

Predicting the CTE & hardness for directed energy deposition of functionally graded WC-SS316L Coatings

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Abstract. One of the key components in the design of wear resistant coatings for directed energy deposition is the analysis of the thermo-mechanical properties within the substrate-coating interface. The coefficient of thermal expansion (CTE) and the mismatch are crucial due to their significant influence on the structural integrity and operational performance of the coating. In this study, a four-layered functionally graded tungsten carbide (WC)-316L stainless steel composite coating was designed using a rule-of-mixtures approach for manufacture by directed energy deposition. Two theoretical models were employed to predict the CTE and micro-hardness as a function of increasing volume fraction of WC particles. According to the graded concentrations predicted from the linear models, the four-layer coating exhibits a good hardness with minimal CTE mismatch.

1 Introduction

Directed energy deposition (DED) is an additive manufacturing (AM) technique that combines laser and powder processing to manufacture fully dense components from Computer Aided Design (CAD) models [1]. The DED process has demonstrated its ability in the fabrication of complex geometries, repair of high value parts, and deposition of protective surface coatings [1, 2]. The technology offers low dilution and formation of a metallurgical bond between coating and substrate due to rapid heating and cooling, making it suitable for coating deposition [2]. Moreover, DED machines may be equipped with multiple powder delivery systems, making it possible to deposit multi-material components such as metal matrix composites, which are deemed difficult to process.

Metal matrix composites are widely used to increase the wear and corrosion resistance in engineering materials because they possess excellent mechanical, chemical, and thermal properties [3]. Tungsten carbide (WC) is used as a reinforcement in metal matrix composite coatings during directed energy deposition to improve the mechanical properties of engineering components [4]. The WC phase has many advantages such as high hardness, low coefficient of thermal expansion, and excellent wear resistance. Studies have shown that the competing thermo-physical properties between the hard WC and soft metal matrix can play

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a critical role in the formation of distortions and cracks [5]. This is especially true in DED due to the localized heating and cooling which leads to non-uniform thermal expansion and contraction. This results in structural defects such as distortions, warping, cracks, and delamination [6]. Such cracks affect the structural performance of the coating and should be minimised.

Wang [7] studied crack initiation and propagation behaviour in WC-Fe based metal matrix composites formed by laser metal deposition. In the study it was reported that thermal stresses originated due to the high temperature gradients and a mismatch in the coefficient of thermal expansion (CTE) between the WC and the Fe-based matrix, and this led to the initiation of cracks inside the WC particles. These cracks were found to propagate along eutectic phases showing high brittleness. Shen [6] studied the effect of increasing wt.% WC in a NiCrSiBC composite coating deposited onto carbon steel substrates using laser cladding. It was observed that as the wt.% WC increased, the degree of cracking increased. Both authors found an increase in the wt.% WC led to the formation of eutectic carbides which facilitated grain refinement and increased the coating hardness, however the stress concentration, and cracking susceptibility also increased

The issues encountered can be attributed to dissimilarities in the material and metallurgical properties of the constituent materials. These differences can be accounted for as 1) solidification cracking, which occurs as a result of the large solidification range between WC and the binder material [8]; 2) chemical incompatibilities that prevent perfect mixing or the formation of brittle intermetallic phases [9]; and 3) high internal stresses generated from the mismatch in the coefficient of thermal expansion and these can lead to macro/microscopic cracking [6, 7].

To overcome these undesirable effects, functionally graded composite coatings have been developed and have shown to effectively alleviate stress concentration in the metal-ceramic interface, release residual stress, and improve adhesion of the coating to the substrate through the relaxation in the thermal mismatch [10, 11, 12]. Functionally graded composite coatings are formed through the mixing of two or more dissimilar materials by gradually varying the composition. They have proven suitable for applications that require varying material properties at different locations within a component. Adequate functionally graded coatings (FGC's) can be attained by optimising the volume fraction distributions of the constituent phases to address thermal stresses and temperature depended material properties like the coefficient of thermal expansion (CTE). Thus, design parameters for the additive manufacturing of functionally graded structures rests in defining the optimum material constituent distributions. The selection of a suitable material distribution is an essential part in tailoring FGC's that will optimally meet the desired performance, loading and boundary conditions [13]. Recently, there have been more studies carried out that are dedicated to material-property estimation techniques using volume-averaging concepts [12, 14, 15]. Different models for estimating these properties in FGC's have been investigated by scholars, and it has become generally accepted that the rule of mixtures (ROM) approach can be used to reasonably predict the behaviour of functionally graded structures [10, 11, 16].

