

Calculation of the effective solidification range and its relationship with hot brittleness of alloys based on Mg–Al and Mg–Zn systems

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Abstract: Magnesium alloys of the Mg–Al and Mg–Zn systems have a wide effective solidification range (ESR), and as a result, have the tendency to hot brittleness during casting. There are several methods for analyzing and calculating the hot brittleness of magnesium alloys, but they are very labor-intensive. In this regard, the objective of the study is to develop a model for calculating the hot brittleness index (HBI) based on the value of the calculated effective solidification range, identifying and analyzing their relationship in binary and multicomponent alloys based on the Mg–Al and Mg–Zn systems. The ESR was calculated using the Thermo-Calc program (TTMG3 database). The ESR was calculated as the difference between the temperature of formation of a given amount of solid phases and the nonequilibrium solidus temperature. The study showed a good correlation between the calculated values of ESR and HBI in both binary and multicomponent magnesium alloys. In the Mg–Al system alloys, the calculated dependences of the ESR at 90 % of solid phases (ESR_{90}) show the best correlation with the experimental values of HBI. In the binary alloys of the Mg–Zn system, a qualitatively similar dependence is observed. However, no clear correlation was noted between the ESR and HBI. The ESR_{65} and ESR_{80} dependences demonstrate the closest nature. According to the relationship between HBI and ESR, the considered multicomponent alloys are divided into two groups as a first approximation: the first one is the Mg–Al–Zn system alloys; the second one is the Mg–Zn–Zr and Mg–Nd–Zr alloys. Within these groups, the dependence of HBI and ESR has a nature close to a linear one. To describe the dependence of all alloys, a single equation can be applied if ESR_{65} is used in the calculations for Mg–Al–Zn alloys and ESR_{90} – for Mg–Zn–Zr and Mg–Nd–Zr alloys. The proposed model will allow for easy and quick calculation of the HBI, which is very important in the development of new high-tech magnesium alloys.

Keywords: magnesium alloys; hot brittleness; effective solidification range; thermodynamic calculation; nonequilibrium solidification.

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INTRODUCTION

Magnesium alloys combine a good level of mechanical properties with low density. Almost all cast magnesium alloys belong to two basic alloying systems – Mg–Al and Mg–Zn [1; 2]. Alloys of the Mg–Al and Mg–Zn systems have a very wide solidification range [3]. In this regard, when selecting the composition of cast alloys, it is necessary to take into account the possibility of narrowing the solidification range, or more precisely, the effective solidification range (ESR) – the range between the temperature of the onset of linear shrinkage and the nonequilibrium solidus of the alloy [3]. It can be said that the ESR value directly determines the tendency of the alloy to form cracks of solidification origin or hot brittleness [3; 4]. The appearance of solidification cracks in castings is an irreparable defect. Experimental determination of the ESR value is a very labor-intensive process [3; 4]. The authors of the work [5] proposed a method for calculating the ESR value using thermodynamic calculations of nonequilibrium solidification for binary and ternary aluminum-based alloys. It was shown that it is the effective solidification range, and not the full solidification interval, that determines the manu-

facturability during casting [6], and the possibility of calculating the ESR and the hot brittleness index (HBI) in multicomponent aluminum-based alloys was identified [7].

The problem of the formation of cracks of solidification origin is no less acute during casting of castings from magnesium alloys [8–10]. There are several methods for analyzing and calculating the hot brittleness of magnesium alloys, but they are very labor-intensive [11–14]. For example, the authors of the work [11] estimated the tendency to form cracks of solidification origin based on the characteristics of mechanical properties in the solid-liquid state and the location of the layers of the last liquid. The results of [12] are based on the optimization of the Clyne – Davies model and use a very narrow parameter for determining the HBI on a 4-point scale. In the work [14], a complex and inaccessible method of neutron diffraction was used to analyze the initiation of hot cracks. The proposed approach is faster and sufficiently accurate.

When designing castings for casting from known magnesium alloys, when developing new casting magnesium alloys, it is necessary to take into account the level of casting properties, especially the HBI. Cracks of solidification

origin can also form in ingots of deformable magnesium alloys. As shown in the example of aluminum alloys, the HBI is directly related to the ESR value. Therefore, there is a need to analyze the relationship between the ESR and the HBI value for magnesium alloys. Identifying patterns similar to those obtained in aluminum alloying systems will allow using this technique for calculating the HBI, which is especially important when developing new magnesium alloys. This study is aimed at adapting the method for calculating the ESR for binary and multicomponent alloys based on the Mg–Al and Mg–Zn systems and analyzing the relationship between the calculated ESR and the experimentally determined HBI.

