Novel General Solution for the Analysis of a Multicomponent Interdiffusion Couple

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Abstract In this paper, we draw attention to the investigation of the novel exact solution [1.Scripta Mat. 210:114430; M.A. Dayananda, in JPED, this issue, (2022);] that is applicable to a multicomponent (n-component) interdiffusion couple where the interdiffusion matrix may change with alloy composition. In the derivation of this solution the interdiffusion flux J_j of a component j is related to (n-1) independent composition gradients for an isothermal, diffusion couple using the well-known continuity equation. Novel exact expressions are then derived for all of the interdiffusion coefficients, $\tilde{D}_{ij}^n$ ($i, j = 1, 2, \dots n-1$), where the partial derivatives of the product $J_j(y - y_0)$ with respect to composition C_i (y_0 is the Matano plane) are used. In this paper, it is shown that the novel solution leads to a computational procedure similar to the Boltzmann-Matano analysis. Note that the derivatives $\partial(J_j(y - y_0))/\partial C_i, i,j = 1, \dots, n-1$ (that are required for the solution) can only be calculated along the diffusion path and therefore, for $n > 2$, a single couple will not be enough to calculate all of them correctly.

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1 Introduction

In 1894, Boltzmann[3] calculated the (single) interdiffusion coefficient analytically for a binary alloy. In 1933, Matano[4] extended Boltzmann's concept by introducing a special plane named the "Matano plane" (plane (x_0) across which equal amounts of atoms diffuse to the left and right hand sides of the plane). This became known as the Boltzmann-Matano (BM) method. Then, Sauer and Freise[5] examined the interdiffusion coefficients for the binary alloy using the interrelations between the atomic fluxes in such a way that the position of the Matano plane was not needed. The resulting method is the modification of the BM method, and it is widely known as the Sauer-Freise (SF) method. Then Hall[6] revised the BM method and claimed that, at the ends of the composition profiles, the resulting method gives more accurate outcomes than the BM and SF methods. The method introduced by Hall is known as the Hall method (HM). Many authors have worked on all these methods from different points of view[7–10] in applications to binary interdiffusion problem (Zhang and Zhao,[11] Kass and Keeffe,[12] Mittemeijer and Rozendaal,[13] Garcia et al.[14] to name a few). In 2013, Belova et al.[15] developed simultaneous measurement of interdiffusion and tracer diffusion coefficients computationally for binary and multicomponent alloys. Furthermore, in 2015, Ahmed et al.[16,17] studied interdiffusion coefficients for binary alloys using the explicit finite difference method. In their study, they used BM, SF, HM and a newly developed method treated as the extended Hall

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method to obtain the interdiffusion coefficients. There, it was found that the original HM does not improve the accuracy of the interdiffusion coefficients at the ends of the composition profiles.

In 1956–1959, Fujita and Gosting[18–20] analysed diffusion in ternary metallic systems using an exact solution of two simultaneous diffusion equations (first presented in Gupta and Cooper[21] and Krishtal et al.[22]) They derived a direct exact analysis for extracting the four interdiffusion coefficients from the fitting parameters of the closed form solution into the experimentally obtained interdiffusion profiles. This analysis consists of obtaining the elements of the inverse interdiffusion matrix and then taking the inverse of this matrix. The resulting equations consist of a set of the second order polynomial equations. The problem with this approach is that it is difficult to extend it to quaternary alloys and almost impossible for the quinary and higher alloys. Very recently, this problem was successfully overcome in.[23–25]

In 1965, Dayanada and Grace[26] worked on ternary diffusion in CuZnMn alloys, whereas Ziebold[27] worked on CuAgAu metallic alloys. In 1985, Malik and Bergner[28] investigated interdiffusion experiment in the ternary system and calculated the constant interdiffusion matrix using various methods. Further, a theoretical overview of ternary diffusion was developed by Vrentas and Vrentas.[29] A new method was developed by Dayananda and Sohn[30] for obtaining constant interdiffusion coefficients in a three-component system. There the authors used experimentally obtained composition profiles to determine the interdiffusion coefficients using a single diffusion couple for CuNiZn and Fe-NiAl alloys and two couples for the NiCrAl alloy. For comparison, in,[31] composition-independent interdiffusion coefficients were determined from composition profiles using several other techniques. In,[32] Day et al. analysed various ternary metallic systems using a single diffusion couple making use of MultiDiFlux software that was developed for calculating ternary interdiffusion coefficients. In general, MultiDiFlux software is used for the analysis of a single ternary diffusion couple and, possibly, for two couples. Recently, Dayananda[33] investigated interdiffusion coefficients using a single couple in ternary diffusion from diffusion constraints at the Matano plane. In 2002, Bouchet and Mevrel[34] developed a numerical inverse method for obtaining the composition-dependent ternary interdiffusion coefficients from a single diffusion couple.

