CORRELATION IN DISTRIBUTION OF INTERSTITIAL ATOMS IN H.C.P. ALLOYS

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The physical properties of crystals are defined to a large extent by materials structure and character of atoms distribution in crystal lattice [1, 2]. The interstitial phases [3-23], as refractory, hard, high-temperature alloys, are of considerable current use in science and technology. The peculiarity of their physical properties is determined by the presence of structural defects induced by interstitial atoms.

In studies of correlation in distribution of atoms of different sort throughout the lattice sites and interstitial sites it can be established, the reality of short-range and local order forming, it can be ascertained the concentration and temperature ranges of phases homogeneity and chiefly it is possible to gain insight into the development of new materials based on old materials, to create new refractory metallic carbides, nitrides, borides without use of too costly alloying elements.

In the present paper the calculation of correlation parameters is carried out at first for h.c.p. alloys AB-C_{IN} with superstructure $B8_1$ and then with B19. The alloys of such type present in Ir-Si-C, Pd-Si-C, Pt-Si-C, Rh-Si-C, Nb-N systems and others.

The arrangement of interstitial atoms C in interstitial sites of two types: octahedral and tetrahedral is taken into account in calculation. It is suggested that C atoms (hydrogen, nitrogen, carbon, boron) are located in the centre of octahedron or tetrahedron.

In ordered state of alloys with $B8_1$ structure the one type of octahedral interstitial site O and two types of tetrahedral interstitial sites T_1, T_2 are distinguished in this lattice. The O interstitial site is surrounded by three sites of first type legal for A atoms and by three sites of second type legal for B atoms, the T_1 interstitial site has three nearest sites of first type and one site of second type and the T_2 interstitial site has one site of first type and three sites of second type.

The alloys with B19 structure have two types of octahedral O_1, O_2 and one type of tetrahedral T interstitial sites. The O_1 interstitial site is surrounded by four nearest sites of first type and by two sites of second type, the O_2 interstitial site has two nearest sites of first type and four – of second type. The T interstitial site has two sites of first type and two sites of second type from the four nearest sites.

For determination of correlation parameters the free energy is calculated. The method of configurations is used, i.e. all possible configurations of A and B atoms around interstitial sites are taken into account. The interaction of AC, BC atomic pairs is taken into consideration in the first coordination sphere.

For $B8_1$ structure the correlation parameters in substitution of sites i = 1, 2 and of interstitial sites O, T_1, T_2 respectively by atomic pairs AC, BC are found to be:

$$\varepsilon^{(10)} = \frac{cP_A^{(1)}P_B^{(1)}e}{K_1^4K_2^3}, \qquad \varepsilon^{(20)} = \frac{cP_A^{(2)}P_B^{(2)}e}{K_1^3K_2^4},$$

$$\varepsilon^{(1T_1)} = \frac{2cP_A^{(1)}P_B^{(1)}e'K_1'}{\left(K_1'^2+K_2'^2\right)}, \quad \varepsilon^{(2T_1)} = \frac{2cP_A^{(2)}P_B^{(2)}e'K_1'^2}{K_2'\left(K_1'^2+K_2'^2\right)},$$

$$\varepsilon^{(1T_2)} = \frac{2cP_A^{(1)}P_B^{(1)}e'K_2'^2}{K_1'\left(K_1'^2+K_2'^2\right)}, \quad \varepsilon^{(2T_2)} = \frac{2cP_A^{(2)}P_B^{(2)}e'K_2'}{\left(K_1'^2+K_2'^2\right)},$$

and for B19 structure and interstitial sites O_1, O_2, T they are equal to

$$\varepsilon^{(10_1)} = \frac{2cP_A^{(1)}P_B^{(1)}eK_1}{\left(K_1^2+K_2^2\right)}, \quad \varepsilon^{(20_1)} = \frac{2cP_A^{(2)}P_B^{(2)}eK_1^2}{K_2\left(K_1^2+K_2^2\right)},$$

$$\varepsilon^{(10_2)} = \frac{2cP_A^{(1)}P_B^{(1)}eK_2^2}{K_1\left(K_1^2+K_2^2\right)}, \quad \varepsilon^{(20_2)} = \frac{2cP_A^{(2)}P_B^{(2)}eK_2}{\left(K_1^2+K_2^2\right)},$$

$$\varepsilon^{(1T)} = \frac{cP_A^{(1)}P_B^{(1)}e'}{K_1'}, \qquad \varepsilon^{(2T)} = \frac{cP_A^{(2)}P_B^{(2)}e'}{K_2'},$$

