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Araştırma Makalesi / Research Article

Investigating the Relationship Between Creep Rate, Thermal Conductivity and Enthalpy of Mixing in Some Magnesium Alloys

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Abstract

Magnesium (Mg) and its alloys are known for their high specific strength, but they have limitations that need to be addressed to expand their range of industrial use. One major limitation is their high temperature performance, which requires improvement for them to be used in industries such as aviation where creep resistance is important. In this study, we investigated the creep behavior of several Mg alloys, including the commercially used AZ series (AZ31, AZ61, and AZ91), as well as the AM50, AX52, and AE42 alloys. We studied the relationship between the creep rate of these alloys and two material properties: thermal conductivity and enthalpy of mixing. By analyzing these properties, we can compare and evaluate newly developed or existing alloys in terms of their creep behavior. When the obtained results are interpreted in general, a linear relationship is found between the creep rate, thermal conductivity and enthalpy of mixing as a general trend.

Keywords: Mg alloys, Creep, Thermal conductivity, Enthalpy of mixing

Bazı Magnezyum Alaşımlarında Sürünme Hızı, Termal İletkenlik ve Karışım Entalpisi Arasındaki İlişkinin İncelenmesi

Öz

Magnezyum (Mg) ve alaşımları, yüksek özgül güçleri ile bilinirler, ancak endüstriyel kullanım alanlarını genişletmek için ele alınması gereken sınırlamaları vardır. En büyük sınırlamalardan biri, yüksek sıcaklık performanslarıdır ve bu, havacılık gibi sürünme direncinin önemli olduğu endüstrilerde kullanılmaları için iyileştirme gerektirir. Bu çalışmada, ticari olarak kullanılan AZ serisi (AZ31, AZ61 ve AZ91) ve AM50, AX52 ve AE42 alaşımları dahil olmak üzere birçok Mg alaşımının sürünme davranışını inceledik. Bu alaşımların sürünme oranı ile iki malzeme özelliği arasındaki ilişkiyi inceledik: termal iletkenlik ve karıştırma entalpisi. Bu özellikleri analiz ederek, yeni geliştirilen veya mevcut alaşımları sürünme davranışları açısından karşılaştırabilir ve değerlendirebiliriz. Elde edilen sonuçlar genel olarak yorumlandığında, sürünme hızı, ısıl iletkenlik ve karışım entalpisi arasında genel bir eğilim olarak doğrusal bir ilişki bulunmuştur.

Anahtar kelimeler: Mg alaşımları, Sürünme, Termal İletkenlik, Karışım entalpisi

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1. Introduction

Creep is a phenomenon that occurs in materials subjected to continuous loads at elevated temperatures over time. This refers to the gradual deformation and eventual damage of the material under these conditions, even if the applied stress is below the yield strength of the material. This is a major concern for materials used in high-temperature applications such as the aerospace, power generation, and automotive industries (Kutty et al., 1996; Maruyama et al., 2002; M. Pekguleryuz & Celikin, 2010; M. O. Pekguleryuz & Kaya, 2003).

Mg alloys stand out for their light weight and excellent strength-to-weight ratios. This makes them an attractive structural metal for use in a range of applications (Jayasathyakawin et al., 2020; Luo et al., 2020; J. F. Nie et al., 2020; Ramalingam et al., 2020; Y. Yang et al., 2021; Z. Zhang et al., 2021). However, they are susceptible to creep under high-temperature and high-stress conditions, which can limit their usefulness in certain situations. Therefore, creep resistance in Mg alloys is important because it determines their ability to withstand high temperatures and loads for a long time without deformation or deterioration. Creep-resistant Mg alloys have the potential to offer enhanced performance in high-temperature applications including engine components, heat exchangers, and other high-stress environments. The creep resistance of Mg alloys is affected by several factors including their composition, microstructure, and machining methods. For instance, the addition of certain alloying elements, such as aluminum, zinc, and rare earth metals, can increase the creep resistance of Mg alloys by forming reinforcing precipitates or stabilizing the microstructure (Gavras et al., 2019; Majhi & Mondal, 2019; D. Zhang et al., 2022).

