

STRUCTURAL PHASE TRANSFORMATIONS IN ALLOYS OF THE Cu–Pt SYSTEM DURING ORDERING

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Using the results of a Monte Carlo investigation, it is established that there are complicated structural phase transformations in the Cu–Pt system during cooling, which combine the order–disorder transitions and structural transformations. In the course of ordering, the high-temperature disordered fcc-phase can also undergo structural transformation. In the ordered state, a wide range of structures can be formed, depending on the alloy composition. In a few cases, there are closely spaced horizontal sections in the curve of the temperature dependence (in a certain temperature range) of the configurational energy. They correspond to different structural phase states of the system, with the energy difference being negligibly small. This is an indication of poor stability of these states to the structural phase transformations. In an approximation of thermodynamic probability of the occurrence of these states, in the case of finite temperatures we can expect them to co-exist in one and the same system or mutually transform under the action of a minimal thermal force.

Keywords: low-stability states, structural phase transformations, role of structure defects.

INTRODUCTION

The Cu–Pt system is the only system [1] in which alloys form a number of crystallographic structures during their order–disorder transition from a regular fcc-solution. The CuPt₃ alloy in the ordered state has an $L1_2$ superstructure based on an fcc-lattice. The equiatomic CuPt alloy during order–disorder phase transition is transformed from an fcc-into an orthorhombic lattice (a state with an $L1_1$ superstructure, where the angles α , β , and γ are other than $\pi/2$). The Cu₃Pt alloy in the course of ordering is transformed from a regular solid fcc-solution into an ordered state with the $L1_2$ superstructure. Earlier the authors of [2] reported a possible occurrence of an ordered phase with 20 at.% Pt, which corresponds to a Cu₄Pt alloy (the superstructure type was not then determined). Moreover, the literature contains predictions of an ordered Cu₇Pt compound with the concentration ~12.5 at.% Pt [3]. All these facts demonstrate that the Cu–Pt alloys occur in the states with a large number of superstructures in a wide range of concentrations of the binary alloy components. Of special mention are the variety of structural phase transformations accompanying the order–disorder transitions, such as fcc – fcc and fct – orthorhombic structure transformations. This process involves a wide range of such superstructures as $L1_2$, $L1_1$, $L1_3$, and $L1_0$. A combination of the order–disorder phase transition with multiple structural phase transformations offers new possibilities of controlling the structure and mechanical properties of metal alloys.

The purpose of this investigation is to study the features of structural phase transformations during their combined occurrence with the order – disorder phase transition.

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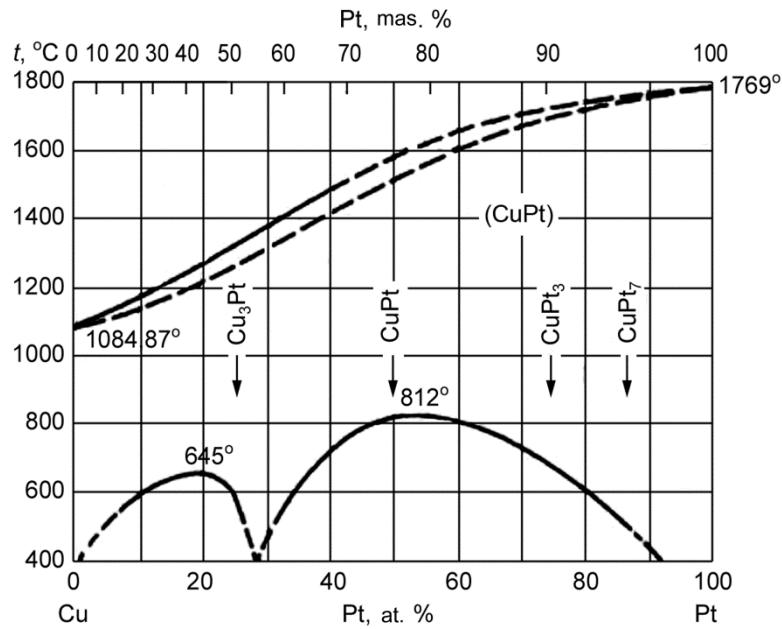


Fig. 1. State diagram of the Cu–Pt system.

