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Metal-Ceramic Interfaces Produced by Laser Melt Injection Processing

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Abstract

This paper concentrates on the mechanical performance of various ceramic coatings of Cr_2O_3 on steel (SAF2205), as produced by CO_2 laser processing. It is concluded that a firmly bonded coating of Cr_2O_3 on steel could be produced by high power laser processing. The actual interface strength of a (Fe,Cr)-spinel applied to stainless steel by laser coating depends strongly on the composition of the substrate and coating materials. The energy release rate was extremely high and delamination occurred by fracture through the coating and partially along the interface, indicating that the interface strength is similar to or higher than the fracture strength of (Fe,Cr)-spinel.

1.0 Introduction

In previous publications (1,2), we reported about the characteristic features of coating a duplex steel SAF 2205, stainless steel 304 and Fe-22 wt% Cr by fusing different powders with a laser beam. In fact, a powder mixture of Cr_2O_3 and pure iron was brought into the laser beam in order to form a spinel structure inside the coating. Further, it became evident that

it is possible to form a thick and homogeneous ceramic coating of Cr_2O_3 on steel by a proper selection of coating parameters such as scanning velocity, overlap and number of layers. A higher laser scan velocity may decrease the convection speed and prevent cracks in the coating induced by the convection. More than 50% overlap is needed in order to compensate for the lack of coating material in the centre of the track being swept away

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by convection (1-4). The work was concentrated on the stabilisation of high temperature distorted spinel phases due to the high quench rates involved as well as on the quantitative crystallographic analysis of the resulting morphologies. This paper concentrates on the actual performance of various coatings produced by laser processing. The relevant physical quantities concern the critical elastic energy release rate G_c and the critical stress intensity factor K_{Ic} (5,6).

2.0 Experimental

A CW-CO₂-laser (Spectra Physics 820) is used for the laser coating process with the following laser parameters: 1.0 kW laser power, a spot size of 1.27 mm, a laser scan velocity of 20 mm/s and an overlap of 75% between the tracks. Coating powders - Cr₂O₃, Cr₂O₃ containing Mn and Cr₂O₃ containing extra Cr and Mo were used. The Mn is added to investigate the influence of Mn on the lattice parameters of the (Fe,Cr)- spinel coating and on the interface between the substrate and the coating. Cr and Mo are added to manipulate the crystal structure of the melt pool in the austenitic stainless steels from fcc to bcc in order to change the thermal expansion coefficient of the substrate.

Dual phase SAF2205 steel was used as the substrate and was of composition: C(0.03), Si(1), Mn(2), Cr(22), Ni(5.5), Mo(3), bal. Fe (in wt%). Colour etching and X-ray diffraction methods indicate that phases in the laser melted substrate are the same prior to and after laser treatment when Cr₂O₃ powder or Cr₂O₃ powder containing Mn is used. After coating with

Cr₂O₃ powder containing extra Cr and Mo, the melt pool in the substrate is completely transformed to bcc. X-ray diffraction is used to identify the phases present in the laser coating. The peak positions of the FeCr₂O₄ and Cr₃O₄ phases (7) are used to calculate the lattice parameters of the (Fe,Cr)-spinel phases in the coating.

The specimen used in the flexure experiments consists of a bimaterial beam with overall dimensions 40x3x3 mm. The sides of the specimen are polished to facilitate optical observations during and after testing but the surfaces of the coatings are as received to prevent the formation of surface cracks due to grinding or polishing. The actual experiments are performed on duplex SAF2205 coated with a 200 mm plasma sprayed Cr₂O₃ coating and with a 100 mm laser applied (Fe,Cr)-spinel coating. Specimen with laser tracks parallel as well as perpendicular to the direction of loading were tested.

The four-point flexure test is based on the storage of a well known amount of elastic energy on bending and a release of this elastic energy on fracture. The steady state energy release rate G_{ss} can be deduced from the difference in the strain energy in the cracked and the uncracked beam. Since there is negligible strain energy in the beam above the crack, i.e. in the delaminated coating, the energy release rate is simply the difference in strain energy of the uncracked section and the section of the lower beam beneath the crack, i.e. of the substrate plus coating and the substrate respectively. Based on the description given by ref.(8) for thin coatings, i.e. $h_1 \ll h_2 \sim h$, the strain energy release rate may be approximated by:

$$G_{ss} = \frac{18M^2(1-\nu_2^2)^2 E_1}{E_2^2(1-\nu_1^2)} \frac{h_1}{h^4} \quad (1)$$

As can be seen from eq. (1) the strain energy release rate depends critically on the measured value of $h \sim h_2$. Interfacial crack propagation occurs when the strain energy release rate G_{ss} equals the critical energy release rate G_c of interfacial failure.

