NON-EQUILIBRIUM THERMODYNAMIC MODEL OF THE AUSTENITIC NON-DIFFUSION TRANSFORMATION IN IRON-BASED ALLOYS

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Abstract/Резюме: *A non-equilibrium thermodynamic model of the austenite non-diffusion transformation in iron and alloys based on it is developed, taking into account internal stresses in the system. Onsager motion equations are found for a model thermodynamic system describing a non-diffusion transformation and kinetic equations for changing deformations and growth rates of the α-phase. A scheme of austenitic non-diffusion transformations is constructed, including normal and martensitic transformations, as limiting cases.*

KEYWORDS: NONEQUILIBRIUM THERMODYNAMICS, NON-DIFFUSION TRANSFORMATION, AUSTENITE, ONSAGER MOTION EQUATIONS, GROWTH RATES OF THE A-PHASE, IRON-BASED ALLOYS

1. Introduction / Введение

The study of phase transformations is one of the most important problems in the physics of metals [1-3]. Phase transformations are divided into diffusion and non-diffusion [1]. If the kinetics of phase transformation in steels and cast irons is determined by the diffusion of carbon, this allows them to be attributed to conversions controlled by diffusion [1-4].

Such transformations in iron-carbon alloys include pearlitic transformation of austenite, and transformations occurring during tempering, graphitization of undoped cementite, separation of carbides in alloyed steels, and others [4-6].

When the rate of transformation of austenite is determined by the rate at which the interface separates, differing only in its crystalline structure, the transformation is called non-diffusion [1]. Kinetically, the normal polymorphic and martensitic transformations of austenite are distinguished. When the temperature of the normal transformation decreases, its velocity first increases, and then decreases. The kinetics of the martensitic transformation is characterized by a very high rate of growth of individual crystals and the maximum space velocity at the initial moment of transformation under isothermal conditions.

In addition to martensite, at least two other structural components are known, which are formed with a shear ("martensitic") morphology of crystal formation - ferrite side-plates (widmanstätten) and acicular ferrite. They can also be attributed, with some simplifying assumptions, to the products of the non-diffusion transformation of austenite. In addition, in some alloys martensitic and normal transformations occur at the same temperature [1]. The consistent theory of non-diffusion transformations should explain this phenomenon.

The purpose of this paper is to analyze the non-diffusion transformation of austenite based on the positions of nonequilibrium thermodynamics, taking into account the influence of internal stresses and finding the main kinetic parameters of the process.

2. Equations of motion and model parameters/ Уравнения движения и параметры модели

Martensite is the basis of hardened steel, so studying the mechanism and kinetics of its transformation is still of extreme interest for the theory and practice of heat treatment.

In the works of G.V. Kurdyumov and co-workers, the martensitic transformation is considered as the usual phase transformation in a one-component system, further complicated by the influence of a strong interatomic interaction, which leads to the development of significant stresses in the martensite crystal and matrix [5].

In accordance with the alternative mechanism, the martensitic transformation takes place by means of an instantaneous shift of atomic planes that does not require thermal activation and is not associated with thermodynamic transformation stimuli [1, 6]. In this case, the stress initiating the transformation is believed to be the stresses arising from the sharp cooling of the sample (quenching) [6].

Considering the martensitic transformation as a thermally activated process, B.Ya. Lyubov used the equations of normal transformation obtained on the basis of the positions of nonequilibrium thermodynamics to describe his kinetics [3].

Changes in a complex or composite system under constant external conditions can be described as the process of increasing entropy. The rate of increase of entropy σ can be represented as the sum of the flux products and the corresponding forces for all transfer substrates in an amount of N [10-13]:

$$\sigma = (dS/dt)_{irrev} = \sum_{\kappa=1}^{N} J_k X_k \quad (k=1,..,N), \qquad (1)$$

In the general case, the flows can be represented in the form [11, 12]:

$$J_i = \sum_{\kappa=1}^{N} L_{ik} X_k \quad (i,k=1,..,N), \qquad (2)$$

The irreversible change in the entropy dS_{irrev} is equal to the sum of entropy changes in the system and the environment:

$$dS_{irrev} = dS + dS_e \qquad (3)$$

Under isothermal conditions, when the released heat is absorbed by the environment and the temperature remains constant:

$$dS_e = - dQ/T, \qquad dQ = dU + PdV.$$
$$dS_{irrev} = dS - (dUa + PdV)/T = (TdS - dU - PdV)/T. \qquad (4)$$

Since $dU + PdV - TdS = dG$, and if we take into account the low compressibility of bodies in the condensed state and relatively small pressures, then

$$(dS/dt)_{irrev} = - T^{-1}dG/dt \approx - T^{-1}dF/dt, \qquad (5)$$

where F is the free energy of the system.

