High Thermal Conductivity Coatings via LENS™ for Thermal Management Applications

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Reviewed, accepted 9/15/09

Abstract

Surface modification has been used to improve wear resistance, corrosion resistance and thermal barrier properties of metals. However, no significant attempts have been made to improve thermal conductivity by surface modification. In this work, we have examined the feasibility of enhancing thermal conductivity (TC) of stainless steel by depositing brass using Laser Engineered Net Shaping (LENS). The coating increased the TC of the substrate by 65% at 100 C°. Significantly low thermal contact resistance was observed between the coating and the substrate due to minimal dilution and defect free sound interface. Our results indicate that laser processing can be used on low coefficient of thermal expansion metal matrix composites to create feature based coatings to enhance their heat transfer capability.

Introduction

Coating is a unique way to tailor the surface properties of a component to suit a specific environment without sacrificing the bulk characteristics of the structure. In recent years, lasers have been extensively used as a surface modification tool for engineering properties of materials and structures [1]. Laser can be used to develop a coating or cladding of a material having a property grossly different from that of the substrate. These coatings are metallurgically bonded, with the substrate providing a sound and adherent interface. The surface structure can be modified in accordance with the service requirement by varying process variables such as laser power, scan speed, beam diameter, precursor composition and precursor feed rate [2, 3]. Till date lasers have been used to deposit alloys and ceramics to create structures with high hardness, excellent wear [4] and erosion resistance [5], and highly encouraging thermal barrier properties [6, 7]. However, there have been no attempts to improve the thermal conductivity (TC) of a material via surface modification, except one preliminary study by our group [8]. Surface modification of an alloy substrate with high thermal conductivity coating can not only improve thermal properties but also thermal performance by providing faster heat dissipating surface layer. Moreover, it has been shown that the laser-based deposition techniques usually ensure good metallurgical bond with uniform intermixed region between the coating and the substrate material [9]. Such structures with enhanced heat dissipating top layer can find potential applications in heat exchanges of automotive and power plant industries. For example, copper or copper alloy coating on stainless steel would provide necessary corrosion resistance on one side and high heat dissipating layer on the other side leading to overall enhanced thermal performance. Moreover, this coating approach can also be implemented on low coefficient of thermal expansion (CTE) metal matrix composites (MMCs), used in electronic components, to improve their thermal performance by providing enhanced heat dispersing surface layer. In line with this, the TC of Mo–Cu alloys has been increased, from ~190 to about 245 W/m K, by bonding these alloy with outer copper layers (Cu/Mo–Cu/Cu) [10]. However, these laminates usually suffer from high interface thermal contact resistance due to sharp interface. On the other hand, laser-based deposition techniques usually ensure good metallurgical bond with uniform intermixed region between the coating and the substrate materials [9, 11]. In addition, laser surface modification is characterized by rapid solidification rates, up to $\sim 10^3$–10^5 K/s, eliminating formation of undesirable equilibrium intermetallic compounds between metallurgically incompatible metals/alloys [12]. This feature is extremely useful when developing a copper-based coating on steel base substrate, which has a high affinity to form cracks due to low solid solubilities, as in the present work.

In this study, high thermal conductivity brass (70Cu-30Zn) coating was prepared on AISI 410 stainless steel substrate to enhance its inherent TC, using laser engineered net shaping (LENS™). The simplicity of the coating process and flexibility in selecting process parameters with a high degree of deposition accuracy for desired physical and mechanical properties at appropriate locations of the part are some of the advantages of LENS™. This feature enables us to fabricate feature based high TC coating for enhanced thermal performance.

