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Response to Comment on "Adiabatic shear instability is not necessary for adhesion in cold spray"

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ABSTRACT

In our original paper, Acta Materialia 158 (2018) 430–439, we showed that adiabatic shear instability is not necessary for impact-induced jetting and bonding to occur in cold spray. We also developed a mechanistic framework to estimate the critical velocity for jetting on the basis of a hydrodynamic spall process. In their comment, Scripta Materialia xx (2018) xx-xx, Assadi et al. raised several questions about the versatility of our framework in capturing cold spray-related physical phenomena. Here, we demonstrate that not only can our mechanistic framework explain cold spray physical phenomena such as particle size effects, strength effects and temperature effects, but also it can be used to quantify them.

In their comment on our original paper (Acta Materialia 158 (2018) 430–439) on the nature of particle-impact-bonding, Assadi et al. [1] repeatedly iterate that we take the critical velocity for particle-impact bonding as being a unique function of the bulk speed of sound. This is a significant mischaracterization of our work [2], which begins with a mechanistic derivation of a very generic model. Upon the application of some special conditions, namely, limiting the discussion only to pure metals, our model simplifies to involve a proportionality to the bulk speed of sound, but Assadi et al. [1] neglect to acknowledge that the more general mechanistic approach is far broader. Indeed, the specific concerns of Assadi et al. [1] about the ability of our model to capture various physical phenomena, including the effect of particle size, strength and temperature, are incorrect. In this letter, we respond by first reiterating our mechanistic derivation, providing more of the steps explicitly, and offering a careful examination of the special conditions under which this approach leads to a proportionality between the critical velocity and the bulk speed of sound. We also present new results and arguments showing that our approach, unlike what Assadi et al. [1] suggest, is capable of explaining mechanistic phenomena in cold spray including particle size effects, strength (hardness) effects and temperature effects.

Clean metallic contact at the atomic level is necessary for metallic bonding, and one way to achieve this condition is through impact-induced large interfacial straining. The large interfacial strain needed for bonding is, however, not known a priori. In our original paper [2], we made explicit what is most commonly a "tacit assumption":

- "it is a tacit assumption in the field (which we continue to make here) that such jetting and the attendant large interfacial strain it involves produce a clean metallic contact capable of bonding…"
- "… as envisioned above, the extreme strain associated with jetting is perceived to produce a clean intimate contact amenable to metallic bonding at the interface."

Notwithstanding these clear statements about our view of the relation between jetting and bonding, Assadi et al. [1] incorrectly claim that a "core statement" of our work is "jetting is the cause of bonding"; we have clearly portrayed jetting as a means of producing the large interfacial strains which in turn permits clean metallic contact, break-up of oxides, and bonding. A more correct set of core statements for our work is:

1. Thermal softening, and as a result, adiabatic shear instability, are not necessary for jetting (or bonding) to occur.

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2. The critical velocity for jetting can be estimated on the basis of a hydrodynamic spall process upon impact, and such jetting permits bonding through the large interfacial strains that it produces.

Our observation that jetting leads to bonding is rooted in recent in-situ studies of the moment of bonding in aluminum [3]. We observed in real time that the particles that undergo jetting bond to the substrate, while rebounding particles do not form jets.

Assadi et al. [1] appear to agree with us on our above statement (1) on jetting, but raised several questions on our mechanistic framework, to which we respond here.

As we noted in the original paper (see Fig. 8 in Ref [2]), there are many areas of research including explosive welding [4,5], micro-droplet impact [6,7], shaped charges [8,9], and asteroid strikes [10,11], in which jetting has been discussed and understood as hydrodynamic phenomenon. Our contribution [2] showed that the same concept can be extended to cold spray. Our finite element simulations [2] suggested that pressure release at the particle edge and reflected tension are the origin of jetting in microparticle impact. We proposed a criterion that if the hydrodynamic tension exceeds the dynamic strength of material, then localized failure of material can produce large interfacial strain needed for bonding:

