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Correlation of splat state with deposition characteristics of cold sprayed niobium coatings

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ABSTRACT

The cold spray technique has a great potential to deposit refractory metals for a variety of potential applications. Cold spraying of different metals have been addressed comprehensively to understand the deposition characteristics of the coatings. Since there is no available data on the deposition characteristics of cold sprayed Niobium, impact behavior of splats at different deposition conditions were simulated and numerically analyzed using Finite Element Modeling (FEM) and correlated with the experimental observations that highlight the role of the velocity and temperature of the particle upon impact on the bonding features. The increase in temperature of the splat drastically reduces the flow stress at the interface leading to best inter-splat bonding state. The synergistic effect of the temperature and the velocity leads to the formation of very dense, defect free niobium coating associated with deformation localization including interface melting. Formation of nanocrystalline grains at the inter-splat boundary was confirmed through TEM and compared with the FEM findings. Finally, understanding the deformation and deposition behavior of refractory metal such as niobium will be helpful to engineer the coatings for potential applications.

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1. Introduction

Niobium is considered to be one of the potential refractory metals in a variety of high-temperature applications as it possesses high melting point, good fabricability, low ductile to brittle transformation temperature etc. [1–3]. Niobium has relatively lower corrosion resistance over Tantalum; however it is an economically viable alternative to tantalum in many applications. One of the potential applications of niobium and its alloys is to fabricate corrosion resistant process equipments including reaction vessels, columns, heat exchangers, and thermo wells [4]. It is also used in the fabrication of sputtering target for semiconductor industry [1,5]. In the traditional powder metallurgy, niobium powder is compacted and subsequent sintering is performed at 2000° C under very high vacuum. Melt metallurgical technique is another route in which exclusive electron beam melting with water cooled crucibles are used. Conventionally, for manufacturing of niobium parts, hot forging (650-950° C) is used which leads to severe oxidation and brittleness of the niobium parts [1].

In contrast, niobium parts can be made in inexpensive way using cold spray process with relatively less oxygen content in the light of the fact that the ductile to brittle transformation temperature is in the range of 200° C [2]. In cold spray, deposition occurs when micron sized metallic particles accelerated to supersonic velocities impact onto a substrate [6–9]. Severe plastic deformation and related mechanisms [10,11] at very high strain rates (~10^7/s) [12,13] play a crucial role in depositing metal powders. In cold spray process, dense coatings can be realized without any oxidation or phase transformation as the coatings are produced without any significant heating of feedstock materials. A wide range of metals [14–19], alloys [20–22], composites [23–25] and nano-structured materials [26–28] have been deposited using this technique.

Since plastic deformation related phenomena dominate cold spray deposition, thermo-mechanical response of niobium to supersonic impact is important. Generally, pure niobium is extremely ductile because of its low strain rate sensitivity. Hence forming can be done without intermediate annealing up to 95% cross-sectional area reduction [29]. Compared to other refractory metals and their alloys, niobium alloys are considered to be most ductile that can be formed into any complicated shapes [2]. In general, the plastic deformation in BCC crystalline solids strongly depends on strain rate and temperature [30–33]. The temperature and strain rate also

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determines the flow stress response in high strain rate deformation. It is reported that the work hardening of the niobium is independent of temperature and weakly dependent on strain rate. However, there is no available scientific study on the deposition characteristics of the niobium using cold spray techniques. A systematic study comprising numerical simulation and deposition of niobium can pave a way to comprehensively understand the deposition characteristics and microstructure of cold sprayed niobium.

For successful deposition of the feedstock materials in cold spraying, critical velocity has been accepted as an important parameter which is a minimum velocity required by the particle to get deposited successfully [10,11,34]. It was clearly demonstrated that high strain rate thermo-mechanical characteristics of the powder-substrate material combination are important in determining the critical velocity of the feedstock [10]. In general, higher impact velocity results in better inter-splat bonding thus leading to lower porosity in cold sprayed deposits. At particle – substrate interface/inter-splat boundaries, adiabatic shear instability induced response of the materials at/above critical velocity has been well-accepted phenomena for successful bonding of the impacting powder [10,11]. This phenomenon is characterized by abnormal temperature/strain rise and flow stress collapse at the impacting interface which is a result of severe plastic deformation at/above critical velocity.

