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PROBLEM ON DIFFUSION OF A PARTIALLY SOLUBLE IMPURITY IN A SOLID MATRIX WITH ALLOWANCE FOR THE DEPLETION OF THE DIFFUSANT SOURCE WITH TIME

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An analytical expression for the concentration profile of a diffusing element partially soluble in the material's sample has been obtained on condition that the diffusion source is depleted with time. Examples of the use of the solution obtained for processing of diffusion experiments carried out with a number of impurities in beryllium have been considered. The use of the present model shows a more accurate agreement of the calculated and experimental concentration profiles, which enables one to refine the characteristics of diffusion mobility of the impurities in the materials under study.

The stability of the structure of solid materials containing impurities is determined, as a rule, by the redistribution of the impurities between the solid solution and the isolated phases. The mobility of impurities in solid materials is limited by diffusion processes; therefore, the diffusion coefficients D_i ($i = 1, 2, ..., n$) of the impurities, which are found by the corresponding experiments [1], are an important characteristic of any impurity-containing material. To increase the migration rate of the impurities one carries out the experiments at a higher-than-average temperature (homogenizes samples) and then extrapolates the result obtained for D to the region of low$^{*)}$ temperatures, using the Arrhenius law [2]:

$$D(T) = D_0 \exp(-B/T) .$$

Clearly, in extrapolating $D(T)$ to lower temperatures, the error of determination of low-temperature diffusion coefficients increases; therefore, to improve the accuracy of their determination one must organize the processing of diffusion experiments so as to minimize the computational error for D.

We recall that, in the course of diffusion experiments, one most often applies a source layer of labeled (radioactive) diffusing atoms to one side of the sample; the sample is annealed isothermally for a certain period; then one successively removes its layers on the source side of the source layer and analyzes the radioactivity of the sample's residue $N(x)$, where x is the distance from the source layer to the sample [3].

To simplify the processing of the experiment the layer applied to the sample is made as thin as possible and the geometry of the sample is selected so that the process of diffusion can be considered to be one-dimensional. The diffusion coefficient D is determined by comparison of the dependences $N_{\text{exp}}(x)$ obtained in the experiment and a certain reference calculated function $N_{\text{calc}}(x)$. The form of the latter depends on conditions that are realized at the boundary of the matrix and the source layer of a diffusing impurity in the process of diffusion. In [4], it has been shown that unreliable data on the boundary conditions reduce the accuracy of determination of the coefficient D several times. Consequently, extrapolating the result for D to low temperatures, one can make a mistake by an order of magnitude or more. In this connection, in processing the experiments, we seek to reconstruct the boundary conditions realized at the source layer–matrix boundary in homogenization of a sample as accurately as possible. A mathematical apparatus describing the diffusion propagation of the impurity atoms in a solid matrix has entirely been borrowed from heat-con-

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$^{*)}$ Here and in what follows in the work, low temperatures are those lower than the homogenization temperatures.

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duction theory [5]. In differential form, the problem on diffusion of the impurity from the boundary into a semiinfinite matrix has the form

$$\frac{\partial}{\partial t} c\,(x, t) = D \frac{\partial^2}{\partial x^2} c\,(x, t) + \delta\,(x)\, f\,(t)\,, \quad x \ge 0\,, \quad t \ge 0\,;$$

$$c\,(x, 0) = 0\,, \quad x \ge 0\,; \quad \frac{\partial c}{\partial x}(0, t) = 0\,, \quad t > 0\,, \tag{1}$$

where $c(x, t)$ is the concentration of the impurity at a distance x from the matrix boundary at the instant of time t (coordinate of the boundary $x = 0$), $f(x)$ is the strength of the impurity source on the sample's surface ($x = 0$), i.e., the amount of the impurity arriving at the matrix per unit area of the boundary in a unit time, and $\delta(x)$ is the Dirac delta function.

In processing most of the experiments, it is assumed that the model of an "instantaneous" source is realized at the boundary when

$$f\,(t) = Q_0 \delta\,(t)\,, \tag{2}$$

where Q_0 is the strength of this instantaneous source. Such an assumption is permissible if the time of the experiment (homogenization time) is much longer than the time of dissociation of the source layer; the solubility of the impurity in the matrix is fairly high and in no way influences the penetration of the impurity into the matrix. For a number of materials containing poorly soluble impurities in the sample and for a number of diffusion sources whose dissociation time is comparable to that of the experiment or exceeds it, we cannot make the latter assumption.

