Particle property impact on its distribution during laser deep alloying processes

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Abstract

Alloying of surface layers could be recently improved by using the laser deep alloying process. For this, a highly focused laser beam is guided on the material surface producing a wide and deep melt pool. Alloying particles are applied as a powder using a powder nozzle. This process allows deep alloying and dispersing of particles. The homogeneity and the penetration depth of the particles in the melt pool vary when using particles with different thermo-physical properties. Therefore, the influence of particle properties, feeding rates and modulation strategies of the laser on the resulting distribution in the weld pool is studied in this work. The modulation strategy and the feeding rate have a minor impact on particle distribution within the observed parameter field. Particle entering and distribution depend mainly on its density and wetting behavior with aluminum. Most homogeneous distribution has been found using TiB\textsubscript{2}-particles.

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1. Introduction

Surface treatment of components and assemblies is used in many industrial applications. Bewilogua et al. (2009) used hard coatings on tools for manufacturing processes and Klocke et al. (1996) managed metal surface hardening by using temperature treatment by alloying or dispersing particles in the melt pool. Aluminum can be treated with alloying elements e.g. for improved corrosion behavior, shown by Neugebauer et al. (2011) or by dispersing hard solid particles into the melt pool to increase wear resistance.

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This process requires a molten surface region in order to insert solid particles into the liquid base material. Dispersing of non-melting material can increase the strength and wear resistance of the surface layer. Laser processes are a good choice to melt the material due to their controlled local heat input at high velocities. Partes et al. (2007) pointed out that the process of heat conduction welding is regularly used to melt the material surface. Almeida et al. (1998) achieved an increased hardness and wear resistance by dispersing niobium in aluminum. A homogeneous distribution in depth could be created by re-melting the weld seam. Majumdar et al. (1999) found positive effects on wear resistance for Si in Ti base material and Cr in Cu base material. However, the depth and width of the melt pools of the laser heat conduction process is limited. In order to achieve surface treatments of larger surface areas a lot of paths are mandatory. Due to the circular shape cross section of the weld seam, a big overlap of the paths is necessary in order to guarantee homogeneous melting of a surface area at the same depth. In addition, Guyenot (2004) found that dispersed powder material is not reliably homogenously distributed in the melt pool.

Compared to melt pools produced by heat conduction laser welding, deeper and wider melt pools can be achieved by using the laser deep penetration process. Using high power lasers that provide high intensities lead to an increased laser energy input into the material and to material vaporization and a forming of a keyhole. This process mode can be used for deep alloying or dispersing of elements as described by Partes et al. (2007). Therefore, Vollertsen et al. (2007) recently developed the deep alloying process at BIAS. Elements are applied as powder using a nozzle pointing on the weld pool surface during the laser deep penetration process. In order to achieve a wider molten pool and an additional stirring of the melt pool for distributing particles or elements in depth, the laser beam is modulated during the process. Stirring of the melt pool is assumed to more effectively distribute element particles in depth. Until now, different kinds of particles have been tested but no systematic evaluation of the influence of powder material on the homogeneity has been done. Therefore, the influence of different powder particles on their distribution is evaluated in this work.

Properties of particles are assumed to influence the particle behavior when hitting the melt pool. Vreeling et al. (2000) found that the kinetic energy of a particle has a significant impact on the entering of the particle into the melt pool in order to overcome the barrier energy of the molten surface. Partes (2008) found that low melting temperature, low heat conduction and good absorption behavior of laser energy can result in melting of the particle surface which can result in improved wetting behavior when the particle hits the melt pool. In case of a solid particle hitting the melt pool the particle behavior is influenced by the wetting properties in combination with the liquid material. Wetting behavior has been studied by Weirauch et al. (2005) in experiments using the sessile drop method of wetting of liquid aluminum on titanium diboride (TiB2) non-porous surfaces. Devyatkin et al. (2000) showed that when the oxide layer is broken off at high temperatures wetting of the TiB2-drop with low wetting angles can be observed. In laser deep penetration processes the temperatures are high enough to achieve an oxide free surface around the keyhole. Oh et al. (1989) found that TiB2 shows a better wetting behavior than boron carbide (B4C). WC has to be already wetted by the binder for production of tungsten carbide (WC) particles. Resulting properties of the WC-binder mixture strongly depend on the binder material as described by Prakash (1980). Co is commonly used as a binder but also Fe, Ni or Al are known for good wetting abilities on WC. Li et al. (2001) found that the oxidation of Co reduces wettability of WC in liquid aluminum.

