Effect of vanadium on Fe-rich phase, mechanical properties and thermal conductivity of hypoeutectic Al–Si alloy

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Abstract

In this paper, the effects of element V addition (0, 0.2, 0.4, 0.6, 0.8 and 1.0 wt%) on microstructures, mechanical properties and thermal conductivity of Al–8Si–0.1Cu–0.6Mg–0.7Fe hypoeutectic alloy in gravity die cast process were investigated. Microstructure analysis indicates that the size of Fe-rich phase in the Al–Si alloy is greatly affected by the contents of element V addition. The modification of element V could decrease the length of needle-like $\beta$-Fe phase which has the adverse impact to mechanical properties, and promote the formation of short rod-like $\alpha$-Fe phase which has the favorable effect on mechanical properties and thermal conductivity. Then, the dependencies of mechanical properties and thermal conductivity on the size of Fe-rich phase of the alloy is revealed. When the content of element V is 0.6%, the length of Fe-rich phase in the alloy reaches the minimum, and correspondingly both the mechanical properties and the thermal conductivity could achieve the optimal level, as comparison with other contents of element V addition. However, excessive V addition would form VSi$_2$ phase and Fe-rich phase with large size, resulting in poor properties of the alloy.

1. Introduction

Due to the excellent process performance, low density and excellent corrosion resistance [1], good thermal conductivity and thermal stability [2] of Al–Si Alloy, it can be used to replace many traditional steel materials to reduce the weight of parts. So far, it has been widely applied in the fields of automobile [3], aerospace and so on. With the development of communication technology, the combination of mechanical properties and thermal conductivity of materials for base station radiator is urgently required, and Al–Si alloy has the ability to meet the requirements. At present, from the perspective of cost saving, energy saving [4] and environmental protection [5], recycled Al alloy has become the main raw material of Al alloy industry, because of the genetic effect of metals, Fe will inevitably enter the Al alloy while melting. It is generally accepted that the presence of Fe in Al–Si alloy will greatly reduce mechanical properties, while the positive effect of Fe on demoulding property [6] and thermal stability [7] in die casting process is often ignored. In the practical process, in order to ensure the demoulding ability of castings, a certain amount of Fe element will be artificially added into the Al alloy to ensure the smooth demoulding of castings. Therefore, it is of great practical significance to investigate the reduction of the adverse effect of element Fe addition on mechanical properties of Al–Si alloy with good demoulding performance.

Because the solubility of Fe in liquid Al is higher than that in $\alpha$-Al matrix in Al–Si alloy, Fe element generally exists in the Fe-rich phase with the formation of forms intermetallic compounds through combination with other elements. The Fe-rich phase with long rod-shape would cause stress concentration in Al–Si alloy, and would seriously split $\alpha$-Al matrix due to its sharp edge. At present, there are two commonly used methods to weaken the adverse effect of Fe on the properties of Al–Si alloys. The first method is physical approaches, such as electromagnetic Fe removal technology [8], filtration Fe removal technology [9], centrifugal Fe removal...
technology, etc. However, it could not retain the beneficial effect of Fe on demoulding property of the alloy. The second method is to add moderate neutralizing elements to modify Fe-rich phase, such as Mn [10], Cr [11], etc. The addition of Mn in Al–Si alloy can transform the needle-like Al5FeSi (β-Fe) [12] with greater harm into the plum-shaped Al15Fe3(Si2 (α-Fe) [13] or Fe8Mg3FeSi6 (π-Fe) [14]. However, there is a serious deficiency, that is, the addition of Mn will increase the amount of Fe phase in Al–Si alloy, and the amount of sludges and the porosity increases, resulting in the great reduction of the ductility of Al–Si alloy. In recent years, in addition, some rare and precious metal elements, such as Co [15], Sc [16] and Y [17], have been gradually used to change the morphology of Fe phase to reduce the harm of Fe phase in Al–Si alloy, even so, due to the cost of these elements, their practical application is greatly limited. Therefore, it is necessary to explore an element that has significant effect and can maintain the cost at a lower level.

