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Composition-Dependent Solidification Cracking of Aluminum-Silicon Alloys during Laser Powder Bed Fusion

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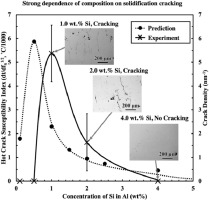
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# Abstract

Consistent manufacturing of volumetrically dense engineering components, free of solidification cracks by laser powder bed fusion (LPBF), has been demonstrated for Al-Si alloys such as AlSi10Mg and Al12Si. The success in LPBF of these alloys is attributed to the near eutectic composition with a small freezing range. To illuminate this observation, cracking susceptibility was examined from Scheil-Gulliver solidification modeling by calculating the hot cracking susceptibility, |dT/dfS1/2|. To validate the findings from hot cracking susceptibility calculations, six binary Al-Si alloys, whose compositions were strategically chosen at hypo-, near-, and hyper-eutectic compositions, were gas atomized into alloy powders, and processed by LPBF. Only Al-Si alloys with 1.0 and 2.0 wt.% Si were found to exhibit cracking, which was predicted by relatively large magnitudes of |dT/dfS1/2|. Either as particles or with a eutectic structure, Si segregation at the intercellular boundaries was observed to define the sub-grain cellular structure. For selected compositions, measurement of the cellular structure allowed for estimation of the cooling rate to be 106 to 107 K•s−1. Excluding the alloys with solidification cracking, an increase in tensile strength and the corresponding decrease in ductility were observed with an increase in Si concentration, which were attributed to the formation of a cellular structure and the amount of Al-Si eutectic found at the intercellular boundaries.

# Graphical abstract



# Keywords

Cracking susceptibility, Scheil solidification, Additive manufacturing, Cooling rate, Cellular structure, Tensile testing

# 1. Introduction

Additive manufacturing (AM) has demonstrated its ability for rapid prototyping and customized manufacturing of complex metallic components [1], [2], [3], [4]. Laser powder bed fusion (LPBF) is an AM technique that utilizes a laser source for selectively melting consecutive layers of a powder bed [2,3]. Many commercial Al-alloys such as high strength AA7075 (Al-Zn-Mg-Cu based), high strength AA6061 (Al-Mg-Si based), and corrosion resistant AA5083 (Al-Mg-Mn based) are desired for applications in military, nuclear, and aerospace industries. These Al-alloys were designed for casting and post thermomechanical processing. Moreover, AA5083 and AA6061 are considered to be weldable alloys [5], showing versatility in localized high temperature melting with rapid solidification, similar to that found in LPBF. However, these Al-alloys are not considered suitable for LPBF without composition modification due to the consequence of solidification cracking (hot tearing) and/or excessive porosity, regardless of processing parameter employed [6], [7], [8], [9], [10], [11], [12].

Without alloy modification, only near eutectic compositions, ~ 12.6 wt.% [13], of Si alloyed with Al, such as Al – 10 wt.% Si – 0.5 wt.% Mg (commonly referred to as AlSi10Mg) and Al – 12 wt.% Si (Al12Si), have exhibited suitable behavior in LPBF with reliable production of volumetrically dense parts, free of solidification cracks [14], [15], [16], [17]. Interestingly, long columnar grains are found in the microstructures of AlSi10Mg and Al12Si similar to unmodified AA7075 and AA5083; however, no cracking is observed [16,18]. Since the near eutectic compositions have a small freezing range, there is a nearly instant transformation from liquid to solid. Most commercial, high-strength, wrought Al-alloys contain multiple alloying additions in low concentrations, < 5 wt.%. Deviations from the eutectic composition result in a temperature freezing range bounded by the liquidus and solidus in the phase diagram. It has been shown that cooling rates in LPBF are on the orders of 105 to 107 K•s−1 [17,19], significantly higher than 101 to 103 K•s−1 estimated for casting and other conventional processes [17,19,20]. Therefore, the temperature decrease from the liquidus to the solidus temperature will occur rapidly, which may result in crack initiation along the mushy zone, as there is little time for crack healing through merging of grains or liquid back flow.

It is likely that the hot cracking tendency in LPBF is dependent on alloy composition, because of variation in solidification characteristics such as the magnitude of the freezing range and solid fraction change between the liquidus and solidus. In this study, Scheil-Gulliver solidification calculations along with a hot cracking susceptibility criterion were carried out to predict and understand the hot cracking susceptibility of the several binary Al-Si alloys. In conjunction, six binary Al-Si alloy powders, their compositions strategically chosen at hypo-, near, and hyper-eutectic compositions, were produced via gas atomization and were subsequently processed with LPBF in order to experimentally observe the dependence of solute concentration on the LPBF solidification behavior. Finally, cooling rate and thermal gradient were estimated from results of microstructural analysis and related to the cracking susceptibility of Al-alloys.

# 2. Criterion for solidification cracking

## 2.1. Background on solidification cracking

Solidification cracking (hot tearing) is thought to occur when the stress and/or strain induced from solidification shrinkage and thermal contraction cannot be accommodated by elastic or plastic deformation of the alloy [21,22]. For an isotropic solid, the accumulated strain, ε based on the change in dimension, δ, can be related to the thermal expansion coefficient, γ, and freezing range of an alloy through the expression [23]:

(1)

where TL and TS are the temperatures of the liquidus and solidus, respectively. Therefore, some earlier studies postulated that the hot cracking susceptibility is directly related to the temperature freezing range,  ΔT of an alloy [24,25]. Even though the freezing range did influence cracking susceptibility, it has been shown that cracking occurs at the end of solidification, when the temperature is near the solidus temperature [24].

Strain-based models focus on the last remaining liquid, when the fraction of solid, fS, approaches 1. They.\ postulate that plastic deformation of the material such as grain bridging to close the cracks or liquid backfilling, only occurs in the final stages of solidification. As fS → 1, the last remaining liquid would wet the grain boundaries with a continuous film before reaching the solidus/eutectic temperature. Strain would develop along this liquid film, and when the strain supersedes the ductility of the material, the liquid film will break, causing grain separation and cracking. Moreover, so little liquid and time would be left to allow for grain coalescence or for liquid back-flow. Therefore, cracking models based on strain rate are becoming more prominent because it considers solidification time, i.e., kinetics, to play a significant role in strain accumulation during solidification [26,27].

Clyne and Davies [28] proposed that the cracking susceptibility criterion (CSC) can be determined by the ratio of the time period when the material is vulnerable to cracking, i.e. when fS = 0.9 to 0.99, to the time period available for stress relief, i.e. when fS = 0.4 to 0.9. Even though the CSC model takes into account the factor of time and last remaining liquid, the oversimplification of the model breaks down at high cooling rates (extremely low freezing solidification times, < 10−3 seconds), where it is difficult to quantify the total freezing time, and the range of fS for both time periods is dependent on an arbitrary range that may not apply to all alloy systems.