where

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$$P_A^{(1)} = a + \frac{1}{2}\eta, \quad P_A^{(2)} = a - \frac{1}{2}\eta,$$

$$P_B^{(1)} = b - \frac{1}{2}\eta, \quad P_B^{(2)} = b + \frac{1}{2}\eta,$$

$$e = \exp\frac{\alpha}{kT} - \exp\frac{\beta}{kT},$$

$$K_1 = P_A^{(1)} \exp\frac{\alpha}{kT} + P_B^{(1)} \exp\frac{\beta}{kT},$$

$$K_2 = P_A^{(2)} \exp\frac{\alpha}{kT} + P_B^{(2)} \exp\frac{\beta}{kT},$$

the e', K_1', K_2' quantities are expressed by formulae as e, K_1, K_2 with α', β' in place of α, β; *a*, b, c are atomic concentrations of alloy components A, B, C; η is the atomic order degree, $\alpha = -\upsilon_{AC}, \beta = -\upsilon_{BC}$ are energies of AC, BC pairs interaction for octahedral interstitial sites, α', β' are identical energies for tetrahedral interstitial sites.

The derived formulae determine the dependence of correlation parameters on alloy composition, temperature and order parameters.

From the derived formulae it follows that correlation parameters are found to be equal to zero, firstly, in the case of completely ordered alloys of stoichiometric composition, secondly, at the equal energetic parameters $\alpha=\beta$, $\alpha'=\beta'$, thirdly, in uncombined metals A (or B), and fourthly, at very high temperatures ($T\to\infty$).

The derived analytical expressions for correlation parameters can be used in studies of many phenomena in alloys, as an example, at the investigation of intensity of diffuse scattering of X-rays, neutrons, electrons.

The regularity knowledge of functional dependences of correlation parameters can permit the prediction of atomic order and the correlation influence on physical characteristics of alloys and on occurred processes in them.

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КОРРЕЛЯЦИЯ В РАСПРЕДЕЛЕНИИ АТОМОВ ВНЕДРЕНИЯ В ГПУ СПЛАВАХ АВ

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Физические свойства кристаллов в значительной степени определяются структурой материала и характером распределения атомов в кристаллической решетке [1,2]. Широко используют в науке и технике фазы внедрения [3-6], какими являются тугоплавкие, твердые, жаропрочные сплавы. Особенность их физических свойств обусловлена дефектностью структуры, создаваемой внедренными атомами.

Исследуя корреляцию в распределении атомов разного сорта по узлам и междоузлиям решетки, можно выявить реальность формирования ближнего и локального порядка, приводящего к образованию зародышей новой фазы, установить концентрационные и температурные интервалы гомогенности фаз, а главное – получить сведения, которые могут помочь разработать новые материалы на основе старых, создать новые тугоплавкие металлические карбиды, нитриды, бориды без использования дорогостоящих легирующих элементов.

В работе выполнен расчет параметров корреляции для ГПУ сплавов АВ-C_{BH} сначала со сверхструктурой $B8_1$, затем B19. Такого типа сплавы проявляются, например, в системах Ir-Si-C, Pd-Si-C, Pt-Si-C, Rh-Si-C, Nb-N и других.

В расчетах учитывается размещение атомов внедрения С в междоузлиях двух типов: октаэдрических или тетраэдрических. Предполагается, что атомы С (водорода, азота, углерода, бора) находятся в центре октаэдра или в центре тетраэдра.

В упорядоченном состоянии сплавов со структурой $B8_1$ в решетке выделяются один тип октамеждоузлия О, окруженного тремя узлами первого типа, законными для атомов А, и тремя второго типа, законными для атомов В, и два типа тетрамеждоузлий T_1, T_2, из которых междоузлие T_1 имеет три ближайших узла первого и один узел второго типа, а междоузлие T_2 – один узел первого типа и три второго.

Сплавы со структурой B19 в упорядоченном состоянии обладают двумя типами октаэдрических O_1, O_2 и одним типом тетраэдрических Т междоузлий. Междоузлие O_1 окружено четырьмя ближайшими узлами первого типа и двумя второго, междоузлие O_2 имеет два ближайших узла первого типа и четыре второго. У тетрамеждоузлия Т из четырех ближайших узлов два являются первого и два второго типа.