The change in free energy in a system with a variable number of particles and internal stresses can be represented in the form [3, p.142]:

$$dF = dF\varepsilon + dFn = \sigma_{ik}d\varepsilon_{ik} + \varphi_l dn_l, \qquad (6)$$

where $dF\varepsilon$ is the change in free energy in the system related to internal stresses; dFn is the change in the free energy in the system, determined by the variable number of particles of type l; σ_{ik} is the stress tensor; ε_{ik} is the strain tensor of the system; φ_l is the chemical potential of the lth element of the system; n_l is the number of particles of the lth element of the system per unit volume, l = 1, N.

We now introduce some simplifying assumptions. First, for the non-diffusion transformation of austenite, only one kind of particles, the α-phase of iron n_α, will be taken into account. Approximately this is also true for alloys of iron with close elements (nickel, chromium, cobalt). Of course, φ is some effective (averaged) chemical potential of the atoms of the alloy.

Secondly, we assume that the deformation of the system is a triaxial compression-expansion, and in the expression for $dF\varepsilon$ only the diagonal components of stress and strain tensors are taken into account:

$$\sigma_{ik} = \varepsilon_{ik} = 0, i \neq k. \ \sigma_{ii} = \sigma, \varepsilon_{ii} - \varepsilon. \qquad (7)$$

The change in internal energy can then be represented as:

$$dF = 3\sigma d\varepsilon + \varphi dn_\alpha, \tag{8}$$

but the change in entropy:

$$(dS/dt)_{irrev} = - T^{-1}(3\sigma d\varepsilon/dt + \varphi dn_\alpha/dt). \tag{9}$$

Thus, in our system, in addition to the particle flow from the γ phase to the α-phase of $J_1 = dn_\alpha/dt$, we will also take into account the change in the strain of the sample with time $J_2 = d\varepsilon/dt$. These flows are related to the driving forces by the equation (2).

If, as charges of the process of non-diffusion transformation of austenite, two quantities-the concentration of α-phase particles and the strain value, then, according to (1), the equations of motion take the form:

$$J_1 = L_{11}X_1 + L_{12}X_2 \tag{10.1}$$
$$J_2 = L_{21}X_1 + L_{22}X_2, \tag{10.2}$$

where $X_1 = \Delta\varphi$ - the thermodynamic force for iron - the change in the chemical potential at the transition of particles from the γ-phase to the α-phase, $X_2 = \Delta\sigma$ - the change in the internal stress during the transition from the γ-phase to the α-phase.

The system of equations (42) describes the contribution of stresses and deformations to the non-diffusion transformation of austenite. However, we do not yet know the coefficients of the equations in it.

We now find expressions for the coefficients of the system of equations (42).

The coefficient L_{11} characterizes the normal transformation:

$$J_1 = dn_\alpha/dt = L_{11}(\varphi_\gamma - \varphi_\alpha). \tag{11}$$

In the normal kinetics of the phase transformation, the formation of the center (particle) of the α-phase occurs through separate (independent) acts of detachment of particles from the γ-phase and the attachment of atoms to the ferrite center. If we consider the process of formation of an α-phase close to the process of self-diffusion of iron in the γ-phase, then the coefficient L_{11} has the form [12]:

$$L_{11} = D_\gamma/RT, \tag{12}$$

where D_γ is the self-diffusion coefficient of iron in the γ-phase (or the effective coefficient of self-diffusion in the γ-phase of the alloy),

T is the transformation temperature,

R is the gas constant [16].