At present, there are no guidelines on material compatibility, mixing range, or a framework for optimal property distribution. Therefore, the purpose of this study was to design a functionally graded DED WC-316L stainless steel composite coating by using ROM to predict the thermal expansion mismatch that will result in reduced cracking susceptibility.

2 Theory and Formulation

During directed energy deposition there exist rapid solidification and cooling rates which are advantageous for the development of fine microstructures that lead to superior mechanical properties, such as micro-hardness [2]. However, these also lead to an accumulation of

internal thermal stresses which are detrimental during the joining of dissimilar materials, such that cracking and delamination result due to significant differences in the coefficient of thermal expansion (CTE) of the materials.

Therefore, to successfully deposit functionally graded composite coatings, novel material design is required to match the CTE between different materials to reduce thermal stress that could create distortions or cracks. It is hypothesised that compositional changes can be used to tailor the CTE and enable a design of FGC's that can mitigate thermal stresses through the minimisation of the CTE mismatch [1].

The CTE has been found to be governed by the rule of mixtures (ROM) and can be generalised by Equations 1 (Voigt) and 2 (Reuss), adapted from [16]. These equations are useful for composites reinforced with randomly dispersed spherical particles in a polymer and metal matrix [16, 17]. For the equations, α_c is the computed coefficient of thermal expansion of the composite layer, α_i and V_i are the theoretical coefficient of thermal expansion and actual volume fraction of each constituent in the matrix, and E_i is the elastic modulus of each constituent.

$$\alpha_c = \sum_{i=1}^n V_i \alpha_i \quad (1)$$

$$\alpha_c = \frac{\sum_{i=1}^n V_i E_i \alpha_i}{\sum_{i=1}^n V_i E_i} \quad (2)$$

Equation 1 describes particles that are suspended in a matrix with the assumption that a combination of the independent phases does not alter the behaviour of each phase. Using Equation 1, Reuss assumed a uniform strain and developed an equation based on the CTE, volume and elastic modulus of each component, as shown by Equation 2.

These equations can also be used to model other effective properties of multi-material composites such as micro-hardness which is a vital mechanical property when designing wear-resistant coatings. Thus, Equations 1 and 2 can be rearranged such that the micro-hardness of the composite coating is assumed to be proportional to the volumetric fraction of the reinforcing particles and re-written as Equations 3 and 4 where H_c is the computed hardness of the composite layer, and H_i is the theoretical hardness of each constituent.

$$H_c = \sum_{i=1}^n V_i H_i \quad (3)$$

$$H_c = \frac{\sum_{i=1}^n V_i E_i H_i}{\sum_{i=1}^n V_i E_i} \quad (4)$$

The properties of the two constituent phases, tungsten carbide (WC) and 316L stainless steel (SS316L) used in the current study are shown in Table 1.

Table 1. Thermo-mechanical properties of the composite and matrix [18, 19].

	CTE ($10^{-6}/^\circ\text{C}$)	Hardness (HV)	Elastic Modulus (GPa)
SS316L	17	190	193
WC	5.5	2400	669-696

The coefficient of thermal expansion and micro-hardness of the resulting WC-316L stainless steel composite coating layers were determined when the volume fraction of WC was varied in the z-direction. It was assumed that there would be perfect mixing between the reinforcing particles (WC) and matrix (SS316L) and that all particles are perfectly bonded together and deform together. Using the data presented in Table 1 and varying the WC phase volume fraction from 0 (substrate) to 1 (pure WC), the effective properties at each volume fraction was estimated from the standard homogenisation models in Equations 1 to 4. The

coefficient of thermal expansion and micro-hardness were plotted as a function of WC volumetric fraction and then the number of FGC layers and composition were deduced from the graphs based on the criterion of minimum thermal mismatch.

3 Results and Discussion

The effective coefficient of thermal expansion (CTE), α and microhardness of the composite were determined by using theoretical CTE and hardness values at 25-500 °C for both SS316L and WC. A series of numerical approximations was conducted by using isotropic thermal deformation properties of both the WC and SS316. The obtained approximations are shown in Table 2.

Table 2: Results obtained from rule of mixture approximations.