The aim of this study is to develop a model for calculating the hot brittleness index based on the value of the calculated effective solidification range, establishing and analyzing their relationship in binary and multicomponent alloys based on the Mg–Al and Mg–Zn systems.

METHODS

The calculation of the ESR was performed using the Thermo-Calc software product (TTMG3 magnesium alloy database). The ESR was calculated as the difference between the temperature of formation of a given amount of solid phases (from 65 to 90 %) and the nonequilibrium solidus temperature. Additionally, the values of the total equilibrium solidification range TSR_{eq} were calculated as the difference between the liquidus temperature and the equilibrium solidus temperature; the total nonequilibrium solidification range TSR_{noneq} – as the difference between the liquidus temperature and the nonequilibrium solidus temperature. The calculation of nonequilibrium solidification is based on the Sheil model, which assumes complete passage of diffusion processes in the liquid and complete suppression of diffusion in the solid phase. To compare the calculated values of ESR and TSR with hot brittleness, experimental data on the HBI of binary and multicomponent alloys from work [3] were used, which are in good agreement with more recent information on binary Mg–Zn alloys [14; 15], Mg–Zn alloys with the addition of Y [16] or Y and Zr [17–19], Mg–Al alloys [20–22]. The HBI of magnesium alloys in work [3] was determined by one of the most common tests for magnesium and aluminum alloys – the ring test. In this test, the melt is poured into a sand mold with a metal rod and a refrigerator on half of the ring, and the critical width of the ring in millimeters is determined.

RESULTS

For binary alloys of the Mg–Al and Mg–Zn systems in the concentration range from 0 to 6 % of Al and from 0 to 10 % of Zn, nonequilibrium solidification curves were constructed. Fig. 1 presents typical dependences of the change in the mass fraction of solid phases during nonequilibrium solidification of Mg–0.75%Al, Mg–1.5%Al, Mg–1%Zn, and Mg–2%Zn binary alloys. The solidification process of the Mg–0.75%Al alloy (Fig. 1 a) begins at a temperature of 646 °C with the formation of a magnesium solid solution (Mg). Under equilibrium condi-

tions, solidification ends at 637 °C, in accordance with the dotted curve. Under nonequilibrium conditions, solidification ends at 553 °C, in accordance with the solid curve. As a result, TRS_{eq} and TRS_{noneq} are 9 and 93 °C, respectively.

As mentioned above, the ESR is the interval between the temperature of the onset of linear shrinkage and the nonequilibrium solidus. The temperature of the linear shrinkage onset usually corresponds to the formation of 65–90 % of solid phases in the alloy. It is for this range of the amount of solid phases that the ESR values were calculated (Table 1). Using the Mg–0.75%Al alloy as an example, Fig. 1 a shows the intervals corresponding to the calculated ESR values for 65, 80 and 90 % of the solid phases. For comparison, the solidification process of the Mg–1.5%Al alloy (Fig. 1 b) begins at 642 °C and ends under equilibrium conditions at 624 °C, and under nonequilibrium conditions – at 437 °C. In this case, the solidification of the Mg–1.5%Al alloy (Fig. 1 b) ends with the formation of the $Mg_{17}Al_{12}$ phase in the eutectic with (Mg). A similar solidification pattern was observed in the Mg–1%Zn (Fig. 1 c) and Mg–2%Zn (Fig. 1 d) alloys. The obtained calculation data were used to construct a nonequilibrium solidus in the Mg–Al and Mg–Zn systems. All critical temperatures of formation of 65, 80 and 90 % solid phases, nonequilibrium and equilibrium solidus and liquidus ($T_{65\%}$, $T_{80\%}$, $T_{90\%}$, $T_{n.s.}$, $T_{eq.s.}$, $T_{liq.}$), calculated ESR values corresponding to formation of 65, 80 and 90 % solid phases (ESR_{65} , ESR_{80} , ESR_{90}), calculated values of equilibrium and nonequilibrium TSR (TSR_{eq} , TSR_{noneq}) for all binary alloy compositions (first column of the table) are collected in Table 1. The calculation results are used to analyze the relationship of ESR and TSR with the experimental HBI values in binary alloys of the Mg–Al and Mg–Zn systems.

