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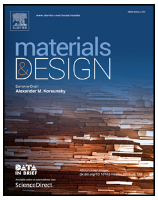

Cold-spray bonding mechanisms and deposition efficiency prediction for particle/substrate with distinct deformability

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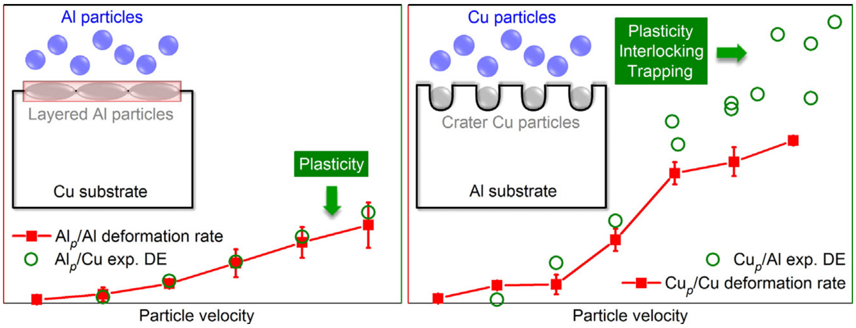
HIGHLIGHTS

- Cold-spray of particle/substrate with distinct deformability has been investigated.
- Deposition is controlled by particle plasticity for soft particle/hard substrate.
- Trapping and interlocking in hard particles/soft substrate is elucidated via numerical simulations.
- The operation and playoff of different bonding mechanisms is demonstrated.
- Predictive assessment of deposition efficiency is in good agreement with experiments.

GRAPHICAL ABSTRACT



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ABSTRACT

Cold-spray involving particle and substrate that are of distinct deformability, i.e., *soft* (particle)/*hard* (substrate) and *hard* (particle)/*soft* (substrate) systems, has been investigated using systematic numerical simulations. For the *soft*/*hard* system, the bonding and deposition is dominantly controlled by particle plasticity. In contrast, physical trapping and mechanical interlocking play an important role for the *hard*/*soft* system. Employing a layered-particle/substrate model and a crater-particles model, we demonstrated the operation and playoff of different bonding mechanisms, and provided predictive assessments of the deposition efficiency that correspond well to experimental measurements. The present study offers key mechanistic insights towards rational process design of cold-spray.

1. Introduction

Cold-spray is a rapid kinetic coating process where feedstock particles are accelerated by an expanding gas stream towards a substrate while maintaining temperatures well below their melting temperatures [1–3]. At or above a critical velocity, particles will experience significant plastic deformation rate that induces adherence of deformed particles on the substrate, and with each other, to form a coating [4–6]. Owing to its low-temperature operation, cold-spray possesses many technological advantages such as minimum modification of material microstructure, and low-degree of oxidation and chemical degradation or reaction [7–10]. These advantages enable the preferable applications of cold-spray in coating temperature-sensitive materials, e.g., polymers [11,12] and nanocrystalline materials [13,14], and oxidation-sensitive materials e.g., aluminum, copper and etc. [15–17]. In addition, cold-

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spray is capable of handling a wide spectrum of feedstock materials. This promises great versatility and possibilities of achieving new functionalities in coating. For instance, cold-spray provides a quick route to manufacture complex and/or heterogeneous coatings that integrate different material systems, e.g., ceramic [18,19] and metal [20–22] matrix composite and polymer/metal [11,23] coating.

In the application of cold-spray technology, one metric people strive to improve is the deposition efficiency (DE), defined as the ratio of the weight of adhered particles to the total weight of sprayed particles. To date, the optimized spraying conditions that yield high DE for cold-spray are mostly determined on a *trial-and-error* basis [1,2,4,24], which not only incurs substantial cost but also time-consuming. In this regard, substantial research has been conducted to investigate detailed deformation behaviors and deposition mechanisms occurring in particle/substrate and particle/particle contacts [6,24–29] in order to identify a rational strategy for the optimization of the spraying parameters to attain high DE. Assadi et al. [6] suggested shear instability as an indication of the occurrence of bonding. Li et al. [27] postulated a model to examine the effects of particle size and spray angle on base of the critical velocity [4–6]. Those studies provide important mechanistic information towards understanding the onset of particle deposition. However, they did not provide predictive assessment of the evolution of DE as spraying conditions (e.g., particle velocity) vary.

In a recent study, we demonstrated that for particle/substrate systems with similar deformability, the evolution of DE can be quantitatively analyzed in terms of the plastic deformation of the particle [30]. In particular, we showed that the experimental DE is linearly correlated with an effective plastic deformation rate through a material-independent correlation parameter [30]. However, the deposition scenario is necessarily different in systems comprising particle/substrate of largely dissimilar deformability where plastic deformation is predominately occurring in either the particle or the substrate [28,31,32]. In this regard, here we probe the detailed deposition process in those systems with distinct deformability during cold-spray using finite-element modeling, aiming to clarify the complexities induced by the deformability mismatch between the particle and substrate. Two different models, namely a layered-particle/substrate model and a crater-particles model, were employed to simulate distinct material deformation behaviors and particle-substrate interactions, and to clarify the playoff of different bonding mechanisms. Based on numerical simulations, predictive assessments of DE curves were obtained, showing good agreements with experimental measurements. The present study provides crucial insights towards predictive modeling of the cold-spray process.