THE MODEL AND EXPERIMENTAL PROCEDURE

When being crystallized, the Cu–Pt system forms a continuous series of solid solutions (Fig.1) [1]. In a solid state at low temperatures, there is a number of ordered structures in a wide range of concentrations. The highest degree of ordering is achieved in the alloys at the concentrations 22.5, 50.0, 72.5, and 86.0 at.% Pt, these corresponding to the following compounds: Cu₃Pt, CuPt, CuPt₃, and CuPt₇, respectively [1]. The Cu–Pt system is the only binary compound that in the case of an equiatomic composition contains an ordered CuPt phase with the L1₁ superstructure (Fig. 2a). The Cu₃Pt alloy in an ordered state has the L1₂ superstructure (Fig. 2b). An L1₃ superstructure was observed in the CuPt₃ alloy (Fig. 2c), which is also present in the Cu–Pt system [2]. The superstructural ordering is exhibited by the CuPt₇ (Fig. 2d) and Cu₃Pt₅ (Fig. 2e) alloys [3].

Let us take the above superstructures as model subjects of investigation. In what follows we are going to use three-dimensional alloy models of the Cu–Pt system with varying component concentrations.

The investigation will be performed by the Monte Carlo method using computer simulation [4], similarly to those conducted earlier [5–14].

The initial configuration is a disordered fcc-solution, the model crystal dimensions being 24×24×24 atomic layers (55296 atoms). The boundary conditions used on the edges of the model grid are periodic, which effectively corresponds to an infinite system of coordinates. In order to activate the diffusion process, let us randomly introduce one vacancy, which would correspond to the concentration of vacancies $\sim 1.81 \cdot 10^{-5}$. Assume the atomic diffusion to follow the vacancy mechanism, much the same as the mechanism used earlier [7–9]. The alloy state would change at discrete points of time; one iteration would refer to a single event of self-diffusion corresponding to an electron hopping into a vacant node. For every iteration we would calculate the probability of the nearest or the next nearest atom *i* to the vacant node to hop to the vacant site. The hopping probability for an atom into the vacant site we would determine from

$$\text{the expression } p_i = A^{-1} e^{-\frac{(E_i^i - E_F^i)}{kT}}, \text{ where } E_i^i \text{ is the atomic energy in its initial position, } E_F^i \text{ is its final energy after}$$

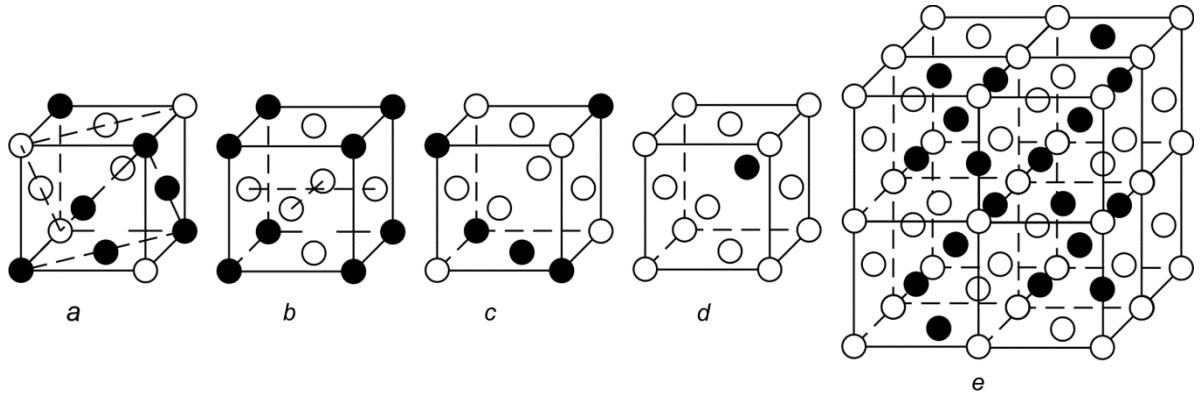


Fig. 2. Unit cell for the superstructures: $L1_1(a)$, $L1_2(b)$, $L1_3(c)$, $CuPt_7(d)$, $Cu_3Pt_5(e)$.

hopping to the vacant site, $E_{\max} = \max_i (E_I^i - E_F^i)$, k is the Boltzmann constant, T is the temperature, A is the normalizing factor, and $1 \leq i \leq M$, M is the number of neighbors on two coordination spheres.

The configurational energy of the crystal is calculated using the following formula: $E = \frac{1}{2} \sum_{i=1}^N \sum_{j=1}^M \phi(r_i - r_j)$,

where $r_i - r_j$ is the spacing between atoms i and j , N is the number of atoms in the crystal, M is the number of neighbors in the two coordination spheres, and $\phi(r_i - r_j)$ is the pair potential. The atomic interaction [5, 6] is prescribed using the following pair interaction potentials:

$$\begin{aligned} \phi(r_i - r_j) = & P_i P_j \times V_{AA}(r_i - r_j) + (1 - P_i)(1 - P_j) \times V_{BB}(r_i - r_j) \\ & + [P_i(1 - P_j) + (1 - P_i)P_j] \times V_{AB}(r_i - r_j). \end{aligned}$$