Fig. 2 shows as an example the dependences of the mass fraction of solid phases in the process of nonequilibrium solidification of multicomponent ML5 (Mg–Al–Zn) and ML10 (Mg–Nd–Zr) alloys. Similar dependences were used to calculate the critical temperatures and intervals for all multicomponent alloys. The solidification process of the ML5 alloy begins at 629 °C with the formation of the Al_8Mn_5 phase (Fig. 2 a). Under equilibrium conditions, solidification ends at 489 °C, and under nonequilibrium conditions – at 412 °C, which corresponds to $TSR_{eq}=140$ °C and $TSR_{noneq}=217$ °C (Table 2). In the process of nonequilibrium solidification, the (Mg), $Al_{11}Mn_4$ and Al_4Mn phases are formed by eutectic and peritectic reactions, and the $Al_{11}Mn_4$ and Al_8Mn_5 phases dissolve (Fig. 2 a). Solidification of the ML10 alloy begins at 737 °C with the formation of the α -Zr phase, then the (Mg) and $Mg_{41}Nd_5$ phases appear (Fig. 2 b). In this case, in the ML10 alloy, $TSR_{noneq}=237$ °C (Table 2), i.e. wider than in the ML5 alloy. However, in this case, ESR_{65} in the ML10 alloy is significantly narrower – 55 °C versus 139 °C in the ML5 alloy (Table 2). Results of calculations of critical $T_{65\%}$, $T_{80\%}$, $T_{90\%}$, $T_{n.s.}$, $T_{eq.s.}$, and $T_{liq.}$ temperatures and ESR_{65} , ESR_{80} , ESR_{90} , TSR_{eq} , TSR_{noneq} intervals for different multicomponent alloys based on the Mg–Al–Zn (ML1, ML2, ML3, ML4, ML5, ML6, ML7-1), Mg–Zn–Zr (ML12, ML15), and Mg–Nd–Zr (ML10) systems are collected in Table 2. The obtained calculated data on the ESR values are analyzed in comparison with the experimentally determined HBI.

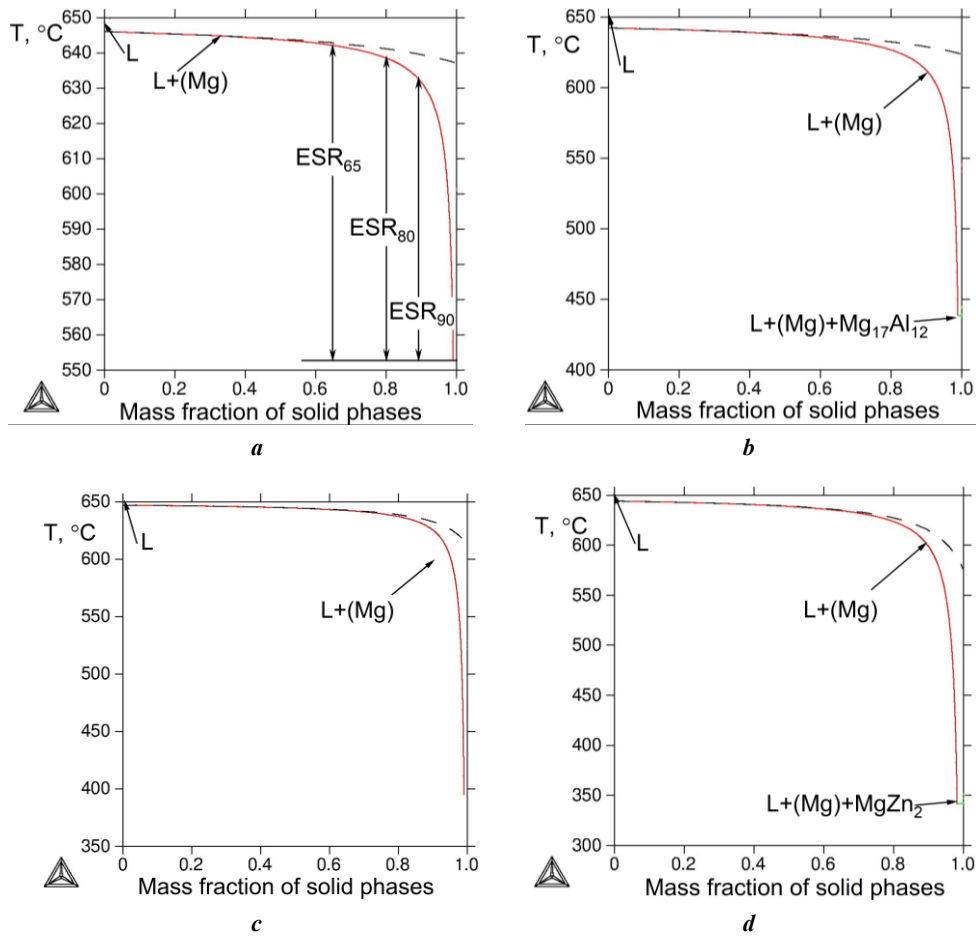


Fig. 1. Change in the mass fraction of solid phases during nonequilibrium solidification (dashed line – equilibrium solidification) of binary alloys: **a** – Mg–0.75%Al; **b** – Mg–1.5%Al; **c** – Mg–1%Zn; **d** – Mg–2%Zn (L – liquid phase)