Precipitates in alloys are primarily known for their strength-enhancing properties (Gladman, 1999; Jian Feng Nie, 2012). In AZ series alloy systems, a type of Mg alloy containing aluminum (Al), Al atoms can form a solid solution with Mg atoms, making it difficult for dislocations to move through the material (Cáceres & Rovera, 2001; F. Liu et al., 2019; Prameela et al., 2020). As the material is subjected to more stress over time, Al atoms can precipitate out of the solid solution and form clumps or precipitates. These precipitates then act as barriers to dislocation motion, leading to strain hardening. This is because dislocations have to move around the precipitates, which requires more energy and makes the material harder. However, as the material continues to experience stress, the precipitates can become coarser and lose their ability to block dislocations effectively (Boyd & Nicholson, 1971; Rometsch et al., 2014; D. K. Xu et al., 2006). This can result in faster creep, as the dislocations can no longer be blocked by the precipitates. The formation of these precipitates during creep is known as dynamic precipitation. Depending on the special conditions of the alloy (regional composition, stress, temperature, etc.), changes in the morphology of these precipitates can be observed.

Thermal conductivity and creep behavior are two prominent properties of alloys that are often associated. Thermal conductivity refers to the ability of a material to conduct heat. Since a change in one of the creep or thermal conductivity properties may affect the other, the thermal conductivity and creep behavior in alloys can be evaluated together in close relation (Li et al., 2000; Y. F. Liu et al., 2019; Rudajevová et al., 1999; Webb et al., 2019). The relationship between the thermal conductivity and creep behavior can be explained by the microstructure of the alloy. The alloy composition also plays a critical role in the thermal conductivity and creep behavior. Alloys with high concentrations of alloying elements tend to have lower thermal conductivity and higher creep because these elements can form secondary phases that inhibit heat transfer and affect deformation. On the other hand, alloys with low alloying element concentrations tend to have higher thermal conductivity and lower creep, as the microstructure is more homogeneous and there are fewer barriers to heat transfer and deformation. In addition to the microstructure and composition, temperature is a parameter that affects the thermal conductivity of alloys as well as the creep behavior. At high temperatures, the thermal conductivity of alloys typically decreases, whereas the creep behavior increases. This is due to thermal expansion and breaks in atomic bonds, which can promote deformation and inhibit heat transfer (Y. Wang et al., 2022; Zheng et al., 2019).

The properties of alloys are highly dependent on the atomic arrangement and interactions between constituent elements. Enthalpy of mixing, which is the energy required to mix the atoms of different elements in a solid solution, plays a significant role in determining the stability, phase transformation, and mechanical properties of alloys (Sluiter & Kawazoe, 2002; T. Xu et al., 2022). In the case of Ni-Co alloys, the combination of Ni with other alloying elements such as Ti, Cr, and Fe causes an increase in enthalpy of mixing, leading to a decrease in elongation and an increase in strength and brittleness. This is attributed to the formation of stronger Ni-metal bonds as Ni replaces Co in the alloy (Anand Sekhar et al., 2019; Yeh, 2015; M. Zhang et al., 2019). Similarly, in MnAlCo alloys, the negative enthalpy of mixing between Co and other atoms, combined with the difference in atomic radii, leads to the preferential occupation of Al crystallographic positions by Co atoms. This results in the shrinkage of the t-phase unit cells and contributes to the improvement in the thermal stability of the t-phase in the alloy system (Xiang et al., 2019). Furthermore, the enthalpy of mixing can be used to predict and introduce second phases in alloys through liquid-phase separation. For instance, in CoCrFeMnNi alloys doped with Nd, the positive mixing enthalpies of Nd with Cr and Fe lead to the separation of Nd from the matrix and precipitation with the miscible elements with nonpositive mixing enthalpies, including Ni, Co, and Mn. Among the possible precipitates, Nd-Ni-Mn has the minimum mixing enthalpy and is, therefore, the most favorable precipitate. These precipitates, with hardness three times that of the matrix, effectively strengthened the alloy (C. Wang et al., 2019). Briefly, the enthalpy of mixing is a critical parameter for evaluating the properties and

behavior of alloys, including their stability, phase transformation, and mechanical properties. Understanding the enthalpy of mixing can aid in the design of alloys with desirable properties and the prediction and introduction of second phases through liquid-phase separation.

In this study, thermal conductivity and calculated enthalpy of mixture data, which directly affect the basic properties of the alloys, were used to obtain information about the creep strength of Mg alloys using a different approach. In this study, in addition to AZ alloy series with a wide range of industrial applications, different Mg alloys were included in the evaluation. The main aim of this study is to determine the appropriateness of using thermal and thermodynamic ("*two-T approximation*") data for a comparative analysis of creep behaviors.