In [6], an analysis has been made of the process of diffusion of the impurity from a thin layer into a solid matrix, in which the finite time of dissociation of the layer α^{-1} was taken into account and the time dependence of the diffusion-source strength and an expression for the concentration profile of the impurity in the sample $c(x, t)$ were obtained. In [7], consideration has been given to the process of diffusion on condition of a constantly acting source but under the assumption that the content of the impurity in the matrix is limited by the solubility limit c^*.

The present work seeks to analyze the diffusion of the impurity from the boundary of a semiinfinite solid body with allowance for the following factors:

(1) depletion of the source layer of the impurity with time;

(2) limitations on the content of the impurity in the matrix — the impurity concentration must not exceed the solubility limit c^*, $c^* > c(x, t)$, $x \ge 0$, and $t \ge 0$.

Let us perform an analysis of the problem of diffusion (1), taking the above factors into account. We write the expression for the concentration profile $c(x, t)$, using the existing [6] general solution of problem (1):

$$c\,(x, t) = \frac{1}{\sqrt{\pi D}} \int_0^t \frac{f\,(t)}{\sqrt{t - \tau}} \exp\left[-\frac{x^2}{4D\,(t - \tau)}\right] d\tau\,, \quad x \ge 0\,, \quad t \ge 0\,. \tag{3}$$

We make the following assumptions specifying the form of the function $f(t)$:

(a) the source layer of the diffusant is so thin that the time it takes any impurity atom from the sample to reach the sample's surface is negligible as compared to the time of the diffusion experiment;

(b) the source–matrix boundary is absolutely permeable;

(c) the equilibrium number of the impurity atoms capable of penetrating in a diffusion manner from the source layer into the matrix, i.e., the number of activated atoms, is in proportion to the total number of atoms left in the source layer by the instant of time t.

If we denote the number of atoms in the source layer at the instant of time t by $Q(t)$, we can write that

$$Q\,(t) = Q\,(0) - \int_0^t f\,(t')\, dt'\,, \tag{4}$$

and the expression for the strength of the impurity source $f(t)$ will take the form

$$f(t) = \alpha Q(t)\,\eta(t)\,, \tag{5}$$

where $1/\alpha$ is the characteristic time of activation of the impurity atoms [5] and $\eta(t)$ is the fraction of vacancies for the impurity atoms in the surface layer of the sample [6]:

$$\eta(t) = 1 - c(0, t)/c^* \,. \tag{6}$$

Substituting the concentration at the boundary $c(0, t)$ into expression (6) in the form (3)

$$c(0, t) = \frac{1}{\sqrt{\pi D}} \int_0^t \frac{f(t')}{\sqrt{t - t'}}\, dt' \,,$$

we obtain an equation for determination of $f(t)$:

$$f(t) = \alpha \left(Q_0 - \int_0^t f(t')\, dt' \right) \left(1 - \frac{1}{\sqrt{\pi D}\, c^*} \int_0^t \frac{f(t')}{\sqrt{t - t'}}\, dt' \right). \tag{7}$$

Thus, the time dependence of the diffusion-source strength is described by the nonlinear integral equation (7). We introduce the dimensionless time $\tau = \alpha t$, flux $\varphi(\tau) = f(t)/(\alpha Q_0)$, and parameter β:

$$\beta = \frac{Q_0}{2c^*} \sqrt{\frac{\alpha}{D}} \,, \tag{8}$$

and rewrite Eq. (7) in the dimensionless form

$$\varphi(\tau) = [1 - A(\varphi, \tau)]\,[1 - B(\varphi, \tau)]\,, \tag{9}$$

where $A(\varphi, \tau) = \int_0^\tau \varphi(\tau')d\tau$ and $B(\varphi, \tau) = \frac{2\beta}{\sqrt{\pi_0}} \int_0^\tau \frac{\varphi(\tau')\, d\tau'}{\sqrt{\tau - \tau'}}$.