In this expression, P_i and P_j are the probabilities for atom A to appear in nodes i and j , respectively, $V_{AA}(r_i - r_j)$ is the pair interaction potential, e.g., for a pair of atoms A at a spacing $R = |r_i - r_j|$. Let us restrict ourselves to considering the interaction of the nearest and next nearest neighbors, and in order to approximate the central pair interaction potential, $V_{AA}(R)$, let us use the Morse potential function given by

$$V_{AA}(R) = D_{AA} \left\{ \exp[-2\alpha_{AA}(R - R_{AA}^0)] - 2 \exp[-\alpha_{AA}(R - R_{AA}^0)] \right\},$$

where D_{AA} characterizes the energy of dissociation of a pair of atoms, A , α_{AA} is the bond ‘rigidity’, R is the interatomic spacing, and R_{AA}^0 is the equilibrium value of R for the $A-A$ pair.

RESULTS AND DISCUSSION

It should be noted that the initial state of the system of every model alloy under consideration is disordered. The parameters of the atomic interaction potentials were taken from [16, 17]. For this initial state we calculated the mean configurational energy and the lattice parameters as a function of copper atom concentration in the Cu–Pt system (Fig. 3). The concentration of Cu in the alloy was varied from 0 to 100% with a step of 5%. The configurational energy value was determined after $5 \cdot 10^6$ iterations at $T = 1900$ K. In so doing we observed a relationship between the component concentration in the alloy and the energy value. The lowest value of the energy corresponds to the alloy with

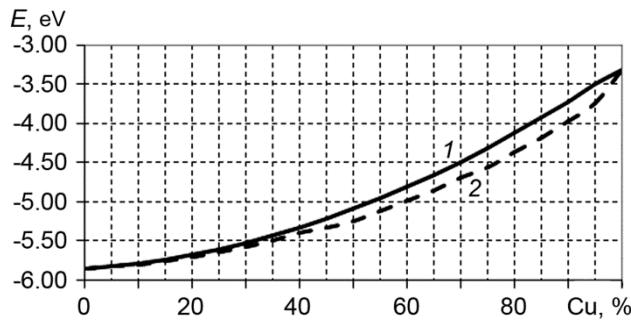


Fig. 3. Dependence of the average configurational energy of the Cu–Pt alloys on the component concentration in disordered state at $T = 1900$ (curve 1) and 200 K (curve 2).

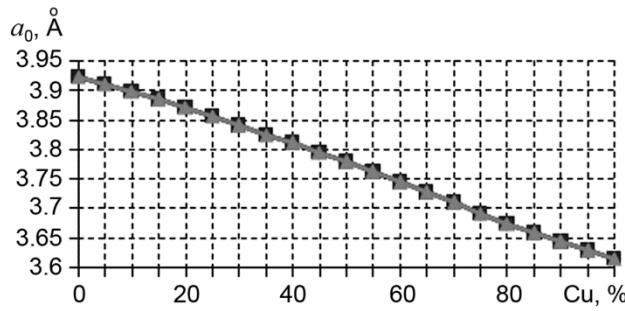


Fig. 4. Dependence of the lattice parameters of the Cu–Pt alloys on the component concentration in a disordered state.

the platinum atom concentration 100%, and the highest energy corresponds to the alloy with the copper atom concentration 100%. These data are quite consistent with the results reported in [17–25]. At $T = 200$ K, the energy values within the entire range of concentrations (with the exception of 100% Cu and 100% Pt) are lower than those at $T = 1900$ K. This implies that in a wide range of concentrations, more favorable states than a disordered state can form in the alloys under study.

It is evident from Fig. 4 that all three lattice parameters are equal and their dependence on concentration is nearly linear. The lower the concentration of Cu atoms, the higher is the lattice parameter value.

In what follows, we look at the process of structural changes in five model alloys of the Cu–Pt system during cooling. The alloys under study are as follows: CuPt, Cu₃Pt, CuPt₃, CuPt₇ and Cu₃Pt₅. Here the calculation grid for every alloy was prescribed using random distribution of Cu and Pt atoms over the nodes of an fcc lattice in accordance with the component concentration. The temperature was varied from 2000 to 200 K with a step of 100 K.

CuPt alloy. In the model calculation, 50% Cu and 50% Pt were randomly distributed over the crystal lattice nodes. From the mean configurational energy we constructed the plot of energy variation as a function of temperature in the course of cooling (Fig. 5). In the CuPt alloy, at the temperature below 1200 K the atoms are distributed over the nodes of a rhombohedral lattice ($\alpha = \beta = \gamma = 91^\circ$). This crystal structure rearrangement is energy-favorable.