The self-diffusion coefficient of iron is taken in the usual notation [17]:

$$D_\gamma = D_0 e^{-\frac{U}{kT}} = 4,58\cdot10^{-4}\exp(-2,52\cdot10^5/RT), \tag{13}$$

D_0 is a multiplier;

U is the activation energy of diffusion;

The coefficient L_{22} characterizes the direct relationship:

$$J_2 = d\varepsilon_\alpha/dt = L_{22}(\sigma_\gamma - \sigma_\alpha). \tag{14}$$

where σ_α is the v stress in the α phase, σ_γ is the stress in the γ phase. Let $\sigma_\gamma = 0$. Let us take into account that for triaxial compression-stretching [28]:

$$\sigma_\alpha = \frac{E}{3(1-2\mu)}\frac{\Delta V}{V} = \frac{E}{1-2\mu}\varepsilon_\alpha, \tag{15}$$

where E is the modulus of elasticity of steel($\sim$2,17$\cdot10^5$ MPa), μ is the Poisson ratio ($\sim$ 0.26). Then expression (14) can be transformed as follows:

$$d\varepsilon_\alpha/dt = L_{22}\sigma_\alpha = L_{22}\frac{E}{1-2\mu}\varepsilon_\alpha = \frac{v}{L}\varepsilon_\alpha, \tag{16}$$

where the following values are entered:

v is the propagation velocity of the microdeformation in sample ($\sim$1000 m/s) [3];

L is the characteristic distance over which the micro-deformation of the shear is propagated (the size of the martensitic strips or plates). At the initial stage of the formation of the shear structure, it has a magnitude of the order of the diameter of the austenite grain ($\sim$ 100 μm), and then decreases with decreasing temperature [1]. From equation (48) we find that the coefficient L_{22} is equal to:

$$L_{22} = \frac{v(1-2\mu)}{LE}. \tag{17}$$

The cross-coefficients $L_{12} = L_{21}$ for a nonequilibrium thermodynamic system are found with sufficient accuracy by the formulas proposed in [5]:

$$L_{12} = \sqrt{L_{11}L_{22}} = \sqrt{\frac{D_\gamma}{RT}\frac{v(1-2\mu)}{LE}} \tag{18}$$

Thus, we obtained simple differential equations for a nonequilibrium thermodynamic system describing the non-diffusion transformation of austenite taking into account the influence of internal stresses.

Let us write the equations of motion of our system in the form:

$$dn_\alpha/dt = L_{11}\Delta\varphi + L_{12}\sigma_\gamma - L_{12}\sigma_\alpha. \tag{19.1}$$
$$d\varepsilon_\alpha/dt = L_{21}\Delta\varphi + L_{22}\sigma_\gamma - L_{22}\sigma_\alpha. \tag{19.2}$$

We first transform equation (19.2) taking into account expression (16). We have:

$$d\varepsilon_\alpha/dt + v\varepsilon_\alpha/L = L_{21}\Delta\varphi + L_{22}\sigma_\gamma, \tag{20}$$

where ε_α is the magnitude of deformations of the α-phase. The differential equation (20) with constant coefficients (temperature) has a solution:

$$\varepsilon_\alpha = \frac{(L_{21}\Delta\varphi + L_{22}\sigma_\gamma)(1-2\mu)}{L_{22}E}(1 - e^{-\frac{v}{L}t}). \tag{21}$$

This kinetic equation describes the change in the magnitude of the deformation of the α-phase in time. At t = 0, ε_α = 0. When the time is counted, a fast ($\sim$ 10^{-6} s) process of transition to deformation occurs:

$$\varepsilon_\alpha = \varepsilon_\gamma + \Delta\varepsilon_\alpha = \varepsilon_\gamma + \frac{L_{21}\Delta\varphi(1-2\mu)}{L_{22}E}. \tag{22}$$

