Title:

A diffusion mobility database for magnesium alloy development

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Abstract:

A database of atomic mobility parameters for single phase Mg-X (X = Ag, Al, Be, Cd, Ce, In, Fe, Ga, La, Mn, Ni, Sb, Sn, U, and Zn) systems was developed through the critical assessment of available Mg diffusion literature. Optimization of the binary diffusion activation energy terms for Ag, Al, Sn, and Zn solutes, as well as the Al-Zn interaction parameter were completed through the coupling of an existing thermodynamic database with the DICTRA software program. In general, agreement between simulated and experimental diffusion profiles was observed, although limitations for extrapolation to higher order systems were seen due to the limited multicomponent diffusion studies in Mg. This work, however, ultimately provides the foundation for the integration of diffusion simulations and predictions in future Mg alloy design. Further research to expand the current database, through either the experimental measurement or simulation of multicomponent diffusion and diffusivity in other phases (e.g. intermetallics, liquid), is expected to increase the database utility.

Keywords: diffusion modeling, magnesium alloys, kinetic database, thermodynamics

Main Text:**1. Introduction**

An integral component to accelerated materials development within the Integrated Computational Materials Engineering framework is the incorporation of fundamental, predictive models throughout the design process [1, 2]. For example, the CALPHAD approach to modeling materials thermodynamics uses experimental descriptions of lower order alloy systems to predict

phase equilibria in higher order systems where limited experimental data may exist [3, 4]. The coupling of these thermodynamic models with minimal experiments then facilitates the prediction of material microstructures and properties, ultimately accelerating the optimization of material performance with reduced development time.

An inherent limitation of phase equilibria calculations, however, is the lack of kinetic information. The integration of diffusivity predictions into materials design approaches enables the simulation of many metallurgical processes, ranging from homogenization to particle coarsening. These phase evolution behavior predictions then allow microstructure induced effects on alloy performance to be predicted. Such materials design approaches have seen success in several alloy fields, such as high performance steels [5-7] and titanium alloys [8]. In contrast, Mg alloys have experienced limited application of these design approaches for alloy development, which have hindered commercial integration despite their significant advantages in weight-conscious industries (e.g. transportation). Currently, Mg alloys have a significant number of studies related to the development of thermodynamic descriptions (e.g. [9, 10]) and their implementation into commercial programs that predict phase equilibria (e.g. ThermoCalc [11], FactSage [12], Pandat [13]), but no database of Mg alloy kinetic descriptions currently exists.

In the present work, an atomic mobility database for single-phase hexagonal close packed (HCP) Mg-X ($X = \text{Ag, Al, Be, Cd, Ce, Fe, Ga, In, La, Mn, Ni, Sb, Sn, U, and Zn}$) alloys were developed using the DICTRA and ThermoCalc software programs [11, 14-16]. A brief summary of the solutes, data type (single crystal vs. polycrystalline), and data references used is given in Table 1. For several solutes (U [17], Ni [17], Mn [18], La [19], Ga [20], Fe [17], Ce [19], Be [21]), only one dataset for experimental diffusion in polycrystalline Mg currently exists. For the solutes with a single dataset, the developed mobility database was not optimized and the reported

activation energy and pre-exponential factors were simply placed in the database. It is important to note that elements for which diffusion data exists but there is no associated thermodynamic description in the TCMG2 database (e.g. Cd, Ga, In, U) were still placed into the kinetic database. For Sn, a single dataset for diffusion in single crystal Mg exists and was optimized [22]. Ag [19, 22], Al [23-29], and Zn [19, 26, 30, 31] solutes, which have thermodynamic descriptions in the current ThermoCalc database and multiple datasets in the literature, were also optimized. Sb [22], In [19, 22], and Cd [22] solute mobilities in HCP-Mg were not optimized because single crystal values are reported and the elements are not assessed in the current TCMG2 thermodynamic database [32]. For convenience, the reported activation energy and pre-exponential factors for diffusion parallel to the c-axis were utilized in the mobility database for Sb and Cd, while the polycrystalline values for In were used. Additionally, for Be, Cd, Ga, In, and Sb solutes, the kinetic parameters in the database serve as reference values, as the incorporation of thermodynamic descriptions for these solutes into the current thermodynamic database would be needed to simulate diffusion. Although additional diffusion data is available from atomistic simulations (e.g. [33, 34]), it is uncertain how this data should be weighed since it lacks statistical comparison and thus these results were not included in the creation of the database. Developing a methodology for the incorporation of atomistic calculations into atomic mobility optimizations in the future, however, will enable more complex alloy systems to be modeled.