$$P^{-} = P_s \tag{1}$$

here P^{-} is the reflected hydrodynamic tension. Inspired by the observation of discernible isolated ejecta in our in-situ experiments [3], as a first step, we proposed that spall strength, P_s, can reasonably represent material strength against shock-induced localized fragmentation. Assuming that the reflected tension after pressure release is proportional to the impact pressure itself, we re-wrote Eq. (1) as:

$$k \times \frac{1}{2}\left(\rho C_0 V_{cr} + s\frac{\rho V_{cr}^2}{2}\right) = P_s \tag{2}$$

with ρ being the density, C_0 the bulk speed of sound, V_{cr} critical velocity for jetting, and s a material constant. The parameter k captures the assumed proportionality between impact pressure and induced tension. Solving Eq. 2 for V_{cr} yields:

$$\frac{V_{cr}}{C_0} = \left(\frac{\sqrt{1+\frac{4sP_s}{kB}}-1}{s}\right) \tag{3}$$

which we can further approximate as

$$\frac{V_{cr}}{C_0} \approx \frac{2}{k} \times \frac{P_s}{B} \tag{4}$$

Eq. (4) relates the critical velocity to the bulk speed of sound, bulk modulus and spall strength of materials. Assadi et al. [1] claim that our "article posits that jetting and particle bonding are governed by the bulk speed of sound, v_s, based on the observed linear correlation between v_s and the critical velocity for four metals". This is a mischaracterization for two evident reasons. First, the bulk speed of sound is not the only material parameter in Eq. (4). Second, the correlation between the critical velocity and the bulk speed of sound is not based on observation. Rather, we have derived the correlation as shown above; the experiments only verify it. Assadi et al. [1] appear to have missed the development and discussion of these equations and in particular, the fact that the spall strength drops out of the equation only when one makes an assumption that ${}^{P_s}/_{B}$ is a constant, which is reasonable only for pure metals. We return to this point in more detail later in this letter.

Assadi et al. [1] claim that we have taken the critical velocity "as a unique function of the bulk speed of sound, a property which does not capture the effects of thermal and plastic properties of the particles". They continue with "an example provided by Krebs et al. [12], where the softer powder is shown to result in better bonding as compared to the harder powder of the same material (i.e. of the same v_s)" and argue that "the bulk speed of sound cannot be a dominant factor in particle bonding during CS deposition". They conclude that the "observed correlation between the critical velocity and v_s should therefore be taken as one that does not imply causation."

Again, Assadi et al. [1] have ignored an important part of our mechanistic derivation and arrived at an incorrect conclusion as a result. Eqs. (1–4) quite explicitly include the dynamic strength of the material in the form of the spall strength P_s. If metal *A* and *B* have the same elastic properties and densities, but different strengths, then according to Eq. (4) the ratio of their critical velocities should be equal to the ratio of their spall strengths:

$$\frac{V_{cr,A}}{V_{cr,B}} = \frac{P_{s,A}}{P_{s,B}} \tag{5}$$

To assess the above premise, we have conducted new in-situ impact experiments with Al6061, Al2024 and Al7075, and add to them our results on pure Al from Ref. [3]. Since pure Al and it alloys have similar speeds of sound but different strengths, this experimental design effectively isolates strength effects on the critical velocity. Details of the experimental procedure can be found elsewhere [3]. We highlight that in our experiments the particle and the substrates are matched materials.

Fig. 1 shows the ratio of the rebound, V_r, and the impact velocity, V_i, as a function of the impact velocity. The transition from the rebound regime to the bonding regime is clean and clear for pure Al; there is no particle with non-zero V_r beyond the critical velocity for pure Al. For the Al alloys, on the other hand, we observe a mixture of bonding and rebounding behaviors at the high velocities, owing most likely to microstructural variations in the alloy particles. For the present analysis, we take the minimum velocity at which we induce bonding as the critical velocity. Unlike Assadi et al.'s argument [1] in using "better bonding" in "softer metals" to imply strength effects, here we have precisely isolated and measured the change in the critical velocity as material strength increases form pure Al to Al6061 to Al2024 and Al7075.