Experimental procedure and Materials

Commercial stainless steel grade 410 flat sheet of 3 mm thickness was used as the substrate material. Brass alloy powder (Sigma-Aldrich) with particle size range between 50-150μm, based on sieve analysis, was used to create the coating. The chemical composition of this particular alloy powder is copper-70wt% and zinc-30wt%. LENS™-750 (Optomec Inc. Albuquerque, NM, USA) fitted with a 0.5 kW Nd-YAG laser system was utilized to satisfactory manufacture the high thermal conductivity brass coating disks with 12.5 mm diameter. A laser power level of 400 W, i.e. corresponding to a laser beam energy density of 226 W/mm^2, was used to partially melt the alloy powder during the deposition process to create the brass coating on the substrate. In this work, scan speed of 15 mm/s and powder feed rate of 22 g/min were employed. The parameters were selected based on previous optimization to create fully dense, good-quality coatings i.e. no voids. The coating thickness was controlled by the number of layers deposited on the substrate. However, post-deposition sample polishing was done to achieve the desired coating thicknesses. The coating sample was segmented and mounted for metallographic analysis. The top surface along with the cross-section of the sample was polished, cleaned and then etched with a solution of $FeCl_3$ (g), HCl (ml), and distilled water (ml) in a ratio of 1:20:100 to reveal the coating's microstructure features. Microstructural characterization of the coating sample was evaluated using a field emission scanning electron microscope (FEI, 200F). Vicker's microhardness measurements (Leco, M-400G3) were performed on the laser processed coating sample using 50 g load for 15 s. An average value of 5 measurements was reported.

To evaluate the thermal properties of the substrate and the brass coating, the sample was polished and cleaned with alcohol, to remove any dirt particles and grease form the surface. The polished sample was lightly coated with graphite to ensure proper testing and accurate results, but mainly to ensure that the emissivity or absorptivity was high so that we can increase the energy absorbed on the flashed side of the sample and also to increase the temperature signal on the back side of the sample. The thermal diffusivity and specific heat were measured using the LFA 447 *Nanoflash*® (NETZSCH). The LFA 447 *Nanoflash*® is based on the well known flash method (ASTM E 1461-07) to obtain the thermal properties. This method uses a high-intensity short duration light energy pulse. The energy of the pulse is absorbed on the front surface of the sample and the resulting rear face temperature rise is measured using an infrared detector. By analysis of the resulting temperature versus time curve, the thermal diffusivity can be determined. When the thermal diffusivity of the sample is to be determined over a temperature range, the measurement must be repeated at each temperature of interest. Simple geometry, small specimen size requirements, fast testing, easiness of handling and high accuracy makes the flash method very versatile and advantageous. Another advantage of the flash method is the fact that specific heat values can be obtained directly from the testing procedure using differential scanning calorimetry and a well know material as the reference. The coating was tested at 25 to 250 °C. Each measurement taken was an average over 10 individual data points assuring accurate results.

Results and Discussion

Coating Microstructure and Hardness. Laser power level of 400 W with 15 mm/s scan speed and a powder feed rate of 22 g/min completely melted the brass powder resulting in a fully dense coating on the AISI 410 stainless steel. As shown in **Figure 1a**, defects such as cracks, residual porosity and voids were also absent in the coating, which could have detrimental effect on thermal properties of the coating/composites.