2. Kinetic Modeling

2.1 Theory

To model the diffusivity of solutes within an alloy, a matrix of mobility values must first be optimized from data in literature. While diffusion coefficients are typically reported in

literature, a kinetic database developed using diffusivity would be overly complex due to the large number of composition dependent values needed [35]. A reduction in the number of kinetic database descriptors can be realized through the evaluation of atomic mobilities, which can be paired with a separate thermodynamic database to predict diffusion [35].

In the lattice-fixed frame of reference assuming a vacancy exchange mechanism, the composition-dependent diffusion coefficient (D_{kj}) can be defined as the product of an atomic mobility factor (M_{ki}) and a thermodynamic factor:

$$D_{kj}^L = \sum_i M_{ki} \frac{\partial u_i}{\partial x_j}. \quad \text{Eqn. 1}$$

where k is the gradient specie, j is the diffusing specie, μ is the chemical potential, and x is the mole fraction [16]. The partial derivative of chemical potential with respect to the composition represents a thermodynamic factor that can be calculated with a multicomponent thermodynamic database.

r the atomic mobility matrix, only the diagonal terms are typically modeled, with the off-diagonal terms assumed to be zero [16]. This assumption is reasonable as the magnitude of the atomic jump correlation effects that give rise to off-diagonal atomic mobility terms are usually small relative to experimental data scatter [35]. The diagonal terms of the mobility matrix can be described by:

$$M_i = M_i^0 \frac{1}{RT} \exp\left(-\frac{Q_i}{RT}\right). \quad \text{Eqn. 2}$$

where M_i is the mobility of element i , M_i^0 is a function of the jump frequency and distance, R is the gas constant, T is the temperature, and Q is the activation energy in the given phase. The frequency factor ($\ln M_i^0$) and activation energy factor (Q_i) are dependent upon composition,

temperature, and pressure. When there is no magnetic effects on atomic mobility, the frequency factor is often set to one and only the composition and temperature dependence of activation energy are considered. Following the CALPHAD approach to thermodynamic free energy modeling, the Redlich-Kister polynomial is used to represent the composition dependence of the diffusion activation energy [3, 16, 36, 37]:

$$Q_i = \sum_j x_j Q_i^j + \sum_p \sum_{j>p} x_p x_j \sum_k A_i^{pj} (x_p - x_j)^k \quad \text{Eqn. 3}$$

where Q_i^j represents the activation energy of component i in pure j and A_i^{pj} is an interaction parameter [3].

2.2 Diffusion Data

To optimize the activation energy and interaction parameter components of atomic mobility, experimental diffusion data from literature must first be evaluated. Tracer diffusivity (D_i^*), determined from diffusion studies using radioactive isotopes, is independent of a thermodynamic factor and can be described by the Einstein relation (Equation 4). Although tracer diffusivity is more difficult to experimentally obtain, it is the preferred data type due to its independence from thermodynamic databases.