Since we can neglect differences in the modulus and bulk speed of sound amongst these four materials, Eq. (4) suggests that an increase in the spall strength should be responsible for the increase in the critical velocity. We have collected experimental measurements of the spall strength for Al and its alloys from the literature [13–23]. Fig. 2 shows the normalized critical velocity as a function of the normalized spall strength following Eq. (5), with the data points being the experimental measurements and the dashed line being the theoretical expectation. We note that although there is scatter in the literature spall strength data for each material due to differences in the experimental conditions, the overall trend is exactly as expected: a linear increase in the critical velocity is found as a function of the spall strength. Thus, Assadi et al. [1] are incorrect that our approach fails to include the effect of materials strength; the analysis is built upon impact deformation exceeding (spall) strength in the first place, and the resulting scaling it predicts is observed in clean experiments.

Assadi et al. [1] claim that our "criterion is also not useful in interpreting the effect of particle size". This statement is also incorrect; while our paper [2] did not focus on particle size effects, Eqs. (1–4) can certainly be used to evaluate them, as described below.

To the best of our knowledge, the only data available in the literature where particle size effects on the critical adhesion velocity were *exclusively* captured, are what we have reported in Ref. [3]. By 'exclusively', we mean that we have measured both particle size and particle velocity *directly*, and we have made sure that no other parameters interfere with the impact-bonding experiments; in most cold spray experiments, particles with different sizes heat up to different temperatures in contact