In 1966–1969, Kim[35,36] investigated interdiffusion in the four-component system by extending the work of Fujita and Gosting.[18,37] They derived a solution to the differential equations using a similar, direct method as used in[18] and obtained the general solution. Utilising an expression from,[35] Kim[36] investigated the combined use of several experimental techniques for obtaining constant interdiffusion matrix in the quaternary system. Kim[38] investigated gravitational stability of diffusion in the four-component liquid system using those mathematical expressions. In[39–44] a square root diffusivity method (SQRD) was developed and applied to various binary, ternary and quaternary alloys to obtain constant interdiffusion matrices. In 2006–2007, Kulkarni et al.,[45,46] investigated a general method for quaternary alloys for the case of CuNiZnMn couples. They used the transfer matrix method (TMM) for obtaining the constant interdiffusion coefficients within the available composition range. The calculated interdiffusion matrix was then used for the generation of composition profiles by TMM followed by comparison with the original profiles. In 2020, Verma et al.[47] investigated the quaternary FeNiCoCr alloy experimentally using a body-diagonal (BD) diffusion couple.[48] In a BD couple all independent concentration differences (at the terminal points) are equal, except for being positive or negative. The advantage of the BDs is that they can be represented with a simple vector notation.

High entropy alloys (HEAs) are a class of multicomponent alloys fabricated with equal or near equal quantities of five or more principal elements. HEAs are usually defined as having four core effects: high entropy, sluggish diffusion, severe lattice distortion and cocktail effects.[49,50] Among these four core effects, sluggish diffusion makes HEAs very competitive for high-temperature strength, impressive high-temperature structural stability, and the formation of beneficial nanostructures. Tsai et al.[51] investigated self-diffusion phenomena of HEAs for the first time for the CoCrFeMnNi alloy. A quasi-binary approach was used to investigate the composition profiles. Zhang et al.[52] studied sluggish diffusion in AlCoCrFeNi and CoCrFeMnNi alloys using the CALPHAD approach. Later, Beke and Erdelyi[53] investigated the composition dependent interdiffusion coefficients of CoCrFeMnNi alloys using semi-empirical rules. In[54] diffusion in the fcc AlCoCrFeNi high entropy alloy was analysed experimentally as well as numerically for several diffusion couples. In that study, tracer diffusion coefficients were determined using the obtained composition profiles from the interdiffusion experiments and fitting method, and results were compared with the data of.[51] Recently, Paul et al.[55] studied the diffusion kinetics behaviour of CoCrFeMnNi HEAs. In that study, various random alloy models were used. Results for all three models showed good agreement with the experimental study. Afikuzzaman et al.[56] studied CoCrFeMnNi HEAs numerically using constant as well as a composition-dependent interdiffusion matrix for several diffusion couples mainly quasi-binary and quasi-ternary. The composition-dependent interdiffusion matrix was calculated using the Darken and Manning theoretical

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formalisms. The obtained composition profiles showed good agreement with the composition profiles obtained experimentally in the previous studies.[51,54]

In[1,2] the novel exact solution that is applicable to a multicomponent (n-component) interdiffusion couple was derived. It was assumed that the interdiffusion matrix may change with composition. In the derivation of this solution, using the continuity equation, the interdiffusion flux J_j of a component j is related to (n-1) independent composition gradients. In the present paper, this solution is tested numerically for the use in binary diffusion where the diffusion coefficient can be a linear or quadratic function of composition. It is tested for the application to the ternary diffusion couple(s) as well. The detailed algorithm for this application is provided. It is shown that the novel solution leads to a computational procedure similar to the Boltzmann-Matano analysis.