$$D_i^* = RTM_i \quad [16] \quad \text{Eqn. 4}$$

Diffusion couples are often used to extract experimental values for interdiffusivity and intrinsic diffusivity where radioactive tracer experiments are not practical or possible. The chemical interdiffusivity ($\tilde{D}_{kj}^n$) is measured in the volume fixed reference frame from diffusion couples and is given by:

$$\tilde{D}_{kj}^n = D_{kj}^V - D_{kn}^V \quad [16] \quad \text{Eqn. 5}$$

where n is the solvent atom. The volume fixed frame of reference diffusivity (D^V), assuming the partial molar volume is constant, can then be represented as:

$$D_{kj}^V = \sum_{i=1} (\delta_{ik} - x_k) x_i M_i \frac{\partial \mu_i}{\partial x_j} \quad [16] \quad \text{Eqn. 6}$$

where δ_{ik} is the Kronecker delta.

Following the Kirkendall experiment, the unequal diffusional fluxes of species create a shift of inert marker locations within a sample in the laboratory fixed reference frame [38]. Determination of the atomic fluxes in the lattice fixed reference frame allows removal of additional advective lattice motion effects, providing a measure of intrinsic diffusivity [39]. This intrinsic diffusivity is given by Equation 1.

The diffusivity equations for intrinsic and chemical interdiffusion include thermodynamic terms that require input from a secondary source. For the DICTRA software implementation used in the present work, the ThermoCalc thermodynamic database provides the necessary information [32]. It is important to note that when intrinsic or chemical diffusivities are used for optimization, the results are dependent upon the accuracy of the thermodynamic database description.

2.3 Optimization Procedure

The DICTRA (Diffusion Controlled TRAnsformations) 27.0.1 software created by ThermoCalc [11] facilitates the connection between a kinetic database and thermodynamic database for the simulation of diffusion. For the current optimization, the TCMG2 thermodynamic database developed by ThermoCalc was used [32]. The PARROT optimization program of DICTRA was used to determine the temperature and composition dependence of the diffusion activation energy. Using initial estimates for atomic mobility parameters, the diffusion coefficients were calculated and compared to experimental data from literature. Through an

iterative optimization procedure in the PARROT module, the atomic mobility fitting parameters were then optimized to reduce the error.

The anisotropic nature of the HCP Mg crystal results in different diffusion coefficients parallel and perpendicular to c-axis. Although the c/a ratio is near ideal, in general the ratio of the perpendicular to parallel diffusivity is approximately 1.1-1.3 [40, 41]. For the current database assessment, the diffusivity values for single crystals in parallel and perpendicular directions, as well as polycrystalline samples were all used for optimization. Therefore, the current results represent an effective average diffusivity. While this could lead to errors in large-grained or single crystal simulations, most Mg commercial applications are concerned more with fine-grained, polycrystalline systems.

3. Material and Methods

To validate the diffusion database, two experimental diffusion couples were prepared and analyzed. The first couple consisted of Mg-Zn-Al and Mg-Zn-Al-Sn alloys with the intent to evaluate Sn diffusivity, while the second and third couples used Mg-Zn-Al alloys bonded to pure Mg to evaluate Zn and Al diffusivity. To prepare the alloys, pure Mg (99.98% Purity), Sn (99.9% Purity), Al (99.9% Purity) and Zn (99.9% Purity) pieces weighed to the desired compositions and placed into graphite crucibles coated with boron nitride. The raw materials were then placed into a resistive furnace in a glove box under positive pressure Ar. The alloys were melted at 725°C for 30 minutes, followed by casting into a graphite mold. The compositions of the alloys were measured by inductively coupled plasma atomic emission spectroscopy and are given along with the alloy designations in Table 2.

The TZA1121 and ZA11 alloys were solutionized at 510°C for 15 hours, while the AZ91 alloy was solution treated at 415°C for 16 hours. The alloys were subsequently quenched in

water after heat treatment. Sections approximately 10 mm in diameter and 5 mm in length were then cut from the TZA1121, ZA11, AZ91, and Mg samples. One circular face for each sample section was then planed and polished using standard metallographic procedures to a final polish of 0.03 microns. The polished faces were placed together and put into Kovar jigs to create TZA1121 – ZA11 and AZ91 – Mg diffusion couples. The pressure applied to the couples was set at the minimum amount needed to prevent sample slippage.