A more complete and prominent model proposed by Rappaz-Drezet-Gremaud (RDG) [29,30]) is based on strain rate and focuses on the growth of columnar dendrites during the final stages within the mushy zone, which is considered to be located at both the interdendritic and grain boundaries. The RDG model also states that there is a uniaxial tensile deformation acting normal to the growth of columnar dendrites, and liquid feeding moving opposite the growth direction. However, the RDG model assumes that crack initiation is caused by formation of a cavity due to a drop in cavitation pressure in the mushy zone. Coniglio and Cross [26] reported, through arc weld experiments, that even though the RDG model considered tensile deformation against the dendrites growth and healing through liquid feeding, the cavitation due to pressure drop is not possible in Al-alloys. Moreover, maximum susceptibility of the RDG is challenging to calculate, and nearly proportional to the nonequilibrium solidification range, according to their original work. Despite the differences, both the RDG and CSC model identify the mushy zone, found near the end of solidification, i.e., fS → 1, is the most important for understanding solidification cracking. To that end, as reviewed well by Easton *et* al. [30], various modified models based on CSC or RDG have been postulated by examining the last moments of solidification.

## 2.2. Cracking criterion calculations

Kou [27] proposed a cracking criterion based on stain rate, which focuses on the final liquid at the grain boundaries near the end of solidification, e.g., fS = 0.99. Cracking is assumed to occur when an increase in strain rate due to cooling during the final stages of solidification, is not accommodated. Cracking is postulated to nucleate due to separation of grains, which can be caused by liquid film fracture or by pulling of growing grains due to accumulated strain. If a sufficient amount of liquid is present at the end of solidification, then the accumulated strain would not be adequate to break the liquid film. Other sites for crack initiation can already be present, such as trapped micropores in the melt or presence of oxides folded into the liquid [26,27]. Cracks can be healed when the grains overcome the strain. Healing can occur through bridging by continual growth towards one another, or through liquid feeding of the open channel to fill the possible crack. For predicting the cracking susceptibility, Kou focused on the last fraction of liquid to solidify and suggesteded that the sensitivity of the fS, or more accurately fS1/2, before reaching the solidus temperature, is critical. Therefore, Kou's prediction of cracking is based on the maximum steepness of the fS1/2 vs. T curves, |dT/dfS1/2|, and can be directly related to the cracking susceptibility of an alloy.

The fraction solidified, fS, can be calculated from the work of Scheil and Gulliver, expressed as [20]:

(2)

where T is the temperature of the melt, TM is the melting temperature of solvent, TL is the liquidus temperature, and k is partition coefficient expressed as:

(3)

where CS is the concentration of the solid and CL is the concentration of the liquid [20]. Eq. (2) assumes infinite diffusion in liquid, no diffusion in the solid, and that both the liquidus and solidus are linear lines in the phase diagram with a constant k [20,27]. The fraction of eutectic formed is then described as [27]:

(4)

where fE is the fraction eutectic, TE is the eutectic temperature, C0 is the concentration of solute, and -mL is the slope of the liquidus defined as [27]:

(5)

In Eq. (5), CE is the eutectic concentration of solute. The term |dT/dfS1/2| can then be calculated as [27]:

(6)

Mathematical derivation for Eq. (6) can be found in the *Supplementary Materials.* Since |dT/dfS1/2| is a function of temperature, Kou suggested that the maximum steepness can be calculated as an average of |dT/dfS1/2| over an arbitrary range near fS = 1. Choosing an appropriate range of fS for all compositions is difficult because not all compositions obtain fS = 1 before reaching the solidus temperature. Therefore, in this study, a maximum |dT/dfS1/2| was determined at a critical value of fS1/2 taken before reaching the solidus temperature as fS →1.

## 2.3. Application of cracking criterion to binary Al-Si

Fig. 1(a) presents the phase diagram of the Al-Si system with a simple, single eutectic reaction including linear liquidus and solidus lines. The maximum solid solubility limit of Si in Al is 1.7 wt.% Si [13], at which the largest freezing range of approximately 76 °C occurs as shown in Fig. 1(b). The binary Al-Si system is an excellent binary system to examine the cracking tendencies based on Kou's criterion model. The TM and TE were taken as 660 °C (933 K) and 572 °C (845 K), respectively. Using Eqs. (2) and (6), fS, fS1/2, and |dT/dfS1/2| were calculated as a function of Si concentration as reported in Table 1 and presented in Fig. 2. The highest |dT/dfS1/2| was found for the 0.5 wt.% Si concentration with |dT/dfS1/2| and fS1/2 of 5874.82 °C and 0.988, respectively. Kou also calculated the maximum |dT/dfS1/2| at 0.4 wt.% [27] and 0.5 wt.% [31] for the binary Al-Si system using Pandat and PanAluminum software packages. Fig. 2(c) presents |dT/dfS1/2| determined as a function of Si concentration in Al.

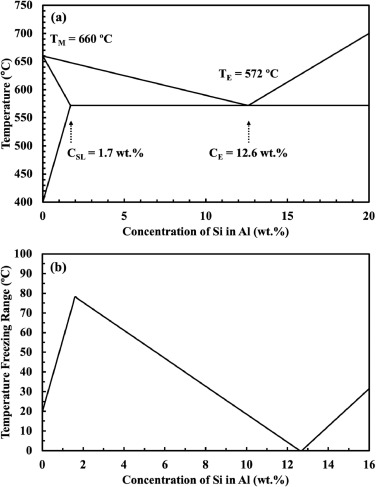


Fig. 1. (a) Equilibrium binary phase diagram for Al-Si system examined in this study; (b) Freezing range as a function of Si concentration in Al.

Table 1. Calculated characteristics of equilibrium solidification for various binary Al-Si alloys.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Concentration of Si in Al (wt.%)** | **Maximum |dT/dfS1/2| (°C)** | **Critical fS1/2** | **Critical fS** | **fE** |
| 0.1 | 1782.82 | 0.990 | 0.980 | 0.020 |
| 0.5 | 5874.82 | 0.988 | 0.976 | 0.024 |
| 1.0 | 2292.17 | 0.970 | 0.941 | 0.059 |
| 1.5 | 1323.62 | 0.950 | 0.903 | 0.097 |
| 2.0 | 944.74 | 0.930 | 0.865 | 0.135 |
| 2.5 | 733.71 | 0.910 | 0.828 | 0.171 |
| 4.0 | 449.00 | 0.850 | 0.723 | 0.277 |
| 10.0 | 94.53 | 0.480 | 0.230 | 0.769 |
| 12.6 | 0.000 | 0.000 | 0.000 | 1.000 |

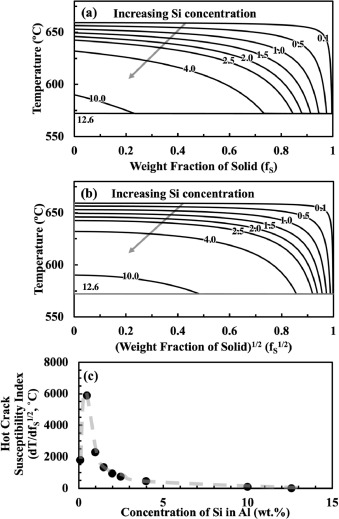


Fig. 2. Temperature vs. (a) fS and (b) fS1/2 solidification curves for binary Al-Si alloys with varying Si content; (c) Maximum steepness, |dT/dfS1/2|, considered as crack susceptibility index, as a function of concentration of Si in Al.