3.0 Results

Cracks in the ceramic coating are observed in the heat affected zone just outside the laser track. In the (Fe,Cr)-spinel coatings on duplex steel SAF2205 it turned out that the cracks are oriented vertically in the coating and as a consequence do not cause delamination of the coating. However, complete delamination is observed when the coating thickness is larger than 200 μm . The vertical cracks run through the amorphous interdendrital phase and are deflected by the α -(Fe,Cr) particles. As the cracks run through the region of intergrowth between the (Fe,Cr)-spinel and the α -(Fe,Cr) particles the crack energy was not dissipated in the metallic particles. Consequently, the cracks run all the way through the coating and are blunted when they reach the substrate (Figure 1) through plastic deformation in the metal substrate. Crack extension along the interface is not observed but crack branching in the ceramic coating is frequently observed in a region of 5 μm near the interface.

A plane view on a polished section of the (Fe,Cr)-spinel coating shows a two-dimensional network of cracks (Figure 2). One type of crack is parallel to the laser track in the heat affected zone. The spacing between these cracks is caused by the

transversal displacement of the laser beam. The other type of cracks makes an angle of 50°-70° with the direction of the laser beam (Figure 2). When the transversal displacement of the laser beam is reversed the crack pattern is mirrored indicating a clear relationship with the geometry of the coating process and not with any irregularity of the Gaussian beam shape (TEM₀₀ mode). These cracks are regularly spaced.

The thickness of the coating that can be applied by the use of lasers is limited to about 200 μm , setting a limit to the maximum strain energy release rate that can be measured before severe yielding occurs. When the specimens having laser applied Cr₂O₃ coatings are set up in 4-point flexure, two modes of failure may occur. First, delamination along the interface is possible resulting in a value for the critical energy release rate and the stress intensity factor. The value obtained is a minimum value because of the large tensile stresses present in laser treated materials. In order to obtain an actual value of the critical energy release rate the strain energy present after laser treatment should be superimposed onto the strain energy stored during bending. The stress state after the laser treatment is however inhomogeneous due to the localized character of the laser melting. When the interface is very strong the cracks through the coating will not extend along the interface but will blunt by plastic deformation of the substrate. In the present case failure occurs by crack blunting in the substrate as can be seen from Figure 3. Here the edge of the specimen is polished before 4-point flexure and the size of the deformed volume can

Figure 1: TEM micrograph of (Fe,Cr)-spinel coating (Sp) on steel substrate (M) showing crack blunting in the substrate.

Figure 2: Plane view of polished (Fe,Cr)-spinel surface showing two dimensional network of cracks. Single laser tracks run downward and displacement of subsequent laser tracks is to the left.

be observed by the plane stress type of necking. Only in some cases delamination occurs after work hardening of the substrate (Figure 4). Assuming that the plastic deformation only occurs in a small volume near a crack through the coating the amount of energy stored, and thus a minimum value for the critical energy release rate, can still be obtained from the maximum load. Observations indicate that the critical strain energy release rate and stress intensity factor are larger than:

$$G_{ss}^{\min} = 270 \text{ [J / m}^2\text{]} \\ |K_c^{\min}| = 8.5 \text{ [MPa}\sqrt{\text{m}}\text{]}.$$

4.0 Discussion

Results in the literature on the laser application of ceramic coatings are scarcely available indicating that laser coating with ceramic materials is rather difficult. The problem is caused by the inhomogeneous heating and subsequent cooling during laser treatment which result in considerable cooling strains. In metals these strains are accommodated by plasticity, likewise in ceramics at high temperatures ($T > 2/3 T_m$). At low temperatures strain results in fracture of the ceramic.

The thermal loading by the laser beam causes stresses being built up. Without melting or yielding all thermal strains will be accommodated elastically, i.e. disappear when returning to the starting temperature. Melting or yielding is necessary in order to attain a residual stress state. Because the cooling during laser treatment starts from the bottom to the surface of the specimen the residual stress state near the

surface of the specimen, i.e. the ceramic coating, will be of tensile character. The residual stress adds up with the mechanically applied stresses causing failure at relatively low levels of applied stress. To study the formation of cracks during and after laser treatment we have scrutinized the temperature field near the laser melt pool to obtain the stress during and after laser treatment based on the work by Cline and Anthony.⁹ The temperature field is calculated as a Gaussian beam sweeping over the surface of a material. By taking derivatives with respect to x and y the thermal gradients $\frac{\partial T}{\partial x}$, $\frac{\partial T}{\partial y}$, and the absolute value of the total gradient is obtained. Although we did not solve the three dimensional temperature field numerically for a bimaterial we may extrapolate the data from the two singular cases of steel and (Fe,Cr)-spinel using the diffusion equation together with the fact that the temperature field has to be continuous across the interface. This implies that the thermal gradients parallel to the interface, i.e. in the x - and y -direction, are equal on both sides of the interface ($z = 0$ on the interface), i.e.:

$$\left(\frac{\partial T}{\partial z}\right)_{x,y,0}^{\text{cer}} \approx 10 \left(\frac{\partial T}{\partial z}\right)_{x,y,0}^{\text{met}} \quad (2)$$

using the fact that the heat flux is continuous over the interface. Observations of the equi-temperature lines on the interface perpendicular to the direction of the laser beam, i.e. in the y -direction, indicate that for the (Fe,Cr)-spinel coating on steel:

$$\left(\frac{\partial T}{\partial z}\right)_{x,y,0}^{\text{met}} \approx 0.26 \left(\frac{\partial T}{\partial y}\right)_{x,y,0}^{\text{met}} \quad (3)$$

Figure 3: Crack blunting in the steel substrate during four-point flexure.

Figure 4: Delamination along the (Fe,Cr)-spinel interface after work hardening of the substrate.

$$\left(\frac{\partial T}{\partial z}\right)_{x,y,0}^{\text{cer}} \approx 2.6 \left(\frac{\partial T}{\partial y}\right)_{x,y,0}^{\text{cer}} = 2.6 \left(\frac{\partial T}{\partial y}\right)_{x,y,0}^{\text{met}} \quad (4)$$

Eq. (3) shows that, in the metal, the thermal gradient in the z-direction is only one

quarter of the gradient in the y-direction validating the use of the temperature field calculated for a single phase steel having a zero thermal gradient in the z-direction on the surface. Eq. (4) indicates that the large

temperature gradient in the y-direction is accompanied by more than twice as large a thermal gradient in the z-direction in the ceramic coating. From the thermal gradient in the y-direction of 1.8×10^6 °C/m in steel a thermal gradient in the z-direction of $4.7 \cdot 10^6$ °C/m is estimated in the (Fe,Cr)-spinel coating. As these large thermal gradients occur at temperatures near the melting point they are accompanied by low stress levels. However, during cooling the temperature difference results in the buildup of residual stresses. Because, in the region of interest, the coating cools from the interface towards the top of the coating the stress is of tensile character in the top of the coating, i.e. a bending moment is set up in the coating that may cause delamination of the ceramic coating.

The residual stress state can be estimated from the solution of a bending plate on an elastic foundation of which the solution is known. When bending is restricted by the substrate, i.e. edges are clamped and delamination does not occur, the stress in the top of the coating is given by:

$$\sigma_{yy}(z) = \frac{\alpha_1 E_1}{(1 - \nu_1)} \cdot \frac{\partial T}{\partial z} \cdot z \quad (5)$$

where index 1 refers to the properties of the coating. Clearly there exists a stress distribution over the thickness of the coating which sets up a bending moment in the coating which can be approximated by:

$$M_1 = \frac{\alpha_1 E_1}{12(1 - \nu_1)} \cdot \frac{\partial T}{\partial z} \cdot h_1^3 \quad (6)$$

Using equations (4) and (5), the bending

moment is found to be 13.7 Nm/m for a coating thickness of 200 µm.

When the coating does not delaminate edge moments have to be applied to prevent bending. These edge moments are effectively produced by stress components in the z-direction over the interface. The stress s_{zz} at the edge of a bending plate is given by Hetenyi (10) and the maximum tensile stress is given by:

$$\sigma_{zz}^{\max} = 2 \cdot \frac{\alpha_1 E_1}{12(1 - \nu_1)} \cdot \frac{\partial T}{\partial z} \cdot \sqrt{\frac{3k_2}{E_1}} \cdot h_1^{\frac{1}{2}} \quad (7)$$

where k_2 is the modulus of deflection of the substrate ($E_1/k_2 = 6.6 \times 10^{-5}$). The process of delamination versus perpendicular fracture is determined by the relative strength of the ceramic and the interface and by the thickness of the coating. As the stress σ_{yy} causing fracture through the coating, scales with h_1 and σ_{zz} , causing delamination, scales with $h_1^{1.5}$ the coating can always be made thick enough so that delamination will occur. Ultimately, when the interface is very strong, delamination may occur by fracture through the ceramic parallel to the interface, instead of along the metal-ceramic interface, and both fracture processes are governed by the strength of the ceramic σ_c^f , being of the order of 1 GPa.