2. Materials and Methods

In this study, creep rate data (specifically, steady-state creep) for Mg alloys were acquired by analyzing the results of creep tests conducted in previous research. The creep rates at 453 K (180 ^{o}C) for three AZ series alloys (AZ31, AZ61, and AZ91) were obtained from T. Chen et al.'s work (Chen et al., 2022). Data for the other three Mg alloys (AM50, AX52, and AE42) were obtained from studies in the literature (Ishimatsu et al., 2006; Terada & Sato, 2007; Zhu et al., 2012). Although creep tests for alloys other than the AZ series were conducted at 448 K (175 ^{o}C), the temperature difference is small, and since the study aims to take a qualitative approach, this difference is not expected to be a problem. The thermal conductivity values for the alloys were gathered from pertinent articles in the literature and obtained by interpolation based on the temperature values utilized in the creep tests (Lee et al., 2013; Y. F. Liu et al., 2019; M. Pekguleryuz & Vermette, 2009; Trojanová et al., 2020). On the other hand, the enthalpy of mixing (ΔH_{mix}) of the alloys was determined by the following equation (Eq 1.) according to the normal solution model using the appropriate values (Takeuchi & Inoue, 2010; X. Yang & Zhang, 2012):

$$\Delta H_{mix} = \sum_{i=1, i\neq j}^{n} 4\Delta H_{AB}^{mix} C_i C_j \quad (1)$$

where, ΔH_{AB}^{mix} is the enthalpy of mixing of constituent binary alloys, C_i and C_j is the atomic percentage of the i^{th} and j^{th} component.

3. Findings and Discussion

Graphs showing the relationship between the creep rate (steady state creep) values at 453 K determined from the creep test data in the literature (Chen et al., 2022) of commercially used AZ series Mg alloys and the thermal conductivity and calculated enthalpy of mixing data of these alloys are given in Fig. 1 and Fig. 2, respectively.

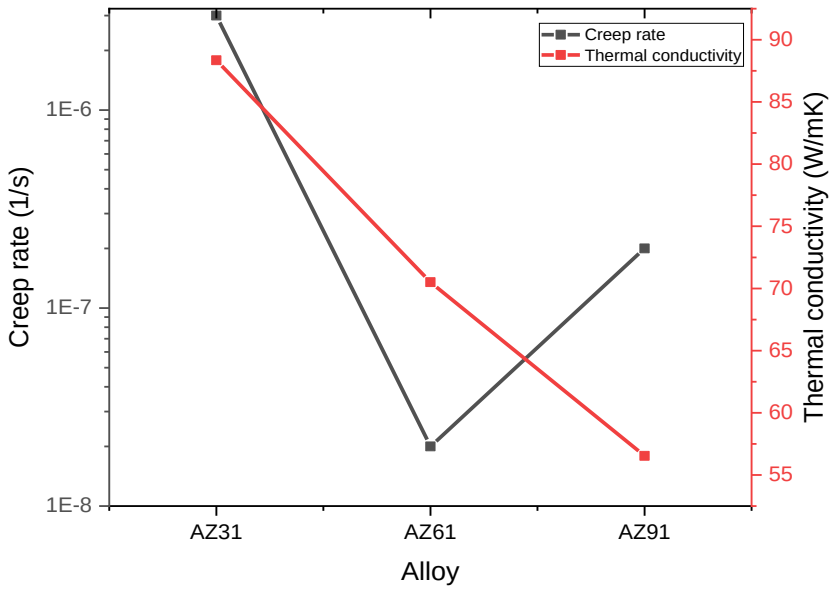

Figure 1. Relationship of creep rate with thermal conductivity in AZ series alloys