It is evident from the plot that up to approximately $T = 1200$ K, the alloy is in a disordered state, whereas as the temperature is decreased to 1000 K the energy is observed to decrease as well, which indicates the beginning of disordering. The energy value is found to stabilize at the temperatures below $T = 700$ K. Shown in Fig. 6 is the variation in the atomic structure of the alloy as a function of temperature.

At the temperatures as high as 1200 K the alloy is disordered. When the temperature is decreased to approximately 1000 K it undergoes ordering in accordance with the superstructure $L1_1$. As the temperature is further decreased, the entire crystal specimen gets ordered in accordance with the superstructure $L1_1$.

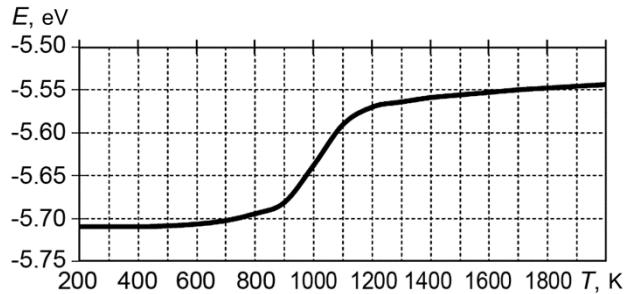


Fig. 5. Temperature dependence of the configurational energy in the volume of the model system calculated per atom of the CuPt alloy.

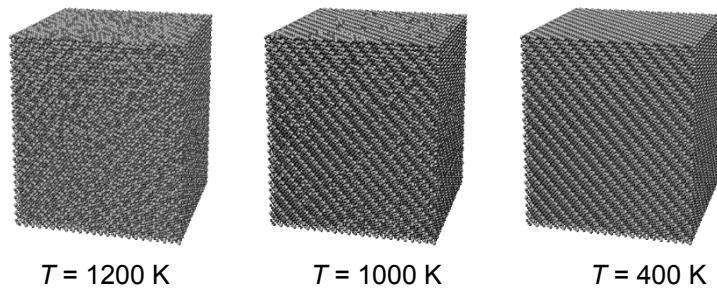


Fig. 6. Atomic structure of the CuPt alloy as a function of temperature.

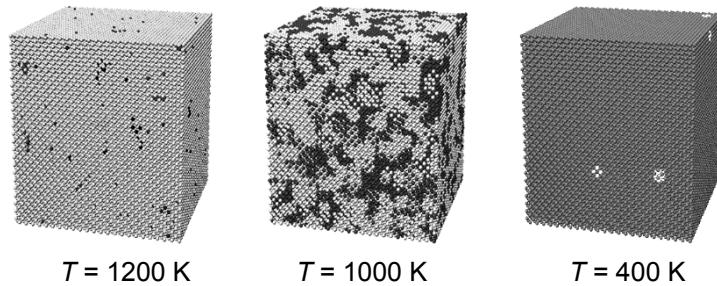


Fig. 7. Distribution of atoms over the ordered and disordered phases of the CuPt alloy in the course of cooling.

Figure 7 illustrates the domain structure variation in the alloy as a function of temperature. For the $L1_1$ superstructure there are four types of antiphase domains along the following directions: $\langle 111 \rangle$, $\langle \bar{1}\bar{1}1 \rangle$, $\langle \bar{1}1\bar{1} \rangle$ and $\langle 1\bar{1}\bar{1} \rangle$. The light-color regions in the figure show the disordered parts and dark-color regions – the domains and clusters of the crystal, respectively (similarly to [7–9]).

At the temperature about 1200 K the alloy is disordered, there are small-size domains and clusters, corresponding to the superstructures $L1_1$, $L1_2$ и $L1_3$. As the temperature is decreased to about 1000 K, the domain corresponding to the superstructure $L1_1$ is observed to predominate. As the temperature is further decreased, the entire crystal transfers to the domain structure of the same type.

Cu₃Pt alloy. In the initial state, 75% Cu and 25% Pt were randomly distributed over the crystal lattice nodes. From the mean configurational energy we constructed the plot of energy variation as a function of temperature in the course of cooling (Fig. 8). There is no rearrangement of the cubic lattice into a rhombohedral one in the Cu₃Pt alloy.