Для определения параметров корреляции рассчитывалась свободная энергия. Использовался метод конфигураций, т.е. учитывались всевозможные конфигурации атомов А и В вокруг междоузлий. Взаимодействие пар атомов АС, ВС учитывалось в первой координационной сфере.

Для структуры $B8_1$ параметры корреляции в замещении парами атомов АС, ВС соответственно узлов типа i=1;2 и междоузлий 0, T_1, T_2 оказались равными

$$\varepsilon^{(10)}=\frac{cP_A^{(1)}P_B^{(1)}e}{K_1^4K_2^3},\qquad \varepsilon^{(20)}=\frac{cP_A^{(2)}P_B^{(2)}e}{K_1^3K_2^4},$$

$$\varepsilon^{(1T_1)}=\frac{2cP_A^{(1)}P_B^{(1)}e'K_1'}{\left(K_1'^2+K_2'^2\right)},\quad \varepsilon^{(2T_1)}=\frac{2cP_A^{(2)}P_B^{(2)}e'K_1'^2}{K_2'\left(K_1'^2+K_2'^2\right)},$$

$$\varepsilon^{(1T_2)}=\frac{2cP_A^{(1)}P_B^{(1)}e'K_2'^2}{K_1'\left(K_1'^2+K_2'^2\right)},\ \varepsilon^{(2T_2)}=\frac{2cP_A^{(2)}P_B^{(2)}e'K_2'}{\left(K_1'^2+K_2'^2\right)},$$

и для структуры B19 и междоузлий O_1, O_2, Т они равны

$$\varepsilon^{(10_1)}=\frac{2cP_A^{(1)}P_B^{(1)}eK_1}{\left(K_1^2+K_2^2\right)},\quad \varepsilon^{(20_1)}=\frac{2cP_A^{(2)}P_B^{(2)}eK_1^2}{K_2\left(K_1^2+K_2^2\right)},$$

$$\varepsilon^{(10_2)}=\frac{2cP_A^{(1)}P_B^{(1)}eK_2^2}{K_1\left(K_1^2+K_2^2\right)},\quad \varepsilon^{(20_2)}=\frac{2cP_A^{(2)}P_B^{(2)}eK_2}{\left(K_1^2+K_2^2\right)},$$

$$\varepsilon^{(1T)}=\frac{cP_A^{(1)}P_B^{(1)}e'}{K_1'},\qquad \varepsilon^{(2T)}=\frac{cP_A^{(2)}P_B^{(2)}e'}{K_2'},$$

где

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$$P_A^{(1)} = a + \frac{1}{2}\eta, \quad P_A^{(2)} = a - \frac{1}{2}\eta,$$

$$P_B^{(1)} = b - \frac{1}{2}\eta, \quad P_B^{(2)} = b + \frac{1}{2}\eta,$$

$$e = \exp\frac{\alpha}{kT} - \exp\frac{\beta}{kT},$$

$$K_1 = P_A^{(1)} \exp\frac{\alpha}{kT} + P_B^{(1)} \exp\frac{\beta}{kT},$$

$$K_2 = P_A^{(2)} \exp\frac{\alpha}{kT} + P_B^{(2)} \exp\frac{\beta}{kT},$$

величины e', K_1', K_2' выражаются формулами для e, K_1, K_2 при замене $\alpha \to \alpha'$, $\beta \to \beta'$; *a*, b, c – атомные концентрации компонентов A, B, C сплава, η - степень атомного порядка, $\alpha = -\upsilon_{AC}, \beta = -\upsilon_{BC}$ - энергии взаимодействия пар AC, BC для октамеждоузлий, α', β' - такие энергии для тетрамеждоузлий.

Полученные формулы определяют зависимость параметров корреляции от состава сплава, температуры и параметра порядка.

Из полученных формул вытекает, что параметры корреляции оказываются равными нулю, во-первых, в случае полностью упорядоченных сплавов стехиометрического состава, во-вторых, при соответственно равных энергетических параметрах $\alpha=\beta$, $\alpha'=\beta'$, в-третьих, в чистых металлах A (или B), и в-четвертых, при очень высоких температурах ($T \to \infty$).

Полученные аналитические выражения для параметров корреляции могут быть использованы при изучении многих явлений в сплавах, например, при исследовании интенсивности диффузного рассеяния рентгеновских лучей, нейтронов, электронов.

Знание закономерностей функциональных зависимостей параметров корреляции может позволить предсказать влияние атомного порядка и корреляции на физические характеристики сплавов и на процессы, протекающие в них.

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