The diffusion couples were then encapsulated in Pyrex tubes that were evacuated to approximately 2 to 4 Pa. The TZA1121 – ZA11 diffusion couple was heat treated at 500°C for 250 hours, while two AZ91 – pure Mg diffusion couples were held at 435 and 390°C for 144 hours. All diffusion couples were quenched in water after heat treatment. The bonded alloys were sectioned perpendicular to the interface using a low-speed diamond saw and then polished to a final polish of 0.03 microns. The composition profiles of the solutes were measured perpendicular to the diffusion zone using a JEOL Superprobe 733 Electron Probe MicroAnalysis (EPMA) with a one-micron step size. The element concentrations were calibrated in EPMA using pure element standards. The diffracting crystals that were used included thallium acid phthalate ($2d = 25.757 \text{ \AA}$, for Mg and Al), lithium fluoride (200) crystal ($2d = 4.0267 \text{ \AA}$, for Zn), and pentaerythritol (002) crystal ($2d = 8.742 \text{ \AA}$, for Sn).

4. Results and Discussion

4.1 Mg Self-Diffusion Assessment

The Mg self-diffusion literature consists of four primary studies. Shewmon [42] initially used extruded polycrystalline Mg alloys with a (0001) fiber texture to study self-diffusion from 468 to 627°C, but later refined his measurements with single crystal studies in a similar temperature range [40]. Combronde studied the anisotropic diffusion coefficients of single

crystal Mg in the temperature range 500 to 630°C using the ^{28}Mg radioactive tracer [41].

Kulkarni *et al.* used secondary ion mass spectrometry to extract concentration profiles of the ^{25}Mg radioactive isotope to determine the tracer diffusivity D^* [43]. The sum of errors in the extraction of tracer diffusivity for the studies is estimated at less than 10%. As seen in Figure 1, there is good agreement between the fitted atomic mobility parameters for DICTRA and the Mg-self diffusion experimental data in literature.

4.2 Ag Diffusion Assessment

Two tracer diffusivity studies of ^{110}Ag in pure Mg exist. Lal [19] studied diffusion in polycrystalline Mg in the temperature range of 476.5 to 621°C. The samples were sectioned using a precision lathe and their activity measured using a scintillator. In contrast, Combronde [22] used Mg single crystals to study diffusion between 479 and 638.9°C by measuring the activity penetration curves after abrading fixed amounts of material. In the temperature range of interest, the polycrystalline tracer diffusion values were intermediate to the single crystal values. Additionally, for Ag there was significant deviation in the parallel to perpendicular diffusivity ratio compared to Mg self-diffusion, as the reported ratio was 1.56 - 2.37 [22]. This Ag heterodiffusivity ratio was also much greater than the ratios observed for Cd, In, Sn, and Sb tracer diffusion studies. The change in diffusion anisotropy was attributed to the local electrostatic potential arising from valence differences between solute and solvent [22]. The results of the optimization for Ag are shown in Figure 2.

4.3 Al Diffusion Assessment

While there are several experiments for the diffusion of Al into Mg, none of them include tracer diffusion results, as the ^{26}Al isotope is very expensive and not readily available. Brennan [24, 25] deposited Al thin films onto Mg and measured the compositional profile after diffusion

using secondary ion mass spectrometry. An inherent flaw in that measurement technique is related to the surface roughness that results from sputtering. At small grain sizes, the increased roughness can lead to an artificial overestimation of the diffusion coefficient [24].

