NON-METAL DIFFUSION COEFFICIENTS FOR THE Ta–C AND Ta–N SYSTEMS

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Abstract—The diffusivity of carbon in tantalum and tantalum carbides was investigated in the temperature range 1700°C–2200°C, and that of nitrogen in tantalum and tantalum nitrides between 1700°C and 1950°C. The concentration-independent diffusion coefficients were obtained in all phases by investigating the enhanced layer growth in wedge-shaped specimens. In the nonmetal-rich phases having a broad homogeneity range, i.e. in δ-TaC$_{1-x}$, β-Ta$_2$C$_{1-x}$ and β-Ta$_2$N$_{1-x}$, also the concentration-dependent diffusion coefficients were calculated and compared with the concentration-independent diffusion coefficients. The calculation of the concentration-dependent diffusion coefficients was performed by fitting a modified error function on the measured concentration profiles assuming the nonmetal diffusivity being an exponential function of the nonmetal concentration. Strong dependence of the diffusion coefficient on the nonmetal concentration was found for the δ-TaC$_{1-x}$ phase, whereas the nonmetal diffusion coefficients were nearly concentration-independent for β-Ta$_2$C$_{1-x}$ and β-Ta$_2$N$_{1-x}$. © 1998 Acta Metallurgica Inc.

1. INTRODUCTION

The tantalum carbides and tantalum nitrides are important materials for high-temperature applications. The melting point of the δ-TaC$_{1-x}$ phase, which lies near 4000°C [1], is one of the highest among any material. In the temperature range 1700°C–2150°C, two further carbide phases are known: β-Ta$_2$C$_{1-x}$ and ζ-Ta$_4$C$_{3-x}$. At 2175°C, only δ-TaC$_{1-x}$, β-Ta$_2$C$_{1-x}$ and α-Ta(C) are stable — the ζ-Ta$_4$C$_{3-x}$ phase decomposes at 2160°C [2]. The homogeneity range of ζ-Ta$_4$C$_{3-x}$ is very narrow in the whole temperature range as it was established recently by electron probe microanalysis (EPMA/WDS) [2]. In the Ta–N system, four phases are stable up to 1860°C: ε-TaN, δ-TaN$_{1-x}$, β-Ta$_2$N$_{1-x}$ and α-Ta(N). Above 1860°C, the ε-TaN phase does not form, and a steep increase of the homogeneity range in the δ-TaN$_{1-x}$ phase occurs. The revised phase diagrams of the Ta–C and Ta–N systems are given in [2].

Two approaches were applied for evaluation of nonmetal diffusion coefficients in tantalum, tantalum carbides and tantalum nitrides. First, the diffusion coefficients were calculated in all phases from layer growth assuming that the diffusivity is independent of the nonmetal concentration within each phase. Second, in the δ-TaC$_{1-x}$, β-Ta$_2$C$_{1-x}$ and β-Ta$_2$N$_{1-x}$ phases the concentration-dependent diffusion coefficients were obtained from the concentration profiles measured by EPMA/WDS. In both

cases, it was assumed that the diffusivity of the nonmetal is much larger that the diffusivity of the metal atoms. Therefore the diffusion process could be described as the diffusion of the nonmetal in a rigid metal grid.

2. EXPERIMENTAL

Reaction diffusion samples were prepared both as planar diffusion couples and as wedge-shaped diffusion couples. Tantalum sheets and wedges were brought into contact either with graphite or nitrogen, respectively, and annealed. Details on the sample preparation are given elsewhere [2]. Plane-sheet samples were used mainly for measurement of the concentration profiles in the main diffusion direction (perpendicular to the sample surface), from which the concentration-dependent diffusion coefficients were calculated. The concentration profiles were measured by use of EPMA/WDS in the step scan mode [3]. The wedge-shaped samples were used to investigate the homogeneity ranges of individual phases and to calculate the diffusion coefficients from the layer growth.