Although the current research on the change of Fe phase mainly focuses on the effect of Mn modification, some researchers [18] have analyzed element V from the perspective of valence electron. Since the atomic radius of Fe and V are very close, V atom can replace the Fe atom in the ternary Al–Si–Fe phase without causing structural changes. In addition, the bond energy of each atomic bond network will directly affect the stability of crystal structure, and it is well known that the bond energy of V atomic network is higher than that of Fe atomic network. Therefore, the substitution of V atom for Fe atom in ternary Al–Si–Fe phase can stabilize the structure of Fe-rich phase. Prasada Rao [19] found that adding V to A319 alloy can change the morphology of coarse flake Fe-rich phase and make it tend to be equiaxed in morphology, which is vital to improve mechanical properties of A319 alloy. According to the study of Cameron et al. [20], the addition of a small amount of V in Al-6063 alloy affects mechanical properties of the alloy by promoting the precipitation kinetics of β and β′. Nevertheless, the effect of V on the morphology and properties of the second phase remains to be explored.

For thermal conductive metals, both strength and thermal conductivity are crucial properties [21] although they are often mutually exclusive. For example, the thermal conductivity of pure aluminum is as high as 235 W/(m·K), but its tensile strength is only 80–100 MPa. In order to improve the strength, alloying is generally used, which will lead to the decrease of thermal conductivity. As a result, in the selection of heat dissipation parts, it is often necessary to make a compromise between strength and thermal conductivity [22]. At this stage, the most commonly used radiator materials are A6061 or A6063, whose thermal conductivity can reach 180 W/(m·K) [23], however, there is a serious problem that the alloy has poor casting performance and cannot be cast to produce thin-walled heat dissipation components with slightly complicated structure. The thermal conductivity of ADC12 aluminum die casting alloy is only 96 W/(m·K) [23], which greatly limits the application of this alloy in thermal conductivity.

It is a significant goal to develop an Al–Si foundry alloy with high strength and high thermal conductivity, which also has fine demoulding performance due to Fe content. The current research mainly focuses on the effect of Fe-rich phase on mechanical properties of Al–Si alloy, but the literatures about its effect on thermal conductivity are very few. For the sake of it, this work aims to investigate the relationship of the morphology of Fe-rich phase in Al–8Si–0.1Cu–0.6Mg–0.7Fe–xV alloy to mechanical properties and thermal conductivity of the alloy.

2. Materials and experimental details

Industrial pure Al (99.99 wt%) and Al-24.4Si (wt%) were added into the resistance furnace (SG2-7-12, \( \varphi \times 300 \) mm), the furnace temperature is set at 800 °C for melting, after full melting, Al-50Cu (wt%), Al-20Fe (wt%) and Al-5V (wt%) with different ratios were added for melting, then the furnace temperature was reduced to 760 °C, and industrial pure Mg (99.95 wt%) was added to continue smelting, then Al-10Sr (wt%) was added for modification, the hydrogen dissolved in the alloy was removed by high-purity Ar (99.99 wt%) and slag removal agent was added. Finally, the alloy liquid is poured into the #45 steel grinding mold. It is noted that before and after each addition of new ingredients, sufficient stirring should be done, and the temperature should be held for 2–3 min after mixing, and before pouring, the mold should be pre-heated at 250 °C and the pouring temperature of the alloy is 760 °C. The specific composition table obtained from the experiment is shown in table 1.