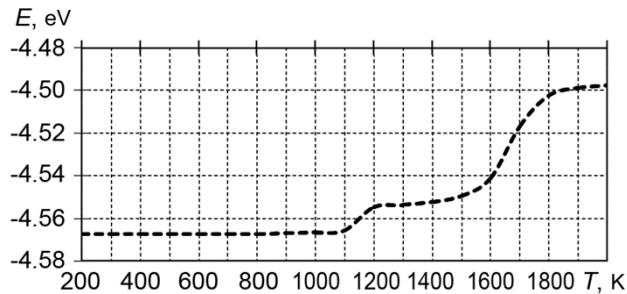


Fig. 8. Temperature dependence of the mean configurational energy in the volume of the model system calculated per atom in the Cu₃Pt alloy.

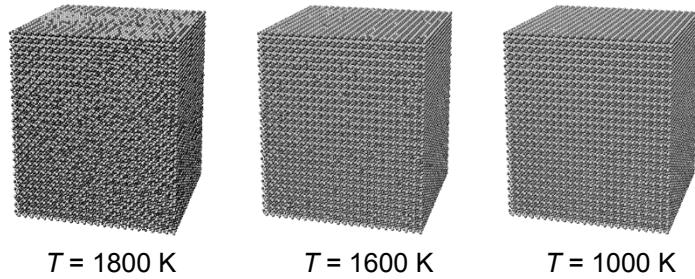


Fig. 9. Atomic structure of the Cu₃Pt alloy as a function of temperature.

It is evident from Fig. 8 that at the temperature above ~1800 K the alloy is disordered. As the temperature is decreased the energy value is also decreased, which is indicative of structural-phase rearrangements into more favorable structural states. For the decreased temperatures, the following sequence of structural states is observed (Fig. 9). Shown in Fig. 9 in dark and light colors are the Pt and Cu atoms, respectively. At the temperatures about ~2000 K, the alloy is completely disordered, while at $T = 1600$ K there are conspicuous regions ordered as the $L1_2$ superstructure. Already at $T = 1000$ K nearly the entire crystal has the $L1_2$ ordering, and as the temperature is further decreased this ordering is maintained.

Two horizontal sections on the curve of mean configurational energy versus temperature (within 1200–1500 K and lower than 1100 K) in the volume of the alloy are worth noting. These sections correspond to different structural states of the system, whose energy difference is quite small. This indicates low stability of these structural-phase states of this system to structural-phase transformations. The completely and partially ordered states that occur in this temperature interval exhibit a very little energy difference and are, therefore, low-stability structural states with respect to mutual transformations. To an approximation of thermodynamic probability of occurrence of these states, in the case of finite temperatures we could expect them to co-exist in a single system or to transform under the least thermal force action.

Figure 10 presents the variation in the domain structure of the alloy as a function of temperature. At $T = 2000$ K the alloy is disordered, it contains the nuclei of antiphase domains with the $L1_2$ superstructure. As the temperature is decreased down to approximately 1600 K, a single domain with the $L1_2$ superstructure is formed, which is sustained when the temperature is further decreased.

CuPt₃ alloy. In the initial state, 25% Cu and 75% Pt were randomly distributed over the crystal lattice nodes. From the mean configurational energy we constructed the plot of energy variation as a function of temperature in the course of cooling (Fig. 11). There is no structural-phase transition from the cubic lattice into a rhombohedral in the CuPt₃ alloy.

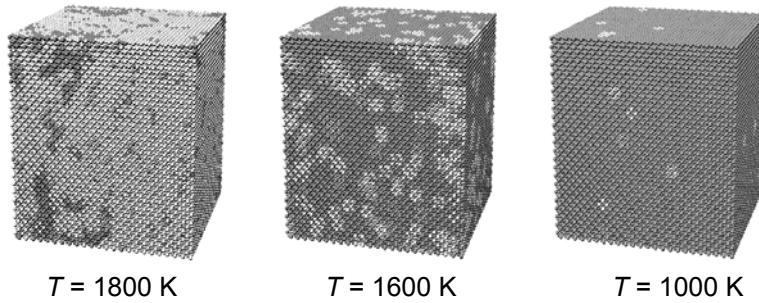


Fig. 10. Distribution of atoms over the ordered and disordered phases in the Cu₃Pt alloy during cooling.

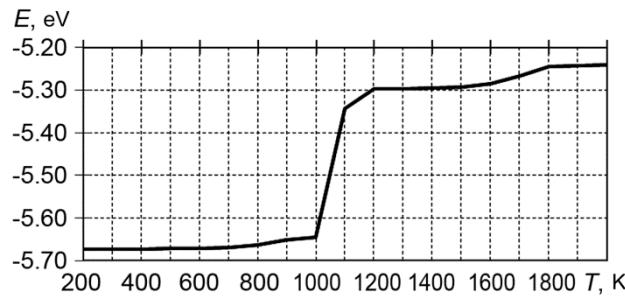


Fig. 11. Temperature dependence of the mean configurational energy in the volume of the model system calculated per atom in the CuPt₃ alloy as a function of temperature during cooling.