3. CALCULATION OF DIFFUSION COEFFICIENTS

The concentration-independent diffusion coefficients can be obtained from layer growth only if the finite sample geometry is employed that causes an enhancement of the layer growth, i.e. a deviation from the parabolic layer growth [4]. In a recent study [5], we showed that the wedge-shaped sample geometry is especially advantageous for investi-

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gation of the diffusion kinetics because a wedge contains apparently a continuous series of samples with different thickness. In such samples, a systematic broadening of layers towards the tip of the wedge is usually observed, which can be used to adjust the diffusion coefficient in the central phase. This is impossible for infinite samples [6]. The computing procedure was performed iteratively by fitting the calculated positions of phase boundaries to the measured phase boundary positions. In the first step, the diffusion coefficients in outer phases except those in the metal (α-Ta(C) and α-Ta(N)) were varied to arrive at the best match in the calculated and observed phase boundary positions. This was done for a single (large) sample thickness with an arbitrary diffusion coefficient in the metal. In the second step, the positions of phase boundaries were calculated using the above set of diffusion coefficients for different sample thickness and compared with the observed phase band structure. If the starting diffusion coefficient in the metal was lower than the correct one, the calculated layer growth enhancement was lower than the observed layer growth enhancement and vice versa. Accordingly, the diffusion coefficient in the metal phase was corrected, and these two steps were repeated until the differences between the calculated and the observed phase boundary positions were lower than a desirable limit (e.g. 1–5 μm, depending on the thickness of the phases). The calculation of the phase boundary positions was performed using the forward finite differences (FFD) algorithm [7]. The procedure for the refinement of diffusion coefficients employed the "regula falsi" routine, and therefore the refinement was very stable despite the initial diffusion coefficient used for the metal. If the initial diffusion coefficient was too high (one or two orders of magnitude above the right value), the metal disappeared usually from the core in the simulated microstructure of the wedge. On the contrary, too low initial diffusion coefficient of non-metal in the central phase (approximately two orders of magnitude below the right value) caused a decrease of the calculated layer thickness towards the tip of the wedge.

The second approach was applied to calculate the diffusion coefficients from concentration profiles by approximating the measured concentration profiles with a modified error function [8]:

tities $y(0)$, $y(\xi)$, y, y' and y'' are expressed in the Boltzmann–Matano variable, $y = x/(2\sqrt{t})$, where x is the spatial co-ordinate and t the diffusion time. The concentration dependence of the diffusion coefficient was described by the well-established exponential function [9] with two free parameters a and D_0:

$$D(c) = D_0 \exp[a(c^+ - c)] \qquad (2)$$

4. RESULTS

4.1. Diffusion coefficients of carbon in tantalum and tantalum carbides

The homogeneity ranges of carbon in individual phases, which were used for calculation of the concentration-independent diffusion coefficients from layer growth, are listed in Table 1 in terms of the maximum and minimum concentrations. All concentrations are expressed in mol C/cm^3 to take the crystal lattice expansion (the volume changes) occurring during the diffusion process into account. The concentration-independent diffusion coefficients of carbon in tantalum (α-Ta(C)) and tantalum carbides are represented by solid symbols in Fig. 1. In δ-TaC$_{1-x}$ and, in particular cases also in β-Ta$_2$C$_{1-x}$, the diffusion coefficients were additionally calculated from the measured concentration profiles (equation (1)) assuming the exponential decrease of carbon diffusivity with increasing carbon concentration (equation (2)). The results of the concentration profile fitting are plotted with open symbols in Fig. 1. It was found that carbon diffuses approximately six times faster in the sub-stoichiometric δ-TaC$_{0.77}$ ($c = 0.0588$ mol C/cm^3) than in the stoichiometric tantalum carbide (δ-TaC$_{1.00}$, $c = 0.0747$ mol C/cm^3). The concentration-independent diffusion coefficients calculated from layer growth lie in-between the diffusion coefficients obtained for the minimum and the maximum carbon concentrations. The strong dependence of the diffusion coefficient in δ-TaC$_{1-x}$ on the carbon concentration is well documented on the shape of the concentration profile (Fig. 2). A simple error function, (i.e. equation (1) with concentration-independent diffusion coefficient) did not approximate the measured concentrations correctly; a substantially better match was obtained taking the concen-

