Offiery sparks and glittering spots: melting-resolidification and spherical particle formation in abrasion

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The curious occurrence of perfectly spherical particle debris when a steel substrate is slid against a hard abrasive was first documented by Robert Hooke in the seventeenth century. Similar observations now abound in other abrasion-type processes, from industrial grinding to sliding rock faults. The prevalent hypothesis, originally proposed by Hooke, is that these particles form due to high local temperatures, resulting in particle ejection, melting and resolidification. In this work, we revisit this hypothesis, using a model steel-abrasive contact, a combination of in situ and post-process investigations, and complementary analytical calculations. Our results reveal two primary findings—firstly, the temperature of particles ejected from the contact zone is far from the melting point, and secondly, exothermic surface oxidation plays a critical role in actually melting the particle. Melting is either complete or partial, leading to spherical particles or ‘slivers’, as described originally by Hooke. Finally, we confirm that resulting particle surface patterns are typical of rapid solidification from the melt. Apart from throwing light on a centuries’ old curiosity, our results precisely quantify the melting-resolidification process, with implications for a variety of applications, ranging from abrasion and powder production to the formation of micrometeorite dust.

1. Introduction

The curious occurrence of substantial quantities of perfectly spherical particles during abrasion of a steel surface was first reported by Robert Hooke in his highly
Figure 1. Images showing the occurrence of spherical particles in different physical contexts: (a) Hooke’s original drawings showing magnified images of the ‘glittering spots’ observed when rubbing steel and flint [1]. (b) Spherical wear particle formed due to repeated burnishing [2]. (c) Spherical particles recovered from sliding rock fault [3] and (d) micrometeorite sample showing perfect spherical shape and dendritic surface patterns [4].

influential treatise [1]. Hooke systematically analysed the sparks formed when a steel specimen was struck against a piece of flint and found ‘... certain very small, black, but glittering spots ...’ that, when examined with his microscope, were

... pretty regularly round ... being fastened onto the end of a small sliver of Iron, which seemed to be nothing else but a long thin chip ... some of these sparks are the slivers of Iron ... [others] are only small slivers of the Iron, made red-hot with the violence of the stroke...

Based on these detailed observations, Hooke put forth a hypothesis on how these spherical particles were formed and, indeed, on the nature of the spark itself. In modern parlance, he essentially proposed that friction between the flint and steel sample, being operative at local asperity contacts, caused large local temperature rise (red-hot). Consequently, he reasoned, the sparks themselves were nothing but these locally hot regions removed in the form of slivers or chips of iron. In certain cases, depending on the size of the chip, the inherently high local temperatures were postulated to cause complete melting, thereby showing up as round glittering spots post-solidification, see figure 1a. He supported this proposal by drawing attention to common experience—that pieces of violently rubbed steel can indeed reach temperatures high enough to cause skin damage.
Subsequent to these ingenious observations, sub-millimetre-sized spherical particles have appeared in numerous abrasion problems ranging from conventional wear to sliding fault planes. A series of papers in the 1970s established the now-common occurrence of spherical wear debris in rolling contact fatigue [2,5,6] and fretting wear of sliding surfaces [7,8]. These systems certainly could not accommodate material melting since temperatures in lubricated contacts seldom exceed the boiling point of the lubricant (approx. 300°C) [9]. Surface damage in such contacts likely occurs during repeated sliding of the two surfaces, either by adhesive wear or by sub-surface cracking [10], leading to the removal of a small particle from the surface and transported by the lubricant. This particle is then trapped locally within the contact (perhaps inside a cavity) and is repeatedly burnished by asperities on the two sliding surfaces or even other, larger and potentially harder, wear particles. The result is a near-perfect spherically shaped particle ejected from the surface, see figure 1b. The process described here is somewhat reminiscent, yet subtly distinct from, the rounding of pebbles by abrasive-laden river water flow [11]. We will henceforth refer to this mechanism as abrasion-burnishing and it represents an alternative route to Hooke’s melting-resolidification process.

On much larger scales, spherical wear particles are also observed on rock faces in the vicinity of active fault planes [3], a seemingly scaled-up version of the abrasion wear problem described above, see figure 1c. Here, the fault plane constitutes the interface, with two sliding (albeit very slowly) rock bodies on either side. The possibility that these spherical wear particles form via the abrasion-burnishing mechanism—involving trapped rock debris burnished into spherical shape—appears to be readily plausible. However, this system represents an unlubricated contact so that significant interface heating is to be expected, with some estimates suggesting that local temperatures can exceed the material’s melting point [12]. An analogous example is seen in cavitation erosion, where large instantaneous local temperatures can result in melting, particle ejection and resolidification [13,14].