Рис. 1. Изменение массовой доли твердых фаз в процессе неравновесной кристаллизации (пунктир – равновесная кристаллизация) двойных сплавов: **a** – Mg–0,75%Al; **b** – Mg–1,5%Al; **c** – Mg–1%Zn; **d** – Mg–2%Zn (L – жидкая фаза)

Table 1. Values of calculated temperatures and ranges for alloys of the Mg–Al and Mg–Zn systems
Таблица 1. Значения расчетных температур и интервалов для сплавов систем Mg–Al и Mg–Zn

Al, %	T ₆₅ %, °C	T ₈₀ %, °C	T ₉₀ %, °C	T _{n.s.} , °C	T _{eq.s.} , °C	T _{liq} , °C	ESR ₆₅ , °C	ESR ₈₀ , °C	ESR ₉₀ , °C	TSR _{eq} , °C	TSR _{noneq} , °C
Mg–Al											
0.1	649	648.5	648	642	648	649.3	7	6.5	6	1.3	7.3
0.5	645	643	638	590	641	647	55	53	48	6	57
0.75	642	638	631	553	637	646	89	85	78	9	93
1	639	635	623	511	632	645	128	124	112	13	134
1.25	636	631	617	464	629	643	172	167	153	14	179
1.5	634	624	611	437	624	642	197	187	174	18	205
2	628	618	598	437	615	640	191	181	161	25	203
4	607	584	535	437	582	630	170	147	98	48	193
6	583	545	465	437	542	618	146	108	28	76	181

Mg–Zn											
0.1	649.2	648.6	647.8	632	646	649.7	17.2	16.6	15.8	3.7	17.7
0.5	646	643	638	540	634	648	106	103	98	14	108
1	643	637	626	396	619	647	247	241	230	28	251
1.5	638	630	6112	341	600	645	297	289	5771	45	304
2	635	623	599	341	577	644	294	282	258	67	303
3	626	608	568	341	548	641	285	267	227	93	300
4	618	596	536	341	502	638	277	255	195	136	297
6	599	563	455	341	424	632	258	222	114	208	291
10	560	485	341	341	341	618	219	144	0	277	277

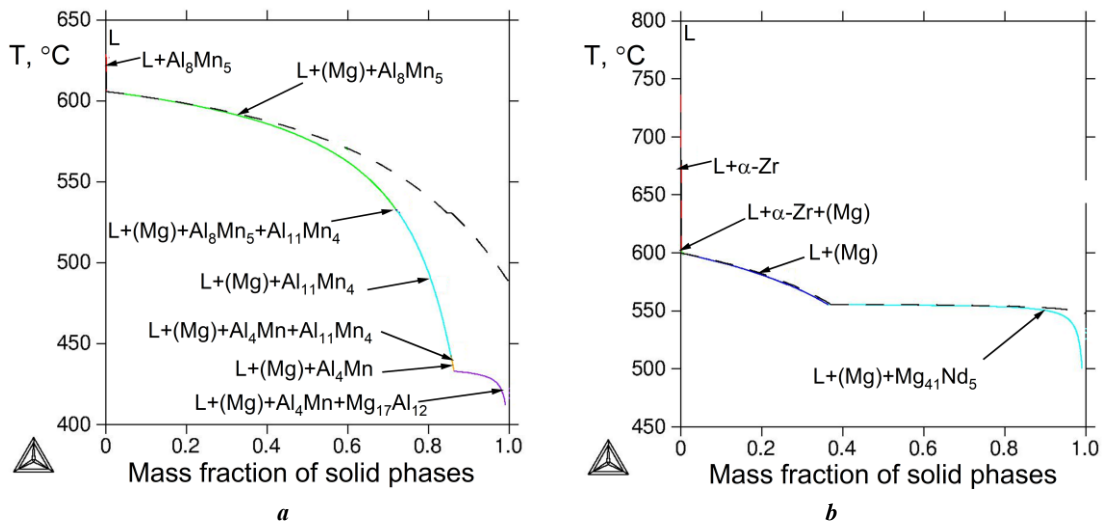


Fig. 2. Change in the mass fraction of solid phases in the process of nonequilibrium solidification (dashed line – equilibrium solidification) of multicomponent alloys: a – ML5; b – ML10