A 10 mm \( \times \) 10 mm \( \times \) 10 mm block was taken from the center of the casting to make metallographic samples, and the corrosion was carried out with 0.5% HF for 30 s. Optical microscope (OM; Carl Zeiss Axio-Imager-A2m, Göttingen, Germany) and scanning electron microscope (SEM; Carl Zeiss EVO-18, Göttingen, Germany) were used to observe the microstructure of the alloy, the phase composition of the alloy was identified by energy dispersive spectrometer (EDS; Oxford INCA-X-Max, England, UK) and x-ray diffraction (XRD; 18 KW/D/Max 2500PC, Rigaku, Tokyo, Japan). Tensile tests are carried on the universal electrical testing machine (SHIMADZU-AGS-100 kN, Japan) for mechanical properties with the tensile speed of 0.6 mm min\(^{-1}\), and the tensile properties were the average values of three tensile samples in each group. The statistical method
of Fe phase length is that five optical microscope photos are randomly selected from each sample, Nano Measure is used to measure the length of iron phase in each photo, and the results with statistical significance are summarized.

Since the heat conduction of metals is mainly accomplished by free electrons, there is an inevitable linear relationship between the electrical conductivity and thermal conductivity of metals. At a fixed temperature, the ratio of thermal conductivity to electrical conductivity of most metals is approximately constant, which is Wiedemann–Franz law [24]. It can be expressed as

\[ \lambda / \sigma = L T \]  

where \( \lambda \) is thermal conductivity, \( \sigma \) is conductivity, \( L \) is Lorentz constant, \( T \) is Kelvin temperature. For Al Si alloy [25]

\[ L = 2.1 \times 10^{-8} + 0.021 \times 10^{-8} - 8 \times \text{wt} \% \text{Si} \text{ W} \Omega \text{m} \]  

This value of \( L \) increases with the increase of Si content in Al–Si alloy, in addition, by modifying Wiedemann–Franz law of Al Si alloy, it is concluded that [26]

\[ \lambda = L T \sigma + c \]  

where \( c \) is 12.6 W/(m · K). The method to test the thermal conductivity of the alloy is to measure the electrical conductivity of the alloy by using the German FORESTER conductivity meter SIGMATEST 2.069, the results of the conductivity of each group are the average values of the conductivity of the five points, the electrical conductivity is transformed into thermal conductivity by Wiedemann–Franz law.

### 3. Results

#### 3.1. Effect of different V content on microstructure of alloy

Figure 1 shows the backscattered electron images of alloys with different V contents, in which three kinds of microstructures can be observed, including two Fe-rich intermetallic compounds and one irregular pattern compound. Table 2 shows the point scan results of the second phase in Al–8Si–0.1Cu–0.6Mg–0.7Fe–xV alloy.

Figure 2 shows the XRD spectra of Al–8Si–0.1Cu–0.6Mg–0.7Fe–xV alloy. The composition of the compound is analyzed by energy spectrum and XRD, it can be concluded that, the needle-like Fe phase is \( \beta \)-Al\(_{4.5}\)FeSi, this phase exists widely in alloys without V addition. But the ratio Fe/Si of \( \beta \)-Al\(_{4.5}\)FeSi is closer to 0.5 instead of 1. The reason is that EDS on the particles is inevitably inaccurate because of their small size, which implies that EDS analyses also detect signals from the matrix. The short rod-like Fe phase is \( \alpha \)-Al\(_{0.7}\)Fe\(_{0.3}\)Si, this phase exists in the alloy containing V, the amount of it increases with the increase of V content. According to the results of XRD, the Al\(_{15}\) (Fe, V) \(_3\)Si\(_2\) phase described in some papers [27] has not been detected in this experiment. In the phase with irregular shape, the V/Si ratio is close to 0.5, so the phase composition should be VSi\(_2\). Because the formation condition of VSi\(_2\) in the alloy is that the content of V is greater than 0.2% [28], this phase mainly exists in the alloy with higher V content. In addition, there is evidence of the existence of Al\(_{4}\)V\(_8\).