$$c(y) = (c^- - c^+)\frac{\displaystyle\int_{y(0)}^{y}(1/D(y'))\exp\left(-\int_{y(0)}^{y'}(2y''\,\mathrm{d}y'')/D(y'')\right)\mathrm{d}y'}{\displaystyle\int_{y(0)}^{y(\xi)}(1/D(y'))\exp\left(-\int_{y(0)}^{y'}(2y''\,\mathrm{d}y'')/D(y'')\right)\mathrm{d}y'} + c^+, \qquad (1)$$

where $y(0)$ and $y(\xi)$ are the beginning and the end of the respective concentration profile, c^+ and c^- are the corresponding concentrations. The quan-

tration dependence of the diffusion coefficient into account. On the contrary, no significant dependence of the diffusion coefficients on the carbon concen-

Table 1. Summary of homogeneity ranges in the Ta–C system between 1700°C and 2200°C used for simulation of the layer growth. The ζ-Ta$_4$C$_{3-x}$ phase did not form at 2175°C. All concentrations are given in mol C/cm^3

T (°C)	Phase							
	δ-TaC$_{1-x}$		ζ-Ta$_4$C$_{3-x}$		β-Ta$_2$C$_{1-x}$		α-Ta(C)	
	c(max)	c(min)	c(max)	c(min)	c(max)	c(min)	c(max)	c(min)
1743	0.0747	0.0588	0.0501	0.0491	0.0476	0.0447	0.0003	0
1915	0.0747	0.0571	0.0501	0.0491	0.0476	0.0447	0.0006	0
2088	0.0747	0.0555	0.0501	0.0491	0.0476	0.0386	0.0011	0
2147	0.0747	0.0549	0.0501	0.0491	0.0476	0.0366	0.0013	0
2175	0.0747	0.0544	–	–	0.0476	0.0349	0.0014	0

tration was detected in the β-Ta$_2$C$_{1-x}$ phase. The reason is the narrow homogeneity range of the β-Ta$_2$C$_{1-x}$ phase in comparison with the δ-TaC$_{1-x}$ phase.

The dependence of the diffusion coefficient on the temperature and nonmetal concentration in δ-TaC$_{1-x}$ can be expressed by the exponential surface:

$$D(T, c) = (0.023 \pm 0.010) \text{ cm}^2 \text{ s}^{-1}$$
$$\times \exp\left(-\frac{(3.6 \pm 0.1) \text{ eV}}{k_B T}\right)$$
$$\times \exp[(105 \pm 2) \text{ cm}^3 \text{ mol}^{-1}$$
$$\times (c^+ - c)], \quad (3)$$

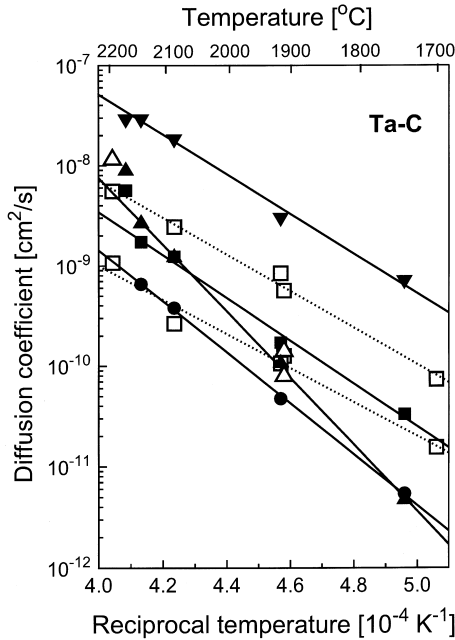with $c^+ = 0.0747$ mol C/cm^3, taking the Arrhenius and concentration dependence (equation (2)) of the diffusion coefficient into account. For the atomic fraction, x in TaC$_{1-x}$, the last term in equation (3) takes the form of:

$$\exp[(7.1 \pm 0.1)x]$$

The concentration dependence of the diffusion coefficient is, in fact, expressed by the pre-exponential factor D_0 in the Arrhenius dependence

$$D = D_0 \exp\left(-\frac{E}{k_B T}\right) \quad (4)$$

This can be explained by a simple model of diffusion through the vacant atomic positions (e.g. [10]). According to this model, the pre-exponential factor consists of several coefficients, one of them being the jump probability of the diffusing species. With decreasing concentration of the in-diffusing nonmetal the jump probability and consequently the pre-exponential factor increase, whereas the activation energy remains constant.