Apart from particle impact velocity, particle temperature upon impact also plays a significant role in bonding process. The concept of thermal boost up zone at the impacting interface was derived and correlated with the adiabatic shear instability and resultant bonding mechanism in cold spray deposition [10]. L. Venkatesh et al. [35] demonstrated that the effect of velocity and temperature of the powder particle influences the bonding process synergistically. Yokoyama et al. [36] reported that the critical velocity of copper decreased significantly with increasing particle impact temperature. Hardness, electrical conductivity, and compressive residual stress were well correlated with powder particle energy (thermal + kinetic) rather than the powder particle velocity (kinetic energy). As a result, the microstructure and the functional properties [35–37] of the cold sprayed copper and nickel [38] coatings correlate well as a function of total energy of the impacting powder particle. It is also documented that the deposition efficiency increased when the particle temperature is increased while keeping the in-flight velocity as a constant [15,38].

Hence, understanding the energy balance during the interaction between the substrate/already deposited layer and impacting powder particle would give an insight of the bonding process in the cold spray deposition. The initial kinetic energy of the powder particle is mainly dissipated into plastic deformation energy and recoverable elastic strain energy. The effect of kinetic and thermal energies on the flow stress, plastic deformation energy and elastic strain energy need to be studied comprehensively for a better understanding of the bonding process. Another important factor deciding bonding is the nature of cold spray impact which is composed of elastic-plastic (compressive) loading and elastic unloading (tensile). The energy required to unload the deposited powder during elastic recovery is called as rebound energy. The adhesion energy at the inter-splat boundary is considered to be the bond generating component at the interface. Wu et al. studied the effect of impact velocity on the bonding and the rebound phenomena in aluminum [12] and Al-Si [39] cold sprayed coatings. Also, the interface parameters such as contact temperature, contact time and contact area would help in understanding the energy conservation and the related bonding process [12,16,40,41]. Since the above factors affect the bonding state of impacting splat, the product of them can be considered as ‘bonding factor’.

The aim of this paper is to provide a comprehensive overview of the deposition mechanism of niobium and microstructure evolution using finite element simulation and experimental data at different process conditions.

2. Finite element modeling

As the deposition mechanism in the cold spray is very short time (in the order of nano sec.) phenomena, investigation of the particle behavior upon impact and deposition characteristics is very useful using a non-linear transient finite element modeling. Micron sized Niobium powder deposition behavior was simulated using a commercial explicit code, Abaqus Explicit [42]. In order to explore the non-linear transient dynamic analysis of the particle impact, the explicit time integration algorithm was chosen. An axisymmetric model was used and fully coupled thermal stress analysis was performed to get the thermo-mechanical response of the impact. A surface-surface penalty contact algorithm with balanced contact pair formulation and a self-contact algorithm were specified for interface region. Four node bilinear axisymmetric quadrilateral mesh elements with reduced integration were used. In order to simulate the plastic deformation of the powder at different impact conditions, the use of strain rate-dependent and temperature dependent constitutive description is required in finite element codes. The flow stress (σ) is expressed as a function of strain (ε), strain rate ($\dot{\varepsilon}$) and temperature (T). Many models exist to describe material behavior in the plastic regime and among which Johnson-Cook model (J-C) which includes strain hardening, strain rate hardening and thermal softening effects upon high velocity impact of the particles was employed [43]. The flow stress is expressed as a function of elastic-plastic response, viscosity and thermal softening character at different temperature and strain rates. In general, the model parameters can be obtained from compression quasi-static and dynamic experiments with different temperatures and strain rates. Since work on high strain rate deformation behavior using Johnson-Cook plasticity model for niobium deformation is missing in the literature, the parameters have been taken from other specific literature which are dedicated separately for strain hardening, strain rate hardening and thermal softening. In the Johnson-Cook plasticity model, constant ‘A’ represents yield stress in a quasi-static tension or compression test. For niobium, the yield stress varies from 105 MPa to 247 MPa [1] based on the manufacturing conditions and post treatments. For this work, 150 MPa was used. The strain hardening term in Johnson-Cook equation was compared with the power law type Holloman equation. Zamiri et al. [33] obtained the values of ‘B’ (360 MPa–374 MPa) and ‘n’ (0.2894–0.3059) for different angles (0° - 90°) and the value at 45° was taken for this simulation since the shear direction upon impact of the cold sprayed powder is obtained at 45°. The values of ‘B’ and ‘n’ were taken as 368 MPa and 0.29 respectively. Alcorta et al. [44]

Table 1
Simulation parameters for materials.

Parameter/Material	Niobium
Density (Kg/m^3)	8600
Young's modulus (GPa)	105
Poison's ratio	0.38
Heat capacity (J/Kg K)	272
Melting temperature (K)	2750
A (MPa)	150.0
B (MPa)	368.0
n	0.29
C	0.042
m	0.251
Reference temperature (K)	298
Reference strain rate (1/s)	1

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