# 3. Experimental methods

## 3.1. Powder production and characterization

Starting from Al-20 wt.% Si and commercially pure Al master alloys, binary Al-Si gas atomized powders were produced using an in-house laboratory-scale gas atomization system. A melt temperature of 900 °C and gas pressure of 2 MPa were employed. A total of six binary Al-Si compositions were gas atomized: hypo-eutectic compositions of 0.5, 1.0, 2.0, and 4.0 wt.% Si, near eutectic composition of 12.6 wt.% Si, and hyper-eutectic composition of 16.0 wt.% Si. The atomized powders were sieved using a 106 µm mesh size screen for 15 mins per ASTM B214-16 standard for sieving metal powders. Powder size distribution was measured with a Beckman Coulter LSTM 13 320 laser powder diffractometer.

## 3.2. LPBF processing strategy

For LPBF, a SLM 125HL (SLM Solutions Group AG, Lubeck, Germany) equipped with a continuous wave Yb 400 W IPG fiber-laser with a 1070 nm wavelength and Gaussian spot size of approximately 70 μm [32], was employed. The Al-alloy build plate was pre-heated to 100 °C, and the builds were performed in an inert N2 atmosphere in which the O content was kept below 0.5 %. In order to examine the effects of composition variation and LPBF processing parameters on development of pores/flaws and crack susceptibility, 12 mm x 12 mm x 12 mm cubes were built with the SLM 125HL for each composition as functions of LPBF parameters. The cubes were built 4 mm above the build plate, supported underneath with a typical block support structure. No re-melting strategies such as bordering or contouring were employed.

Since AlSi10Mg has shown repeatedly to behave well with LPBF, a parameter set optimized for AlSi10Mg by the SLM Solutions Group AG (Lubeck, Germany) was used as the basis: a laser power, scan speed, hatch spacing, slice thickness, and scan rotation of 350 W, 1650 mm/s, 0.13 mm, 0.03 mm, and 67°, respectively. However, this particular parameter may not be optimum for different Si content, so for each alloy composition, the laser power and scan speeds were varied independently to build cubic samples. High and low laser powers of 350 and 250 W were employed. At a laser power of 350 W, the scan speed was varied as 1200, 1600, 2000, 2400, and 3000 mm/s. At a laser power of 250 W, the scan speed was varied as 800, 1200, 1600, 2000, and 2400 mm/s. Hatch spacing and slice thickness were held constant at 0.13 mm and 0.03 mm, respectively, since laser power and scan speed have shown to be the most influential factors on buildability [17,33].

Cube samples were then cross-sectioned both parallel and perpendicular to the build direction, mounted in epoxy, and metallographically prepared for a final finish with 0.05 µm colloidal silica. Each sample was examined with optical microscopy for quantitative analysis of pores/flaws and crack density measurements. For microstructural analysis, samples were etched with Keller's reagent (2.5 vol.% nitric acid, 1.5 vol.% hydrochloric acid, and 1.0 vol.% hydrofluoric acid in distilled water).

## 3.3. Microscopy and microstructural analysis

Optical microscopy, Field-Emission Scanning Electron Microscopy (FE-SEM) equipped with X-ray Energy Dispersive Spectroscopy (XEDS) were employed to characterize the powders and cube samples. A Nikon Metaphot optical microscope was utilized to examine the XZ cross-sections (the plane parallel to the build direction) of the cubes to determine the amount pores/flaws and extent of cracking. Six optical micrographs were taken at 100X for each sample, and were quantitatively analyzed utilizing the ImageJ software (National Institutes of Health). A relative density was determined based on the pore/flaw content; cracks were included in the pore/flaw determination. Linear intersection method was utilized to determine crack density. Five linear lines, approximately 1000 µm each in length, were superimposed on each of the five optical micrographs, and crack-intersect counting was performed with ImageJ. For detailed microstructural characterization, both secondary and backscatter electron contrasts were employed with a Zeiss Ultra-55 FE-SEM operated at 20 kV. Composition was measured with a Si-drift Thermo-Scientific XEDS equipped on the Zeiss FE-SEM. Quantitative compositional analysis was carried out using Thermo-Scientific Noran System Seven (NSS) software, employing the phi-rho-z (PROZA) standardless quantification method.

## 3.4. Mechanical behavior

Tensile specimens with a gauge length of 25 mm in accordance with ASTM E8M-3856, were built for all six compositions. A LPBF parameter set, consisting of a laser power, scan speed, hatch spacing, slice thickness, and scan rotation of 350 W, 1600 mm/s, 0.13 mm, 0.03 mm, and 67˚, respectively, was used to build all the tensile samples, because this parameter set produced the highest volumetric density for all compositions. The tensile samples were built with a 0° tilt with respect the build plate (e.g., X-Y build) so that solidification cracks typically forming in the build direction (e.g., Z) would be oriented normal to the tensile direction, i.e., mode I crack opening. All surfaces of tensile specimens were ground up to 1200 grit SiC paper before tensile testing.

Three tensile samples were tested for each composition with an MTS™ instrument employing a quasi-static strain rate of 4 × 10−4 s−1. Tensile deformation was measured and recorded by a digital image correlation (DIC) camera positioned perpendicular to the loading direction. The DIC system consisted of a Tokina AT-X Pro macro 100 mm −f/2.8−d lens with a resolution of 2448 × 2048 and VIC-2D 2009 software by Correlated Solutions, Inc. The capture frequency was 1 Hz. Engineering stress and strain were determined based on load recorded, cross-sectional area of the gauge section measured, and displacement captured by the DIC system.

# 4. LPBF experimental results

## 4.1. Powder characterization

Powder size distributions for the gas atomized binary Al-Si alloys are presented in Fig. 3(a). Corresponding D10, D25, D50, D75, D90, and mean particle sizes are reported for each composition in Table 2. In general, the powder size distribution after sieving ranged between 20 to 90 µm with a mean particle size of approximately 50 µm. Secondary electron micrographs of the powder morphology and backscatter electron micrographs of a typical powder cross-section are shown in Fig. 3(b) and 3(c), respectively. Overall, the powders were spherical, but with some satellites. The powder cross-sections revealed Si partitioning to the interdendritic regions. Zhou *et* al. [34] performed both SEM and TEM on gas atomized AlSi10Mg powder, and reported that the interdendritic regions consisted of an Al-Si eutectic structure. Moreover, with an increase in Si concentrations, the extent of segregation appeared to increase since the eutectic structure becomes more pronounced in 12.6 and 16.0 wt.% Si alloy powders. Composition of the powders measured by XEDS is also presented in Table 2. The compositions were near the nominal Si concentrations with minimum deviation.