Volume fraction	CTE X10⁻⁶/°C		Hardness [HV]	
	SS316L	WC	Reuss	Voigt
1	0	16,0	16	190,0
0,9	0,1	13,0	14,95	209,3
0,8	0,2	11,0	13,9	232,9
0,7	0,3	9,6	12,85	262,5
0,6	0,4	8,6	11,8	300,8
0,5	0,5	7,8	10,75	352,1
0,4	0,6	7,1	9,7	424,6
0,3	0,7	6,6	8,65	534,6
0,2	0,8	6,2	7,6	721,5
0,1	0,9	5,8	6,55	1109,5
0	1	5,5	5,5	2400

3.1 Coefficient of Thermal Expansion Model

The results from the application of the Voigt and Reuss approximations are shown graphically in Figure 1. By using the volume fraction of WC particles and associated CTE's of the constituents of the composite, both the Voight and Reuss approximations provided an excellent prediction of the WC-SS316L composite coating.

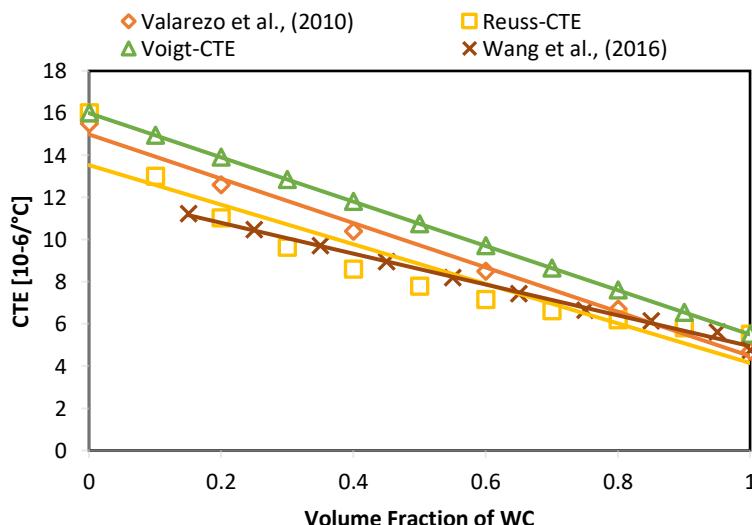


Fig. 1. Theoretical approximation of the coefficient of thermal expansion.

The approximated CTE's were compared to that of a WC-Co graded coating deposited on a SS316L substrate [12, 1] and where the authors also used the rule of mixtures to predict the CTE and compared the results to experimental values. Valarezo et al., concluded that the CTE decreased as the volume fraction of the composite material was increased and a linear trend like the Voigt approximation was found, except for limited deviations at high WC-Co volume fractions of 60 and 80 vol.%, which was attributed to the formation of non-equilibrium phases that are not generally accounted for in the model. This agreed with results obtained by Wang et al., where a linear trend at low binder composition was observed and as the binder is increased, the prediction of CTE become more complex. This can be attributed to the formation of crystal structures and brittle intermetallics [9].

The Reuss approximation showed a deviation from linearity because of the significant differences in the elastic modulus between WC and SS316L. In a metal matrix, the Voigt approximation provides a close approximation, while the Reuss approximation provides a less accurate approximation when the elastic moduli are substantially different. This is especially true for the WC-SS316L composite as the constituent phase elastic moduli are 193 and 669GPa, respectively. Given the overall consistency between the approximations and experimental results from the literature, the Voigt model appears to be a valid prediction method for estimating multiphase alloy CTEs as established by several authors [7, 17, 14].

3.2 Micro-hardness Model

The micro-hardness was computed according to the rule of mixtures by using the Voigt and Reuss approximations by varying the volumetric fraction of the WC particles according to Equations 3 and 4. To assess the validity of the approximation models, the effective micro-hardness values were compared with experimental results found in literature [12, 20]. Figure 2 (a) shows a simple volumetric rule of mixture analysis of the micro-hardness of the composite, assuming a micro-hardness of 190 GPa and 2400 GPa for SS316L and WC respectively. The results are compared to experimentally determined micro-hardness of single layers and FGC of SS316L and WC-Co deposited using HVOF, Figure 2 (b).

A linear correlation between the micro-hardness and volume fraction of the reinforcement particles was observed. This compliments the work of Valarezo et al., [12] and Kim [20]. Valarezo et al., [12] found that experimentally measured values agreed with the analytical predictions as well as single layer coatings, as shown in Figure 2(b). On the other hand, Kim [20] calculated the effective hardness using ROM and compared their results to those obtained by Finite Element Analysis (FEA). It was found that the FEA results fit better with the Voigt approximation at high-volume fractions of the hard phase.

From this result, it can be concluded that the effective hardness can be approximated using the rule of mixtures and that the compositional grading does not affect the mechanical characteristics of each layer in the coatings despite the different CTE values.