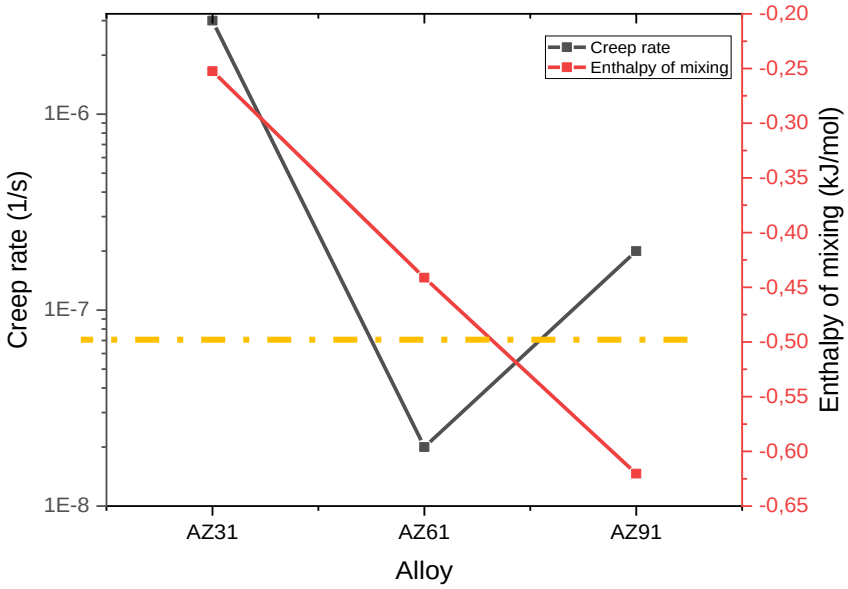

Figure 2. Relationship of creep rate with enthalpy of mixing in AZ series alloys

The higher the creep rate of any metallic material, the lower the creep resistance. From this perspective, when the three AZ series alloys were examined, the order in terms of creep resistance was AZ61, AZ91, and AZ31, from largest to smallest. It is expected that AZ91 will have a higher creep strength than AZ61. However, T. Chen et al. (Chen et al., 2022) showed the opposite in their study. It is obvious that the major factor here is the $Mg_{17}Al_{12}$ intermetallic. The intermetallic compound enhances creep resistance up to a specific threshold owing to its physical and mechanical properties. However, the incrase in Al concentration within the system results in grain coarsening of the $Mg_{17}Al_{12}$ intermetallics as can be observed in Fig. 1 (> ~0.35 µm), which in turn has an adverse impact on creep behavior (Zha et al., 2017). The $Mg_{17}Al_{12}$ intermetallic observed in the AZ alloy series is unstable at high temperatures owing to its low melting point and diffusivity. It has also been reported in the literature that with increasing temperature, the formation of dynamic discontinuous grain boundary precipitation of $Mg_{17}Al_{12}$ and subsequent migration to neighboring grains during grain growth facilitates grain boundary sliding (Hort et al., 2006; Srinivasan et al., 2010).

Literature also suggests that in the AZ91 alloy, the dominant types of precipitates are dynamic discontinuous and coarse continuous particles. Although the pre-creep strengthening mechanism has a significant effect on the strength of the AZ91 alloy, the increasing fraction of dynamic precipitates during creep indicates that the effect of precipitate strengthening is lost as the dissolved alloying elements rapidly decrease. Moreover, with the increasing Al content, larger Mg17Al12 particles form. In tandem with the growth of these particles, there is a greater accumulation of dislocations on them, resulting in an increased overall strength of the alloy. However, this phenomenon leads to stress concentration in adjacent phases, creating a favorable environment for crack initiation. In conclusion, it becomes evident why AZ91 exhibits lower creep strength compared to AZ61 (Chen et al., 2022).

Considering the thermal conductivity and the mixing enthalpy data, it can be seen that it is useful for evaluating the creep behavior of alloys. The thermal conductivity can affect the creep rate in two ways. First, a higher thermal conductivity can lead to better heat dissipation and lower temperatures, thereby reducing the creep rate. Second, thermal conductivity can affect the stress distribution within the material, which in turn affects the creep rate (Dong et al., 2022; Terada et al., 1997). For example, AZ31 has a high thermal conductivity, which means it can dissipate heat quickly and prevent local heating, which can cause creep deformation. Similarly, AZ91 has a low thermal conductivity, which can cause local warming and contribute to its high creep rate; however, as stated above, as in the study of T. Chen et al. (Chen et al., 2022), the data they obtained do not match the expected situation. In other words, the reason for this discrepancy is directly related to the amount of secondary phases formed in the structure. The reason for this discrepancy is actually seen when the mixture enthalpy graph (Fig. 2) is examined. The limit, which is indicated by the yellow dashed line on the graph and roughly coincides with the mixture enthalpy of -0.5, actually allows us to evaluate

this situation more clearly. Thus, besides the thermal conductivity capability of the alloy, it is understood that thermodynamic examination of the formation of stable secondary phases in the structure is also appropriate. The concept of enthalpy of mixing is a thermodynamic property that describes the energy required to mix different elements or compounds. Since this concept relates to the chemical interactions between components, it undoubtedly has an impact on the microstructure and mechanical properties of alloys. Since the concept of creep is directly related to the specified properties, the determination of the enthalpy of the mixture will allow us to create a comparison system from a different perspective on the creep behavior of the relevant alloys.