Рис. 2. Изменение массовой доли твердых фаз в процессе неравновесной кристаллизации (пунктир – равновесная кристаллизация) многокомпонентных сплавов: а – МЛ5; б – МЛ10

DISCUSSION

Based on the results of calculating the nonequilibrium solidification of binary alloys, the nonequilibrium solidus lines were plotted in the magnesium angle of the Mg–Al and Mg–Zn systems (Fig. 3 a, 3 c). During solidification of the Mg–1.5%Al and Mg–2%Zn alloys, a nonequilibrium excess of the Mg₁₇Al₁₂ phase appears at a temperature of 437 °C (Fig. 1 b) and MgZn₂ at a temperature of 341 °C (Fig. 1 d). Fig. 3 b, 3 d shows the dependences of the calculated values of ESR (for different amounts of solid phases), TSR_{eq}, TSR_{noneq} and the experimental values of HBI for binary alloys of the Mg–Al and Mg–Zn systems. The calculated values of TSR_{eq} do not correlate with the values of ESR and, accordingly, with HBI. In this case, as in aluminum alloys of eutectic systems [5; 6], the TSR_{noneq} values are in good agreement with the ESR value. In the Mg–Al

system alloys (Fig. 3 b), the calculated dependences of ESR₉₀ show the best correlation with the experimental values of HBI. The rate of decrease in HBI practically coincides with the rate of decrease in the calculated value of ESR₉₀. In binary alloys of the Mg–Zn system, a qualitatively similar dependence is observed (Fig. 3 d). However, no clear correlation was noted between ESR and HBI. The ESR₆₅ and ESR₈₀ dependences demonstrate the closest nature (Fig. 3 d).

The position of the HBI maximum somewhat diverges from the maximum calculated ESR value. Thus, in the Mg–Al system, an alloy with 1 % of Al has the maximum HBI, which is confirmed by the data of work [9; 10], and the Mg–1.5%Al alloy has the highest calculated ESR. It is worth noting that in works [9; 10], alloys containing more than 1 but less than 2 % of Al were not considered.

Table 2. Values of calculated temperatures and ranges for multicomponent alloys
Таблица 2. Значения расчетных температур и интервалов для многокомпонентных сплавов

Alloy	T _{65 %} , °C	T _{80 %} , °C	T _{90 %} , °C	T _{n.s.} , °C	T _{eq.s.} , °C	T _{liq} , °C	ESR ₆₅ , °C	ESR ₈₀ , °C	ESR ₉₀ , °C	TSR _{eq} , °C	TSR _{noneq} , °C
ML3	610	589	540	363	564	631	247	226	177	67	268
ML4	562	503	405	350	464	612	212	153	55	148	262
ML5	551	489	431	412	489	629	139	77	19	140	217
ML6	531	451	426	389	460	630	142	62	37	170	241
ML7-1	579	542	445	400	487	640	179	142	45	153	240
ML10	555	553	548	500	548	737	55	53	48	189	237
ML12	612	585	514	339	482	732	273	246	175	250	393
ML15	608	573	498	339	477	730	269	234	159	253	391