The Arrhenius plots of the carbon diffusion coefficients for tantalum and tantalum carbides (Fig. 1) yielded the activation energies and pre-exponential factors, which are given in Table 2 and 3. Table 2 contains the activation energies and pre-exponential
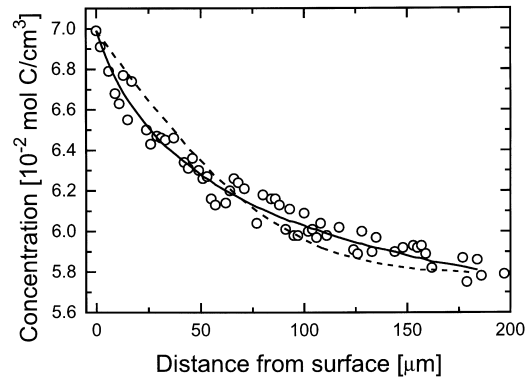

Fig. 1. Arrhenius plots for diffusion coefficients of carbon in the Ta–C system ($\square$: δ-TaC$_{1-x}$, $\bigcirc$: ζ-Ta$_4$C$_{3-x}$, $\triangle$: β-Ta$_2$C$_{1-x}$ and $\triangledown$: α-Ta(C)). Solid symbols are for the concentration-independent diffusion coefficients calculated from layer growth; open symbols are for the concentration-dependent diffusion coefficients obtained from the concentration profile fitting. The Arrhenius plots for the concentration-dependent diffusion coefficient, in TaC (low diffusion coefficients) and in TaC$_{0.77}$ (high diffusion coefficients), are plotted by dotted lines.

Fig. 2. The measured concentration profile in δ-TaC$_{1-x}$ (open circles) was approximated by an error function assuming a concentration-independent diffusion coefficient (broken line) and by the modified error function (equation (1)) with the concentration-dependent diffusion coefficient (solid line). The plain error function could not fit the measured concentration profile correctly as the observed concentration gradient is very high at high carbon concentrations. This is the first indicator for the concentration dependence of the diffusion coefficient.

Table 2. Activation energies and pre-exponential factors for the diffusion of carbon in δ-TaC$_{1-x}$ calculated from the diffusion coefficients obtained by profile fitting

Phase	δ-TaC $c = 0.0747$ mol/cm^3		δ-TaC$_{0.77}$ $c = 0.0588$ mol/cm^3	
E (eV)	3.6 $\pm$ 0.1		3.6 $\pm$ 0.2	
D_0 (cm^2/s)	0.023	$+0.016$ -0.009	0.14	$+0.25$ -0.09

Table 3. Activation energies and pre-exponential factors for the carbon diffusion in tantalum and tantalum carbides calculated from the diffusion coefficients obtained from layer growth. In β-Ta$_2$C$_{1-x}$, the activation energy and the pre-exponential factor were calculated from diffusion coefficients as obtained from both layer growth and profile fitting

Phase	δ-TaC$_{1-x}$		ζ-Ta$_4$C$_{3-x}$		β-Ta$_2$C$_{1-x}$		α-Ta(C)	
E (eV)	4.2 $\pm$ 0.2		5.0 $\pm$ 0.1		6.5 $\pm$ 0.3		3.9 $\pm$ 0.2	
D_0 (cm^2/s)	1.1	$+2.8$ -0.8	20	$+11$ -7	1.1×10^5	$+4.2$ -0.9	4.2	$+6.1$ -2.5

factors calculated from the maximum and the minimum diffusion coefficients (as obtained by the concentration profile fitting). The activation energies and the pre-exponential factors calculated from the concentration-independent diffusion coefficients, which were obtained mainly from layer growth but also from concentration profile fitting in the β-Ta$_2$C$_{1-x}$ phase, are given in Table 3.