In addition, Mg alloys containing different elements with similar compositions were also included in the discussion for better analysis and understanding of the situation. Therefore, an evaluation was performed by adding AM50, AX52 Mg and AE42 alloys, which have been studied within the scope of creep, together with the AZ alloy series (see Fig. 3). Additionally, the compositions of all alloys are shared in Table 1.

Table 1. Chemical compositions of alloys

		Alloying element composition (wt%)					
		Al	**Zn**	**Mn**	**Ca**	**RE**	**Mg**
Alloy name	**AZ31**	3,12	0,75	0,3	x	x	Bal.
	AZ61	5,96	0,86	0,22	x	x	Bal.
	AZ91	8,92	0,9	0,18	x	x	Bal.
	AM50	5,42	x	0,28	x	x	Bal.
	AX52	4,98	x	0,29	1,72	x	Bal.
	AE42	3,9	x	0,3	x	2,2	Bal.

Table 2. Creep rate, thermal conductivity and enthalpy of mixing values of alloys

Alloy name	Creep rate (1/s)	Thermal conductivity (W/mK)	Enthalpy of mixing (kJ/mol)
AZ31	3,00E-06	88,35	-0,2524
AZ61	2,00E-08	70,5	-0,4412
AZ91	2,00E-07	56,54	-0,6204
AM50	3,00E-02	65	-0,2784
AX52	6,00E-06	69,76	-0,6312
AE42	5,00E-10	88,6	-0,4003

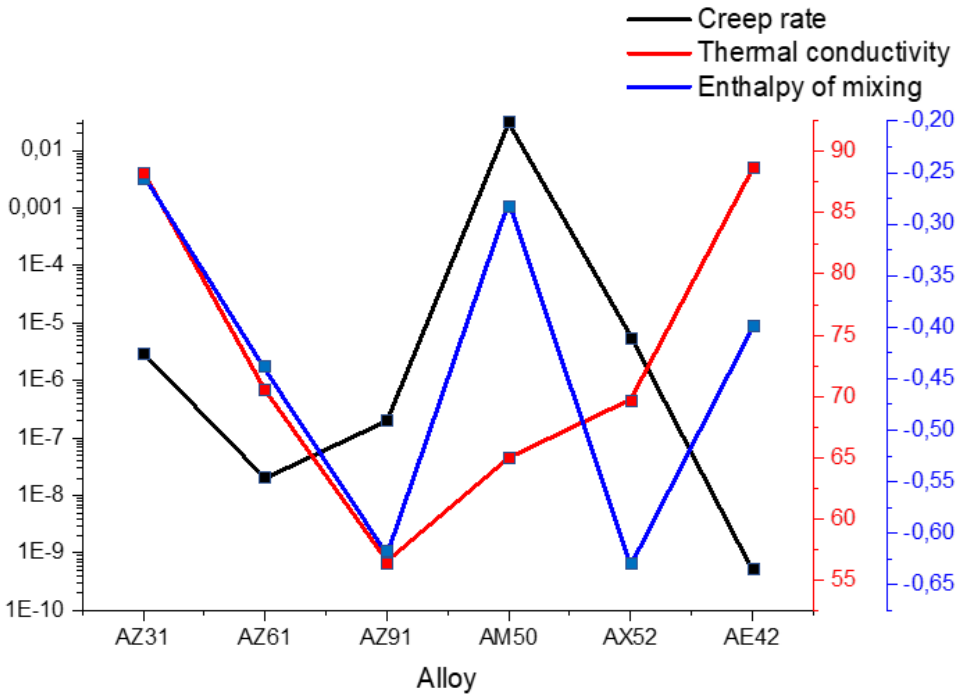

Figure 3. The relationship between thermal conductivity and enthalpy of mixing versus creep rate of all alloys