Figure 3 shows the optical microscope images of alloys with different V contents and the corresponding statistical distribution of \( \beta \)-Fe phase length frequency. Figure 4 shows the variation of \( \beta \)-Fe phase length with the increase of V addition. These microstructures indicate that the Fe phase in the alloy is mainly distributed on the grain boundary, and some larger Fe phases will cross the \( \alpha \)-Al matrix, the average length of the \( \beta \)-Fe phase first decreases with the increase of V content, and then increases with the increase of V content. When V is not added, the length of \( \beta \)-Fe phase is mainly about 20 \( \mu \)m, with the increase of V content, the main distribution of \( \beta \)-Fe phase length gradually approaches to 10 \( \mu \)m, when the content of V is increased to 0.6%, the length of \( \beta \)-Fe phase reaches the minimum, and the length is mainly concentrated at about 10 \( \mu \)m, with an average length of 11.88 \( \mu \)m, which is 9.35 \( \mu \)m shorter than that without V addition, accounting for 44.04% of the original length, with the continuous increase of V content, the length of \( \beta \)-Fe phase continues to grow, when the content of V is 1.0%, the maximum length of \( \beta \)-Fe phase reaches more than 70 \( \mu \)m, and the average length reaches 28.81 \( \mu \)m, even
7.58 μm more than that without V addition, accounting for 35.7% of the original length. Therefore, it can be seen from the experiment that the content of V has a great influence on the length of β-Fe phase.

3.2. Effect of different V content on mechanical properties of alloy

Figure 5 shows the broken line diagram of mechanical properties of alloys with different V content, and the law is similar to the effect of different V content on the length of β-Fe phase. The yield strength, tensile strength and elongation of the alloy without V addition are 106.4 MPa, 195.4 MPa and 4.57%, with the increase of V content, the yield strength, tensile strength and elongation of the alloy increase in the mean time, and reach the maximum value when the V content is 0.6%, the yield strength, tensile strength and elongation of the alloy are 132.3 MPa, 225.6 MPa and 7.91% respectively, which are 24.3%, 15.5% and 73.1% higher than those of the alloy without V addition. With the continuous increase of V content, the mechanical properties began to decrease, when the V content reaches 1.0%, the elongation is even lower than that of the alloy without V, but the yield strength and tensile strength are still higher than that of the alloy without V addition.

3.3. Effect of different V content on electrical and thermal conductivity of alloys

Figure 6 shows the electrical conductivity and thermal conductivity of alloys with different V content, it can be seen that the two laws are basically consistent. The electrical conductivity and thermal conductivity of the alloy without V addition are 23.1 MS m⁻¹ and 172.7 W/(m · K), with the addition of V, the electrical conductivity

Table 2. Composition of the second phase of Al–8Si–0.1Cu–0.6Mg–0.7Fe–xV alloy (at%).

| Alloy | Second Intermetallics | Phase  | Al  | Fe  | V  | Si  |
|-------|-----------------------|--------|-----|-----|----|-----|
| 0 V   | β-Fe                  | Al₄FeSi| 72.03| 10.41| —  | 17.56|
| 0.2 V | β-Fe                  | Al₄FeSi| 59.85| 11.17| 0.06| 28.92|
| 0.4 V | β-Fe                  | Al₄FeSi| 57.36| 27.46| 1.13| 14.05|
| 0.6 V | β-Fe                  | Al₄FeSi| 58.70| 11.68| 0.06| 29.56|
| 0.8 V | β-Fe                  | Al₄FeSi| 57.83| 26.87| 1.56| 13.74|
| 1.0 V | β-Fe                  | Al₄FeSi| 58.34| 13.48| 0.07| 28.11|
|       | α-Fe                  | Al₄FeSi| 58.52| 26.27| 1.85| 13.36|
|       | α-Fe                  | Al₄FeSi| 59.88| 11.75| 0.06| 28.31|
|       | α-Fe                  | Al₄FeSi| 59.66| 25.15| 2.24| 12.95|
|       | VSi₂                  | VSi₂   | 13.68| 5.72 | 24.29| 56.30|
and thermal conductivity of the alloy decrease sharply at first and then decrease slowly, when the content of V is 0.6%, it will increase slightly, the electrical conductivity and thermal conductivity of the alloy are 17.3 MS m$^{-1}$ and 132.2 W/(m · K), then, with the increase of V content, the electrical conductivity and thermal conductivity of the alloy continue to decline. It can be seen that the addition of V has an adverse effect on the overall conductivity and thermal conductivity of the alloy, which will reduce by 20.4% to 27.1%.