2. Experimental

A laser deep alloying process is used in this work (Fig. 1a). The beam of a disk laser (Trumpf TruDisk8002, max. power of 8 kW) is focused on the aluminum (AA6082 alloy) surface using a 3-D-Scanner-Optic (Trumpf PFO, f-theta lens with a focal length of 450mm). The laser is moved with a constant welding speed of \( v_s = 0.5 \text{ m/min} \) and a laser power of 4 kW. The powder nozzle points on the keyhole at an angle of 45° and the particles pass through the laser beam before hitting the molten material in the direct vicinity of the keyhole. Circles, “8-shapes” (forward and backward) and linear modulation at a frequency of 5 Hz at a diameter/radius \( r \) of 0.5 mm are used in this work (Fig. 1b). Three powder particle materials (WC-Co, TiB2 and B4C), commonly used for improving wear resistance of surface layers, are applied to the process at different powder feed rates from 0.8 cm³/min to 3.6 cm³/min.
Powder feed rates have been evaluated for each powder material with a high accuracy scale by measuring the delivered powder mass per time unit. Additionally, high-speed-videos (4000 frames/s) have been taken to observe the particles penetrating the laser beam and measuring the velocity of the particles. By measuring the distance the particle moves in an amount of time the particle velocity $v_p$ can be calculated.

For evaluation, cross section polishes of the seams have been made. In order to evaluate the homogeneity of the seam the amount of dispersed material in relation to the base material is measured in different sections in depth (Fig. 2). The homogeneity $H$ of the particle distribution is evaluated using a particle detection routine in MatLab (Version R2009a). Six elements of the melt pool in different depths at same size have been chosen to calculate homogeneity.

The homogeneity $H$ for a cross section is defined using eq. 1 containing the number $N$ of elements used, the number of particle pixels $D_i$ found in the section $i$ and the average particle number of all sections $\overline{D}$. The formula divides the average of the particle concentration by the standard deviation. A high value of $H$ implies a homogeneous distribution. For statistical certainty at least three cross sections of one parameter set have been evaluated.
3. Results and discussion

3.1. Particle entry into the melt pool

For the trials single paths are produced using the deep alloying process at varying modulation strategies, powder feeding rates and powder materials. Fig. 3 shows cross sections of the dispersed surface region when using “8-forward” modulation.

Compared to the cross sections evaluated with no powder application melt pool size is slightly reduced when applying powders to the process. As described by Partes (2008) the powder penetrates the laser beam before striking the melt pool surface and laser energy is absorbed and reflected on the particles leading to a reduced energy input on the material surface.

Entering of the particle into the melt pool requires energy in order to overcome the surface tension of the aluminum melt. A single particle needs kinetic energy to overcome that barrier energy.

\[
E_{\text{kin}} = \frac{1}{2} \cdot m_p \cdot \left(v_p \cdot \sin(\phi)\right)^2
\]  

In order to calculate the kinetic energy (eq. 2) of the particle, mass of a single particle \( m_p \), velocity of a particle \( v_p \) and the nozzle inclination angle \( \phi \) are needed (Fig. 4). The velocity of the particles is measured from high-speed-video pictures (table 2). Depending on the incident angle of the nozzle \( \phi \) the vertical velocity can be calculated which is contributing to the kinetic energy of the particle.
The particle mass \( m_p \) can be derived from eq. 3 with the particle volume and the density \( \rho \) (table 1) assuming a spherical particle shape. The density \( \rho \) and the average particle radius \( d \) of each particle material used are taken from table 2.

\[
m_p = \rho \cdot \frac{4}{3} \cdot \pi \cdot d^3
\]

Knowing about the kinetic energy of the particle, Vreeling et al. (2000) calculated the minimum velocity of a particle to overcome the melt surface barrier energy \( E_{\text{bar}} \) depending on the wetting behavior of the materials (eq. 4).

\[
v_{\text{min}} = \sqrt{\frac{3}{2 \cdot \gamma_h \cdot d \cdot \rho} \cdot \left( \gamma_h + \gamma_l - \gamma_{pv} \right)}
\]

\( \gamma_l \) denotes the surface tension between the liquid and the vapor, \( \gamma_p \) the surface energy between the liquid and the particle and \( \gamma_{pv} \) between the particle and the vapor. Values for the surface energy of the powder materials \( \gamma_{pv} \) are estimated by using equations for binary mixtures from Butler (1932) with the substance amounts from table 2. Resulting values are listed in table 2.