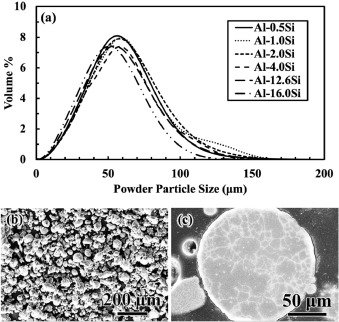


Fig. 3. (a) Powder size distribution for the Al-Si alloy powders with varying compositions; (b) Secondary electron micrograph and (c) cross-sectional backscatter electron micrographs of typical alloy powders.

Table 2. Powder size and composition determined for the six binary Al-Si alloy powders along with the composition determined for the LPBF cube samples.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | **Atomization Charge Composition (wt.% Si)** |  |  |  |  |  |
|  | 0.5 | 1.0 | 2.0 | 4.0 | 12.6 | 16.0 |
| Alloy powder size distribution (µm) |  |  |  |  |  |  |
| **D10** | 20.9 | 21.0 | 21.0 | 19.9 | 18.5 | 16.5 |
| **D25** | 33.8 | 34.2 | 34.6 | 33.9 | 30.6 | 27.8 |
| **D50** | 50.2 | 51.6 | 51.9 | 52.6 | 47.3 | 43.3 |
| **D75** | 67.5 | 69.8 | 70.3 | 73.3 | 65.3 | 60.0 |
| **D90** | 84.1 | 89.2 | 89.1 | 107.6 | 83.0 | 75.5 |
| **Mean** | 51.9 | 54.1 | 54.0 | 52.6 | 49.5 | 45.0 |
| Alloy powder composition (wt.% Si) |  |  |  |  |  |  |
| **Powders** | 1.0 ± 0.9 | 0.9 ± 0.6 | 2.7 ± 1.3 | 3.6 ± 1.0 | 12.5 ± 3.5 | 16.3 ± 0.9 |
| **LPBF Cubes** | 0.9 ± 0.3 | 1.5 ± 0.2 | 2.2 ± 0.2 | 4.4 ± 0.2 | 14.9 ± 0.3 | 18.2 ± 0.5 |

## 4.2. Influence of LPBF processing parameters

Fig. 4 shows the relative density measured from image analysis as a function of scan speed and laser power for each alloy composition. Generally, use of faster scan speeds yielded lower relative densities, below 99 % dense. Moreover, little difference in relative density was observed between samples produced with high (350 W) and low (250 W) laser power for the scan speed range investigated. According to Fig. 4, for all alloy compositions, a relative density greater than 99 % was observed when the LPBF parameter set with laser power, scan speed, hatch spacing, and slice thickness of 350 W, 1600 mm/s, 0.13 mm, and 0.03 mm, respectively, was used for producing cube samples.

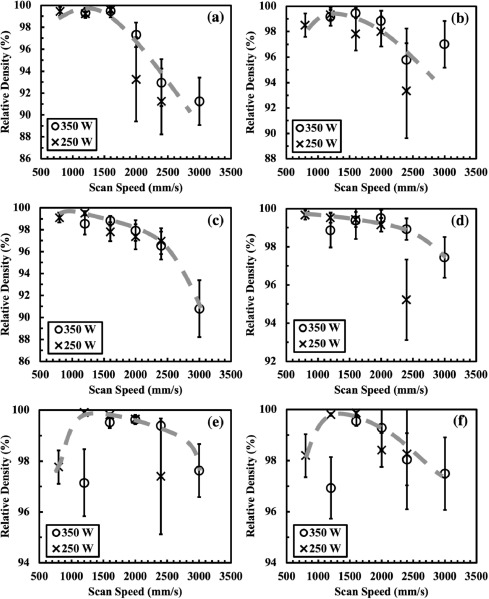


Fig. 4. Relative density determined by image analysis of optical micrographs from the binary Al-alloys with Si content in wt.%: (a) 0.5, (b) 1.0, (c) 2.0, (d) 4.0, (e) 12.6, and (f) 16.0.

Fig. 5 presents optical micrographs from cube samples with six binary Al-Si compositions. Generally, at 350 W or 250 W, there are three regimes observed over varying scan speed: (1) pore formation from keyhole mechanism due to high energy input at low scan speeds, (2) an intermediate regime where the highest relative density is achieved, and (3) flaw formation due to lack of fusion from low energy input at high scan speeds. Similar observations have been reported in previous studies [10,17,33]. High density (> 99 %) could be achieved for all alloy compositions, however, alloy samples with *1.0 and 2.0 wt.% Si always had solidification cracks regardless of LPBF parameters employed*. The alloy with 0.5 wt.% Si exhibited, minor, hairline cracks, but were too fine to resolve and quantify. Solidification cracking was not observed in samples with 4.0, 12.6 and 16.0 wt.% Si.

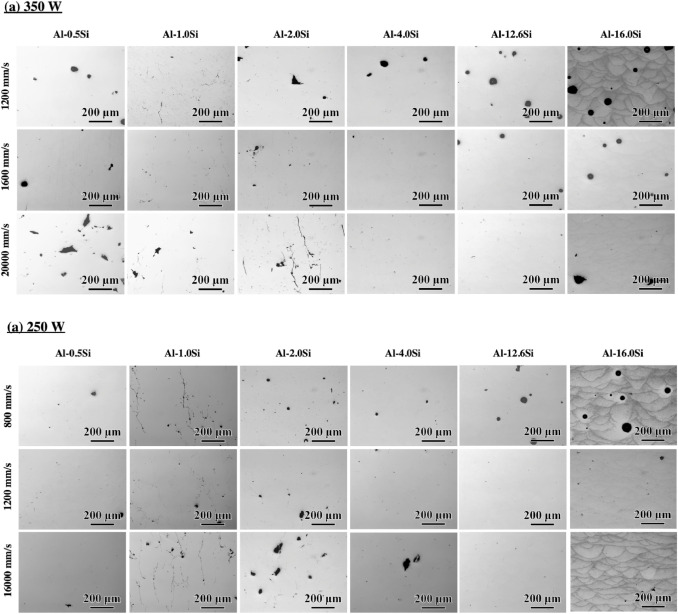


Fig. 5. Optical micrographs from the XZ cross-section of the six binary Al-Si compositions built with different laser powers of (a) 350 W and (b) 250 W as a function of scan speeds.

Presented in Fig. 6(a) and 6(b) are the measured crack density for all cube samples built with 1.0 and 2.0 wt.% Si alloy powders. In general, crack density variation as a function of scan speed varied similar to the relative density determined for these alloys shown in Fig. 4(b) and 4(c). At low scan speed, low crack density is observed along with lower density due to formation of pores. At high scan speed, low crack density is observed corresponding to the low density due to lack-of-fusion flaws. Maximum crack density, as seen in Fig. 6 occurred when the relative density of cube samples was high as presented in Fig. 4. Also, in general, use of lower laser power yielded more solidification cracks.

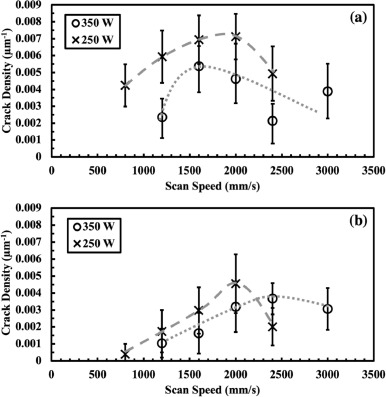


Fig. 6. Crack density measured from LPBF cube samples produced as a function of laser power and scan speed for Al-alloy with Si concentration, (a) 1.0 and (b) 2.0 in wt.%.