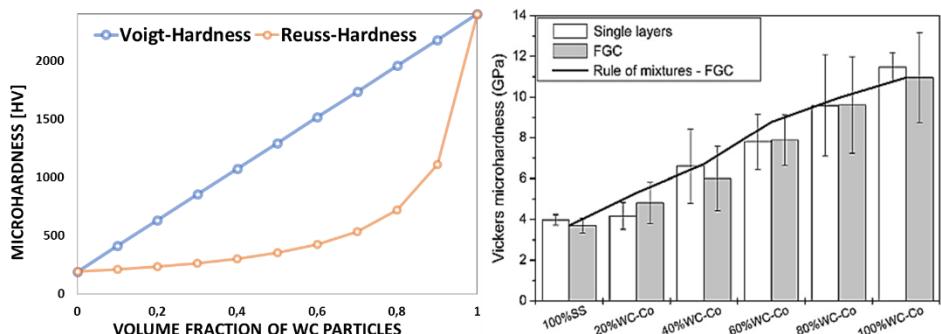


Fig. 2. Theoretical approximation of hardness (a) compared to experimental results (b) adapted from [12].

3.3 Design of the functionally graded coating

The design of functionally graded coatings for DED follows additive manufacturing workflows incorporating materials and their behaviour [13]. The prediction of overall chemical compositions and characteristics in a final component is important for material design and processability. In this study, this was achieved by correlating the effective thermo-physical properties of the composite based on the properties of the constituents. This was achieved by using the Voigt approximation based on the rule of mixtures approach to approximate the CTE of the coating layers at different compositions. It was shown in [20] that the thermal expansion coefficient is the main factor affecting distortion and residual stresses of DED parts. Hence, it is important to determine a compositional window at which the CTE difference between successive layers is reduced. Previous studies have found that material combinations with little or no CTE mismatch demonstrate the least interfacial cracks [6, 7, 21].

The thermal stresses generated as the coating solidifies during DED can be approximated by Equation 5, where σ is the stress generated, E is the elastic modulus, $\Delta\alpha$ is the coefficient of thermal expansion difference between the layers and ΔT is the temperature difference between the substrate and coating during deposition [9].

$$\sigma_{max} \propto E \times \Delta\alpha \times \Delta T \quad (5)$$

The coating was designed such that the total thickness will not exceed 5mm and the number of layers would range from 2 to 5 layers. Thus, CTE approximations were used for the prediction of the number of layers with an average reduced CTE mismatch. The Voigt approximation was applied and used to predict the number of layers and composition of the graded coating that would exhibits reduced thermal mismatch.

Table 3 shows the composition of the substrate, each layer, and their corresponding CTE, hardness and CTE mismatch. The composition of the FGC was chosen such that the CTE mismatch is $\sim 2.4 \times 10^{-6}/^{\circ}\text{C}$ per layer. This was achieved through a gradual increase in WC vol.% to achieve a final coating layer containing 85 vol.% WC which is deemed sufficient to provide good wear properties. The predicted number of coating layers are expected to exhibit good adhesion because the substrate material has the same chemical composition as the matrix, thus good wettability is expected in the coating/substrate interface [22]. The minimised thermal mismatch is expected to benefit the FGC by preventing distortion and cracking.

Table 3. Parameters used in the design of the FGC: layer composition, CTE and hardness by rule of mixtures interpolation.

Layer composition (vol.%)	CTE ($10^{-6}/^{\circ}\text{C}$)	Hardness (HV)	ΔCTE ($10^{-6}/^{\circ}\text{C}$)
100% SS316L (substrate)	17.00	190	-
79%SS316L-21%WC	14.58	654	2.4
59%SS316L-41%WC	12.29	1096	2.3
38%SS316L-62%WC	9.87	1560	2.4
15%SS316L-85%WC	7.23	2068	2.6

4 Conclusion

This paper presented a conceptual design of a functionally graded WC-SS316L coating for deposition using DED. This was achieved by applying the rule of mixtures approach to minimise coefficient of thermal expansion mismatch which is one of the main factors affecting residual stress formation during laser deposition. The Voigt rule-of-mixtures approach predicted thermo-mechanical properties which were in agreement with experimental results found in literature. Whereas, the Reuss approximation was seen to deviate from experimental trends found in literature. By using the effective CTE predicted, a functionally graded composite coating with a composition ranging from 21 vol.% to 85 vol.% WC in a SS316L metal matrix was predicted. The graded coating consists of 4 layers with a CTE mismatch of $\sim 2.4 \times 10^{-6}/^{\circ}\text{C}$ and an excellent approximated hardness range capable of providing good wear properties.

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