The functions $A(\varphi, \tau)$ and $B(\varphi, \tau)$ have bounds following from the form of Eq. (9):

$$0 \le A(\varphi, \tau)\, B(\varphi, \tau) < A(\varphi, \tau)\,, \quad B(\varphi, \tau) < 1\,, \quad \tau \ge 0\,.$$

To simplify further analysis we drop the nonlinear term $A(\varphi, \tau)B(\varphi, \tau)$ in Eq. (9), i.e., will analyze the equation for $\varphi(\tau)$ in the form

$$\varphi(\tau) = 1 - (A(\varphi, \tau) + B(\varphi, \tau))$$

or

$$\varphi(\tau) = 1 - \int_0^\tau \left[1 + \frac{2\beta}{\sqrt{\pi}} \frac{1}{\sqrt{\tau - \tau'}} \right] \varphi(\tau')\, d\tau' \,. \tag{10}$$

As has been shown by the calculations carried out by the authors of the present paper, neglect of the nonlinear term in Eq. (9) leads to a slight distortion of the resulting curves for the flux $\varphi(\tau)$ and the impurity-concentration profile $c(x, t)$, whereas the shape of the curve is invariant and the results obtained are consistent with the physical meaning of the processes initially incorporated in the mathematical model (1), (7).

Equation (10) represents the linear integral Volterra equation with its kernel in the form of a convolution. The solution of (10) can easily be obtained using the Laplace transformation

$$\varphi\,(t) = \frac{1}{(y_1 - y_2)}\left[y_1 \Phi\,(y_1 \sqrt{\tau}) - y_2 \Phi\,(y_2 \sqrt{\tau})\right], \quad \tau \geq 0\,, \tag{11}$$

where $y_{1,2} = \beta \pm \sqrt{\beta^2 - 1}$ and $\Phi(\nu) = \exp\,(\nu^2)$ erfc (υ), and erfc $(\nu) = 1 - \frac{2}{\sqrt{\pi}} \int\limits_0^{\nu} \exp\,(-y^2) dy$ being the additional probability integral. The function $\Phi(\nu)$ is monotonically decreasing for all real nonnegative values of the argument $\Phi(\nu = 0) = 1$ and $\Phi(\nu \to \infty) = 0$.

Substituting the solution for $\varphi(\tau)$ (11) into Eq. (3), we determine the expression for description of the concentration profile of the impurity:

$$\frac{c\,(z,\,\tau)}{c^*} = 2\beta \exp\,(-\,z^2)\left[\frac{\Phi\,(z + y_2 \sqrt{\tau}) - \Phi\,(z + y_1 \sqrt{\tau})}{y_1 - y_2}\right], \quad z \geq 0\,, \quad \tau \geq 0\,. \tag{12}$$

The set of expressions (11) and (12) is the solution of problem (1), (7) in the approximation of the linearized equation for the impurity flux (10).

It is noteworthy that the parameters $y_{1,2}$ and the function $\Phi(y_{1,2})\ \sqrt{\tau}$ acquire complex values in the case $\beta < 1$; therefore, we give the expressions for the impurity flux $\varphi(\tau)$ and the concentration profile $c(z,\ \tau)$ for $\beta < 1$ in a somewhat different form:

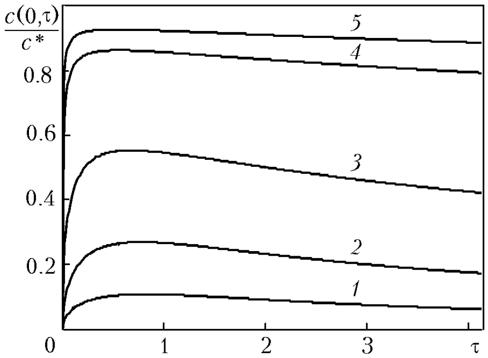
$$\varphi\,(\tau) = \frac{1}{\sqrt{1 - \beta^2}}\,\mathrm{Im}\left\{y_1 \Phi\,(y_1 \sqrt{\tau})\right\}, \quad \tau \geq 0\,;$$

$$\frac{c\,(z,\,\tau)}{c^*} = \frac{2\beta}{\sqrt{1 - \beta'}}\,\mathrm{Im}\left\{\Phi\,(z + y_2 \sqrt{\tau}\,)\right\}, \quad z \geq 0\,, \quad \tau \geq 0\,. \tag{13}$$