That the melting-resolidification route is a generic and more common mechanism is also borne out by its appearance in yet another geological phenomenon—micrometeorite dust, see figure 1d. Early observations of dust collected from meteor showers revealed the presence of so-called cosmic spherules [15], formed due to frictional heating, break-up and melting of larger micrometeorite objects during atmospheric entry [16]. A simple energy balance calculation reveals the maximum size of spherules to be less than 10 µm [17]. Cosmic spherules are geologically important because their occurrence and chemical composition are linked with their travel history, enabling back-calculation of the composition of the earth’s primitive atmosphere [18]. The evidence to support solidification in this context is quite indisputable [4,19].

Taken in toto, these observations appear to suggest that the melting-resolidification mechanism is far more widespread than the abrasion-burnishing route. However, while this mechanism has been well established in the contexts stated above, its utility in explaining Hooke’s original observations is ironically hindered by two fundamental problems. Firstly, more recent studies have shown that local temperatures in dry sliding contact simply cannot exceed the melting point, especially under the pressures and shear stresses normally associated with rubbing a ductile metal (steel) against an abrasive (flint) [20–23]. Secondly, the occurrence of a spherical particle is almost always accompanied by a spark—so much so that in a nitrogen atmosphere, neither sparks nor any spherical particles are observable [24,25]. In fact, sparks generated when grinding steels are now known to possess characteristic features that can be exploited for identification of elemental content [26]. In Fe-based alloys, frequency and intensity of bursts are related to the carbon content—non-carbide-forming elements such as nickel, cobalt and manganese suppress spark burst whereas carbide-forming elements such as molybdenum, chromium and tungsten change the colour of spark patterns [27]. This strongly hints at the important role played by oxygen in effecting the formation of spherical particles via melting, a thesis that has been explored before, albeit with little quantitative analysis [25,28–30]. Finally, indubitable evidence for the resolidification mechanism has hitherto not been provided. This process must happen rapidly and, consequently, leave its own signature in the final spherical particle’s properties.
In this light, we revisit the melting-resolidification mechanism for the specific problem of an abrasive (flint) removing material from a ductile substrate (steel). Our experiment uses an abrasive wheel and a sliding steel sample which, apart from being controllable, also closely resembles Hooke’s original configuration, see §2. We put forth our hypotheses concerning the details of the melting-resolidification process in §3 and present supporting experimental evidence and accompanying analytical calculations in §4. This latter section follows the life of a single metal chip as it is ejected from the contact zone. Our results and their implications are discussed in §5 with concluding remarks in §6.

2. Experimental configuration

A surface grinding configuration [31] was used to generate spherical particles, analogous to Hooke’s original steel-flint abrasion experiments. The workpiece and wheel were AISI 52100 high carbon steel and alumina abrasive wheel of 170 mm diameter, respectively. Spindle-wheel angular velocity was kept constant at 2800 r.p.m. The workpiece was held over an auto-feed bed with controlled translation velocity. Material ejected from the wheel-workpiece contact zone (grinding swarf) was collected at different distances using a magnetic container. Experiments were performed without the magnetic container as well to ensure that there were no magnetic field effects on the particles produced. No coolant was used during the grinding process. The following parameters were used for the experiments, unless specified otherwise: wheel velocity $25 \text{ m s}^{-1}$, workpiece (bed-translation) velocity $0.15 \text{ m s}^{-1}$, nominal depth of wheel engagement $50 \mu\text{m}$, with a workpiece contact width of $10 \text{ mm}$.

Simultaneous full-field temperature measurements were made using an infrared (IR) camera (Optrix PI 640) placed at the side ($xz$-plane) of the grinding zone. The camera optical resolution was $640 \times 480$ pixels, with spectral range 8–14 $\mu\text{m}$. The resulting accuracy in the temperature field measurement was $\pm 2^\circ\text{C}$. The field of view of the camera was $83 \text{ mm} \times 62 \text{ mm}$, and it recorded images at 32 frames per second (FPS). Prior to making the measurements, the workpiece emissivity was first evaluated by matching IR data with temperatures measured using a K-type thermocouple. The resulting emissivity values were set equal to 0.1 for the metal surface being imaged.