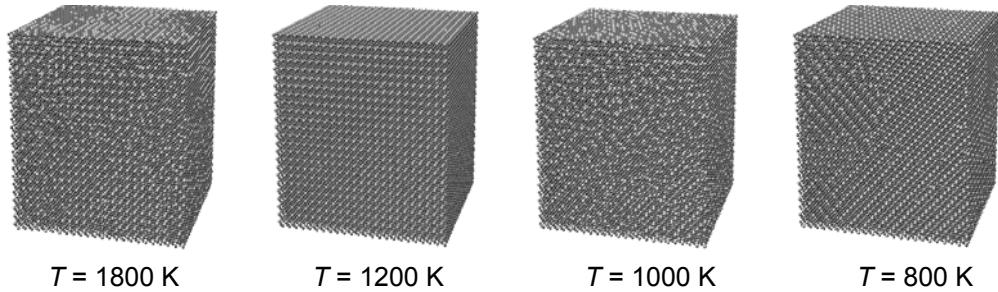


Fig. 12. Atomic structure of the CuPt₃ alloy as a function of temperature.

It is evident from Fig. 11 that down to approximately 1800 K the temperature is in a disordered state (Fig. 12), while as the temperature is decreased down to 1600 K there is a decrease in the energy values, which is evidence of partial ordering of the system. Within the range of temperatures 1600–1200 K (horizontal section of the curve), a certain structural-phase state is realized which is demonstrated in Fig. 12 at the temperature 1200 K (ordered state with the L1₂ superstructure). The completely disordered and partially disordered states hardly differ in their energies and therefore represent low-stability structural states with respect to transformations. To an approximation of thermodynamic probability of occurrence of these states, in the case of finite temperatures we could expect them to co-exist in a single system or to mutually transform under the least thermal force action.

The energy is stable to approximately 1200 K; it is then observed to decrease, which indicates a structural-phase rearrangement into a more energy-favorable structure (Fig. 12). At T = 1000 K, the alloy again transforms into its

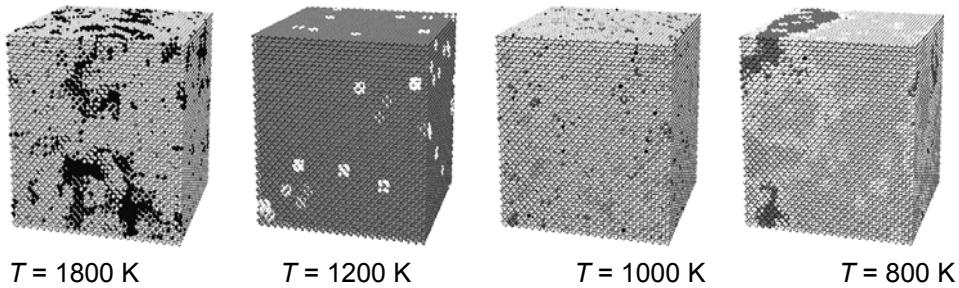


Fig. 13. Distribution of atoms over the ordered and disordered phases in the CuPt₃ alloy during cooling.

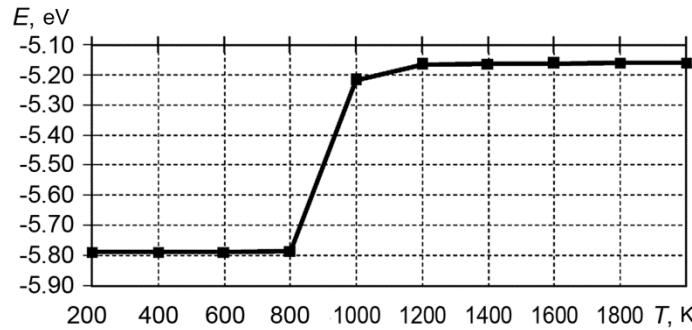


Fig. 14. Temperature dependence of the mean configurational energy in the volume of the model system calculated per atom in the CuPt₇ alloy as a function of temperature during cooling.

disordered state and starts rearranging into an $L1_3$ superstructure (Fig. 12). The energy is stabilized at the temperatures below 800 K, which is also evident from Fig. 12. At $T = 800$ K, we clearly observe the regions ordered as the $L1_3$ superstructure. When the temperature is further decreased, the entire model alloy has the $L1_3$ superstructure ordering.