1.1 Theory

In[1,2] a novel solution to the multicomponent interdiffusion problem was derived. The final relations for the components of the matrix of the interdiffusion coefficients $\left(\tilde{D}_{ij}^n\right)$ are given as follows. For the diagonal terms, we have:

$$\tilde{D}_{ii}^n = \frac{(x - x_0)^2}{2t} - \frac{\partial\{\tilde{J}_i(x - x_0)\}}{\partial C_i}, \; i = 1, \ldots, n - 1. \quad \text{(Eq 1)}$$

and for the off-diagonal terms we have:

$$\tilde{D}_{ij}^n = -\frac{\partial\{\tilde{J}_i(x - x_0)\}}{\partial C_j}, \; i \neq j, i, j = 1, \ldots, n - 1 \quad \text{(Eq 2)}$$

where x is the diffusion direction, x_0 is the position of Matano plane and $\tilde{J}_i$ are the interdiffusion fluxes of component i:

$$\tilde{J}_i(x) = \frac{1}{2t} \int_{C_i^-}^{C_i(x)} (x - x_0) dC_i. \quad \text{(Eq 3)}$$

2 Tests for Binary Systems

The solution, Eq 1, 2, 3, can be applied to a binary diffusion couple with a constant interdiffusion coefficient, as was demonstrated in.[1] Here we consider interdiffusion coefficient as a linear and quadratic functions of composition. All the profiles below were calculated using the finite difference numerical procedure described in detail in.[16,17]

For the interdiffusion flux $\tilde{J}(x)$ at any x-position in the diffusion zone we have the following expression:

$$\tilde{J}(x) = \frac{1}{2t} \int_{C^-}^{C(x)} (x - x_0) dC. \quad \text{(Eq 4)}$$

and for the given problem this flux can be calculated (similar to[1]) and then application of Eq. 1, 2, 3 gives a resulting value for the interdiffusion coefficient $\tilde{D}$ as a function of composition, C_1. The resulting interdiffusion coefficients (as function of composition) are presented in Fig. 2(a).

Similarly, for the quadratic $\tilde{D} = D_0(1 - 2C_1(1 - C_1))$ where D_0 is a scaling constant, with the boundary condition $C_1^- = 1.0$ and $C_1^+ = 0.0$ we have the interdiffusion profile that is shown in Fig. 1(b).

Application of Eq. 1, 2, 3 gives resulting value for the interdiffusion coefficient $\tilde{D}$ as function of composition, see Fig. 2(b).

Obviously, in both these cases the agreement between the input and output values of the interdiffusion coefficient is excellent.

3 Test for Ternary Systems

In the ternary system (C_1, C_2, C_3 where C_3 is chosen as the reference component) the direct application of expressions Eq. 1, 2, 3 to a single couple is not possible. This is clear if we look at the available functional dependences of the two compositions, C_1, C_2. Their profiles are available only along the diffusion path which can be represented by a line, v, in the composition space. Therefore, along v, C_1 must be related to C_2 in such a way that $C_1^v = C_1^v(C_2)$. Therefore, if we take a derivative of $\tilde{J}_1(x)(x - x_0)$ with respect to C_1 along v, we will have that:

$$\frac{\partial[\tilde{J}_1(x)(x - x_0)]}{\partial C_1}\bigg|_v = \frac{\partial[\tilde{J}_1(x)(x - x_0)]}{\partial C_1} + \frac{\partial[\tilde{J}_1(x)(x - x_0)]}{\partial C_2}\frac{\partial C_2^v}{\partial C_1} \quad \text{(Eq 5)}$$

where the derivative on the left-hand side is taken straight from the corresponding profiles, and the derivatives on the right-hand side are the "true" derivatives that are to be used in the Eq. (1, 2). Similarly, for the other three derivatives we have the following expressions:

$$\frac{\partial[\tilde{J}_1(x)(x - x_0)]}{\partial C_2}\bigg|_v = \frac{\partial[\tilde{J}_1(x)(x - x_0)]}{\partial C_2} + \frac{\partial[\tilde{J}_1(x)(x - x_0)]}{\partial C_1}\frac{\partial C_1^v}{\partial C_2} \quad \text{(Eq 6)}$$
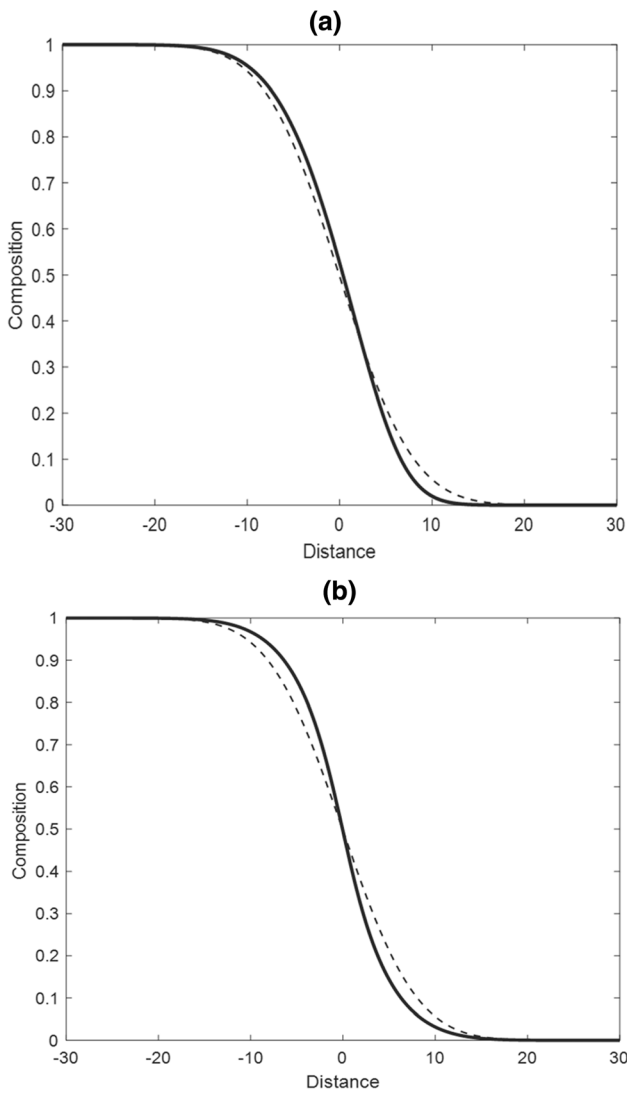
Fig. 1 Interdiffusion profiles in binary alloys (see text for details). With (a) linear interdiffusion coefficient; and (b) quadratic interdiffusion coefficient (dashed lines are for the constant interdiffusion coefficient for comparison)
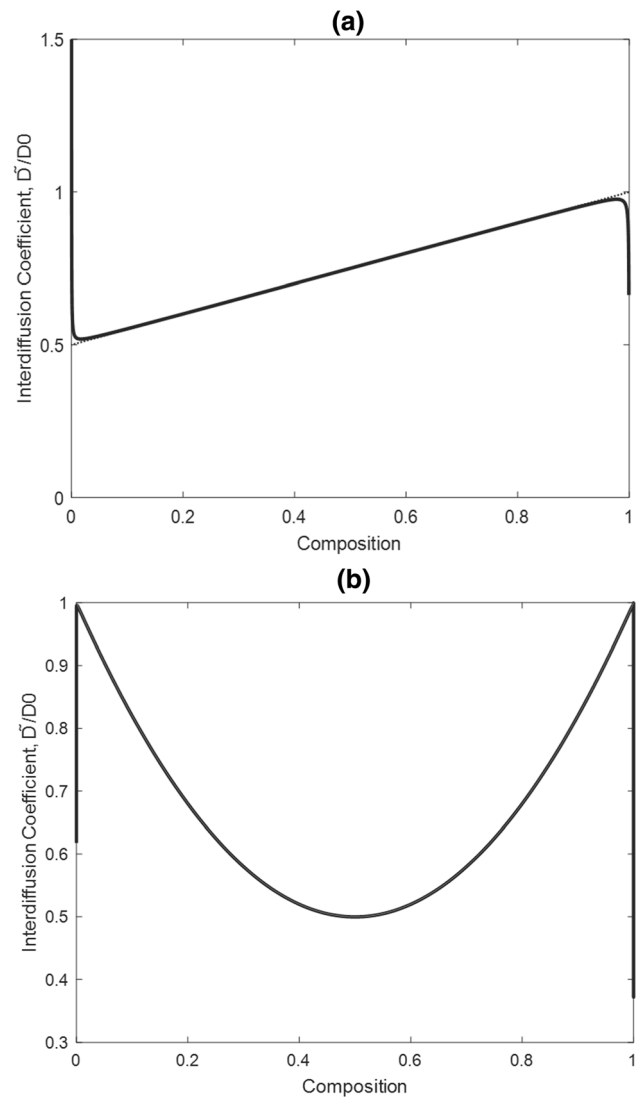