The calculated hot cracking susceptibility shown in Fig. 2(c) for the binary Al-Si alloy indicated that the maximum cracking would be observed at a composition of 0.5 wt.% Si without any diffusion in solid. Maximum severity of cracking was experimentally observed at 1.0 wt.% Si as shown in Fig. 7. The crack density for the cubes presented in Fig. 7 were processed with a laser power and scan speed of 350 W and 1600 mm/s, respectively, because these cube samples had the highest density regardless of composition. Clearly this discrepancy in composition, i.e., 0.5 vs. 1.0 wt.% Si, for cracking would be due to many factors including composition measurements, resolution of optical microscopy employed for crack density determination, among many. Therefore, to further assess the relationship between crack susceptibility/density and composition, mechanical behavior in tension was examined for all alloy compositions.

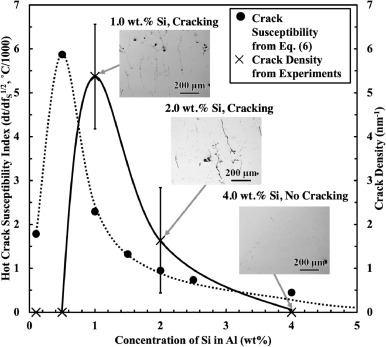


Fig. 7. Hot cracking susceptibility prediction vs. measured crack density for the binary Al-Si compositions examined.

## 4.3. Mechanical behavior and microstructure

Engineering stress-strain curves from tensile testing are presented in Fig. 8 for all six compositions. Three tensile samples were built utilizing a laser power and scan speed of 350 W and 1600 mm/s, where high density was consistently observed for all samples. As expected, during tensile testing, alloys with 1.0 and 2.0 wt.% Si fractured prematurely as presented in Fig. 8. The measured tensile strength (from 0.2% offset), ultimate tensile strength (UTS), strain at failure, and Young's modulus for all eighteen tensile samples are reported in Table 3. The average values of these mechanical properties are presented as a function of Si concentration in Fig. 9. A significant magnitude of hot cracking susceptibility shown in Fig. 2(c) and high crack density observed for 1.0 and 2.0 wt.% Si alloys in Figs. 5 and 6 can be clearly related to the low strengths, ductility, and modulus. A minimum in mechanical integrity appears to be at 1.0 wt.% Si, corresponding to the maximum crack density observed experimentally. Of the compositions that did not fracture prematurely due to solidification cracking, Al-0.5 wt.% Si alloy with some hairline cracks, had the lowest/inconsistent strength along with the highest elongation. The Al-16.0 wt.% Si alloy had the highest strength, but lowest elongation as presented in Figs. 8 and 9.

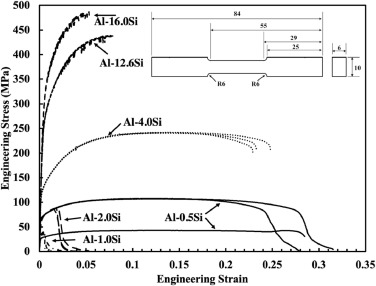


Fig. 8. Tensile stress-strain behavior of binary Al-Si alloys. Inset is an engineering drawing of tensile bar specimen with all units in mm.

Table 3. Tensile properties of the six binary Al-Si alloys produced by LPBF. All tensile specimens were produced with laser power, scan speed, hatch spacing, and slice thickness of 350 W, 1600 mm/s, 0.13 mm, and 0.03 mm, respectively, and had greater than 99 % relative density based on image analysis of optical micrographs.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Composition (wt.%)** | **Sample** | **0.2 % Yield Strength (MPa)** | **UTS (MPa)** | **Strain at Failure** | **Young's Modulus (GPa)** |
| Al-0.5Si | 1 | 71.5 | 107.6 | 0.279 | 53.7 |
|  | 2 | 70.9 | 107.1 | 0.315 | 51.5 |
|  | 3 | 28.0 | 42.9 | 0.286 | 17.1 |
|  | Avg. ± Dev. | 56.8 ± 20.37 | 85.9 ± 30.41 | 0.293 ± 0.0155 | 40.8 ± 16.78 |
| Al-1.0Si | 1 | 37.7 | 38.5 | 0.004 | 19.3 |
|  | 2 | 39.6 | 41.0 | 0.005 | 17.5 |
|  | 3 | 36.1 | 36.2 | 0.004 | 15.7 |
|  | Avg. ± Dev. | 37.8 ± 1.43 | 38.6 ± 1.96 | 0.004 ± 0.0006 | 17.7 ± 1.46 |
| Al-2.0Si | 1 | 69.6 | 86.4 | 0.015 | 38.5 |
|  | 2 | 70.2 | 89.9 | 0.020 | 40.0 |
|  | 3 | 69.6 | 88.3 | 0.017 | 36.5 |
|  | Avg. ± Dev. | 69.9 ± 0.24 | 88.2 ± 1.45 | 0.017 ± 0.0019 | 38.4 ± 1.42 |
| Al-4.0Si | 1 | 128.8 | 242.1 | 0.248 | 62.5 |
|  | 2 | 128.8 | 241.4 | 0.229 | 64.9 |
|  | 3 | 128.0 | 240.6 | 0.233 | 59.6 |
|  | Avg. ± Dev. | 128.5 ± 0.37 | 241.4 ± 0.63 | 0.237 ± 0.0081 | 62.4 ± 2.16 |
| Al-12.6Si | 1 | 272.7 | 438.6 | 0.077 | 66.5 |
|  | 2 | 269.7 | 436.8 | 0.079 | 69.6 |
|  | 3 | 269.7 | 348.0 | 0.079 | 69.8 |
|  | Avg. ± Dev. | 270.7 ± 1.42 | 407.8 ± 42.28 | 0.078 ± .0008 | 68.7 ± 1.49 |
| Al-16.0Si | 1 | 325.7 | 487.1 | 0.050 | 71.7 |
|  | 2 | 323.2 | 486.5 | 0.054 | 70.9 |
|  | 3 | 328.3 | 481.1 | 0.051 | 73.0 |
|  | Avg. ± Dev. | 325.8 ± 2.07 | 484.9 ± 2.69 | 0.052 ± .0016 | 71.91 ± 0.89 |

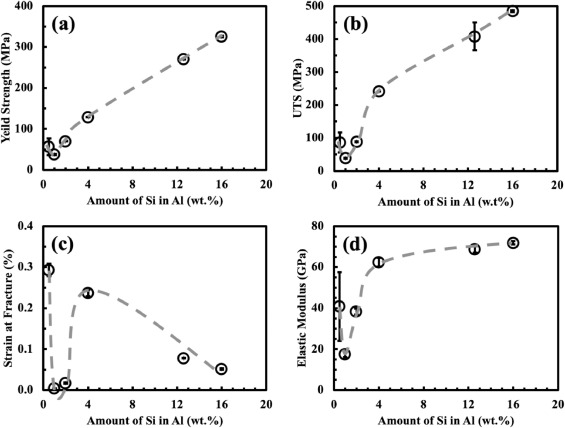


Fig. 9. Average (a) yield strength (0.2% offset), (b) ultimate tensile strength, (c) strain at fracture, and (d) elastic modulus as a function of Si concentration in binary Al-Si alloys.