Equation (22) shows that the residual deformation of the α-phase after the transient process consists of the deformation of the austenite $\varepsilon\gamma$ and the additional deformation $\Delta\varepsilon_\alpha$. This additional deformation determines the change in the volume of the sample as $\gamma \to \alpha$-transformation:

$$\Delta V_{sam}/V_{sam} = 3n_\alpha\Delta\varepsilon_\alpha. \tag{23}$$

Then substituting expression (21) into equation (19.1), we find:

$$dn_\alpha/dt = L_{11}\Delta\varphi + L_{12}\sigma_\gamma - \frac{L_{12}(L_{21}\Delta\varphi + L_{22}\sigma_\gamma)}{L_{22}}(1 - e^{-\frac{v}{L}t}) = L_{11}\Delta\varphi + L_{12}\sigma_\gamma$$

$$-(L_{11}\Delta\varphi + L_{12}\sigma_\gamma)(1 - e^{-\frac{v}{L}t}) = (L_{11}\Delta\varphi + L_{12}\sigma_\gamma)e^{-\frac{v}{L}t}. \tag{24}$$

It can be concluded from expression (24) that the growth rate of α-phase particles depends on the stresses in the γ-phase. The greater the value of tensile stresses in the γ phase, the higher the growth rate of ferrite particles. The rate of growth of the α-phase particles at a constant temperature very rapidly (exponentially) decreases in time, determining the incompleteness of the transformation.

Integration of equation (24) with time-independent coefficients L_{11} and L_{12} allows us to obtain the kinetic equation for n_α:

$$n_\alpha = \frac{(L_{11}\Delta\varphi + L_{12}\sigma_\gamma)L}{v}(1 - e^{-\frac{v}{L}t}). \tag{25}$$

In accordance with equation (25), the amount of α-phase formed depends not only on the thermodynamic force $\Delta\varphi$, but also on the magnitude of the stresses in the γ-phase.

3. Scheme of the non-diffusion transformation of austenite / Схема бездиффузионного превращения аустенита

Before discussing the equations obtained, we introduce some more useful relations characterizing the $\gamma \to \alpha$ transformation.

With the $\gamma \to \alpha$ transformation, the effective atomic volume of the iron lattice changes in the sample under

consideration, characterized by $\Delta V_{\gamma \to \alpha}$ and the relative volume change $\Delta V_{\gamma \to \alpha}/V_{\gamma}$.

According to the data of [3]:

$$\Delta V_{\gamma \to \alpha} = 0{,}268 - 1{,}62*10^{-4}T, \; \text{sm}^3/\text{mol}. \qquad (26)$$

We will assume that with the formation of the α-phase, the relative change in volume is determined by the additional deformation: $\Delta V_{\gamma \to \alpha}/V_{\gamma} = 3\Delta\varepsilon_{\alpha}$, and the compressive stress arising in the α-phase has the value

$$\sigma_{\alpha} = \frac{E}{1-2\mu}\Delta\varepsilon_{\alpha} \cdot \qquad (27)$$

When the alloy sample is cooled by ΔT, a deformation occurs in its surface layer: $\varepsilon_{\gamma} \sim \alpha\Delta T$ and the tensile stress σ_{γ} corresponding to this deformation:

$$\sigma_{\gamma} = \frac{E}{1-2\mu}\alpha\Delta T \cdot \qquad (28)$$

Comparing the values of thermodynamic forces among themselves, it is possible to classify the types of non-diffusion transformation according to the kinetic criterion. As shown in [1, p. 208], for small deviations of the system from equilibrium, the growth of crystals is more likely, controlled by self-diffusion, at large - cooperative growth. The same phase transition in a single-component system under different external conditions can take place with an independent (or slightly dependent) temperature growth rate (martensitic kinetics) and with a rate that exponentially depends on the temperature at an activation energy close to the activation energy of self-diffusion (normal kinetics).

The parameter characterizing the deviation of the system from equilibrium is the supercooling of the alloy $\Delta T = \text{Ac}_3 - T$, where Ac_3 is the temperature of the end $\alpha \to \gamma$ of the conversion upon heating, and T is the transformation temperature. The transformation scheme for the constructed model is shown in Fig. 1