Brennan later refined the diffusion measurements using a Boltzmann-Matano analysis of pure Mg - Al diffusion couples annealed between 300 and 400°C [23]. The limited diffusion distance in the Mg solid solution region at the lower temperatures in this study led to lower statistical confidence in the concentration gradients and thus the interdiffusivity calculations. Therefore, only the data for 400°C was used in the current optimization. Das [28, 29] also used diffusion couples to extract the anisotropic diffusion coefficients for Al in Mg single crystals using multiphase modeling coupled with a thermodynamic database. The composition dependence of Al diffusion in Mg polycrystals was determined using diffusion couples by Kammerer [26] and Kulkarni [27]. Relative errors in the reported diffusion coefficients were obtained from each respective paper when available or from comparison to similar experimental methodologies.

In general, there is good agreement between the modeled diffusivity and literature data (Figure 3). The modeled diffusivity of Al at low concentrations, however, overestimates the values reported by Kulkarni [27]. This is a result of the influence of the Kammerer dataset at the lower concentrations in the optimization.

4.3 Sn Diffusion Assessment

For the Mg-Sn binary system, only one experimental study has been performed [22]. While first principles density functional theory calculations of dilute Sn diffusivity in Mg have been completed [33], preference was given to experimental data for the current optimization. Combronde [22] diffused the ^{113}Sn tracer into Mg single crystals at temperatures between 475

and 629.3°C. The authors determined the concentration profiles by radioactivity measurements of cuttings made with a precision lathe [22]. Combronde extracted an experimental activation energy of 149.9 kJ/mol for diffusion parallel to the Mg c-axis but was unable to calculate the value for diffusion perpendicular to the c-axis. The parallel value is close to the optimized activation energy of 151.3 kJ/mol in the current study, with the differences attributable to the inclusion of the perpendicular values during optimization. The DICTRA optimization of atomic mobility reproduced the temperature dependence of diffusivity well (Figure 4).

4.3 Zn Diffusion Assessment

The Zn diffusion literature includes four primary experimental studies. Lal [19] and Cermak [31] diffused the radioactive ^{65}Zn isotope into polycrystalline Mg samples and measured the depth profile using standard serial sectioning and residual activity methods. Kammerer [26] fabricated diffusion couples between polycrystalline Mg and Mg-6wt%Zn alloys to study the diffusion of Zn between 623 and 723°C. The composition profiles obtained from EPMA were used to extract the interdiffusion and impurity diffusion coefficients.

Das [30] fabricated diffusion couples between Mg single crystals and polycrystalline Zn. To extract the anisotropic diffusion coefficients of Zn in Mg from the measured composition profiles, the authors used multiphase modeling via solutions to Fick's second law. This type of diffusion modeling requires an accurate thermodynamic description of the system and the FTlite FactSage database was used. As the Das data required a separate thermodynamic description that could introduce greater error and their diffusivity data is almost one order of magnitude lower than the other three studies, it was excluded from the current optimization. As tracer diffusion is expected to be more accurate than interdiffusion, the error associated with the Kammerer data was set at 20%. From Figure 5, it can be seen that the diffusion of Zn in Mg in both the dilute

limit and at higher concentrations is reproduced well by the DICTRA simulations using the optimized kinetic database.

4.5 Al-Zn Interaction Parameter Assessment

The ^{65}Zn tracer diffusion series of experiments completed by Cermak [31] also reported the Zn diffusivity in three Mg-Al solid solutions at temperatures between 575 and 375°C. The Zn diffusivities reported at the highest temperatures exhibited values that deviated significantly from the expected values, which was likely a result of partial melting at grain boundaries. Removing this anomalous data and optimizing the Al-Zn interaction parameter yielded good agreement with literature (Figure 6).

4.6 Database Optimization Summary

The relevant parameters in the developed kinetic database are shown in Table 3T5o1. The activation energies and interaction parameters that were optimized, designated MQ in DICTRA, are also given.