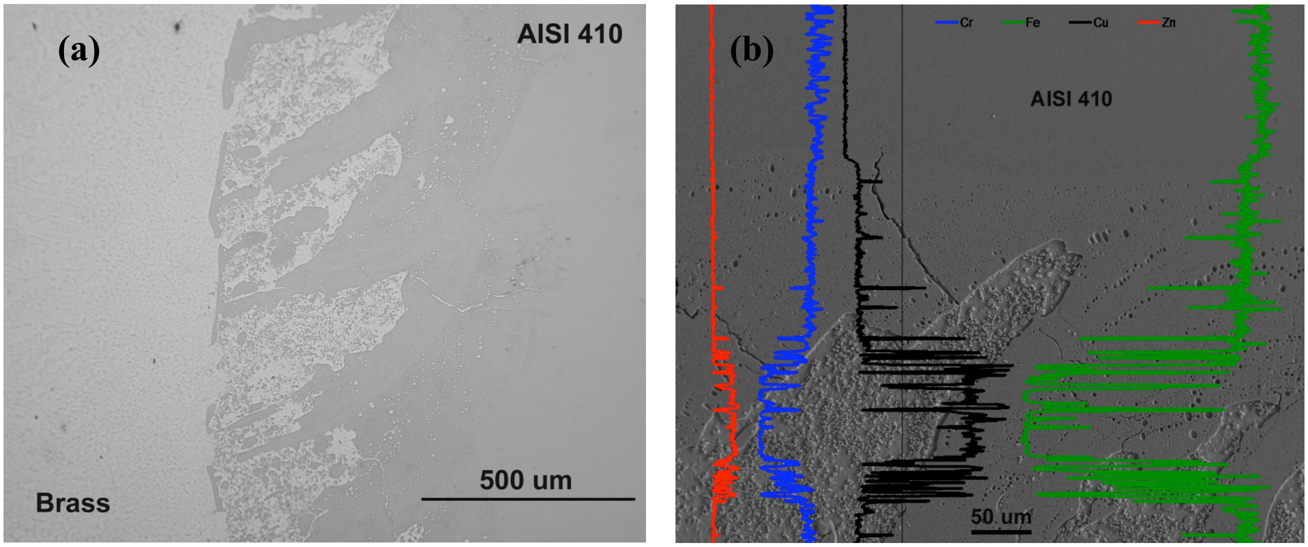

Figure 1: (a) Microstructure showing defect free and fully dense brass coating on stainless steel substrate, (b) High magnification SEM microstructure of coating-substrate interface and composition variation across the interface.

During laser processing the material's laser absorptivity plays a special role, because it controls the energy input from the laser to the sample. The laser absorption coefficient of the material is directly proportional to its electrical resistance and inversely proportional to the wavelength of the laser [13]. The relatively high electrical conductivity of brass than AISI 410 steel decreases its laser absorption coefficient. Under identical processing conditions of laser power and scan speed, the total energy absorbed by the brass will thus be lower than the energy absorbed by the AISI 410 steel substrate, which can lead to incomplete melting at the interface of individual layers and consequently poor interlayer bonding. However, in the present coating, complete melting between individual layers and between the coating and the substrate indicates that the laser energy input used to fabricate the coating was sufficient to completely melt the brass leading to fully dense product.

Another important factor that is of great importance is the interface region between the brass coating and the substrate. The interface was observed to be crack free and contained an intermixed region with uniform distribution of brass and steel throughout the region. A magnification SEM microstructure of the interface is shown in **Figure 1b**. During LENS™

processing, initially the substrate is partially melted and then the powders are injected into the molten pool, this creates an intermixed/diffuse interface region rather than a sharp interface during plasma spraying or roll bonding to create such composites. The absence of sharp interface between the coating the substrate would reduce the thermal contact resistance at the interface and improves heat transfer capability between the two materials. The thermal contact resistance of the laser processed composite was measured between 25 and 250°C using the two layer multi-material mode available in LFA 447 *Nanoflash*®. It was observed that the temperature has no effect on the thermal contact resistance over the studied temperature range. An average value of 1.982×10^{-11} m^2K/W was recorded for the 0.45 mm brass coating on AISI 410 stainless steel. The extremely low contact resistance values were presumably arises due to the absence of sharp interface between the coating and the substrate. In addition, the concentration profile across the coating-substrate interface, shown in **Figure 1b**, illustrates that the laser deposition did not result in any sharp interface or any intermetallic formation within the coating sample. The EDS line scan also shows how the Cu-Zn infiltrates the AISI 410 stainless steel substrate more than the stainless steel on the Cu. This can be attributed to the melting of the substrate as we introduce the brass powder.