As reported by Zhou *et* al. [34], LPBF AlSi10Mg contains a sub-grain cellular structure defined by the web-like Al-Si eutectic structure at the intercellular boundaries in an otherwise α-Al matrix. Fig. 10 presents representative backscatter electron micrographs from the XY cross-sections of the LPBF Al-Si alloys processed with a laser power and scan speed of 350 W and 1600 mm/s, respectively. Similar to the powder microstructure, Si partitioning is observed in all six compositions. As the concentration of Si in Al increases, the web-like Al-Si eutectic structure became better defined. An increase in strength and a decrease in ductility observed as a function of Si concentration may be attributed to the amount of eutectic structure, which can act to impede dislocation motion [35].

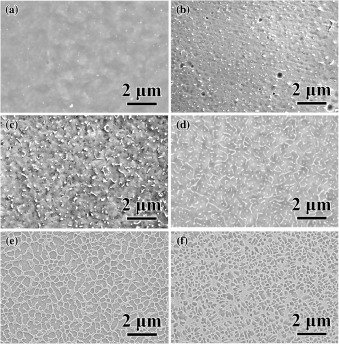


Fig. 10. High magnification backscatter electron micrographs from the XZ cross-sections of (a) Al-0.5Si, (b) Al-1.0Si, (c) Al-2.0Si, (d) Al-4.0Si, (e) Al-12.6Si, and (f) Al-16.0Si alloys.

# 5. Discussion

## 5.1. Cracking of binary Al-Si alloys

Fig. 7 compares the measured crack density to the hot crack susceptibility index, |dT/dfS1/2|, calculated from Eq. (6) for the binary alloys examined. The Al-0.5 wt.% Si alloy had the highest crack susceptibility, whereas experimental results showed that 1.0 and 2.0 wt.% Si alloys exhibited the highest severity of solidification cracking. Investigation by Singer and Jennings [25] using ring cast and clamped weld found that the maximum crack density was observed for alloys with 1.5 to 2.0 wt.% Si after the ring casting, and 0.6 to 1.0 wt.% Si after clamped welding. Pumphrey and Lyons [36] reported maximum cracking in alloys with 0.6 to 0.7 wt.% Si from ring cast and restrained weld experiments. Kimura *et* al. [37] examined various binary Al-Si alloys (i.e., 1.0, 4.0, 7.0, 10.0, 12.0 and 20.0 wt.% Si) with LPBF, and reported cracking in Al-1.0 wt.%, although more attention was given to the densification, mechanical behavior, and thermal properties. However, they suggested that compositions of Al-alloys with approximately 1.0 wt.% Si should be avoided due to cracking.

In order to account for the difference between the predicted and observed Si content corresponding to the most severe cracking, Liu and Kou [38] suggested the effect of solid-state diffusion on composition-dependent solidification cracking. Since the Eq. (2) assumes no diffusion in the solid, the work of Kurz and Fisher [39] included the addition of a dimensionless parameter to include the effect of diffusion. This parameter, α, is described by:

(7)

where DS is the diffusion of the solute in the solid matrix, tf (freezing range/cooling rate) is the freezing time, and λ2 is the secondary dendrite arm spacing (SDAS). The value of α is usually < 0.3, and α = 0 corresponds to no diffusion in the solid. The Scheil equation can then be modified to include the diffusion parameter, α, as follows:

(8)

where α’ is expressed as:

(9)

To demonstrate the effect of diffusion on the cracking susceptibility of the binary Al-Si system, values of α were calculated using Eq. (7) for varying solid-state diffusion coefficients. Values of tf and λ2 were held constant at 1×10−6 s and 300 nm, respectively, based on the average cell size and calculated cooling rate adopted from Hyer *et* al. [17] for LPBF of AlSi10Mg alloy. The values of DS chosen included 1 × 10−12 m2·s−1 found true at 873 K [40], right below the melting temperature, and 1 × 10−9, 2×10−9, and 9 × 10−9 m2·s−1, consistent with self-diffusion coefficients of Al above the melting temperature > 933 K, where 9 × 10−9 m2·s−1 is considered to be extreme of the solid-state diffusion coefficient [41,42]. Understandably, no solid should be observed above the melting temperature. However, the high cooling rates and associated thermal gradients found in LPBF processing would allow for di- and tri-vacancies present in the solid, near the solid-liquid interface, thus allowing for faster diffusion since the probability of atom jumping is higher [40,43,44].

Utilizing the modified Scheil equation in Eq. (8), the fS1/2 was calculated for the different α values calculated for the chosen DS. Fig. 11(a) presents fS1/2 vs. temperature curves for the 0.5 wt.% Si composition, where the maximum cracking susceptibility was observed in Fig. 2(c) for the case of no diffusion in the solid. For DS of 1 × 10−12 m2·s−1 that yields the magnitude of α = .0004, the influence of diffusion was negligible, i.e., similar to no diffusion in the solid. However, with increasing DS, i.e., increasing influence from solid-state diffusion, the steepness, |dT/dfS1/2| was observed to decrease as shown in Fig. 11(a). Even with a minor increase in the DS from 1 × 10−9 to 9 × 10−9 m2·s−1, the solidification behavior is shown to change significantly. Moreover, the melt fully solidified before reaching the eutectic temperature when DS = 9 × 10−9 m2·s−1.

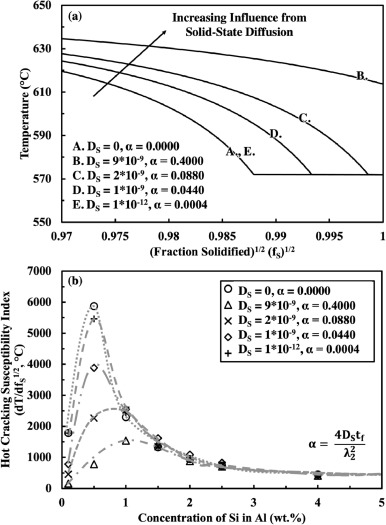


Fig. 11. (a) Temperature vs. fS1/2 for Al - 0.5 wt.% Si alloy with varying influence of solid-state diffusion; (b) |dT/dfS1/2| as a function of Si concentration in Al for various diffusion coefficients. The tf and λ2 were kept constant at 1 × 10−6 s and 300 nm, respectively.