4.7 Diffusion Couple Validation

To validate the current optimized database, experimental diffusion couple composition profiles after various diffusion anneals were compared to simulated profiles calculated using the newly optimized DICTRA database. For the TZA1121 – ZA11 diffusion couple, the concentration profiles for all three solutes (Al, Sn, Zn) were reproduced well by the current simulations (Figure 7). In particular, the distance that the Sn diffused experimentally was in good agreement with the predicted value. This indicates that it is unlikely that there is an interaction parameter for Sn solute diffusivity in a Mg-Al or Mg-Zn alloy.

Despite the fact that the Zn and Al concentrations are relatively similar in each section of the initial TZA1121 – ZA11 diffusion couple, the chemical potential gradient and the driving

force for diffusion are not. During the diffusion anneal, diffusive flux of Zn and Al can therefore occur. The Zn and Al profiles indicate that the diffusivity of Zn and Al may potentially be overestimated, but it is still within the range of error of the EPMA measurements.

To further evaluate the rate of Al and Zn solute diffusion in HCP-Mg, the experimental composition profiles for two experimental AZ91 – Mg diffusion couples were compared to their expected profiles calculated using the atomic mobility database (Figure 8, Figure 9). Both couples appeared to simulate the Al and Zn profiles well. There was some minor differences in the Zn profiles, as the simulated profiles appeared to potentially overestimate the Zn diffusivity. This result was not unexpected, as only one study was found for multicomponent diffusion in the single phase HCP-Mg system with Al and Zn solutes [31]. Thus, the difference could be attributable to the need for modification to the interaction parameter for Al and Zn solute diffusivity. This information could be obtained with further experiments.

4.8 Diffusion in Mg

Due to limited experimental diffusion data available in literature, the current kinetic database was optimized for Mg self-diffusion and four solutes (Ag, Al, Sn, Zn). For the other solute elements, the diffusivity was obtained from literature. In Figure 10, a summary of the solutes with commercial relevance that have reported diffusion values in single-phase HCP Mg is given. It is important to note that, for many of these elements, only a single diffusion experiment exists.

This highlights one of the greatest challenges towards building a comprehensive kinetic database for Mg alloys: the lack of fundamental diffusion studies. As seen in Figure 10, there are many technologically-relevant solutes missing, including a majority of the rare earth elements. The missing data has profound implications for alloy design, as not enough information exists to

make informed decisions. Furthermore, there are even fewer experiments detailing the composition dependence of solute diffusivity. The close grouping of diffusivity values for the various solutes (Figure 10) suggest that slight modifications to diffusivity due to compositional changes may have significant effects. Additionally, while some interdiffusion experiments exist in literature (e.g. $\text{Mg}_{17}\text{Al}_{12}$ phase [44]), more experiments on other precipitate phases would be beneficial. This would enable the prediction of particle growth during aging treatments, which would affect alloy strength.

5. Conclusions

A kinetic database for single-phase Mg alloys has been developed for Ag, Al, Be, Cd, Ce, In, Fe, Ga, La, Mn, Ni, Sb, Sn, U, and Zn solutes. Critical assessment of the available diffusion literature for Ag, Al, Sn, and Zn allowed optimization of the atomic mobility activation energy terms for the binary systems. The optimizations were assessed through comparisons to several experimental diffusion couples. The results highlight that more fundamental diffusion work is needed to provide data for further optimization, as many commercially relevant solutes (e.g. rare earths) have not been studied and there are limited multicomponent diffusivities reported. The incorporation of predictive, accurate first-principles calculations is expected to ease the experimental burden and greatly assist the expansion of the current database. Further, it is anticipated that continued database development would enable more practical simulations of industrially-relevant processes, accelerating alloy development.

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Figures:

Figure 1: Comparison between the optimized and experimental values for Mg self-diffusion. Data taken from Shewmon '54 [42], Shewmon '56 [40], Combronde '71 [41], and Kulkarni '13 [43].

Figure 2: Comparison between the optimized and experimental values for Ag solute diffusion in HCP-Mg. Data taken from Lal '66 [19] and Combronde '72 [22].

