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Conditions and mechanisms for the bonding of a molten ceramic droplet to a substrate after high-speed impact

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ABSTRACT

In this study, the mechanism controlling the formation of a chemical bonding between a ceramic melt and a solid body prior to the melt solidification was investigated using the thermal spraying process as an example. The formation of a chemical bonding between TiO_2, LCO, LZO, YSZ splats and ceramic substrates of the same material was investigated focusing on the effect of the deposition temperature. The bonding state was examined by studying both fractured cross-section samples and samples prepared utilizing the focused ion beam (FIB) technique. The microstructure at the interface of typical FIB-prepared cross section samples was examined by high-resolution transmission electron microscopy. The results show that an effective bonding at the splat interface can be formed only when the deposition temperature is higher than a critical bonding temperature. The critical bonding temperature linearly increases with the melting point of the ceramic material. The interface temperature, directly influencing the bonding formation, was calculated utilizing a one-dimensional heat transfer model. The maximum interface temperature corresponding to the critical bonding temperature was found to be close to the glass transition temperature of the splat material. Thus, the concept of an intrinsic bonding temperature and a sufficient condition for the formation of an efficient bonding are proposed. Furthermore, a model for the bonding mechanism at the melt/solid interface is established to explain the bonding formation and the microstructure at the interface.

1. Introduction

Bonding or adhesion is an essential problem of any two materials system which arises when these two materials are brought in close contact. In order to join two solid bodies together, a number of techniques have been developed, e.g., cold pressure welding [1], friction welding [2], friction stir welding [3] and cold spraying [4], which are now widely applied in many industrial fields. Theoretically, when two atoms with electronic band structures suitable for bond formation are brought into intimate contact, a chemical bond can be formed and the atoms are joined together. It has been shown that, when two solid bodies with clean and sufficiently flat surfaces are just brought into close proximity, the formation of direct bonds across the interface can pull the bodies together even in ambient air at room temperature (RT) [5–7]. However, in practice, high pressure [1,8,9], high-velocity impact [4], or friction heating [3,10,11] processes are generally utilized to form an effective solid/solid bonding through the formation of interatomic bonds [8], an inter-diffusion layer [10] or intermetallic compounds [9,11]. It is because that the quality of the direct bonding usually declines with increasing surface roughness [6,7], since the surface roughness affects the inter-contact conditions of the atoms at the interface. When the surface roughness is too high, the solids cannot be bonded together. Thus, plastic deformation processes, induced by either a high pressure or a high-velocity impact, are employed to bring the two bodies with a rough surface into an intimate contact which allows for the formation of a strong joint.

When a high temperature melt is brought into contact with a solid surface, the formation of an intimate contact between the atoms in the melt and the solid surface might be promoted by the good flowability of the liquid. Thus, during the processes, such as liquid phase sintering (LPS), brazing and thermal spraying, a strong bonding at the interface between the solidified melt and the solid surface is usually expected [12–18]. However, in some cases, such as melt spatters usually generated in welding and metallurgical melting processes, the melt should not bond to the target surface

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[19]. Moreover, even in thermal spraying processes, the inter-splat bonding should be limited for coatings intended to act as a thermal barrier because an increase of the inter-lamellae bonding ratio is positively related to a high thermal conductivity [20]. Therefore, methods for controlling the bonding formation between the melt and the solid become essential when a liquid melt is in contact with a solid surface.

During LPS or brazing processes, the bonding formation at the melt/solid interface occurs under equilibrium conditions through atomic diffusion or melt/solid reactions [12–16]. Generally, a diffusion or reaction layer with a certain thickness is required to achieve the desired bonding between the solidified melt and the substrate. For example, in case of brazing, the thickness of an effective reaction layer usually ranges from a few up to ten micrometers [15,16]. Considering the diffusion or reaction rates during the LPS and brazing processes, the melt and the substrate are always kept at a relatively high temperature over a time period from several seconds to several hours in order to form the desired bonding.

During thermal spraying, molten droplets impact on a cold substrate with a high velocity. Following the impact, the flattening droplets cool down at a very high cooling rate (possibly exceeding 10^6 K/s for metals) and solidify within tens of microseconds [18,21,22]. Because the splat remains in the liquid state for only a very short period of time, the formation of a diffusion or reaction layer at the melt/solid interface is generally considered to be impossible. However, it is worth noting that, when a substrate with a lower melting point is locally melted by the liquid droplet after impact, interdiffusion across the splat/substrate interface can easily occur [23]. For instance, when an Fe-based substrate is locally melted due to the impact of a Mo droplet, the Fe and Mo atoms were demonstrated to interdiffuse across the interface, thus forming an intermetallic layer at the interface [24]. However, when a molten ceramic splat is deposited on a substrate with the same compositions as splat material, e.g., in case of a successive stacking of splats to form a coating with a certain thickness, local melting cannot be expected through molten droplet impact. Thus, the interdiffusion or reaction between the molten splat and the substrate during the rather short spraying process is of no practical significance.

