = I_1/\tau_1 + I_2/\tau_2. \tag{1}$$

Above, $\tau_2 = \lambda_2^{-1}$ represents the trapped positron lifetime in a monovacancy. It is supposed that vacancies and/or dislocations could be trapping centers depending on the annealing time.

It can be seen that the annealing behaviour is equal for both alloys and both deforming processes. In Ref. [4] an annealing temperature of 160°C has been assumed to be sufficient for obtaining an ordered and defect free state. However, even after our final annealing (300°C/60 d) it is obvious (see Fig. 2) from the presence of a long-living component τ_2 with an intensity I_2 of about 40% that no defect free state of the alloys has been reached. The average positron lifetime $\bar{\tau}$ for all samples in the final state amounts to 130 ps.

The lines connecting measuring points in Figs. 2–4 presented in this work have been drawn as a guide for the eyes only. Typical errors are given in Fig. 3 and Fig. 4 for some points only.

To study the changes of the defect structure in more detail, isochronal annealing (20 min) up to 620°C has been performed. The results for the average positron lifetime $\bar{\tau}$ are presented in Fig. 3.
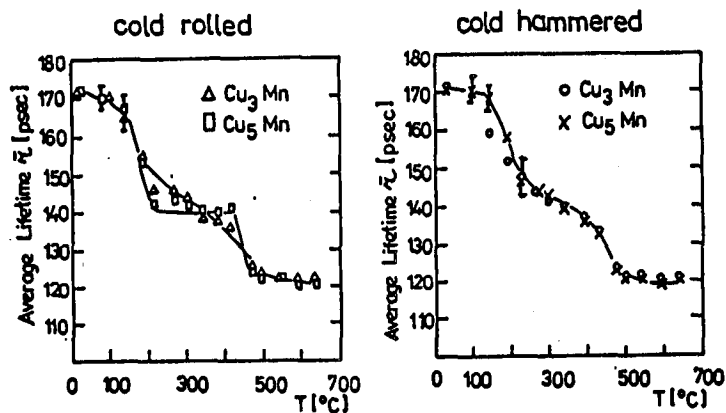

Fig. 3.   Average positron lifetime $\bar{\tau}$ as a function of isochronal annealing temperature.

The development of $\bar{\tau}$ due to isochronal annealing exhibits the existence of two annealing steps. The first one at about 150°C should be connected with vacancies, whereas the second one might be due to dislocations. For all samples finally $\bar{\tau}$ reached a value of 120 ps. This value is close to the average positron lifetime for Cu known from the literature ($\bar{\tau}_{Cu} = 122$ ps [10]). Because the studied compositions Cu_3Mn and Cu_5Mn belong to the Cu-rich part of the Cu–Mn system this agreement seems to be reasonable. Saturation trapping in vacancies is reported to increase the positron lifetime in Cu by about 60 ps [10], i.e. our interpretation of the first annealing step (Fig. 3) as being due to vacancies seems to be supported too.

A decomposition of the measured positron lifetime spectra due to isochronal annealing up to 620°C is presented in Fig. 4.

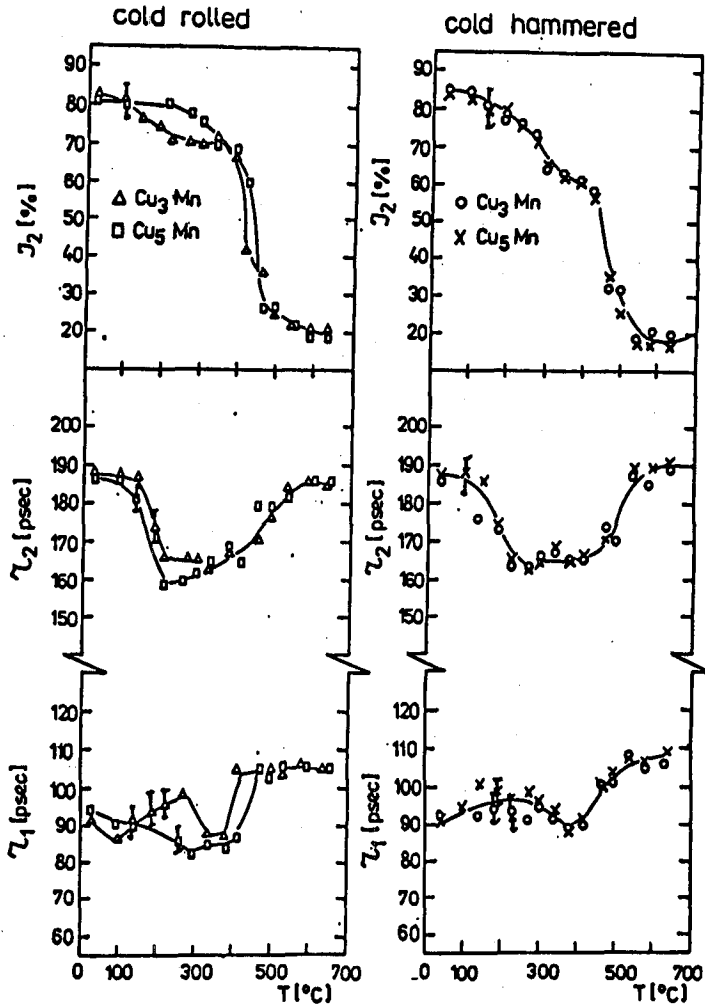Generally, a decrease in the intensity I_2 from about 80% at the beginning

Fig. 4. Positron lifetime parameters τ_1, τ_2 and I_2 as a function of isochronal annealing temperature.

of the annealing procedure to about 20% in the final state indicates the disappearence of lattice defects, which is not complete even due to the applied heat treatment. The interpretation of the changes in the lifetime component τ_2 is not so straightforward. Their decrease to a value of about 160 ps seems to really indicate the annealing of vacancies up to about 200°C. The again observed increase in τ_2 at higher temperatures might be connected with the formation of Mn-precipitations due to the transition from the ordered γ-CuMn region to the CuMn-phase. However, this guess needs further clarification. It is also planned to use Kossel-technique to enlighten the role of dislocations, which has been assumed from the changes of the mean lifetime $\bar{\tau}$ (Fig. 3).

4. Conclusions

Structural information about the lattice parameters of the alloys has been revealed from X-ray measurements.

The results from positron lifetime measurements do not support the conclusions from the literature that an annealing temperature of 160°C is sufficient to obtain the defect free state of these alloys.

Further work is necessary in order to clarify the annealing behaviour and defect characteristics of the above-mentioned alloys.

Acknowledgment

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