Table 1. Properties of dispersed particle materials.

| Material and substance amounts | Density \( \rho \) | Particle radius/ average particle radius \( d \) | Melting temperature \( ^\circ \text{C} \) | Heat conduction \( \text{W/(m*K)} \) | Source |
|-------------------------------|------------------|-----------------------------|-------------------|----------------|--------|
| WC/Co (88/12) W: 94%; C: 6%   | 15.6             | -25 +10 / 17.5              | 2850              | 42             | Heigl (2004) |
| TiB\(_2\) (95%) Ti: 69%; B: 31% | 4.4              | -37 +16 / 27                | 2900              | 27             | Vollertsen et al. (2008), Rudolph (2001) |
| B\(_4\)C (99%) B: 78%; C: 22% | 2.52             | -29 +11 / 20                | 2445              | 30 to 42       | Lipp (1966), Cotton (2014) |

The interface energy \( \gamma_{lp} \) is calculated with Young’s equation (eq. 5) while contact angles are taken from literature (table 2, WC/Co by Panov et al. (2009); TiB\(_2\) by Rhee (1970); B\(_4\)C by Fox et al. (2010)).

\[
\gamma_{lp} = \gamma_{pv} - \gamma_h \cdot \cos(\Theta)
\]

The minimum particle velocity needed to overcome the barrier energy is reached for TiB\(_2\)- and B\(_4\)C-particles but not for WC-particles (table 2). The small measured contact angle and therefore good wetting behavior of WC-particles seems to lead to a particle input although the minimum particle velocity is not reached. Therefore, it can be assumed that WC-particles rest on the melt pool surface until the wetting process leads to the entry of the particle. Oh et al. (1989) observed good wetting behavior when using TiB\(_2\)-particles and in combination with the particle velocity higher than the calculated minimum velocity, particle input seems to be increased. TiB\(_2\)-particles seem to easily access the aluminum melt pool. As mentioned before, the oxide layer is broken off at the high process temperatures and therefore good wetting behavior of TiB\(_2\)-particles with liquid aluminum can be assumed like it has been found by Devyatkin et al. (2000) and Weirauch (2005) (see chapter 1).
Table 2. Interface energies and particle velocities.

| Material and substance amounts | Contact angle $\theta$ (°) | Calculated surface energy $\gamma_{pv}$ (Butler 1932) mN/m | Surface tension unoxidized aluminum $\gamma_v$ (Kaptay 1996) mN/m | Interface energy $\gamma_p$ (eq. 5) mN/m | Minimum particle velocity $v_{min}$ (eq. 4) m/s | Measured particle velocity $v_p$ m/s |
|------------------------------|-----------------------------|-------------------------------------------------------------|---------------------------------------------------------------|----------------------------------------|------------------------------------------|----------------------------------|
| WC/Co (88/12) W: 94%; C: 6% | 35                          | 2690 (WC)                                                  | 1100                                                         | 1679                                   | 4.5                                      | 0.97 (±0.27)                      |
| TiB$_2$ (95%) Ti: 69%; B: 31% | 55                          | 1938                                                       | 1100                                                         | 1949                                   | 3.9                                      | 4.15 (±0.5)                       |
| B$_4$C (99%) B: 78%; C: 22% | 80                          | 1500                                                       | 1100                                                         | 1406                                   | 2.9                                      | 6.4 (±0.35)                       |

In addition, chemical reactions can change the wetting behavior. As Young's equation (eq. 5) assumes non-reactive wetting, chemical reactions are not considered but can influence the surface tension values. Fox et al. (2010) found that the products of the reaction between aluminum and B$_4$C reduce the surface tension of the melt leading to a better wetting behavior of the B$_4$C-particle. As mentioned, Li et al. (2011) found that Co from the WC-Co-particles is oxidized during the process which leads to a reduced wetting behavior of WC and aluminum.