Fig. 11(b) shows the |dT/dfS1/2| vs. Si concentration curves utilizing calculated values of α for the chosen DS. For the case of no diffusion in the solid, based on the assumption of the Scheil equation, where DS = 0 and α = 0, the maximum cracking susceptibility is observed at a concentration of 0.5 wt.%, just as presented in Fig. 2(c). Just below the melting temperature at 600 °C where DS = 1 × 10−12 m2·s−1, the effect of diffusion is found to be almost negligible, accruing larger magnitudes of |dT/dfS1/2|, similar to that of case for no diffusion. For the extreme case when the solid-state diffusion is high assuming that additional diffusion mechanisms such as di- and tri-vacancies are operative, e.g. DS = 9×10−9 m2·s−1 and α = 0.4, the overall magnitude of |dT/dfS1/2| decreased significantly as seen in Fig. 11(b). Moreover, the concentration of Si at which the maximum |dT/dfS1/2| occurs shifts from 0.5 wt.% Si to a higher concentration, at 1.0 wt.% Si. As the magnitude of DS is decreased, i.e. 2×10−9, and 1×10−9 m2·s−1, the magnitude of α decreases, and the magnitude |dT/dfS1/2| increases. Moreover, the relatively small decrease in the DS, i.e. 9×10−9 to 2×10−9 m2·s−1, decreases the magnitude of α considerably.

## 5.2. Cell size and cooling rate

Si partitioning was observed in the six binary Al-Si compositions as shown in Fig. 10, similar to that found in LPBF AlSi10Mg [17], where the cell size was related to the cooling rate. Even though the Si partitioning is observed in all six Al-Si compositions, microstructure with well-defined sub-grain cells with distinctive boundaries can only be identified in alloys with 12.6 and 16.0 wt.% Si alloys. But for Al-16.0 wt.% Si alloy, presence of primary Si and thick intercellular boundaries made it challenging to quantify the cell size. Therefore, for further analysis, easily identifiable cellular structure in the Al-12.6 wt.% Si alloy was employed.

Fig. 12 shows backscatter electron micrographs from the XY cross-sections of the Al-12.6 wt.% Si alloy samples produced with various laser powers and scan speeds. At constant power, i.e., 250 or 350 W, larger cells were observed with slower scan speeds. To quantify, the cell size measurement was performed via ASTM E1382 procedure using ImageJ. The measured cell sizes are reported in Table 4 and are presented as a function of scan speed in Fig. 13(a). The cell size decreased with an increase in scan speed.

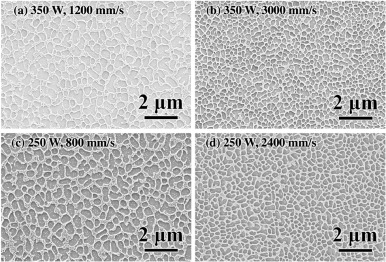


Fig. 12. Backscatter electron micrographs from the XY cross-sections of Al - 12.6 wt.% Si alloy samples produced by LPBF with (a) 350 W at 1200 mm/s, (b) 350 W at 3000 mm/s, (c) 250 W at 800 mm/s, and (d) 250 W at 2400 mm/s.

Table 4. The measured cell size and corresponding cooling rate estimated for LPBF Al - 12.6 wt.% Si cube samples.

|  |  |  |  |
| --- | --- | --- | --- |
| **Scan Speed (mm/s)** | **Cell Size (µm)** | **Cooling Rate from Eq. (10) 107 (K•s−1)** | **Cooling Rate from Eq. (12) 107 (K•s−1)** |
| 350 W |  |  |  |
| 1200 | 0.182 ± 0.044 | 2.8 ± 1.33 | 0.43 |
| 1600 | 0.163 ± 0.011 | 3.1 ± 0.64 | 0.58 |
| 2000 | 0.141 ± 0.011 | 4.9 ± 1.16 | 0.72 |
| 2400 | 0.115 ± 0.011 | 9.1 ± 2.30 | 0.86 |
| 3000 | 0.124 ± 0.008 | 7.1 ± 1.46 | 1.08 |
| 250 W |  |  |  |
| 800 | 0.202 ± 0.018 | 1.6 ± 0.41 | 0.40 |
| 1200 | 0.123 ± 0.018 | 7.8 ± 2.56 | 0.60 |
| 1600 | 0.133 ± 0.010 | 5.8 ± 1.43 | 0.81 |
| 2000 | 0.138 ± 0.013 | 5.3 ± 1.57 | 1.01 |
| 2400 | 0.129 ± 0.011 | 6.5 ± 1.70 | 1.21 |

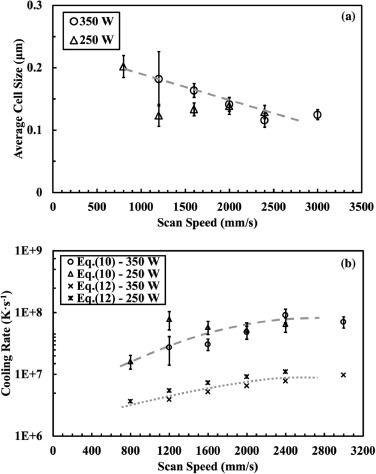


Fig. 13. (a) Average cell size measured by image analysis and (b) the corresponding cooling rate estimated as a function of scan speed for Al - 12.6 wt.% Si alloy.

Assuming that the cell size determined can be substituted for the secondary dendrite arm spacing (SDAS), λ2, cooling rate, T˙, can be estimated using the expression [20]:

(10)

where A = 43.2 and n = 0.324 for Al-alloy, reported by Matyja *et* al. [45]. Cooling rate was alternatively calculated using the analytical solution of Rosenthal equation, which models the heat produced by a moving point-heat source as found in the LPBF process [46]. An analytical solution to Rosenthal equation can be expressed as [46]:

(11)

where T is the final temperature, Q is the input energy usually estimated as the product of laser power and absorption coefficient (i.e., estimated as 0.35 for AlSi10Mg from [47]), R is radial distance from the laser beam position, κ is the thermal conductivity, ξ is the distance from the heat source, and φ is the thermal diffusivity. Thermal conductivity and thermal diffusivity were estimated utilizing the rule of mixtures for Al and Si, i.e., κAl and κSi of 221.75 and 83.68 W•m−1·K−1, respectively [5]; φAl and φSi of 9.7 × 10−5 and 8.8 × 10−5 m2•s−1, respectively [48]. In one dimension, a cooling rate, T˙, can be described as [46]:

(12)

where v is the scan speed, T is taken as the melting temperature (660 °C, 933 K) and To is the initial temperature, taken as 100 °C (373 K) of the build plate. Both Eqs. (10) and (12) are highly dependent on composition and the corresponding materials properties.

Cooling rates calculated using Eq. (10) based on measured cell size and using Eq. (12) based on the Rosenthal equation are reported in Table 4 and presented in Fig. 13(b). Overall, the cooling rate was estimated to be between 106 to 107 K•s−1, similar to that obtained by Hyer *et* al. [17] who also estimated the cooling rate for AlSi10Mg produced by LPBF using the same methods. The cooling rate calculated based on Eq. (10) was an order of magnitude higher than that calculated using (12), (13). Eq. (10) was specifically developed for dendritic structure, but the estimation made in this study used cellular structure with a substantial thickness of the boundaries, which can affect the overall outcome. Aforementioned, as the scan speed increased, the cell size decreased, which corresponds to a decrease in cooling rate. Therefore, given the sufficient energy input for melting, faster scan speeds corresponding to lower energy input would cause a faster cooling and finer cellular structure. Variation in cell size and cooling rate were not significant between the two laser powers used, 250 and 350 W. Therefore given the dense, crack-free, as-built LPBF samples, as per the Hall-Petch relationship [35], variation in scan speed may influence the mechanical behavior, while the power may not, as long as a completing melting occurs by incident laser beam.