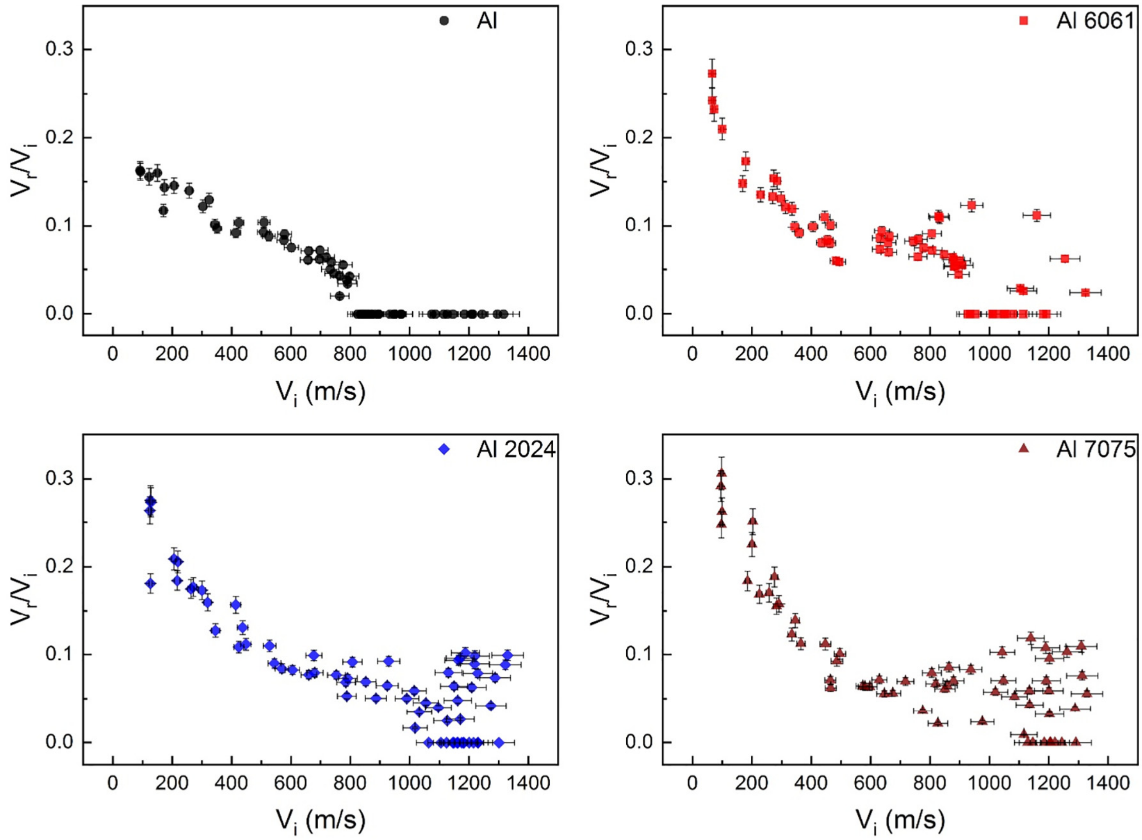

Fig. 1. The ratio of the rebound, V_r, and the impact velocity, V_i, as a function of the impact velocity for Al (adapted from Ref. [3]), Al6061, Al2024, and Al 7075. The particle sizes for these experiments are 14, 15, 14, and 14 microns, respectively.

with the carrier gas, and are accelerated to different velocities. In Ref. [3], we measured a critical velocity of 810 m/s for 14-μm Al particles, and a critical velocity of 770 m/s for 30-μm Al particles. We observe a decrease in the critical velocity by a factor 0.95 with an increase in the particle size by a factor of 2.14. In what follows, we show that Eq. (4) can explain and quantify this size effect.

In the Eq. (4) the term, P_s, is a temperature- and strain-rate-dependent parameter, which we called attention to explicitly in Eq. (3) in the original paper [2]. Since the two particles we have conducted the impact experiments with are on the same order of magnitude in their size (14 and 30 μm) and the critical velocities are close to each other (810 and 770 m/s), they experience about the same strain rate. Therefore, it is more likely that the size effect manifests from the different adiabatic temperature rise in particles of different size.

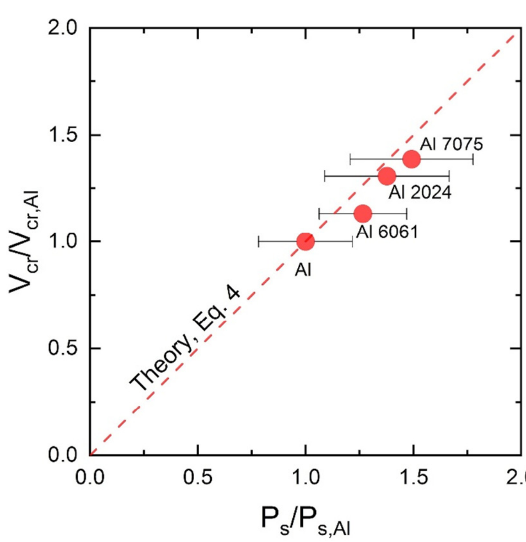

Fig. 2. Normalized critical velocity as a function of the normalized spall strength for Al and its alloys, using pure Al as the reference material following the form of Eq. (5). The experimental data points closely follow the trend suggested by the Eq. (4).

By conducting finite element simulations for Al particles with different particle sizes (see Refs. [2, 24] for method details), we have shown that larger particles experience greater adiabatic heating at the periphery of the particle where jets form [24]. For particles of 14 and 30-μm diameter, we simulate maximum homologous temperatures of 0.48 and 0.58, respectively. This, in turn, leads to greater softening in the larger particle, because the spall strength of Al decreases with temperature. On the basis of Ref. [13], we would expect to see a spall strength reduction by a factor of ~0.93 in the larger particle, and by extension because of the proportionality of Eqs. (4) and (5), the critical velocity of the larger particle should be lower than the small particle by a factor of 0.93. The experimental value is $770/810 = 0.95$, in good agreement with the model.

Clearly, Eq. (4) can be used to model particle size effects, contradicting the assertions of Assadi et al. [1]. In this example, we have assessed the size effect through its effect on the temperature dependency of the spall strength. Interestingly, and by extension, Eq. (4) can capture the effects of temperature too, since P_s is temperature dependent.