4.2. Diffusion coefficients of nitrogen in tantalum and tantalum nitrides

The concentration-independent diffusion coefficients of nitrogen in tantalum and tantalum nitrides obtained from layer growth are given in Fig. 3 (solid symbols). The maximum and minimum nitrogen concentrations expressed in mol N/cm^3 are given in Table 4. As the diffusion coefficients for δ-TaN (large solid squares) showed a curvature above 1900°C, the maximum nitrogen concentration in δ-TaN was also refined to improve the linearity of the Arrhenius plot. Table 2 contains the refined maximum nitrogen concentrations in δ-TaN; the corresponding diffusion coefficients are plotted by small solid squares in Fig. 3. The capability of this approach is discussed in Section 5.3.

Because of the narrow homogeneity ranges in ε-TaN, δ-TaN and α-Ta(N), the concentration profiles in these phases were not appropriate for calculation of the diffusion coefficients. Only the diffusion coefficients in β-Ta$_2$N could be obtained from the measured concentration profiles (open triangles in Fig. 3). By concentration profile fitting no significant dependence of the diffusion coefficients on the nonmetal concentration was found in the β-Ta$_2$N$_{1-x}$ phase. This was also confirmed by the excellent agreement with the diffusion coefficients obtained from layer growth (solid triangles in Fig. 3). The diffusion coefficients of nitrogen in β-Ta$_2$N$_{1-x}$ given here are quite reliable, as they were verified by using two independent approaches.

The activation energies and pre-exponential factors calculated for individual phases from the temperature dependence of the diffusion coefficient are listed in Table 5. Whereas the activation energy could be determined with a sufficient accuracy, the estimated standard deviation of the pre-exponential factor is comparable with its nominal value. The reason is the narrow temperature range in which the diffusion coefficients could be investigated. Consequently, the pre-exponential factor, which is obtained by extrapolating the Arrhenius dependence to zero, cannot be determined with a higher precision.

5. DISCUSSION

5.1. Problems in the calculation of diffusion coefficients

The accuracy of diffusion coefficients calculated from the layer growth enhancement is influenced principally by the precision in reading the thickness of individual layers and by the precision of the homogeneity range measurement. The precision of the layer thickness measurement is rather high; the relative error is below 1% in the majority of cases.

Table 4. Summary of homogeneity ranges in the Ta–N system between 1810°C and 1955°C used for simulation of the layer growth. The ε-TaN phase did not form above 1869°C. All concentrations are given in mol N/cm^3

T (°C)	N$_2$ pressure (bar)	Phase							
		ε-TaN		δ-TaN$_{1-x}$		β-Ta$_2$N$_{1-x}$		α-Ta(N)	
		c(max)	c(min)	c(max)	c(min)	c(max)	c(min)	c(max)	c(min)
1812	25	0.1470	0.1455	0.0648	0.0618	0.0424	0.0274	0.0110	0
1869	25	–	–	0.0684	0.0614	0.0424	0.0273	0.0115	0
1903	25	–	–	0.0720	0.0611	0.0424	0.0272	0.0119	0
1955	10	–	–	0.0650	0.0607	0.0424	0.0271	0.0124	0

Table 5. Activation energies and pre-exponential factors for the nitrogen diffusion in tantalum and tantalum nitrides calculated from the diffusion coefficients obtained from layer growth. In β-Ta$_2$N$_{1-x}$, also the diffusion coefficients obtained from profile fitting were taken into calculation

Phase	δ-TaN$_{1-x}$	β-Ta$_2$N$_{1-x}$		α-Ta(N)	
E (eV)	~3.6	3.30 ± 0.09		2.8 ± 0.5	
D_0 (cm^2/s)	~3.4	1.4	$+0.9$ -0.6	0.03	$+0.40$ -0.02

Larger errors are to be considered only if the layers are extremely narrow or if the interfaces are uneven. A special case of the latter is a dominant diffusion along the grain boundaries, which occurs at low temperatures or if the in-diffusing species penetrates into the host material through a porous surface.

The accuracy of the homogeneity range measurement influences strongly the accuracy of the diffusion coefficients calculated from layer growth. This is because that the calculated positions of phase boundaries are proportional to the respective width of the homogeneity range multiplied by the diffusion coefficient [5, 6], and therefore the inaccuracy in the homogeneity range measurement is compensated through the variation of the respective diffusion coefficient. This phenomenon is crucial for δ-TaN$_{1-x}$.