Figure 13 presents the variation in the domain structure of the alloy as a function of temperature. For the $L1_3$ superstructure, there are three types of antiphase domains along the $\langle 100 \rangle$, $\langle 010 \rangle$ and $\langle 001 \rangle$ directions. The light regions in Fig. 13 correspond to the disordered states, the dark regions represent the $L1_2$ domains, and the grey ones represent the antiphase domains with the $L1_3$ superstructure. At the elevated temperatures (~ 2000 K), the alloy is disordered, there are fine-grained domains and clusters with the $L1_2$ and $L1_3$ superstructures. As the temperature is decreased to approximately 1800 K, the domain corresponding to the $L1_2$ superstructure is observed to predominate. Their structure (high-temperature disordered state and that within the 1800–1200 K temperature interval) exhibits a range of low stability- to mutual transformations or their co-existence in one and the same system at finite temperatures due to the entropy factor, which follows from Figs. 12 and 13. These states are thermodynamically similar but different in terms of symmetry. As the temperature is decreased down to approximately 1200 K, the entire crystal gets ordered as the $L1_2$ superstructure. At the temperature ~ 1000 K, the system again transforms into the disordered state followed by the formation of nuclei of the $L1_3$ -superstructure antiphase domains, i.e., it undergoes a structural-phase transformation from the $L1_2$ into $L1_3$ superstructure. As the temperature is decreased down to 800 K, the number of disordered regions becomes smaller and all the three types of the $L1_3$ superstructure antiphase domains are observed simultaneously. When the temperature is further decreased, the crystal is occupied by the domains of one type.

CuPt₇ alloy. In the initial state, 12.5% Cu and 87.5% Pt were randomly distributed over the crystal lattice nodes. From the mean configurational energy, we constructed the plot of energy variation as a function of temperature in the course of cooling (Fig. 14). There is no structural-phase transition from the cubic lattice into a rhombohedral in the CuPt₇ alloy.

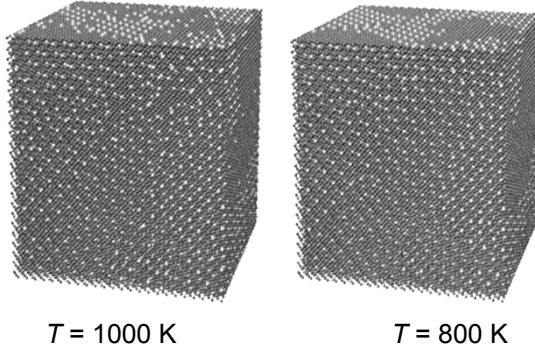


Fig. 15. Atomic structure of the CuPt₇ alloy as a function of temperature.

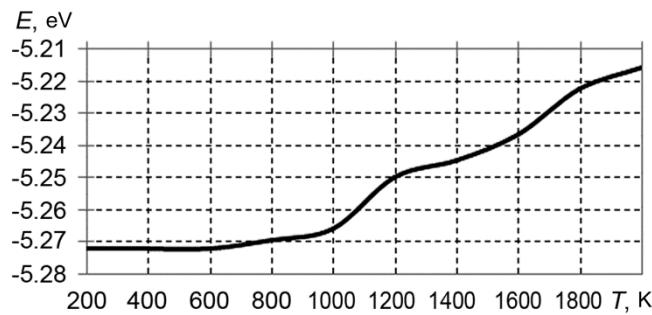


Fig. 16. Temperature dependence of the mean configurational energy in the volume of the model system calculated per atom in the Cu₃Pt₅ alloy.

It is evident from Fig. 14 that above 1200 K the alloy is in a disordered state. As the temperature is decreased there is a decrease in the energy values, which evidences the occurrence of structural-phase transformations into thermodynamically favorable structures. The variation in the alloy atomic structure is given in Fig. 15. The dark color indicates Pt atoms and the light color – Cu atoms.

At $T = 2000$ K, the alloy is completely disordered. As the temperature is decreased from 1200 to 1000 K, the atomic structure is rearranged according to the superstructural type of CuPt₇. At $T = 800$ K, there are conspicuous regions ordered in accordance with the CuPt₇ superstructure, which is maintained as the temperature is further decreased.

Cu₃Pt₅ alloy. In the initial state, 37.5% Cu and 62.5% Pt were randomly distributed over the crystal lattice nodes. From the mean configurational energy we constructed the plot of energy variation as a function of temperature in the course of cooling (Fig. 16). There is no structural-phase transition from the cubic lattice into a rhombohedral in the Cu₃Pt₅ alloy.

It is evident from Fig. 16 that at the temperature above 1800 K the alloy is disordered. As the temperature is further decreased the energy values are also decreased, which suggests the occurrence of structural-phase transitions into thermodynamically more advantageous structures. At $T = 2000$ K, the alloy is completely disordered. When the temperature is decreased from 1400 to 1200 K, the system undergoes a structural-phase transition into a superstructural state Cu₃Pt₅. This structural type is maintained as the temperature is further decreased.