The main difficulty in calculations from formulas (13) is presented by computation of the function $\Phi(\nu)$ for a complex value of the argument because of the presence of the additional probability integral erfc (υ) in $\Phi(\nu)$. Using the approximate formula for computation of erfc (υ) for complex argument values [7], we can write expression (13) in a form more convenient for computation. Thus, when $\beta < 1$, we have

$$\begin{aligned}\varphi\,(\tau) = {} & \left\{\left[\Phi\,(\Delta_1) - \frac{1}{2\pi\Delta_1} - \frac{\Delta_1}{\pi}\sum_{n=1}^{\infty}\frac{\exp\,(-\,(n/2)^2)}{(n/2)^2 + \Delta_1^2}\right]\left[\cos\,(2\Delta_1\Delta_2) + \frac{\Delta_1}{\Delta_2}\sin\,(2\Delta_1\Delta_2)\right] + \frac{1}{2\pi\Delta_1}\right\}\exp\,(-\,\Delta_2^2) + \\ & + \frac{1}{2\pi}\frac{\Delta_1}{\Delta_2}\sum_{n=1}^{\infty}\left[\frac{\exp\,(-\,(n/2 - \Delta_2)^2)\,(\Delta_2 - n/2) - \exp\,(-\,(n/2 + \Delta_2)^2)\,(\Delta_2 + n/2)}{(n/2)^2 + (\Delta_1)^2}\right], \quad \tau > 0\,;\end{aligned} \tag{14}$$

$$\begin{aligned}\frac{c\,(z,\,\tau)}{c^*} = {} & \frac{2\Delta_1}{\Delta_2}\exp\,(-\,z^2)\,\Bigg\{\exp\,(-\,\Delta_2^2)\sin\,[2\,(z + \Delta_1)\,\Delta_2]\Bigg[\frac{1}{2\pi\,(z + \Delta_1)} + \frac{z + \Delta_1}{\pi} \times \\ & \times \sum_{n=1}^{\infty}\frac{\exp\,(-\,(n/2)^2)}{(n/2)^2 + (z + \Delta_1)^2} - \Phi\,(z + \Delta_1)\Bigg] + \frac{1}{2\pi}\sum_{n=1}^{\infty}\frac{n/2}{(n/2)^2 + (z + \Delta_1)^2}\Bigg\{\exp\left[-\left(\frac{n}{2} - \Delta_2\right)^2\right] -\end{aligned}$$



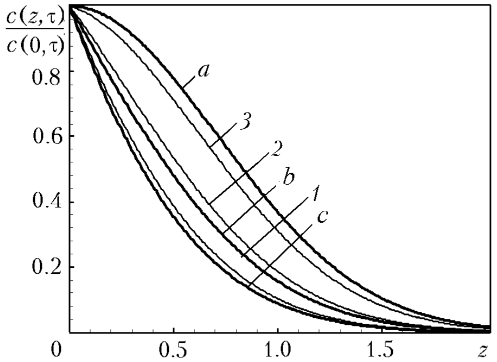


Fig. 1. Concentration of the impurity at the matrix boundary $c(0, \tau)/c^*$ as a function of the dimensionless time τ: 1) $\beta = 0.1$; 2) 0.3; 3) 1; 4) 3; 5) 10 (above curve 5, solubility limit).

Fig. 2. Concentration profiles of the impurity ((12) and (14)) when $\beta = 3$ for different values of the dimensionless time: 1) $\tau = 0.1$; 2) 10; 3) 1000 [a) instantaneous source ($\tau \to \infty$); b) impurity concentration; c) constant flux ($\tau \to 0$)].

$$-\exp\left[-\left(\frac{n}{2}+\Delta_2\right)^2\right]\Bigg]\Bigg\}, \quad z \ge 0, \quad \tau > 0,$$

where $\Delta_1 = \beta\sqrt{\tau}$ and $\Delta_2 = \sqrt{\tau(1-\beta^2)}$.

It is noteworthy that expressions (11), (12), and (14) have an indeterminacy of the 0/0 type for $\beta = 1$. However, the analysis made has shown that the bounds of the functions $\varphi(\tau)$ and $c(z, \tau)$ exist and are equal when $\beta \to 1 + 0$ and $\beta \to 1 - 0$, i.e., the functions are continuous for $\beta = 1$. Evaluation of the indeterminacy leads to the following results for $\beta = 1$:

$$\varphi(\tau) = (1 + 2\tau)\,\Phi(\sqrt{\tau}) - 2\sqrt{\tau}/\sqrt{\pi}\,,$$

$$\frac{c(z,\tau)}{c^*} = \frac{4\sqrt{\tau}}{\sqrt{\pi}}\exp(-z^2)\,[1 - \sqrt{\pi}\,(z + \sqrt{\tau})\,\Phi(z + \sqrt{\tau})]\,, \quad z \ge 0\,, \quad \tau > 0\,. \tag{15}$$

Let us illustrate the results obtained. Figure 1 gives the time dependence of the concentration of the impurity at the matrix boundary $c(z = 0, \tau)/c^*$ ((12), (14) and (15)) for different values of the parameter β (8). It is clear that the presence of the solubility limit is insignificant when $\beta \ll 1$, since the impurity source has time to be exhausted before the impurity concentration in the matrix attains values comparable to c^* [6]. When $\beta \ge 1$, conversely, the source strength is fairly high and the solubility limit turns out to be a dominant factor, since it limits the arrival of the impurity from the source layer at the surface layer of the matrix [7].