Assadi et al. [1] repeatedly suggest that our criterion does not lead to a "correct prediction of the critical velocity as a function of materials and process parameters." In terms of materials parameters, we have density, bulk modulus, and spall strength explicitly included in Eq. (4), and any "materials and process parameters" that one would like to consider can be related to these inputs either directly (as we show for strength in Section 3) or indirectly (as we show for particle size affecting temperature and thus spall strength in Section 4). We thus believe Eq. (4)

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explicitly and implicitly includes the most critical material and process parameters that play a role on impact-induced jetting and bonding.

Of course, in future work we expect there will be many efforts to connect the physics of jetting to other 'process parameters' used in cold spray:

- Gas-related parameters (G): pressure, temperature, density
- Nozzle-related parameters (N): length, shape, material, throat diameter, exit dimeter, etc.
- Particle-related parameters (P): particle size, material mechanical/physical/thermal properties, location in the nozzle, density, etc.

It has been shown that particle impact velocity, V_i, and particle temperature, T_p, right before impact are unique functions of the above process parameters [25]: $V_i = V_i(G,N,P)$, and $T_p = T_p(G,N,P)$. Eq. (4) provides us with the critical velocity as a function of material parameters as discussed above. Therefore, it can be used as a guideline for designing process parameters. One can thus build the connection to process parameters needed for successful bonding using Eq. (6):

$$V_i(P,N,G) \geq V_{cr}(P,T(P,N,G)) \tag{6}$$

The discussion above shows that the dynamic strength of material (which we propose to represent by spall strength), is a central piece of our mechanistic view. Assadi et al. [1] are incorrect when they neglect this aspect of our work and repeatedly suggest that our equation "is a unique function of the bulk speed of sound". The development of Eqs. (1–4) from this letter (Eqs. (2–4) in the original paper [2]) includes a detailed discussion about when it is possible to simplify the expression further to eliminate the spall strength *for pure metals*. As noted in the paper, there are several ways to *estimate* spall strength for *pure metals*. The simplest is Orowan's sinusoidal representation of intermolecular potential that leads to a theoretical spall strength of $P_{s,\ Orowan} \approx B/\pi$ [26]. Second, from the Morse potential one can express the spall strength as a function of density, bulk modulus and the specific cohesive energy U_{coh} i.e. $P_{s,Morse} = \sqrt{\rho U_{coh} B/8}$ [27]. Third, Grady et al. [27,28] developed an energy-based prediction by equating the energies associated with the tensile loading and the fragmentation energy dissipated in spallation, giving $P_{s,Grady} = \sqrt{2BY\varepsilon_c}$. Here Y is the flow stress and $\varepsilon_c = 0.15$ is a material-independent strain. We have calculated all three of these spall strength estimates for pure metals by collecting the specific cohesive energies, densities and bulk moduli from [29] and by using a flow stress from [30] considering a grain size of 1 μm—relevant to our microparticle impact experiments.

In Fig. 3, we plot all three theoretical spall strengths as a function of the bulk modulus on a double logarithmic plot that highlights the

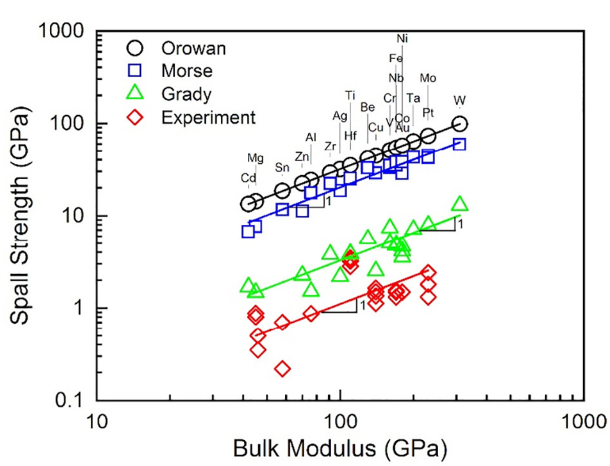

Fig. 3. Spall strength of pure metals as a function of the bulk modulus.