Ac₃	
	$L_{11}\Delta\varphi > L_{12}\sigma_{\alpha} > L_{12}\sigma_{\gamma}$, normal transformation, polygonal ferrite
Ac₁	$L_{11}\Delta\varphi \sim L_{12}\sigma_{\alpha} > L_{12}\sigma_{\gamma}$, shear + normal transformation, ferrite side-plates
Mni	$L_{11}\Delta\varphi < L_{12}\sigma_{\alpha}, \sigma_{\alpha} = \sigma_{\gamma}$
Mna	$L_{11}\Delta\varphi < L_{12}\sigma_{\alpha}, \sigma_{\alpha} < \sigma_{\gamma}$, shear transformation isothermal martensite, acicular ferrite
Мκ	$L_{11}\Delta\varphi \ll L_{12}\sigma_{\gamma}$, shear transformation athermal martensite

Fig. 1. Scheme of non-diffusion transformations from the constructed model. Ac_1 is the temperature of the beginning of $\alpha \to \gamma$ transformation when the alloy is heated; Mni is the temperature of the onset of the formation of isothermal martensite upon supercooling of the alloy. Mn is the temperature of the onset of athermal martensite formation upon supercooling of the alloy. Mk is the temperature of the end of martensite formation upon supercooling of the alloy.

Thus, for small ΔT:

$$L_{11}\Delta\varphi > L_{12}\sigma_{\alpha} > L_{12}\sigma_{\gamma}, \qquad (29.1)$$

then the growth of α-phase crystals is determined by self-diffusion by the normal mechanism. However, as follows from equation (19), in this case too, the contribution of deformations (and stresses) to the conversion kinetics is very significant. In order that the condition (29.1) is satisfied, it is necessary that the stress level in the γ- and α-phases be small; for the α-phase this is possible only in the case of relaxation of internal stresses in the alloy at high temperature by the mechanism of recrystallization.

With increasing supercooling of the alloy, the thermodynamic stimulus and the rate of normal transformation increase.

For a larger ΔT:

$$L_{11}\Delta\varphi \sim L_{12}\sigma_{\alpha} > L_{12}\sigma_{\gamma}. \qquad (29.2)$$

The existing thermal stresses in the γ-phase (28) contribute to the formation of the α-phase by the shear mechanism; the stresses arising in the α-phase compensate thermal stresses in the γ-phase. With a certain amount of α-phase, the stress equals $\sigma_{\alpha} = \sigma_{\gamma}$ arises and the further formation of the α-phase occurs according to the normal mechanism with the relaxation of the arising stresses by recrystallization. Consequently, the condition (29.2) corresponds to the transformation of the γ-phase by a mixed mechanism, and also to the formation of a ferrite side-plates (widmanstätten), followed by the release of the α-phase by the normal mechanism [1].

With a certain supercooling of ΔTi, stress compensation occurs only when the γ-phase is completely transformed into ferrite by a shearing mechanism. In this case:

$$L_{11}\Delta\varphi \sim L_{12}\sigma_{\alpha}, \; \sigma_{\alpha} = \sigma_{\gamma}, \qquad (29.3)$$

The temperature corresponding to this supercooling is the starting point for the formation of the isothermal martensite Mni ($M_{\text{н}}^{\text{из}}$ on Fig. 2). Below the point Mni, the formation of the α-phase occurs by a shearing mechanism; However, the normal component of the process still has a significant value, affecting the morphology of the resulting precipitates.

When supercooling a greater ΔTi:

$$L_{11}\Delta\varphi < L_{12}\sigma_{\alpha}, \; \sigma_{\alpha} < \sigma_{\gamma}, \qquad (29.4)$$

At temperatures below Mni, isothermal martensite or acicular ferrite is formed with a "reticular" or acicular morphology of precipitates.

Finally, for large ΔT below Mna ($M_{\text{н}}^{\text{am}}$ on Fig. 2):

$$L_{11}\Delta\varphi \ll L_{12}\sigma_{\gamma}, \qquad (29.5)$$

Inequality (29.5) determines the condition for the formation of "athermal" martensite, when the normal component does not affect the formation of the shear structure. The main effect on the rate of the $\gamma \to \alpha$ transformation, in accordance with expression (24), is due to thermal stresses in the γ phase.