When we assume a possible transition from the cubic to rhombohedral lattice, the situation is somewhat different. Shown in Fig. 17 are the results of two model cases. The first case deals with the order – disorder transition in the cubic lattice only, while the second model case addresses the order – disorder transition combined with a possible structural transformation from the cubic to face-centered rhombic lattice. We can readily see that in the course of

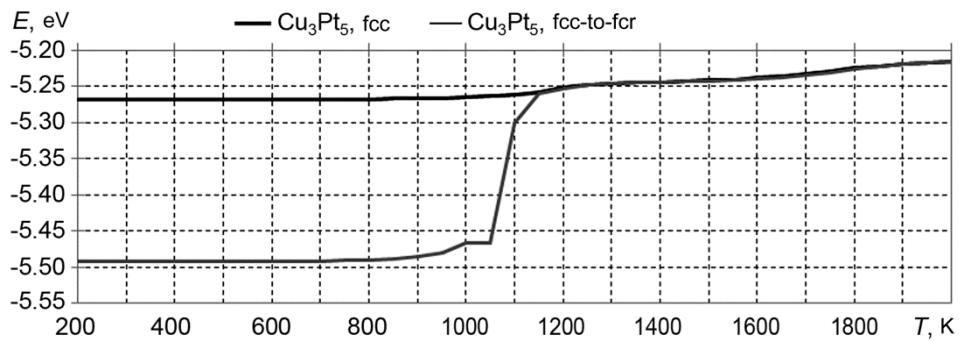


Fig. 17. Temperature dependence of the mean configurational energy in the volume of the crystal calculated per atom in the Cu_3Pt_5 alloy. The upper curve represents the system with an fcc-lattice, and the lower curve corresponds to the system allowing for an fcc-to-fccr transformation.

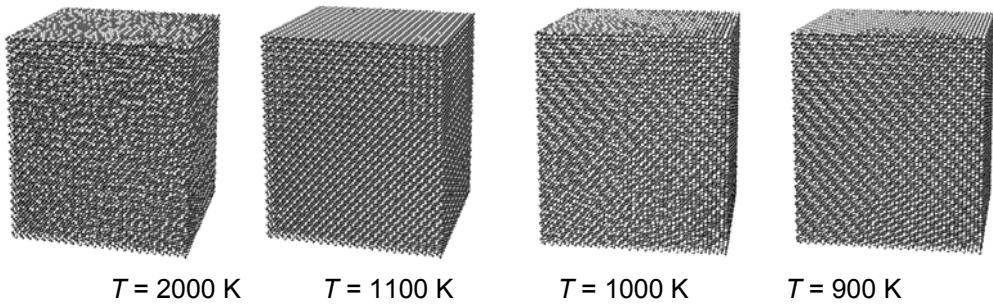


Fig. 18. Atomic structure of the Cu_3Pt_5 alloy as a function of temperature.

cooling the crystal structure undergoes ordering combined with its transition into a more energy-favorable structure based on a rhombohedral lattice.

The variation in the alloy atomic structure is given in Fig. 18. The dark color indicates Pt atoms and the light color – Cu atoms. It is evident from Fig. 18 that down to $T = 1100$ K the alloy undergoes ordering according to the Cu_3Pt_5 superstructural type, while as the temperature is further decreased the superstructural type is changed. In the first case, the planes of the atoms of one kind lie along one of the following directions: $\langle 100 \rangle$, $\langle 010 \rangle$, $\langle 001 \rangle$ and in the second case – the superstructure is formed by alternation of the same kind atoms (Pt atoms) in one of the directions of the $\langle 111 \rangle$ planes. Thus, a new structure is formed, which is probably a version of the Cu_3Pt_5 superstructural type.

SUMMARY

We have established that in the course of cooling, the Cu–Pt system undergoes a complicated structural-phase transformation combining an order-disorder phase transition and a structural transformation. As the temperature is decreased, the high-temperature disordered fcc-phase, depending on the composition, can, in addition to the atomic ordering, undergo a structural change so that a number of different superstructures can be realized, whose variety is controlled by the composition.

In certain cases, the configurational energy – temperature plot contains horizontal sections (within a certain range of temperatures). These sections represent different structural states of the system, whose energy difference is quite small. This suggests a poor stability of these structural states with respect to mutual structural-phase transformations. In an approximation of thermodynamic probability of the occurrence of these states in the case of finite

temperatures, they are expected to co-exist in one and the same system or undergo mutual transformations under the action of a minimum thermal force.

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