A survey into the market shares of different processes for the deposition of advanced ceramic coatings, including thermal spraying, physical vapor deposition (PVD) and chemical vapor deposition (CVD), revealed that about a two-third share is taken by thermal spraying [25], which indicates the important role of thermal spraying in the production of advanced ceramic coatings. Compared with solid-state joining processes, the better migration ability of liquid atoms should facilitate the formation of an intimate contact with the solid atoms, thereby promoting the bonding to these atoms. Since two solids with a sufficiently flat surface can be directly bonded together, it is inferred that the liquid body with its superior wetting ability can be well-bonded to the substrate through the formation of direct bonds across the interface. However, the maximum average bonding ratio at the inter-splat interfaces of thermal-sprayed ceramic coatings was determined to just about 0.32 when the substrate temperature was kept at a low level by cooling [26]. Moreover, many experimental studies [20,27–30] have clearly demonstrated that the occurrence of a lamellar structure, which is associated with a limited interface bonding, dominates the coating's properties. This limited bonding makes the design of coatings only possible to a certain extent, i.e., within the maximum bonding ratio range. If one were able to flexibly deposit thermal spray coatings with bonding ratios between 0.32 and 1 by adjusting the spraying conditions, the potential of the spray materials could be fully utilized, and the application range of thermal spray coatings could be substantially improved. Therefore, the development of an effective approach to control the inter-splat bonding by understanding the bonding mechanism is highly desirable.

Elevating the deposition temperature is an alternative approach to improve the inter-splat bonding ratio of thermal spray coatings [23,30–32]. In our previous study, the concept of a critical bonding temperature (T_c) for an effective bonding was proposed [26] and verified through examining the effect of the deposition temperature on the bonding state at the splat/substrate interface of Al_2O_3 and YSZ [33]. T_c denotes the lowest deposition temperature for which the whole splat was found to be well-bonded to the substrate. Thus, the bonding between the splat and the substrate can be controlled by selecting an appropriate deposition temperature based on the concept of the critical bonding temperature. However, the existence of the critical temperature, the bonding mechanism and the effect of the deposition temperature on the bonding mechanism are still not fully understood.

Up to now, many studies have investigated the mechanism controlling the formation of bonded and unbonded interfaces between adjacent lamellae. McPherson suggested that an unbonded interface can be attributed to the gas released during the droplet spreading process [34]. Sobolev argued that the splat/substrate micro-adhesion significantly depends on the pressure developed upon the high-speed impact [35]. Following the report on the effect of adsorbates on the bonding published by Li et al. [36], Jiang et al. [37] proposed that a poor contact between the droplet and the substrate may be traced back to the entrapment of gas caused by the vaporization of adsorbates when the hot liquid droplet impacts onto a cold substrate surface. Moreau et al. suggested that the bonding is affected by the wetting of the substrate surface by the liquid splat [38]. Due to the rapid solidification of the molten splat, there is not enough time for the splat to fully wet the surface. Thus, the splat and the substrate are only partially bonded together at the interface. However, none of the above-mentioned hypothesis can fully explain the occurrence of the lamellar interface bonding during the spraying of ceramic droplets.

In this study, in order to reveal the bonding mechanism, isolated splats of ceramic materials with a wide range of melting points were deposited on polished substrates of the same material at different deposition temperatures. The bonding states at the splat/substrate interfaces were clarified by investigating the cross-sectional morphologies of fractured samples and cross-sections of selected samples prepared utilizing the focused ion beam (FIB) technique. Then, the T_c values of the ceramic materials were systematically determined to explore the relationship between T_c and the characteristic material properties. Utilizing a simulation model, the key factor controlling the bonding process was revealed by examining the relationship between T_c and the interface temperature just prior to melt solidification. Furthermore, the mechanism behind the formation of a direct bonding between the molten splat and the substrate is discussed by introducing the concept of an intrinsic bonding temperature.

2. Experimental

2.1. Materials

Typical ceramic materials with different melting points, including TiO_2, $La_2Ce_2O_7$ (LCO) and $La_2Zr_2O_7$ (LZO) and 8 mol% yttria-stabilized zirconia (8YSZ) (the deposition of Al_2O_3 was systematically investigated in our previous study [33]), were used to prepare single-layer splats on polished substrates of identical chemical composition to simulate the formation of interlamellar bonding. The particle size of the fuse-crushed TiO_2 powders