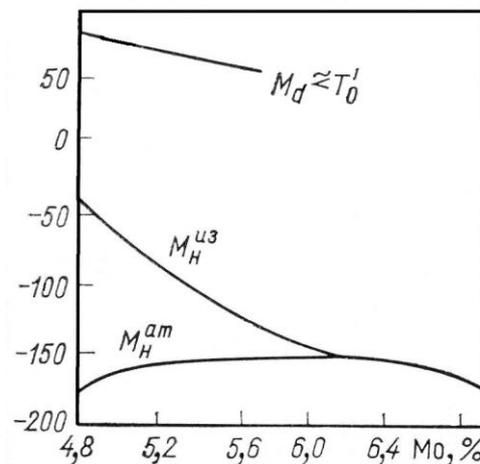The temperature region for the formation of this structural component for real alloys is also shown in Fig. 2.

Fig. 2. Temperature regions of the athermal $M_{\text{н}}^{\text{am}}$ and isothermal $M_{\text{н}}^{\text{из}}$ kinetics of the formation of martensite in Fe-24.5%Ni-Mo alloys with different Mo content [1].

In order that the transformation of austenite into martensite occurs with a temperature-independent growth rate of the α-phase, it must be assumed that the coefficient L_{12} is practically independent of temperature; this is possible under the following conditions:

$$D_{\gamma}/(TL) = \text{const}. \qquad (30)$$

We will perform some estimated calculations. Assume that at $T_0 = 400\,°C \approx 673\,°K$, the size of the martensite plates is $L_0 = 100\,\mu\text{m}$. Substitution of the values of L_0 and T_0 in (30) makes it possible to obtain an equivalent dependence of the martensite plate size on the temperature (Fig. 3). At 580 °K, the length of the martensitic plates corresponding to the condition (30) is only 0.084 μm.

Consequently, with a further decrease in temperature, the formation of martensite will occur with a decrease in the rate of

the $\gamma \rightarrow \alpha$ transformation (but not the rate of shear deformation), since the contribution of the diffusion component to the L_{12} coefficient is very significant. The rate of martensitic transformation in this case is many orders of magnitude greater than the rate of normal transformation.

As the direct calculations show, with the indicated parameters: $L_{11} = 4,3 \cdot 10^{-29}$; $L_{22} = 18,4$; $L_{12} = 0,89 \cdot 10^{-14}$. The rate of martensitic transformation at $T_0 = 400\,°$ C is approximately 10^{14} times greater than the rate of the normal (self-diffusion) transformation component.
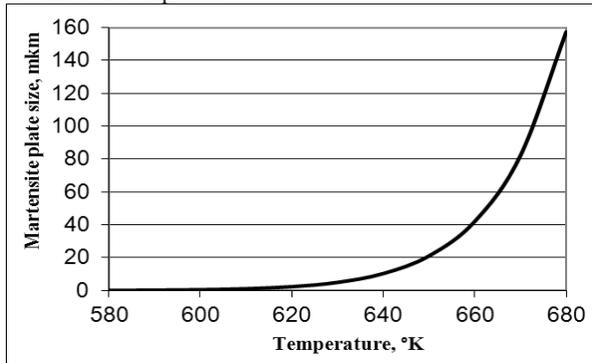

Fig. 3. *The calculated equivalent dependence of the martensite plate size on temperature*

The temperature at which the rate of martensite formation begins to decrease can characterize the temperature of the end of martensite formation Mk, if the amount of the residual γ phase is small (less than 1.0%).

Reduction in the rate of martensite formation at low temperatures should be taken into account in cryogenic technologies for increasing the wear resistance of steels and alloys.

Thus, the constructed model of the non-diffusion austenite transformations allows us to consider the normal and martensitic transformations, as limiting cases.

CONCLUSIONS

1. The application of nonequilibrium thermodynamics to the analysis of the non-diffusion transformation of austenite made it possible to obtain a system of equations for the thermodynamic system and to generalize the results obtained earlier by B.Ya.Lyubov the equations for a normal transformation.
2. The theoretical expression for the growth rate of the α-phase, obtained in this paper, takes into account the influence of stresses on the process of austenite transformation. It is shown that the rate

of growth of α-phase particles at a constant temperature very rapidly (exponentially) decreases in time, determining the incompleteness of the transformation.
3. According to the constructed model, a scheme of non-diffusion austenite transformations was developed, including normal and martensitic transformations, as limiting cases.

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