Figure 3: (A) Comparison between the computed temperature-dependent diffusivity of Al solute in HCP-Mg and literature data. (B) Composition dependence of Al solute diffusivity in HCP-Mg compared to literature data. Data taken from Kammerer '13 [26], Brennan JPED '12 [24], Brennan '10 [25], Brennan MMTA '12 [23], Das '13 [28, 29], and Kulkarni '13 [27].

Figure 4: Optimization results for Sn tracer diffusion in HCP-Mg. Data taken from Combronde '72 [22].

Figure 5: Modeled Zn diffusion into Mg compared to experimental data. (A) Diffusion in the dilute limit, (B) Concentration dependent Zn diffusion. Data taken from Lal '66 [19], Cermak '06 [31], and Kammerer '13 [26].

Figure 6: The current Al-Zn interaction parameter optimization results compared to the Cermak tracer diffusion results [31].

Figure 7: A comparison between the measured compositional profiles (markers) and the DICTRA simulations (lines) for the TAZ1121 – ZA11 diffusion couple annealed at 500°C for 250 hours. Following the figure orientation, the ZA11 alloy is on the left and the TZA1121 alloy is on the right. (A) Good agreement for Sn diffusivity was observed. (B) Zn and Al diffusion composition profiles were within the measurement error for EPMA.

Figure 8: A comparison between the measured compositional profiles (markers) and the DICTRA simulations (lines) for the AZ91 – Mg diffusion couple annealed at 390°C for 144 hours. Following the figure orientation, the Mg sample is on the left and the AZ91 alloy is on the right. Both the Al (A) and Zn (B) profiles were reproduced well.

Figure 9: A comparison between the measured compositional profiles (markers) and the DICTRA simulations (lines) for the AZ91 – Mg diffusion couple annealed at 435°C for 144 hours. Following the figure orientation, the Mg sample is on the left and the AZ91 alloy is on the right. (A) The Al experimental profile was in agreement with the DICTRA simulation. (B) The Zn diffusivity appeared to be slower in the experimental couple relative to the simulation.

Figure 10: Summary of current experimental diffusion data from the present optimization and literature for relevant solutes in Mg.

Tables:

Table 1. Summary of experimental solute diffusion literature in single phase HCP-Mg alloys.

Solute	Data Type		References
	<i>Polycrystalline (PC), Single Crystal (SX)</i>		
Ag	PC, SX		[19, 22]
----- Al	PC, SX		[23-29]
Be	PC		[21]
Cd	SX		[22]
Ce	PC		[19]
Fe	PC		[17]
Ga	PC		[20]
In	PC		[19, 22]
La	PC		[19]
Mn	PC		[18]
Ni	PC		[17]
Sb	SX		[22]
Sn	SX		[22]
U	PC		[17]
Zn	PC, SX		[19, 26, 30, 31]

Table 2: Measured compositions (in wt%) for the alloys used in the diffusion couple validation.

Alloy	Mg	Sn	Al	Zn
TZA1121	86.5	11.0	0.8	1.7
ZA11	97.9	0	0.7	1.4
AZ91	89.8	0	9.1	1.1

Table 3: Optimized mobility parameters in the kinetic database.

DICTRA Notation	Mobility Parameter	Value (J/mole)	Reference
<i>Mg Diffusivity</i>			
MQ(HCP&Mg,Mg:VA)	Q_{Mg}^{Mg}	$-1.2508E5 + RT\ln(2.48E-5)$	Present Work
MQ(HCP&Mg,Al:VA)	Q_{Mg}^{Al}	$-9.4176E5 - RT\ln(5.01E-4)$	Present Work
MQ(HCP&Mg,Mg,Al:VA)	${}^0A_{Mg}^{Mg,Al}$	$-2.8842E5 + 1.967E3*T$	Present Work
MQ(HCP&Mg,Zn:VA)	Q_{Mg}^{Zn}	$-8.2169E7 + RT\ln(1.29E-11)$	Present Work
MQ(HCP&Mg,Mg,Zn:VA)	${}^0A_{Mg}^{Mg,Zn}$	$+8.2831E7 + 3.785E3*T$	Present Work
<i>Ag Diffusivity</i>			
MQ(HCP&Ag,Mg:VA)	Q_{Ag}^{Mg}	$-1.3991E5 + RT\ln(6.13E-4)$	Present Work