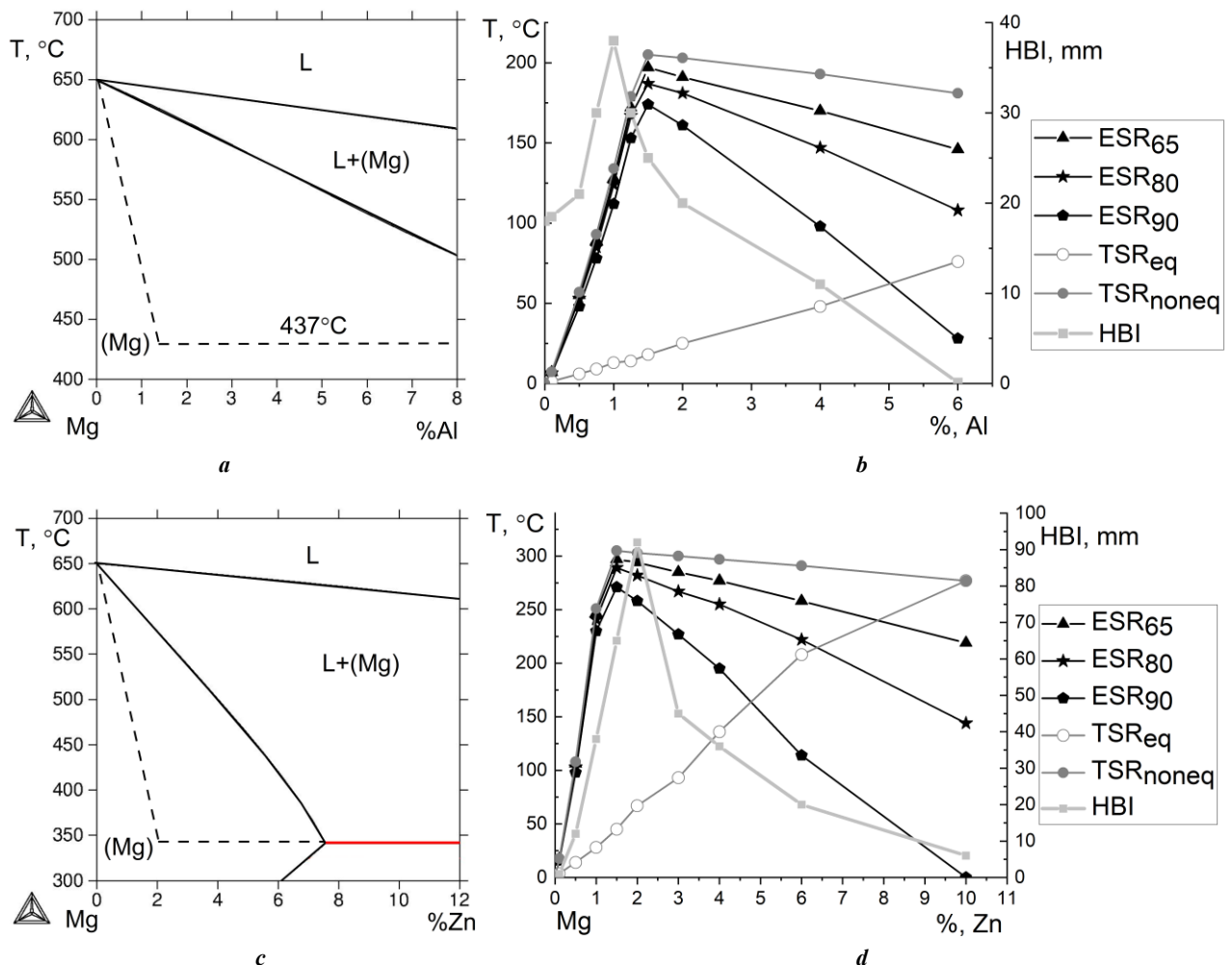


Fig. 3. Magnesium angle of the phase diagrams of the Mg–Al (a) and Mg–Zn (c) systems (dotted line – nonequilibrium solidus) and the dependences of the calculated values of effective solidification range ESR (for different amounts of solid phases), total equilibrium solidification range TSR_{eq}, total nonequilibrium solidification range TSR_{noneq}, and experimental values of the hot brittleness index HBI for binary alloys of the Mg–Al (b) and Mg–Zn (d) systems

Рис. 3. Магнийевый угол фазовых диаграмм систем Mg–Al (a) и Mg–Zn (c) (пунктир – неравновесный солидус) и зависимости расчетных значений эффективного интервала кристаллизации ESR (для разного количества твердых фаз), полного равновесного интервала кристаллизации TSR_{eq}, полного неравновесного интервала кристаллизации TSR_{noneq} и экспериментальных значений показателя горячеломкости HBI для двойных сплавов систем Mg–Al (b) и Mg–Zn (d)

In the Mg–Zn system, the maximum hot brittleness index is found at 2 % of Zn [3], however, according to the data of works [9; 11], the most hot-brittle alloy is Mg–1.5%Zn, which has the highest calculated ESR (Fig. 3).