4. Discussion

By analyzing the EDS results shown in table 2, it can be found that the content of V dissolved in $\beta$-Fe is very small, all less than 0.1% [29], and it does not rise with the increase of V content in the alloy. The formation rate of
β-Fe phase is determined by the mobility of Fe atoms, and V is highly diffused in the liquid phase, which hinders the growth of β-Fe phase to a certain extent, and makes the β-Fe phase refined. The V content in α-Fe increases with the increase of V content slightly, which is because Fe phase can capture more V with the increase of V content in the alloy, but the maximum value of V content is generally not more than 3%, this shows that V atom can replace Fe atom in Fe phase. Because needle-like β-Fe crystal has special growth advantages in a single direction, with the addition of V content, V element gradually dissolves into acicular Fe phase, which hinders the growth advantage in single direction and makes it extend to other directions, this modification principle is similar to that of Mn on Fe phase. When the content of V is more than 0.6%, the β-Fe phase in Al Si alloy will aggregate, which will lead to the intersection or partial overlap of β-Fe phases, as a result, the length of β-Fe phase grows. When the content of V is more than 1%, superfluous V will cause a large number of unconnected β-Fe phases to gather together, which making the length of β-Fe phase even longer than that of the alloy with no V. In addition, the addition of V can increase the melt temperature [28], and the formation temperature of α-Fe phase is higher than that of β-Fe phase, the formation of α-Fe phase promotes the consumption of Fe in the alloy, and the decrease of Fe content will reduce the precipitation temperature of β-Fe phase [14], so the solidification temperature range of α-Fe phase is increased to a certain extent. Therefore, the addition of V not only inhibits the formation of β-Fe phase, but also promotes the transformation of β-Fe phase to α-Fe phase. However, there are only still a few substituted Fe atoms, which is not enough to completely transform ternary Al–Si-Fe phase into quaternary Al–Si-V-Fe phase.
As shown in figure 5, the addition of V has a great influence on the mechanical properties of the alloy, obviously improving the yield strength, tensile strength and elongation of the alloy. The solubility of Al-V compound Al$_x$V$_5$, whose lattice parameters should be less than 1nm can reach 0.9at% Si [30], V-containing compounds phase can pin dislocations and hinder the nucleation and growth of recrystallization [31]. Therefore, when the V content increases from 0% to 0.2%, the mechanical properties of the alloy are greatly improved due to its strengthening effect. Although the length of $\beta$-Fe phase in the alloy also decreases, its contribution to mechanical properties is a secondary factor. When the content of V is more than 0.2%, the solution strengthening effect of V tends to be stable, and the influence of Fe phase plays a major role in the change of mechanical properties. The needle-like Al$_{4.5}$Fe$_2$Si phase is hard brittle phase [32], they exist in the alloy matrix in the form of discrete particles, which are highly multi-faceted, their sharp edges will cause serious stress concentration problems in the matrix. Therefore, the bonding strength between $\beta$-Fe and matrix is relatively low, microcracks are very easy to occur in the interface area between them, and the splitting effect of such microcracks on the matrix is very significant. However, the $\alpha$-Fe phase is relatively dense [33] and forms a rough interface with the matrix, this kind of bonding strength is high, which reduces the probability of microcracks on the interface between Fe phase and matrix [34]. Even if cracks appear, it can effectively prevent the crack propagation during the tensile process. It can be seen from figure 4 that when the V content is 0.6%, the average length of $\beta$-Fe phase is the lowest, the bonding strength with grain boundary is improved [35], and the splitting effect on the alloy is the least. In addition, even if the second dendrite arm spacing seems greater than that of the low V content sample, its mechanical properties are still higher, which shows that the length of $\beta$-Fe phase has a great positive impact on the mechanical properties of the alloy. When the content of V exceeds 0.6%, with the continuous increase of V content, the content of $\alpha$-Al$_5$Fe$_2$Si increases gradually and the content of $\beta$-Al$_{4.5}$FeSi decreases gradually, but the size of Fe phase in the alloy increases sharply. Because the $\beta$-Fe phase precipitates before eutectic solidification, the coarse $\beta$-Fe phase will block the flow of alloy liquid and block the feeding channel between dendrites. As a result, the $\beta$-Fe phase is often a potential site for the formation of shrinkage cavity and porosity [36]. As shown in figure 7(a), when there is no V element addition, there is shrinkage cavities exist around $\beta$-Fe phase, and the fracture path may preferentially goes through the shrinkage porosity, which leads to a decrease in mechanical properties [37, 38]. However, when the content of V is 0.6%, as shown in figure 8(b), the length of $\beta$-Fe phase reaches the minimum value, no shrinkage cavity was found, and even some dimples appeared. Moreover, a large block-like VSi$_2$ phase formed when the content of V reaches 1%, the solubility of VSi$_2$ can even reach 5.4 at% Al [30], which leads to the significant cleavage effect of $\beta$-Fe and VSi$_2$ phase on the alloy.