<i>Al Diffusivity</i>			
MQ(HCP&Al,Mg:VA)	Q_{Al}^{Mg}	$-1.5231E5 + RT\ln(5.74E-4)$	Present Work
MQ(HCP&Al,Al,Mg:VA)	$A_{Al}^{Al,Mg}$	$-7.0918E5 + 1.167E3*T$	Present Work
<i>Be Diffusivity</i>			
MQ(HCP&Be,Mg:VA)	Q_{Be}^{Mg}	$-1.5700E5 + RT\ln(8.06E-4)$	Yerko '66 [20]
<i>Cd Diffusivity</i>			
MQ(HCP&Cd,Mg:VA)	Q_{Cd}^{Mg}	$-1.4070E5 + RT\ln(1.29E-4)$	Combronde '72 [21]
<i>Ce Diffusivity</i>			
MQ(HCP&Ce,Mg:VA)	Q_{Ce}^{Mg}	$-1.7580E5 + RT\ln(4.50E-2)$	Lal '66 [18]
<i>Fe Diffusivity</i>			
MQ(HCP&Fe,Mg:VA)	Q_{Fe}^{Mg}	$-8.8800E4 + RT\ln(4.00E-10)$	Neumann '11 [17]
<i>Ga Diffusivity</i>			
MQ(HCP&Ga,Mg:VA)	Q_{Ga}^{Mg}	$-1.3430E5 + RT\ln(1.20E-4)$	Stloukal '03 [19]
<i>In Diffusivity</i>			
MQ(HCP&In,Mg:VA)	Q_{In}^{Mg}	$-1.1882E5 + RT\ln(5.20E-6)$	Combronde '72 [21]
<i>La Diffusivity</i>			
MQ(HCP&La,Mg:VA)	Q_{La}^{Mg}	$-1.0220E5 + RT\ln(2.20E-6)$	Lal '66 [18]
<i>Mn Diffusivity</i>			
MQ(HCP&Mn,Mg:VA)	Q_{Mn}^{Mg}	$-1.5400E5 + RT\ln(0.76E-4)$	Fujikawa '92 [17]
<i>Ni Diffusivity</i>			
MQ(HCP&Ni,Mg:VA)	Q_{Ni}^{Mg}	$-9.5900E4 + RT\ln(5.00E-10)$	Neumann '11 [17]
<i>Sb Diffusivity</i>			
MQ(HCP&Sb,Mg:VA)	Q_{Sb}^{Mg}	$-1.3730E5 + RT\ln(2.57E-4)$	Combronde '72 [21]
<i>Sn Diffusivity</i>			
MQ(HCP&Sn,Mg:VA)	Q_{Sn}^{Mg}	$-1.5132E5 + RT\ln(4.56E-4)$	Present Work
<i>U Diffusivity</i>			
MQ(HCP&U,Mg:VA)	Q_U^{Mg}	$-1.1470E5 + RT\ln(2.50E-9)$	Neumann '11 [17]

<i>Zn Diffusivity</i>			
MQ(HCP&Zn,Mg:VA)	Q_{Zn}^{Mg}	$-1.2552E5 + RT\ln(9.38E-5)$	Present Work
MQ(HCP&Zn,Zn,Mg:VA)	${}^0A_{Zn}^{Zn,Mg}$	$-2.5024E6 + 3.350E3*T$	Present Work
MQ(HCP&Zn,Al,Mg:VA)	${}^0A_{Zn}^{Al,Mg}$	$+3.6151E4 + 5.061E1*T$	Present Work