If this is the case the fracture process, i.e. delamination vs. perpendicular fracture, is solely determined by the coating thickness and the critical coating thickness is determined by the elastic properties of the coating and the substrate. Whether or not fracture will occur is of course related to the thermal gradient in the coating. Assuming that the thermal

gradient is large enough for fracture to occur delamination is the governing process if:

$$h_1 > \frac{3E_1}{k_2} \quad (8)$$

From the calculated bending moment in the ceramic coating with a thickness of 200 μm , the critical energy release rate G_c of delamination can be calculated:

$$G_c = \frac{M_{lc}^2(1-\nu_1)^2}{2E_1} \cdot \frac{12}{h_1^3} \quad (9)$$

which is equal to 320 J/m². If delamination does not occur the stress σ_{yy} is reduced due to crack opening. The wavelength of the stress reduction is related to the extend of the stress field across the interface and scales with $h_1^{3/4}$. As the maximum value of the stress σ_{yy} scales with h_1 , the distance over which the fracture stress is reached scales with $h_1^{1/4}$ indicating that the crack spacing is only weakly dependent on the thickness of the coating. For a coating of 100 μm the extension of the stress field over the interface is about 200 μm indicating a crack spacing of the same size. This is in agreement with experimentally observed crack spacings. However, these crack spacings also scale with the transversal displacement of the laser beam. This is partially because the thermal gradient peak in the metal causes an extended gradient peak in the ceramic. In this way a region of large stress parallel to the laser track is developed where fracture takes place preferentially. This effect is augmented by the fact that the ceramic coating is somewhat thicker at the edge of the metallic melt pool, i.e. at the

same place where the gradients are largest. It can thus be concluded that the crack spacing is more strongly related to the stress peaks in the coating, i.e. to the transversal displacement of the laser beam, rather than to areas where the stress is reduced due to a prior crack.

A similar explanation can be given for the stress development in the x-direction, i.e. in the direction of movement of the laser beam. Although the thermal gradients in the z-direction in the coating can be very large due to the thermal gradients in the x-direction, delamination is never observed in the x-direction because the coating thickness is always much smaller near the middle of the last laser track that is applied. However, the stress in the x-direction scales with the thermal gradient in the z-direction near the middle of the laser track so that fracture through the coating, more or less perpendicular to the direction of movement of the laser beam is always observed. For all other cases where the heat flow is not purely in the x- or y-direction the stress in the coating is determined by the total magnitude of the thermal gradient. Because of the low thermal conduction coefficient the flow of heat from the coating is however mainly in the y-direction making delamination in the y-direction the most significant process.

Another point to consider is the stress development during laser treatment: (i.e. influence of thermal expansion difference $\Delta\alpha$). A difference in thermal expansion coefficient between the coating and the substrate material can result in both normal and shear stresses. In the present case of coating ceramic material on steel using a high power laser, shear stresses will be set

up due to the asymmetry of the process. As the laser track is restricted by the surrounding material, a tensile stress will be present in both materials near the interface but, as no macroscopic shrinkage can occur the stress is not transmitted over the interface. However, because during laser treatment the material closer to the centre of the laser track is at a higher temperature, yielding will cause a net flow of material in the outward direction. The magnitude of the net displacement of material will scale with the thermal expansion coefficient α of the corresponding material.

Four-point flexure appeared to be an appropriate method to test the interfacial properties of bimetals. However, there are also a few limitations, one of which is the maximum energy release rate that can be measured when investigating thin coatings on ductile substrates. For a 100 μm (Fe,Cr)-spinel coating on duplex the maximum energy release rate is of the order of 160 J/m² for a specimen thickness of 3 mm. The value of 270 J/m² as obtained experimentally involves some substrate yielding making work hardening of the substrate and interface sliding possible. For a reliable measurement of the critical energy release rate this should be prevented but it can be used to obtain an indication of the quality of the interface. The elastic energy stored in a 100 μm thick coating due to the laser treatment is only 37 J/m² so in order to get delamination during 4-point flexure about 280 J/m² should be applied to the coating by bending. However, yielding of the substrate limits the elastic energy stored in the coating to 160 J/m² making delamination of a 100

μm (Fe,Cr)-spinel coating by 4-point flexure impossible.

5.0 Conclusion

It is concluded that a firmly bonded coating of Cr₂O₃ on steel could be produced by high power laser processing. The actual interface strength of a (Fe,Cr)-spinel applied to stainless steel by laser coating depends strongly on the composition of the substrate and coating materials. The strength of bimaterial interfaces was tested by 4-point flexure. The energy release rate was extremely high and delamination occurred by fracture through the coating and partially along the interface, indicating that the interface strength is similar to or higher than the fracture strength of (Fe,Cr)-spinel.

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