5.2. The Ta–C system

If the diffusion coefficient depends on the concentration of in-diffusing species and if the width of

the homogeneity range varies with temperature as observed in δ-TaC$_{1-x}$, a careful interpretation of the concentration-independent diffusion coefficients (as obtained from layer growth) is necessary. The dependence of diffusion coefficient on the temperature T and the nonmetal concentration c can be expressed by an exponential surface given by the generalized form of equation (3):

$$D(T, c) = D_0 \exp[a(c^+ - c)]\exp\left(-\frac{E}{k_B T}\right), \quad (5)$$

where D_0 is the pre-exponential factor, a the coefficient describing the dependence of the diffusion coefficient on the concentration, c^+ the maximum concentration, E the activation energy and k_B the Boltzmann constant. The concentration-independent diffusion coefficient is, however, calculated for the mean concentration within the respective phase. If the mean concentration in a phase shifts with temperature, the Arrhenius plot for the concentration-independent diffusion coefficients does not represent an iso-concentration cut in the exponential plane (equation (5)). This causes a systematic deviation in the calculated activation energy.

As the mean carbon concentration in δ-TaC$_{1-x}$ shifts to lower values [2] (see also Table 1) and as the concentration-independent diffusion coefficient becomes higher with decreasing carbon concentration (equation (3)), an apparent increase of the activation energy occurs. A lower activation energy was indeed obtained from the concentration-dependent diffusion coefficients, which were re-calculated for equal concentrations. Such a discrepancy in the results can also be found in the literature. Resnick *et al.* [11] reported an activation energy for carbon diffusion in δ-TaC of 3.7 eV as investigated by carburizing TaC$_{0.9}$ and by evaluating the measured concentration profile. In this case, the mean carbon concentration was kept constant, and therefore the activation energy corresponded to an iso-concentration cut in the exponential plane (equation (3)). On the contrary, the activation energy given by Brizes [12] and Fromm *et al.* [13] was 3.9 eV and 4.3 eV, respectively. This was obtained from layer growth in multiphase diffusion experiments and therefore the activation energy was higher as it corresponds to a non-identical mean composition. Both sets of data correspond excellently with our findings.

This effect is striking only if the diffusion coefficient depends strongly on the nonmetal concen-
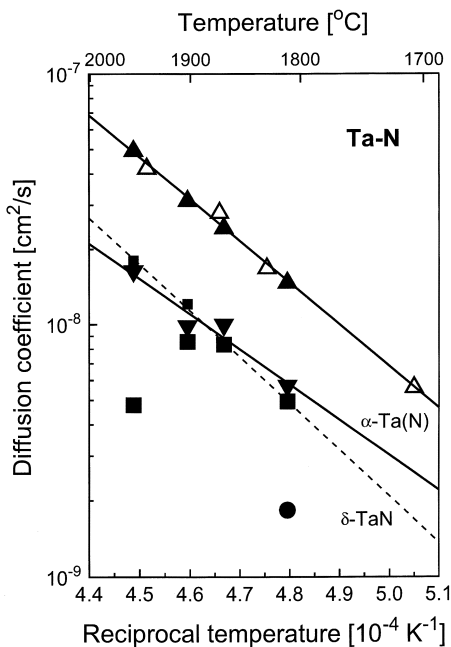

Fig. 3. Arrhenius plots for diffusion coefficients of nitrogen in the Ta–N system (○: ε-TaN, □: δ-TaN$_{1-x}$, △: β-Ta$_2$N$_{1-x}$ and ▽: α-Ta(N)). Solid symbols denote the concentration-independent diffusion coefficients calculated from layer growth, open symbols the concentration-dependent diffusion coefficients obtained from profile fitting. Small squares represent the diffusion coefficients in δ-TaN corresponding to the refined surface concentration.

tration and if the average concentration changes with the temperature. In the phases, in which no concentration dependence of the diffusion coefficient was observed (e.g., in the β-Ta$_2$C$_{1-x}$ and β-Ta$_2$N$_{1-x}$ phases), this effect vanishes. Therefore, it is useful to compare the diffusion coefficients obtained from layer growth with the diffusion coefficients calculated from concentration profiles (and re-calculated to the same concentrations). Unfortunately, the combined approach can only be applied in phases with a broad homogeneity range. In phases having a narrow homogeneity range, the calculation of diffusion coefficients from the concentration profiles is nearly impossible.