According to Table 2 and Fig. 3, the AM50 alloy exhibits the lowest creep performance. This is supported by its low thermal conductivity value. Although the mixing enthalpy value of AM50 alloy is as high as that of AZ31 alloy, the relevant phase diagram (see Fig. 4) shows that there is no Mg-Mn based intermetallic formation in the alloy system. The lower level of creep strength of AM50 compared to AZ31 is attributed to an increase in the fraction of $Mg_{17}Al_{12}$ intermetallic in the microstructure due to the higher Al content. The presence of the Mn element, which can be found at a low level in both alloy structures in the literature (Hamu et al., 2009), causes the formation of Al8Mn5 intermetallic in the microstructure. However, an evaluation based on the solid solution concept shows that approximately 1% Mn solubility is observed in α-Mg according to the relevant phase diagram (see Fig. 4) (Nayeb-Hashemi & Clark, 1985). It is evident from the creep rate data that the solid solution does not have a striking effect due to the low Mn content. In fact, the tendency of some alloying elements to form a short range order (SRO) structure can contribute significantly to creep resistance by remarkably limiting dislocation movement (Abaspour & Cáceres, 2015). Indeed, there is no Mn element in the order Miedema suggested in terms of forming SRO with Mg (Kaya, 2020). Moreover, no data on this subject has been reported in the literature.

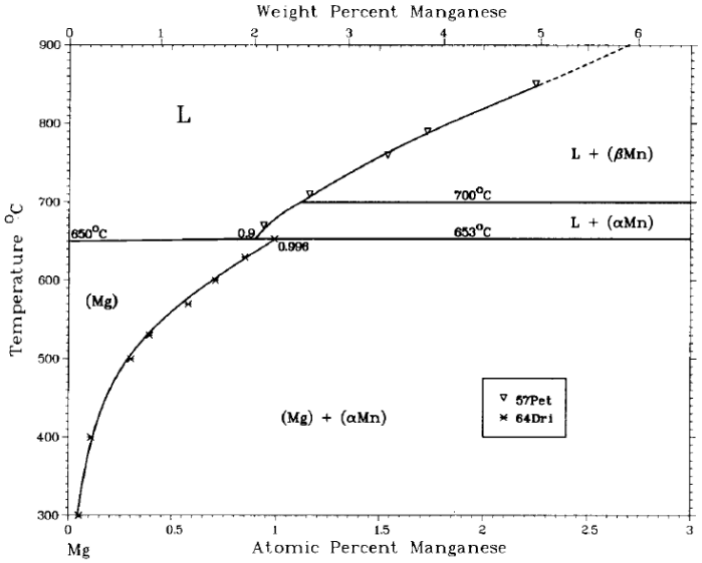

Figure 4. Mg-Mn phase diagram (Nayeb-Hashemi & Clark, 1985)

When the AX52 alloy is examined, it is seen that the thermal conductivity value is higher than AM50. This alloy has approximately the same thermal conductivity as that of AZ61. However, since the enthalpy of mixing value of AX52 is much smaller than that of AZ61, the amount of intermetallic compounds in the structure will be higher. Therefore, there is a significant increase in the creep rate of AX52 compared to that of AZ61. Grain boundary sliding serves as the predominant creep mechanism at temperatures below 190°C. At such temperatures, MgAlCa alloys exhibit notable creep resistance. This can be attributed to the inhibitory effect of Al_2Ca and $(Mg,Al)_2Ca$ eutectic phases (see Fig. 5), which effectively counter grain boundary sliding (Sakai et al., 2020).

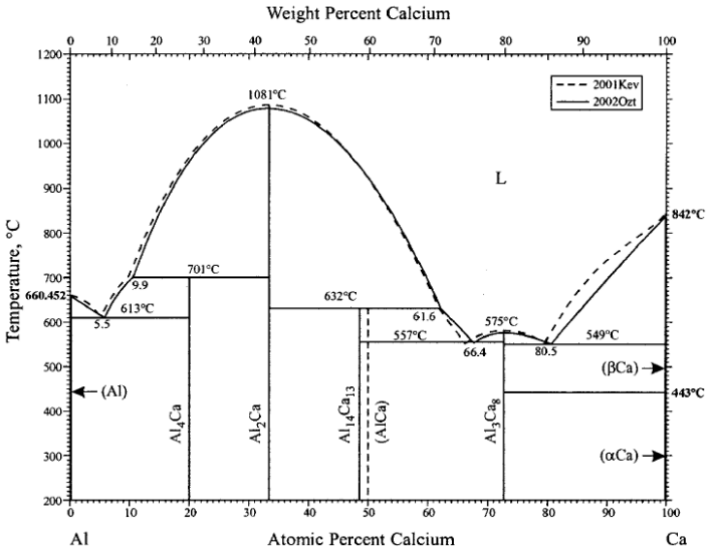

Figure 5. Al-Ca phase diagram (Okamoto, 2003)