## 5.3. Crack susceptibility dependence of thermal gradient

Avoiding specific Si concentration to minimize potential solidification cracking may be challenging. For example, some 6xxx series Al-alloys, such as AA6061, include additions of Si up to 1.0 wt.%. As Martin *et* al. [8] and Uddin *et* al. [12] reported, AA6061 is prone to solidification cracking when processed with LPBF, as demonstrated by Fig. 7. In order to remedy solidification cracking in LPBF AA6061, Martin *et* al. [8] suggested coating the AA6061 powder with Zr-hydride, that allows for inoculation of the AA6061 with Zr-aluminide during the LPBF, a methodology also adopted by Zhou *et* al. [6] by pre-alloying the powders. On the other hand, Uddin *et* al. [12] took a different approach, heating the build plate to 500 °C, which yielded a maximum volumetric density of 98.7% without any solidification cracking for AA6061.

From the Rosenthal equation in Eq. (11), the thermal gradient can be expressed as:

(13)

and the solidification front velocity, V, can be expressed as:

(14)

In Eq. (13), G is dependent solely on the laser power, and not the scan speed. On the other hand, according to Eq. (14), V is directly related to the laser scan speed. Heating the build plate to 500 °C would reducing the temperature range (T-To) in the Rosenthal solutions, i.e., Eqs. (11)–(13), thus reduce the cooling rate and thermal gradient significantly. As the cooling rate decreases to the order of 103 to 104 K•s−1, the diffusion parameter, α in Eq. (7) would also increase and would help mitigate solidification cracking. Of course, the more significant benefit of a heated build plate has been discussed by Uddin *et* al. [12] who postulated that the reduction in the (T-To) would allow relief of thermal residual stress.

As shown in Fig. 6, a higher cracking density was observed at a lower laser power (250 W), consistently for both the Al-1.0 and Al-2.0 wt.% Si alloys. Simulations by Vasinonta *et* al. [49,50] for welding of stainless steel 304 (SS304) reported that an increase in the thermal gradient increased the overall thermal residual stress. Since the thermal gradient increases with decreasing laser power, to help relieve thermal residual stress, welding typically utilizes [24,46] a larger laser power and higher energy input to decrease the thermal gradient. Ali *et* al. [51,52] systematically studied the changes in residual stress as a function of build plate temperature during LPBF Ti-6Al-4V, and reported that an increase in build plate temperature decreased the magnitude of residual stress. Xu *et* al. [53] attempted a similar strategy for Ni-based superalloy IN738, previously shown to crack during LPBF processing due to its high Ti+Al content. They found a significant reduction in solidification cracking with higher build plate temperature, however, were unable to eliminate the cracks completely.

The thermal gradient and G/V terms also control the solidification characteristics and microstructure. A higher thermal gradient can lead to formation of columnar grains, whereas a lower thermal gradient would yield a more equiaxed structure. In general, higher cooling rates would reduce the grain size, just as smaller cell size was observed with higher cooling rate as shown in Fig. 13. Development of smaller equiaxed grains would tend to lower the cracking tendency. Kou and Le [54] studied the grain structures of gas-tungsten-arc (GTA) welded AA6061 at different laser powers and scan speeds. They found that a higher energy input (W), at a given scan speed, yielded more equiaxed grains. Even though Uddin *et* al. [12] did not present microstructure, it is likely that reducing the thermal gradient helped to mitigate the development of columnar grains. Zhou *et* al. [10], who recently published work on LPBF of ternary Al-6.0wt.%Zn-2.0wt.%Mg and quinary Al-6.0wt.%Zn-2.0wt.%Mg-0.7wt.%Sc-0.3wt.%Zr, modeled after AA7075 alloy, reported that equiaxed grains were observed more so with the use of laser power at 350 W than at 250 W. They too attributed the equiaxed structure found in samples with lower thermal gradient at 350 W than at 200 W.

# 6. Conclusions

To better understand the LPBF of Al-alloys, effects of solute concentration on the hot cracking susceptibility, microstructural development and mechanical behavior in tension were examined with six binary Al-Si alloys with hypo-, near-, and hyper-eutectic compositions. Gas atomization was carried out to produce alloy powders of respective compositions, and samples for microstructural analysis and tensile testing were manufactured with LPBF as functions of laser power and scan speed. Findings from this study can be summarized as:

* Utilization of the “hot cracking susceptibility index” based on the magnitude of |dT/dfS1/2|, suggested a strong dependence of hot cracking susceptibility on composition. Lower concentrations of Si in the binary Al-Si alloys exhibited larger magnitudes of |dT/dfS1/2|, corresponding to a higher cracking susceptibility. This approach may be applicable to other alloy systems and can assist in the designing of alloys suitable for LPBF.
* The maximum cracking susceptibility, |dT/dfS1/2|, was found to decrease significantly in magnitude and shift to a higher concentration of solute with the contribution from diffusion. In addition to stress relief by high temperature build plate and inoculation for grain refinement, understanding the role of diffusion in solid during solidification, despite the rapid cooling rate, may be an important aspect to explore to mitigate solidification cracking.
* Nearly full volumetric density (> 99 %) was observed for all six binary Al-Si alloys produced by LPBF. However, alloys with 1.0 and 2.0 wt.% Si exhibited solidification micro-cracking, regardless the LPBF parameter employed. Observation of these cracks in alloys with 1.0 and 2.0 wt.% Si corresponded well to the composition-dependent “hot crack susceptibility index” calculated based on the equilibrium binary phase diagram, i.e., |dT/dfS1/2|.
* Tensile mechanical behavior exhibited an increase in strength and a decrease in ductility as the concentration of Si increased, except for the alloys with 1.0 and 2.0 wt.% Si, because presence of solidification cracks caused premature fracture. Si was found in all alloys, either as a fine Si particle with low Si concentration, or a web-like network of Al-Si eutectic for alloys with high Si concentration. An increase in Si concentration led to an increase in the amount eutectic network that would act as barriers to dislocation motion.
* The cellular size quantified for the near-eutectic Al-12.6 wt.% Si alloy was found to decrease with an increase in scan speed of laser, powered either at 350 W or 250 W. Cooling rate was estimated from the cell size and Rosenthal equation to be on the order of 106 to 107 K•s−1. The increase in cooling rate corresponded with faster scan speed and smaller cell size.
* For cases where the alloy composition cannot be changed or modified, reduction of the thermal gradient may help reduce the grain size and residual stress, which would lead to mitigation of solidification cracking. Moreover, reduction in the thermal gradient would unilaterally reduce the cooling rate, per the Rosenthal equation, allowing for crack mitigation.

# Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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