2. Computational methodology

The particle/substrate deformation and subsequent deposition process is simulated using the non-linear finite element analysis (FEA) package from Abaqus/Explicit [33]. Aluminum and copper are the two materials chosen for modeling the particle/substrate systems, i.e., Al_p/Cu and Cu_p/Al, where the subscript p indicates particle. The above choice is based on the availability of reliable and systematic experimental DE vs. particle velocity data [1,4]. Given that coating is produced by the successive deformation and layering up of particles, there are two key stages in the spraying process: (*i*) the formation of a thin "*seeding*" layer that involves particles impacting on the substrate, followed by (*ii*) subsequent incoming particles impacting on pre-deposited particles. Eventually, when there is sufficient build-up of pre-deposited particles, it essentially becomes a cold-spray process where the particle and substrate are of the same material. However, depending on the relative deformability of particle and substrate, the deformation behaviors can be vastly different in the initial stage (i.e., stage (*i*) mentioned above). For a system where particles exhibit higher deformability than the substrate, i.e., *soft* particles versus a *hard* substrate (denoted as *soft*/*hard* below), e.g., Al particles impacting a Cu substrate, the deformation mainly occurs in the particle, and thus flattening of particles during the initial impact is expected, as illustrated in Fig. 1a. On the other hand, for a system involving hard particles and a soft substrate (denoted as *hard*/*soft* below), e.g., Cu particles impacting an Al substrate, embedding of hard particles (see Fig. 1d) in the soft substrate is observed during the initial deposition, where particles are only slightly deformed. Please note that the above classification of the dissimilar deformability, i.e., *soft*/*hard* for Al_p/Cu and *hard*/*soft* for Cu_p/Al, is based on the actual deformation behaviors of materials. That is, for cold-spray of the *soft*/*hard* system, the deformation will dominantly occur on the particle side with relative small amount of deformation on the substrate side (see, e.g., Fig. 1c), while for cold-spray of the *hard*/*soft* system, there will be substantially more deformation on the substrate side, leading to the particle embedded into the substrate after the impact deformation (see, e.g., Fig. 1f) (also see Supplementary material Section S1 for details on the comparison of deformed configurations between Al_p/Cu and Al_p/mild steel as well as Cu_p/Al and mild steel_p/Al).

Considering the distinct deformation behaviors in *soft*/*hard* and *hard*/*soft* systems, we constructed two different finite element models to simulate the corresponding impacting processes, as schematically illustrated in Fig. 1b and e. For the *soft*/*hard* system, because of excessive deformation (and flattening) of particles, we adopted an axisymmetric layered-particle/substrate model (see Fig. 1b) initially proposed by Yin et al. [34], where successive particles are envisioned to impact on a particle build-up film of variable thickness h, bonded to the substrate (i.e., tie constraint, no relative motions between). Note that a separate set of simulations implementing a degradable interfacial material between the particle build-up film and substrate were also performed, yielding nearly identical results as those obtained above using the tie constraint (see Supplementary material Section S2 for details). It is however, important to realize that the treatment of adhesive surface/interface in modeling cold-spray process can be non-trivial, and even may be important when ceramic or composite particles are involved [35,36]. For the *hard*/*soft* system, the deposition can be viewed as being contributed from two parts: *a*) the plasticity-induced bonding and particle build-up, which can still be captured by the layered-particle/substrate model, and *b*) the effects of particle embedding in the initial stage of deposition that are incorporated by considering particles sitting in the substrate craters, termed as crater-particles hereafter (see Fig. 1e). These crater-particles are of the same material as the incoming particles (in light of the minimal deformation of the crater-particles), which effectively correspond to the embedded particles in the "*seeding*" layer (i.e., the first layer illustrated in Fig. 1d). The depth of embedding is denoted as λ and different densities (ρ_E) of embedded particles are considered in this model by varying the average spacing (t) between neighboring crater-particles, i.e., $\rho_E = 1/t$. The impact process of an incoming particle directed towards the crater-particle decorated substrate is then simulated. There are two typical scenarios, i.e., the incoming particle (*i*) is initially positioned directly above the middle of two crater-particles (see Fig. 1e), or (*ii*) impacts the crater-particle. Noting that in the second scenario, preliminary studies found that the impact process is in close proximity to the layered-particle model. Therefore, we focus our discussion on the first scenario below.

The particle diameter, d, is set as 30 μm, considering the particle size range of commercially available cold-spray powders (with mean powder size ~25–35 μm) [31,37–39], and the substrate is square in shape with edge length in 1 mm. Four-node plane strain elements with element size 1/50d and reduced integration (CPE4R) [33] are created for all parts of the model. During simulation, the bottom of the substrate is fixed, while its side edges are only allowed to move vertically, and the particle is given an initial velocity to impact the substrate. The subsequent deformation under two-dimensional (2D) plane strain is examined. For contact interactions, a surface-to-surface contact interaction with penalty algorithm between the particle and the particle build-up film (Fig. 1b) and among the particle, the crater-particles, and the substrate (Fig. 1e) are applied [33], while the relative motion between the build-up film and the substrate in Fig. 1b is inhibited by a tie constraint