Average hardness of the laser processed brass coating was 92 ± 6 HV and that of steel substrate was 360 ± 5 HV, while the average hardness of the intermixed region was around 146 HV. In actuality the hardness of the interface gradually increased from 100 HV to around 195 HV as the substrate was approached. Throughout the coating cross section no abrupt increase in the hardness was observed. The fact that we melt and then rapidly quench (fast cooling rate) does not allow enough time for the brass and stainless steel to form any intermetallics or cracks at the interface region. The same explanation holds for the low contact resistance measured from the experimental procedure for the high TC brass coating materials. There was a noticeable increase of hardness value of the substrate near the intermixed region. This was due to the melting and rapid solidification of the steel resulting in martensite in this steel. Laser surface melting has been shown to increase the hardness of AISI 410 stainless steel due to the formation of martensite during cooling [14]. The high hardness of substrate near the interface could also be due to solid solution strengthening of Cu in steel.

Thermal Conductivity. The thermal conductivity, k, of the coating, and substrate was calculated using the following formula: $k = \alpha \cdot c_p \cdot \rho$, where α is the thermal diffusivity (m^2/sec), c_p is the specific heat (J/(kg·K), and ρ is the bulk density (kg/m^3) of the material. The bulk density of the samples was calculated from sample geometry and mass. The thermal diffusivity and specific heat of the samples were experimentally determined. **Figure 2** shows that the thermal conductivity increases with increasing temperature for the brass coated AISI 410 steel sample. However, the TC of AISI 410 substrate sample initially increased up to 100°C and remained unchanged up to 250°C. The TC values varied from 33 to 40 W/mK for the 0.45 mm thick brass coating and 20 to 23 W/mK for the SS410 substrate over the temperature range of 25 to 250°C. The TC of the substrate increased between 60 and 75% due to the deposition of the brass coating and test temperature. As experimentally determined specific heat and thermal diffusivity increased with increasing temperature for brass-steel composites the thermal conductivity of these samples also increased with temperature. The increase in the TC of the composites with the deposition of the brass coating is understandable. The ratio of coating thickness over the substrate thickness was 0.4. This high ratio means high volume of higher TC brass in the composites, enabling faster heat removal from the steel substrate and faster heat dissipation from the top surface to the environment.