Fig. 2 Interdiffusion coefficients obtained from the profiles in Fig. 1 using Eqs. 1, 2, 3, presented as function of composition. For (a) linear composition dependence; and (b) quadratic composition dependence. Dashed lines are for the actual interdiffusion coefficients, in (b) this line almost perfectly coincides with the result of the analysis

$$\frac{\partial\left[\tilde{J}_2(x)(x-x_0)\right]}{\partial C_2}\bigg|_v = \frac{\partial\left[\tilde{J}_2(x)(x-x_0)\right]}{\partial C_2} + \frac{\partial\left[\tilde{J}_2(x)(x-x_0)\right]}{\partial C_1}\frac{\partial C_1^v}{\partial C_2} \qquad \text{(Eq 7)}$$

$$\frac{\partial\left[\tilde{J}_2(x)(x-x_0)\right]}{\partial C_1}\bigg|_v = \frac{\partial\left[\tilde{J}_2(x)(x-x_0)\right]}{\partial C_1} + \frac{\partial\left[\tilde{J}_2(x)(x-x_0)\right]}{\partial C_2}\frac{\partial C_2^v}{\partial C_1}. \qquad \text{(Eq 8)}$$

Two sets of Eq. (5) and (6); and Eq. (6) and (7) cannot be solved to retrieve four derivatives $\frac{\partial\left[\tilde{J}_i(x)(x-x_0)\right]}{\partial C_j}, i, j = 1, 2$. This is because the matrix of the coefficients for each set is:

$$\begin{bmatrix} 1 & \dfrac{\partial C_2}{\partial C_1} \\ \dfrac{\partial C_1}{\partial C_2} & 1 \end{bmatrix}. \qquad \text{(Eq 9)}$$

and it is clearly a singular matrix with a determinant of zero if it is applied to a single diffusion couple.

To be able to resolve the situation, another couple is obviously needed. As usual, the new couple should have a diffusion path that intersects with the diffusion path of the first couple. Then, at the point of intersection, the Eq. (5) and (7) can be taken from the first couple, and Eq. (6) and (8) can be taken from the second couple. The interdiffusion

coefficient is then available at one composition—at the intersection point. This is similar to the application of the Boltzmann-Matano analysis to ternary interdiffusion couples.

Here we consider two ternary interdiffusion couples where the interdiffusion matrix for both of them is chosen as:

$$[\tilde{D}] = D_0 \begin{bmatrix} 1.0 & 0.5 \\ 0.1 & 0.5 \end{bmatrix} \qquad (Eq~10)$$

where again D_0 is the scaling constant.

The middle composition for both couples is $C_1 = C_2 = C_3 = \frac{1}{3}$. This composition will be accepted (approximated) as the composition of interception where the interdiffusion matrix can be calculated.

In the couple 1, the end compositions are: $C_1^- = \frac{1}{6}; C_2^- = \frac{5}{12}; C_3^- = \frac{5}{12}$ and $C_1^+ = \frac{1}{2}; C_2^+ = \frac{1}{4}; C_3^+ = \frac{1}{4}$.

In the couple 2, the end compositions are: $C_1^- = \frac{5}{12}; C_2^- = \frac{1}{6}; C_3^- = \frac{5}{12}$ and $C_1^+ = \frac{1}{4}; C_2^+ = \frac{1}{2}; C_3^+ = \frac{1}{4}$.

The interdiffusion profiles were calculated for the time $t = 20$ using closed form solution and discretised for the step in the diffusion direction equivalent to about 2 μm. The profiles for the first and second couples are presented in Fig. 3(a), (b).