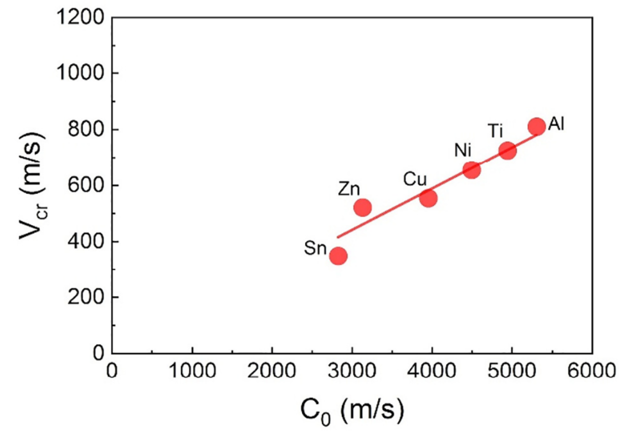

Fig. 4. The proportionality between the critical velocity and the bulk speed of sound for six pure metals.

scaling between these parameters. We also superimpose the experimental measurements of spall strength for a number of pure metals [31] on the plot. While there are expected dependencies upon cohesion energy, density, bulk modulus, and yield strength in the Morse and Grady relations, it is an interesting outcome of the way these parameters combine that the net dependence upon bulk modulus turns out to be linear when spanning pure metals. We also note that there is larger scatter in the experimental data (which can be attributed to differences in the experimental conditions, and the strain-rate, peak pressure, orientation, and grain size dependencies of spall strength [32–34]). Nonetheless, the experimental data show the same scaling that all three theoretical models do: as shown by the fitted lines with a slope of unity in Fig. 3, the spall strength is roughly linearly proportional to bulk modulus in the set of pure metals. It is this correlation in pure metals that provides a basis for further simplification of Eq. (4) by assuming $\frac{P_s}{B} \approx c$ a constant. This leads to a proportionality between the critical velocity and the bulk speed of sound strictly when discussing pure metals, all other things being equal.

Assadi et al. [1] present finite element simulations of the impact-induced deformation and discuss that the deformation is not governed by the elastic properties. This is correct, especially in a regime that involves large plastic strains. However, to use this observation as an argument against our position is inappropriate, since the mechanistic piece, i.e., the process of jetting and fragmentation as a result of dynamic strength of material being exceeded in tension, is absent in their finite element model. In fact, the fundamental mechanisms of the process of spall, itself, are not captured by our finite element modeling either, and we view it as inappropriate to use such models to study the process of spall specifically. However, our Eulerian finite element simulations capture the hydrodynamic and thermomechanical conditions of material in the jet region more accurately than do the Lagrangian simulations of Assadi et al. [1], which helped us identify conditions *where the material is prone to spall*.

In Ref. [2] we showed that the proportionality between critical velocity and the speed of sound appears to hold for four pure metals, in line with the scaling of Eq. (4) developed specifically for pure metals. Here we add our recent results on tin and titanium [35] and extend the proportionality to encompass six pure metals (see Fig. 4). As we pointed out in the original article, the ordering of these data points is not monotonic in virtually any simple property of these metals, including, notably, melting point, modulus, density, or strength/hardness.

In summary, we have discussed various aspects of the theoretical framework to estimate the critical velocity for jetting that we had originally developed in Ref. [2] on the basis of a hydrodynamic spall process. Our mechanistic framework relates critical velocity to the spall strength, bulk speed of sound, and bulk modulus, along with the dependencies of

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these variables upon temperature and deformation during the process of impact. Contrary to the assertions of Assadi et al. [1], our mechanistic framework can explain and quantify fundamental physical phenomena related to impact-bonding, including strength effects, size effects, and temperature effects. Our additional analysis in this paper further bolsters the key argument in our original paper: jetting upon microparticle impact is a hydrodynamic phenomenon that does not require any material softening mechanism, *specifically adiabatic shear localization*. The ability of our simple hydrodynamic spall criterion to capture particle size, strength and temperature effects in line with experiments should further advance the discussion of hydrodynamic effects as being critical in cold spray.

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