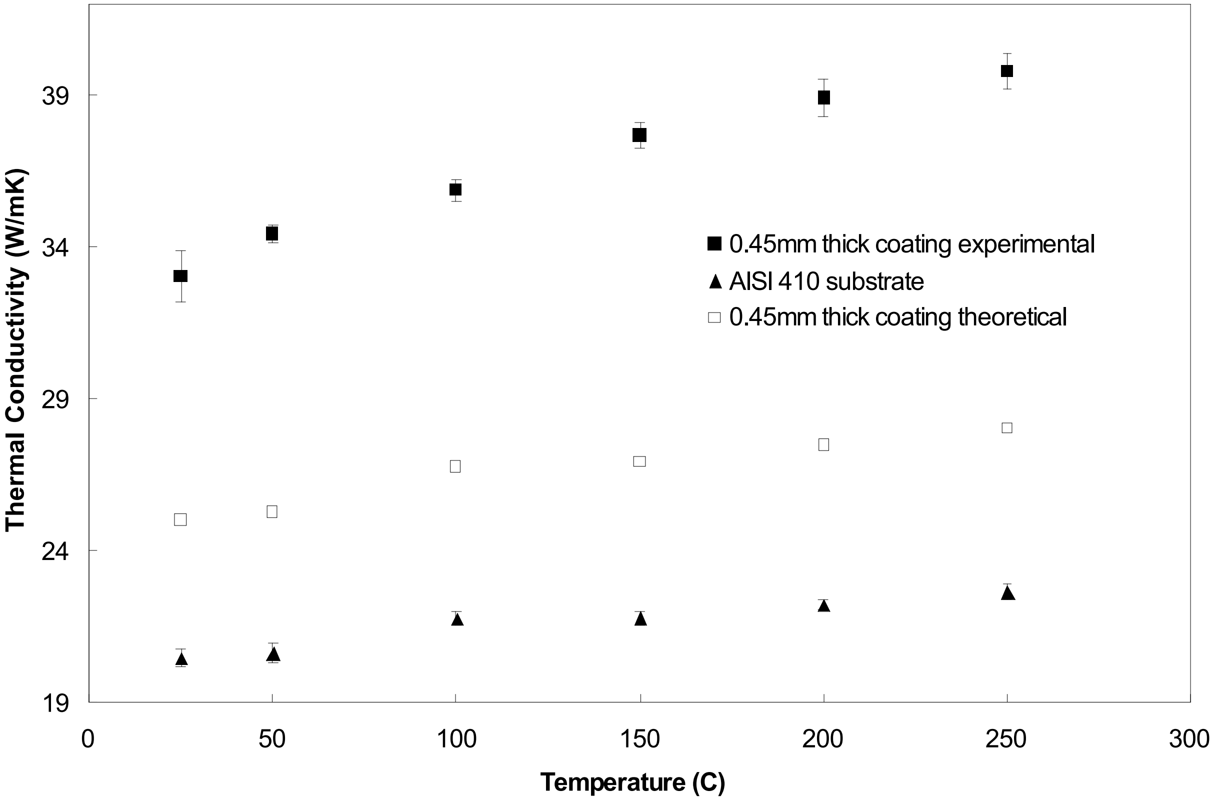

Figure 2: Thermal conductivity of laser deposited high thermal conductivity brass coating both the theoretical and experimental values, and AISI 410 substrate.

The main objective of the high TC brass coating on AISI 410 substrate is to enhance the heat dissipation in the direction perpendicular to the AISI 410 substrate surface. The lower bound TC of multilayer materials (k_{ML}) can be calculated using a series heat-transfer model assuming slab geometry and negligible interfacial resistance [15]: $k_{ML} = \frac{k_1 \cdot k_2}{V_1 k_2 + V_2 \cdot k_1}$, where V is the volume fraction, k is the thermal conductivity, and the 1 and 2 subscripts refer to the individual material layers. As shown in **Figure 2**, the calculated multilayer TC values for the brass coating varied from 25 to 28 W/mK over the temperature range of 25 to 250 °C. The experimentally determined average TC of AISI 410 with brass coatings is higher than the theoretically estimated values, which supports the absence/extremely low interfacial thermal contact resistance in these laser processed composites. In multilayer materials, if the interfaces play a role in reducing heat conduction, then the experimentally determined TC values of AISI 410 steel with brass coating will be lower than those estimated using series heat-transfer model. However, experimentally determined average TC of AISI 410 with the brass coatings is 32 to 42% higher than the theoretically estimated values. This supports the absence of interfacial thermal contact resistance in the present composite structures due to high degree of bonding, diffuse interface region and lack of additional porosity at the substrate–coating interface. The observed high TC could also be due to the supersaturated solid solution of Cu with steel near the substrate–coating interface, which would have eliminated any interfacial thermal resistance between brass and steel. It has been shown that the formation of supersaturated solid solutions during LENS™ processing is very common due to associated rapid cooling rates [16]. The results in this investigation are consistent with recent work on plasma-sprayed multilayer coatings [17], where the largest decrease in the TC of the coatings has been due to splat interfaces within each layer. More efficient coatings can be fabricated via LENS™ by providing fins projecting out form the coating surface. Being a CAD based process LENS™ enables to create feature based high thermal conductivity coatings for enhanced thermal performance in various applications.

Conclusions

Laser surface modification has been successfully applied to show that by depositing high thermal conductivity material the thermal performance of substrate can be improved. The thermal conductivity and performance of AISI 410 stainless steel has been improved up to a range of 60 to 75%, due to defect free diffuse interface and dense coating structure. The LENS™ in combination with the coating approach can be employ to deposit other high thermal conductivity materials on different substrate material eliminating the interfacial problems associated with the sharp interface present in conventional processes.

Acknowledgments

Authors acknowledge the financial support from National Science Foundation under the Grant number CMMI 0728348.

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