Figure 2 gives the impurity concentration ((12) and (14)) as a function of the dimensionless penetration depth z for different values of the dimensionless time τ. The figure shows that the concentration profiles that are the solutions of the mathematical model (1), (10) are located between two curves corresponding to the well-known conditions at the source–matrix boundary: between the instantaneous source ($\tau \to \infty$) and the constant flux ($\tau \to 0$). Here the concentration profile is presented under the assumption that a constant impurity concentration is maintained at the matrix boundary [9].

We give an example of processing of experimental data with the use of the results given in the present work. As has been noted above, the integral activity of the sample's residue $N(x, t)$ and not the concentration of the impurity $c(x, t)$ is recorded in diffusion experiments carried out by the radioactive-isotope method [3]:

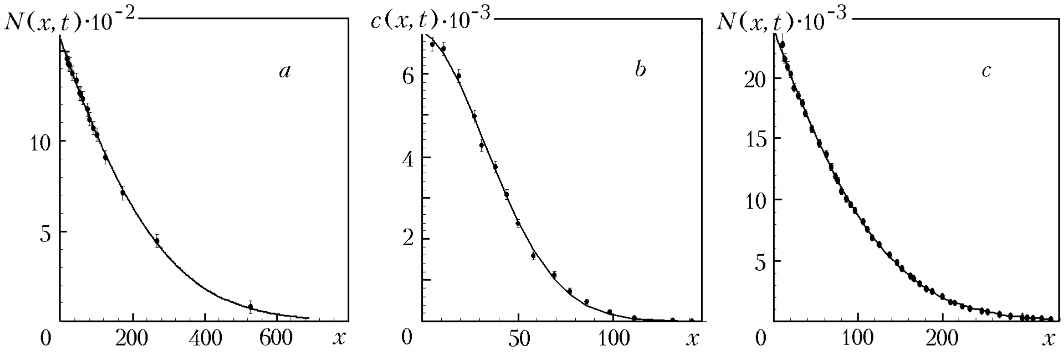


Fig. 3. Diffusion of ^{90}Y (a) and ^{63}Ni (b) in beryllium and of ^{59}Fe (c) in as-cast beryllium: a) T = 1220 and t_{hom} = 67.5, b) 900 and 492, and c) 990$^{\text{o}}$C and 30 h [a, c) processing according to expression (18); b) according to (13)].

$$N(x,t) = A \int_x^L c(x',t) \exp[-\mu(x'-x)]\,dx' . \tag{16}$$

In processing the experiments, we may assume that the sample thickness is infinite for a number of materials and the radiation absorption is slight at distances comparable to the depth of diffusion penetration of the impurity into the sample L_{d}, i.e., $L >> L_{\text{d}}$ and $\mu L_{\text{d}} << 1$. Then the expression for the integral activity (16) is simplified:

$$N(x,t) = A \int_x^{\infty} c(x',t)\,dx' , \quad x \ge 0 , \quad t > 0 . \tag{17}$$

Substituting the equations for the impurity concentration (12) and (13) into formula (17), for the integral activity we obtain

$$\frac{N(z,\tau)}{AQ_0} = \begin{cases} \operatorname{erfc}(z) + \dfrac{\exp(-z^2)}{y_1 - y_2}\,[y_2\Phi(z + y_1\sqrt{\tau}) - y_1\Phi(z - y_2\sqrt{\tau})] , & \beta > 1 ; \\ \operatorname{erfc}(z) + \dfrac{\exp(-z^2)}{\sqrt{1-\beta^2}}\,\operatorname{Im}[y_2\Phi(z + y_1\sqrt{\tau})] , & \beta < 1 , \quad z \ge 0 , \quad \tau > 0 . \end{cases} \tag{18}$$

In the case of strong radiation absorption by the matrix material on the diffusion length $\mu L_{\text{d}} >> 1$ from (16) we have

$$N(z,\tau) = \frac{A}{\mu}\,c(z,\tau) , \quad z \ge 0 , \quad \tau > 0 , \tag{19}$$

i.e., the integral activity of the sample's residue is in proportion to the concentration of the impurity [9].