The effect that particles are heated up while they penetrate the high power laser beam before hitting the surface is not taken into account in the calculation. Nevertheless, particles are heated up by the absorbed laser energy and it is possible that the surface of the particles is partly melted as described by Partes (2008). If that happens the particle entry into the melt pool should be improved as there is liquid particle material floating into liquid aluminum and it is not a wetting of a liquid and a solid. Assuming similar absorption of the laser energy on the particle surface the different melting temperatures of the particle materials (table 1) should lead to different surface properties after being heated by the laser beam. While TiB$_2$ and WC have melting temperatures above the vaporization temperature of aluminum (2470°C) they are assumed to remain solid during the absorption of laser energy and the exposition to the aluminum vapor plume from the keyhole. B$_4$C-particles melt at a lower temperature (2445°C) and have a low heat conduction value (table 2) compared e.g. to aluminum (235 W/(m*K)) which is assumed to support the creation of a liquid film around the particle before hitting the surface. Along with the particle velocity exceeding the minimum velocity to enter the melt pool the access of B$_4$C-particles is assumed to be improved by the particle heating process.

3.2. Particle distribution

The distribution of particles entering the melt pool differs when using different particle materials as can be seen in Fig. 5. The differences are assumed to originate from the particle properties. The observed different modulation strategies lead to similar particle distributions in depth while linear and circular modulations transport more particles in deeper melt pool regions (Fig. 5).

When using WC-particles it seems that they accumulate on the melt pool surface and a high number of WC-particles can be found in the upper part of the seam while some particles are transported in deeper regions (Fig. 5b). B$_4$C-particles can be mainly found in the upper regions of the melt pool while TiB$_2$-particles can be found at similar amounts in all depths of the melt pool (Fig. 5b). The same tendencies can be found when using different modulation strategies.

Homogeneity has been evaluated for different modulation strategies and particle materials (Fig. 6). Homogeneity values do not significantly change when using different modulation strategies for all observed particle materials e.g. for B$_4$C-particles (Fig. 6a). But the distribution of the particles in the melt pool differs when using different particle materials (Fig. 6b). While some of the WC-particles entering the melt pool are distributed by the melt flow to the melt pool tip B$_4$C-particles stay in the upper parts of the melt pool (Fig. 5b).
Therefore, the homogeneity of seams produced with B₄C-particles is calculated lower than for TiB₂. The homogeneity of B₄C-seams is calculated slightly higher than for WC-seams which results from the higher WC-particle accumulation in the upper regions of the seam compared to B₄C (Fig. 5b). TiB₂-particles are distributed in the whole melt pool. Therefore, these seams show higher homogeneity values. TiB₂- and WC-particles can be found in all depths while B₄C-particles remain in the upper parts of the melt pool.

Particles seem to be distributed in the melt pool by fluid flow. By using laser beam modulations additional flow patterns are introduced into the melt pool while for the chosen parameter set the modulation strategy has a minor influence on the homogeneity. As WC-particles show a delayed entry behavior they are assumed to stay in the upper melt pool region and cannot be distributed by the melt flow due to the fast process and the resulting fast solidification of the melt pool. The high density of WC can lead to a downward movement of the particles in the melt pool due to gravity forces and some particles reach the lower melt pool regions. The low density of B₄C can be one of the reasons for the remaining of the B₄C-particles in the upper melt pool regions due to buoyancy forces. Influence of particle distribution on mechanical properties will be discussed in future works.

4. Conclusions

The influence of different particle materials with different properties on its distribution in the weld seam during laser deep alloying has been investigated. Experimental results show that the distribution of particles in the melt pool varies depending on the particle material. Particle entry depends on the kinetic energy of the particle and its
wetting behavior with the molten material. Distribution in the melt pool takes place due to the melt flow and the influence of buoyancy forces depending on the density of the particle. TiB$_2$-particles are well distributed in the melt pool and show highest homogeneity values compared to WC-Co and B$_4$C. Different fluid flows introduced by using different laser modulation strategies result in similar homogeneity values of the particle distribution when using the same particle material.

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