From Fig. 3, AE42 alloy has the lowest creep rate. High thermal conductivity and slightly higher mixing enthalpy values directly support this situation. The performance demonstrated by AE42 is actually a good indication that thermal conductivity and enthalpy of mixing values can be effectively used when evaluating creep resistance. The thermally stable dendritic intermetallic phases $Al_{11}RE_3$ and Al_2RE are responsible for the excellent creep behavior of the AE42 alloy. The phase diagrams of the Al-Re system, with a focus on cerium (Ce) and lanthanum (La) in terms of concentration, are presented in Fig. 6. Moreover, the rare earth (RE) elements have a positive effect on the creep strength by reducing the aluminum concentration to be used in the formation of the $Mg_{12}Al_{17}$ intermetallic in Mg-Al alloy systems, thereby decreasing the formation of $Mg_{17}Al_{12}$, which has a negative impact on creep resistance (Zhu et al., 2008).

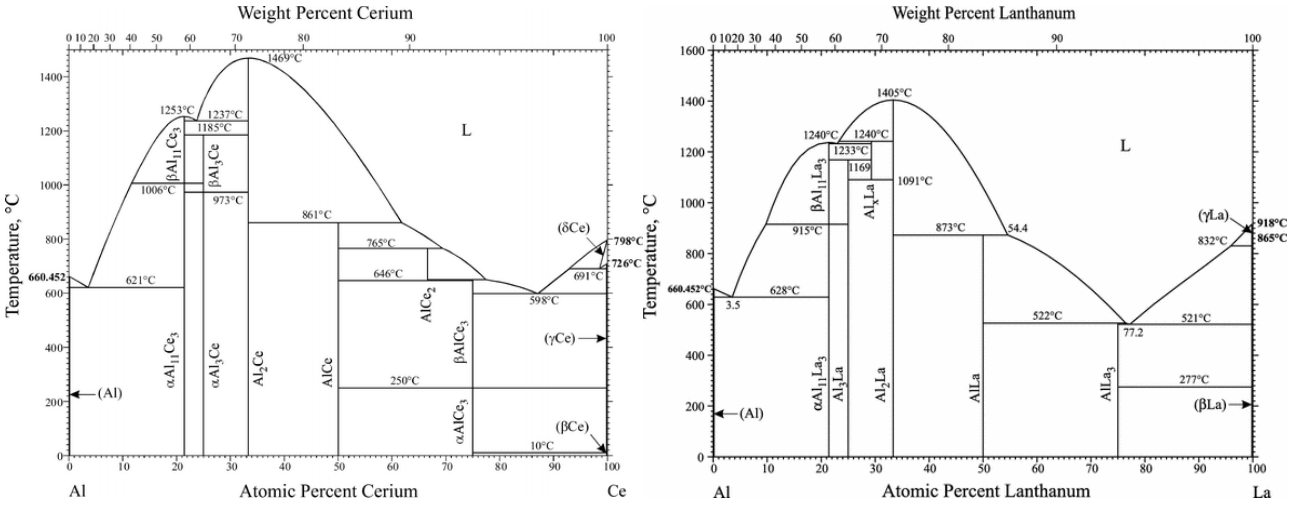

Figure 6. Al-Ce and Al-La phase diagrams (Okamoto, 2007, 2011)

4. Conclusions and Recommendations

In this study, the creep rates of AM50, AX52 and AE42 alloys and the commonly used AZ series alloys were evaluated using the concepts of thermal conductivity and enthalpy of mixing. These two included parameters were found to be appropriate for the qualitative evaluation of alloys in terms of creep. With this approach, the reasons for the differentiation in AZ series alloys have been revealed more clearly. Considering the future, data on different alloys will be obtained based on the approach presented here. As the number of these data increases, it will be possible to obtain information about the creep behavior by processing the mixing enthalpy and thermal conductivity data of the alloy with machine learning-based software.

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Statement of Conflicts of Interest

There is no conflict of interest between the authors.

Statement of Research and Publication Ethics

The author declares that this study complies with Research and Publication Ethics.

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