Figure 3 gives experimental data on the diffusion of impurities in beryllium at different temperatures and calculated curves for the integral activity. The diffusion coefficients of the impurities D that have been calculated from these experimental data within the framework of the model presented in the work are given in Table 1. The values of the standard deviation of the calculated curve from the experimental one allow the conclusion that the mathematical model proposed in the present work makes it possible to achieve a more complete agreement between theory and experiment than the models used earlier and enables one to obtain more reliable data on the diffusion coefficients of impurities.

Table 2 gives the values of the parameters τ, β, and ϑ, calculated in processing the experiments illustrated above (Fig. 3). Based on the data obtained, we may draw conclusions on the influence of the solubility limit on the penetration of the impurity into the matrix during the experiment and the degree of dissociation of the source layer

TABLE 1. Comparison of the Results of Experimental Processing ($D\cdot10^9$, cm^2/sec, and $\sigma\cdot10^{-2}$, rel. units)

Isotopes of the impurity	T, °C	t_{hom}, h	Parameters	Model				
				Instantaneous source	Impurity concentration	Impurity flux	Present work	[9]
^{90}Y	1220	67.5	D	1.2 ± 0.2	2.1 ± 0.2	3.0 ± 0.4	1.8 ± 0.2	2.8 ± 0.3
			σ	0.0319	0.0180	0.0245	0.0169	—
^{59}Fe	990	30.0	D	0.32 ± 0.04	0.88 ± 0.08	1.12 ± 0.14	0.72 ± 0.06	0.068
			σ	7.88	6.19	9.01	5.73	—
^{63}Ni	990	492.0	D	0.0034 ± 0.0003	0.004 ± 0.003	0.013 ± 0.004	0.0042 ± 0.0002	0.0042 ± 0.0003
			σ	2.3	25.6	39.4	1.65	—

TABLE 2. Values of the Parameters τ, β, and ϑ for Three Experiments

Isotope	T, °C	τ	β	ϑ
^{90}Y	1220	$(5.7 \pm 0.4)\cdot10^2$	$(1.7 \pm 0.2)\cdot10^1$	48 ± 4
^{59}Fe	990	$(3.4 \pm 0.3)\cdot10^2$	$(9.2 \pm 0.2)\cdot10^1$	58 ± 4
^{63}Ni	900	8.9 ± 0.3	$(1.6 \pm 0.2)\cdot10^{-1}$	1.0 ± 0.3

over the period of homogenization. It is clear that the influence of the solubility limit of the impurity is the largest in diffusion of ^{90}Y, whereas the solubility limit for the diffusion of ^{63}Ni is of little significance (see Fig. 1). The degree of dissociation of the source $\vartheta = Q(t)/Q_0$ has been determined from formula (4). It is noteworthy that, in diffusion of ^{90}Y and ^{59}Fe, the degree of dissociation of the impurity source is small despite the significant dimensionless homogenization time τ ($\tau >> 1$), since the presence of the solubility limit in the above isotopes exerts a substantial influence on the process of migration ($\beta >> 1$), keeping the impurity from penetrating into the matrix from the source layer.

Thus, in this work, we have obtained for the first time the solution of the problem on diffusion from a depleting source with allowance for the solubility limit. The model can be recommended for the processing of results throughout the range of the homogenization time and for any rate of depletion of the source. The use of the solutions found has allowed refinement of the data for the diffusion of impurities in beryllium.

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NOTATION

A, constant relating the activity of the sample to the concentration of the diffusant, pulses/(cm^2·sec); B, constant, K; c, concentration of the impurity, atoms/cm^3; D, diffusion coefficient, cm^2/sec; D_0, preexponential factor, cm^2/sec; L, sample's length, cm; L_{d}, characteristic depth of diffusion penetration of the impurity into the sample, cm; $N(x)$, integral activity of the sample's residue, pulses/sec; T, absolute temperature, K; t, time, sec; Q_0, diffusion-source strength, atoms/cm^2; x, coordinate of the removed layer, μm; x', integration variable, μm; z, dimensionless coordinate of the removed layer; α^{-1}, characteristic activation time of the impurity atoms; β, dimensionless parameter; φ, dimensionless flux; μ, linear coefficient of absorption of the radiative radiation of the isotope, cm^{-1}; τ, dimensionless homogenization time; ϑ, degree of dissociation of the source, %. Subscripts: d, diffusion; hom, homogenization; exp, experiment; calc, calculation.

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