Application of the approach described above gives the following interdiffusion matrix:

$$[\tilde{D}] = D_0 \begin{bmatrix} 0.99 & 0.49 \\ 0.098 & 0.50 \end{bmatrix} \qquad (Eq~11)$$

Comparing with Eq. (10), this is very accurate result, only maximum of 1% relative error for the diagonal terms and 2% for the off-diagonal terms. As can be seen from this example, in the ternary system, the new method works very well for two couples. However unfortunately, the extended (more than at one point) compositional dependence of the $[\tilde{D}]$ is not accessible with the new method, even when using two couples.

4 Quaternary and Higher Component Systems

Dealing with quaternary and higher components systems is similar to the dealing with the ternary system. It is easy to prove that for the n-component $(n > 1)$ systems, the $(n - 1)$ couples will be needed for the current analysis to be applicable. Results will be obtained only for one point where the diffusion paths of all the couples intersect. This is consistent with the general theories of interdiffusion in multicomponent systems.

However, it should be added that in the case of quaternary and higher component systems, the building of
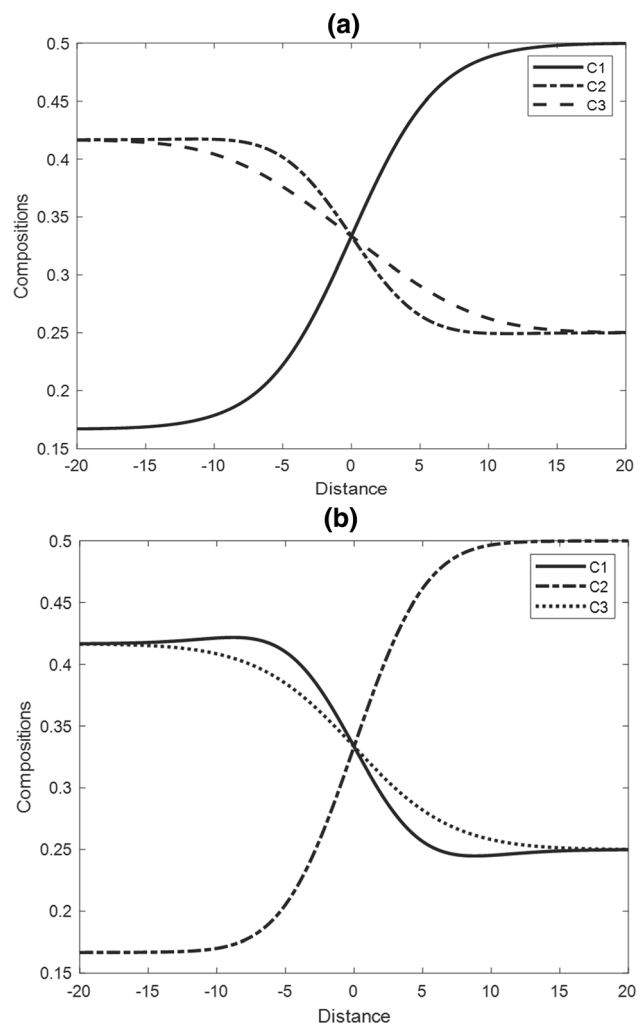

Fig. 3 Interdiffusion profiles in two ternary alloy couples (see text for details). (a) – couple 1; and (b) – couple 2

three and higher number of couples that will have a point of common intersection is very difficult.

5 Conclusions

In this paper, we have investigated the novel exact solution[1,2] that is applicable to a multicomponent (n-component) interdiffusion couple where the interdiffusion matrix may change with alloy composition. Various computational approaches have been used. Here, it was shown that the novel solution leads to a computational procedure similar to the Boltzmann-Matano analysis. For multicomponent diffusion couple, the derivatives $\partial(J_j(y - y_0))/\partial C_i, i, j = 1, \ldots, n - 1$ (that are required for the solution) can only be calculated along the diffusion path and, therefore, a single couple will not be enough to calculate all of them correctly. As expected in the multicomponent interdiffusion analysis, $n - 1$ diffusion couples

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is essential for the correct application of the method. The full interdiffusion matrix can be then obtained, but only at one composition where all diffusion paths intersect.

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