5.3. The Ta–N system

With respect to the curvature in the Arrhenius plot for δ-TaN presented in Section 4.2, the accuracy of the nitrogen diffusion coefficients in δ-TaN is disputable. We supposed that this is due to the limited accuracy of the concentration measurement. In the samples annealed below 1860°C, where the ε-TaN phase is still stable (and grows at the sample surface), the nitrogen-rich boundary of δ-TaN$_{1-x}$ was well accessible for concentration measurements, whereas in samples without the ε-TaN layer an accurate measurement of the maximum nitrogen concentration was impossible. This difficulty was intensified by the high increase of the maximum nitrogen concentration with increasing temperature and nitrogen pressure [2]. In addition, a very steep concentration gradient was observed near the sample surface in δ-TaN, which also decreases the accuracy of the concentration measurements because of a limited lateral resolution of EPMA [3]. The high concentration gradient at the sample surface is, moreover, an indicator for the concentration dependence of the diffusion coefficient [8]. Unfortunately, the concentration profiles measured in δ-TaN were not suitable for calculation of the diffusion coefficients, and therefore this hypothesis could not be confirmed yet.

The observed deviation of the diffusion coefficient (the non-linearity of the Arrhenius plot) for δ-TaN indicated clearly the problems with the maximum concentration measurements in the Ta–N system. However, as the surface concentration and the diffusion coefficient in the surface phase are highly correlating parameters [5], it is impossible to calculate both concurrently. In the standard course of calculation, the diffusion coefficients are refined by using the known limit concentrations. Oppositely, the surface concentration can only be refined if the diffusion coefficient in the outer phase is known. For instance, by extrapolating the Arrhenius dependence from lower temperatures, where no problems with the concentration measurements in δ-TaN$_{1-x}$ occurred, to higher temperatures. In this particular case we assumed that two diffusion coefficients calculated at low temperatures (1812°C and 1869°C)

are rather reliable and therefore they were used to extrapolate the Arrhenius dependence to higher temperatures. Then the surface nitrogen concentrations were modified to arrive at the desired (extrapolated) diffusion coefficients.

However, it is extremely difficult to prepare δ-TaN$_{1-x}$ in a broad temperature range, and therefore only two data points were available for this procedure. Thus, the maximum nitrogen concentrations at higher temperatures, which were obtained by using the extrapolation of diffusion coefficients, are not really reliable. The main gain of this approach is that a discrepancy in the maximum measured concentrations was indicated and that a better value of the surface nitrogen concentration could be estimated that should be compared with an improved maximum nitrogen concentration obtained experimentally.

Still, the product of the homogeneity range width and the diffusion coefficient, which is proportional to the diffusion flow determining the positions of the phase boundaries [5], was determined with a sufficient accuracy at all temperatures because the diffusion coefficient in the neighboring β-Ta$_2$N$_{1-x}$ phase agrees well with that calculated from the concentration profiles. This result has an important methodological consequence. A poor accuracy of one diffusion coefficient calculated from layer growth does not necessarily influence the accuracy of the diffusion coefficients in other phases.

6. CONCLUSIONS

Diffusion coefficients of carbon and nitrogen in tantalum carbides and nitrides were calculated using a combined refinement. This approach yielded two independent sets of diffusion coefficients that were compared. One set was calculated from concentration profiles, the other one from the layer growth enhancement in wedge-shaped diffusion couples. In δ-TaC$_{1-x}$, the concentration-independent diffusion coefficients lie between the maximum and the minimum diffusion coefficients obtained from the concentration profile fitting, but a higher activation energy was evaluated from the concentration-independent diffusion coefficients than from the concentration-dependent diffusion coefficients. The reason is a strong decrease of the diffusion coefficient with increasing carbon concentration, which was confirmed by the concentration profile fitting. The diffusion coefficients obtained using different calculi were identical if the diffusivity of nonmetal did not depend on the concentration. This was the case in the β-Ta$_2$C$_{1-x}$ and β-Ta$_2$N$_{1-x}$ phases.

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