Figure 8 is the schematic diagram of electron flow in the alloy (In the figure, the gray phases represent Fe phase, V represents the solid solution V and V-containing compounds). The alloys with 0, 0.2% and 0.6% V content are taken as examples, as shown in figure 8(a), when V is not added, the electron blocking effect in the alloy mainly comes from the long needle-like Fe phase. Although the size of Fe phase is large and has a certain scattering effect on electrons, the electron still has a higher free path for migration. With the addition of V, as shown in figure 8(b), due to the solubility of V in Al matrix, when the addition of V is 0.2%, a part of V will be dissolved in the Al matrix, at the same time, Al$_x$V$_5$ is generated, these discrete solid solutions and Al$_x$V$_5$ greatly shorten the free path of free electron migration [39]. Although the length of $\beta$-Fe phase is reduced compared
with that of the alloy without V, the gain effect caused by the change of Fe phase is far less than the weakening effect of V added in solid solution, which is the reason for the sharp decrease of electrical conductivity and thermal conductivity. With the increase of V content to 0.6%, the content of V-containing compounds tends to be stable, and the solubility of V in aluminum matrix has reached saturation. From now on, the biggest factor affecting the electrical conductivity and thermal conductivity of the alloy is not the content of V in aluminum matrix anymore, but the change of $\beta$-Fe phase length. As shown in figure 8 (c), the shorter the $\beta$-Fe phase is, the greater the free path of free electron migration is, and the smaller the scattering effect of Fe relative to electrons is. As a result, when the length of $\beta$-Fe phase reaches the smallest, that is, the V content is 0.6%, the electrical conductivity and thermal conductivity of the alloy will be improved. When the content of V continuously increases, the length of $\beta$-Fe phase increases greatly, the electron scattering effect increases, and the electrical conductivity and thermal conductivity of the alloy decrease.

5. Conclusions

In this paper, effects of V on modification of Fe-rich phases and mechanical properties and thermal conductivity in hypoeutectic Al–Si alloys was studied, the following conclusions can be drawn.

1. The existence of V will obviously effect on the improvement of Fe phase size in the alloy. The addition of V refines the $\beta$-Fe phase and promotes the transformation from $\beta$-Fe phase to $\alpha$-Fe phase. The length of Fe phase in the alloy with 0.6% V content is 44.04% shorter than that without V addition.
2. There are positive effects of the addition of V on the mechanical properties of the alloy, when the content of V reaches 0.6%, the yield strength, tensile strength and elongation of the alloy reach 132.3 MPa, 225.6 MPa and 7.91%, which are 24.3%, 15.5% and 73.1% higher than those of the alloy without V addition.

3. The addition of V will bring negative influence on the thermal conductivity of the alloy. However, due to the refining effect of V on the Fe phase, the thermal conductivity of the alloy with 0.6% V content is the highest locally.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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