The nature of solidification of multicomponent alloys is somewhat more complicated (Fig. 2). Mg–Zn–Zr system alloys (just as Mg–Zn) have a low nonequilibrium solidus, and accordingly, a very wide ESR, but at the same time approximately the same tendency to form solidification cracks as Mg–Al–Zn alloys, whose ESR is approximately 100 °C narrower (Table 3). The ML10 alloy (Mg–Nd–Zr system), having a high temperature of nonequilibrium soli-

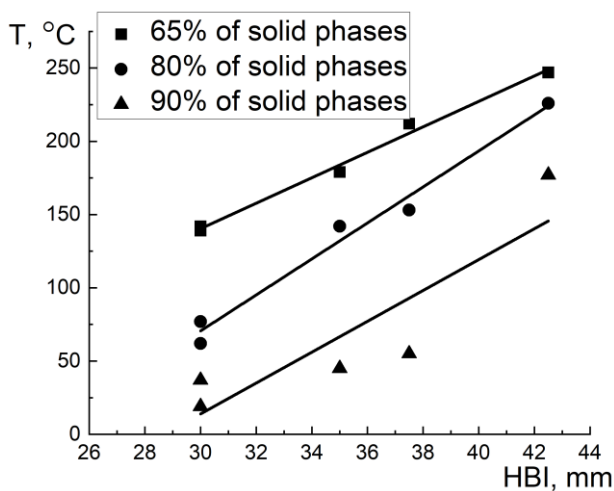
us, is distinguished by a very narrow ESR (48–55 °C) and low HBI (Table 3). The calculated values of solidification intervals in comparison with the experimental HBI for multicomponent magnesium alloys are collected in Table 3.

According to the relationship between HBI and ESR, the considered multicomponent alloys are divided into two groups: the first one is the Mg–Al–Zn system alloys; the second one is the alloys of the Mg–Zn–Zr and Mg–Nd–Zr systems. Within these groups, the dependence of HBI and ESR has a nature close to a linear one, hot brittleness index increases with an increase in the ESR (Fig. 4). At the same time, for the Mg–Al–Zn system alloys, all points lie as

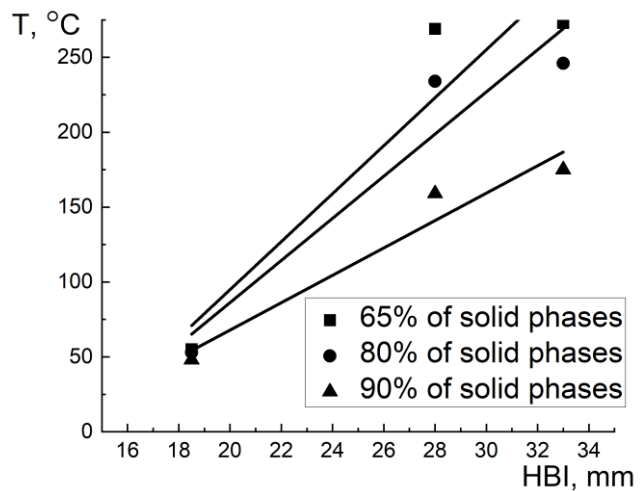
Table 3. Effective solidification range ESR and total equilibrium solidification range TSR values in comparison with the experimental hot brittleness index for multicomponent alloys

Таблица 3. Значения эффективного интервала кристаллизации ESR и полного равновесного интервала кристаллизации TSR в сравнении с экспериментальным показателем горячеломкости для многокомпонентных сплавов

Alloy	ESR ₆₅ , °C	ESR ₈₀ , °C	ESR ₉₀ , °C	TSR _{eq} , °C	TSR _{noneq} , °C	HBI, mm
ML3	247	226	177	67	268	42.5
ML4	212	153	55	148	262	37.5
ML5	139	77	19	140	217	30
ML6	142	62	37	170	241	30
ML7-1	179	142	45	153	240	35
ML10	55	53	48	189	237	18.5
ML12	273	246	175	250	393	33
ML15	269	234	159	253	391	28



a



b

Fig. 4. Comparison of calculated effective solidification range ESR values (for different amounts of solid phases) and experimental values of the hot brittleness index HBI for multicomponent alloys based on the systems: **a** – Mg–Al–Zn; **b** – Mg–Zn–Zr and Mg–Nd–Zr

Рис. 4. Сопоставление расчетных значений эффективного интервала кристаллизации ESR (для разного количества твердых фаз) и экспериментальных значений показателя горячеломкости HBI для многокомпонентных сплавов на основе систем: **a** – Mg–Al–Zn; **b** – Mg–Zn–Zr и Mg–Nd–Zr

closely as possible on the linear dependence for the calculated values of ESR₆₅ (Fig. 4 a). The obtained dependence can be described by a simple linear equation with a confidence probability of $R^2=0.99$:

$$\text{HBI}=0.11\times\text{ESR} + 14. \quad (1)$$

For the Mg–Zn–Zr and Mg–Nd–Zr systems, the calculated ESR₉₀ values demonstrate the dependence closest to linear (Fig. 4 b). For this group, the dependence can be described by a simple linear equation with a lower confidence probability of $R^2=0.94$:

$$\text{HBI}=0.1\times\text{ESR}+13.2. \quad (2)$$

Equations (1) and (2) for the two groups have close coefficients. Combining the data from the two groups obtained for ESR for different amounts of solid phases allows distinguishing a linear dependence with a confidence probability of $R^2=0.96$:

$$\text{HBI}=0.12\times\text{ESR}+12.5. \quad (3)$$

Equation (3) can be applied in the first approximation to both groups of alloys, if ESR₆₅ is used in the calculations for Mg–Al–Zn alloys and ESR₉₀ – for Mg–Zn–Zr and Mg–Nd–Zr alloys. However, it is not entirely correct to consider alloys of different systems with very different nonequilibrium solidus within the same group.

In general, the study showed the possibility of assessing the tendency of magnesium alloys of the Mg–Al and Mg–Zn systems to form cracks of solidification origin based on the calculation of the effective solidification range. All deviations of the calculated ESR values and experimental HBI values are due to other factors affecting hot brittleness, such as mechanical properties in the solid-liquid state and modification [3; 4]. For example, zirconium in wide-range Mg–Zn alloys can act as a modifier, which improves casting properties. The proposed equation for calculation requires experimental verification on alloys of other compositions not used in the calculations. However, such a possibility is not currently available. Based on the existing successful experience of applying a similar model using the example of creating new aluminum alloys [23; 24], the proposed method for calculating the hot brittleness index will also work in magnesium alloys.

CONCLUSIONS

The calculations of the temperatures of formation of a given amount of solid phases, effective and total solidification ranges in binary and multicomponent alloys based on the Mg–Al and Mg–Zn systems are carried out using thermodynamic calculations of nonequilibrium solidification. A good correlation of the calculated values of the effective solidification range with the hot brittleness index in both binary and multicomponent magnesium alloys is shown. According to the relationship between HBI and ESR, the considered multicomponent alloys are divided into 2 groups in the first approximation: the first group is the alloys of the Mg–Al–Zn

system; the second group is the Mg–Zn–Zr and Mg–Nd–Zr alloys. Within these groups, the dependence of HBI and ESR has a nature close to a linear one.

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Расчет эффективного интервала кристаллизации и его связь с горячеломкостью сплавов на основе систем Mg–Al и Mg–Zn

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Аннотация: Магниево-цинковые сплавы систем Mg–Al и Mg–Zn имеют широкий эффективный интервал кристаллизации (ЭИК) и, как следствие, склонны к горячеломкости при литье. Существует несколько методик анализа и расчета горячеломкости магниево-цинковых сплавов, но они являются очень трудоемкими. В связи с этим целью исследования – разработать модель расчета показателя горячеломкости (ПГ) по величине расчетного эффективного интервала

кристаллизации, установив и проанализировав их связь в двойных и многокомпонентных сплавах на основе систем Mg–Al и Mg–Zn. Расчет ЭИК проведен с использованием программы Thermo-Calc (база данных TTMG3). ЭИК рассчитывался как разница между температурой образования заданного количества твердых фаз и температурой неравновесного солидуса. Показана хорошая корреляция рассчитанных значений ЭИК с ПГ как в двойных, так и в многокомпонентных магниевых сплавах. В сплавах системы Mg–Al расчетные зависимости ЭИК при 90 % твердых фаз (ЭИК₉₀) показывают наилучшую корреляцию с экспериментальными значениями ПГ. В двойных сплавах системы Mg–Zn наблюдается качественно такая же зависимость. Однако четкой корреляции между ЭИК и ПГ не отмечено. Наиболее близкий характер демонстрируют зависимости ЭИК₆₅ и ЭИК₈₀. По связи ПГ и ЭИК рассмотренные многокомпонентные сплавы в первом приближении разделены на 2 группы: первая – сплавы системы Mg–Al–Zn, вторая – Mg–Zn–Zr и Mg–Nd–Zr. В пределах этих групп зависимость ПГ и ЭИК имеет близкий к линейному характер. Для описания зависимости всех сплавов можно применить одно уравнение при условии использования в расчетах ЭИК₆₅ для сплавов системы Mg–Al–Zn и ЭИК₉₀ для сплавов систем Mg–Zn–Zr и Mg–Nd–Zr. Предложенная модель позволит легко и быстро произвести расчет ПГ, что весьма актуально при разработке новых высокотехнологичных магниевых сплавов.

Ключевые слова: магниевые сплавы; горячеломкость; эффективный интервал кристаллизации; термодинамические расчеты; неравновесная кристаллизация.

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