In order to isolate spherical particles, the collected powder was sieved using both 45 $\mu\text{m}$ and 75 $\mu\text{m}$ mesh sizes. For post mortem analyses, the following systems were employed. A FESEM system (Zeiss Ultra55) with electron dispersive spectroscopy (EDS) was used for imaging and elemental analysis. X-ray diffraction of the bulk sample before grinding and of the collected particles was performed using Cu-Kα1(1.543 Å) source with scan rate $1^\circ$ per minute. A focused ion beam (FIB) with a micro-manipulator was used to section individual spherical particles and make samples for transmission electron microscopy (TEM). A sample of dimensions $8 \times 15 \times 5 \mu\text{m}$ was sliced radially from the spherical particle and thinned down to $40 \text{ nm}$ thickness using FIB milling. TEM and EDS were performed on samples prepared in this manner. High-resolution transmission electron microscopy (HRTEM) images were analysed using GMS-3 DigitalMicrograph software.

3. Hypotheses concerning the melting-resolidification process

We first present three figures showing the nature, size and morphology of particles produced using the experimental configuration described in §2. Firstly, the shape and size distributions of collected particles are summarized in figure 2. Panel (a) shows an SEM image of particles (post-sieving) at a distance of 300 mm from the wheel-workpiece contact zone. The particles are largely spherical in shape with stringy machining-type chips (approx. 200–500 $\mu\text{m}$ in length) also visible. The spherical particles occur in a variety of sizes, with some of them being hollow (at yellow arrows). A few abrasive particles (alumina) are also seen in the image (at red arrows) and were no doubt dislodged from the grinding wheel. The distribution of typical particle diameters is reproduced in figure 2b. Clearly, particles
Figure 2. (a) SEM image of obtained spherical particles showing particles of perfectly spherical shape. Yellow arrow on image shows a hollow sphere, red arrows show alumina particles dislodged from the abrasive wheel. (b) Size distribution of spherical particles showing wide range of diameters ranging from 10 to 70 µm.

Figure 3. Scanning electron microscopy images showing three distinct spherical particle morphologies. (a) Faceted structure resembling the surface of a soccer-ball. (b) Dendritic surface structure with arrow showing point of emergence of multiple primary branches. (c) Mixed surface morphology with broad dendrite arms.

with diameters between 10 and 70 µm appear to be predominant. The mean particle size (approx. 30 µm) is smaller than mean abrasive size (150–200 µm) in the wheel. The resemblance of these particles to those in figure 1 is striking, especially Hooke’s observations (a).

Secondly, spherical particles show three distinct surface morphologies as seen in figure 3. Panel (a) shows a particle with typical cellular morphology and comprised of multiple facets on the surface. They are arranged together and resemble the patches on the surface of a soccer-ball. In panel (b) is a particle possessing dendritic surface morphology, with primary and secondary branches clearly visible. Several branches appear to emerge from a single point in the image (at arrow). Panel (c) shows an intermediate case between those of panels (a) and (b) and appears semi-dendritic with large branch spacing. All particles collected displayed one of these three morphologies.

Finally, cross-sectional images of two spherical particles, obtained using FIB milling, are reproduced in figure 4. The image in figure 4a shows a particle with a hollow core (at arrow) and a branched inner structure, somewhat like the dendritic exterior seen in figure 3b. On the other hand, figure 4b shows a particle with a cellular-type inner structure (at arrow), analogous to the outer structure seen in figure 3a.
Figure 4. Scanning electron microscopy images of two typical cross-sections of spherical particles. (a) Hollow core showing dendrite-like structures (at arrow) and (b) solid core with facet-like structures in the particle interior (at arrow).

The near-perfect spherical shape, along with these morphological structures, are readily suggestive of solidification from a molten drop. In addition to these images, two other auxiliary observations are also noteworthy. Firstly, local temperatures in the grinding zone have never been recorded to exceed the melting point of steel [21,23]. Consequently, the possibility of material melting within or near the contact zone purely due to interaction with the abrasive appears quite remote. Secondly, when performed in an inert environment (N$_2$ or Ar) and with steel sample, the number of spherical particles formed is nearly zero [32]. Hence, it is clear that oxygen is necessary for the production of spherical particles in steel.

Based on these observations, we hypothesize the following mechanism for the formation of a single spherical particle:

1. An abrasive causes material removal from the workpiece via cutting/rubbing action. The corresponding temperature rise is insufficient to cause local melting.
2. As the removed particle exits the contact zone, it oxidizes rapidly in ambient air. For Fe, this is a highly exothermic reaction that causes intense heat release, leading to melting of the particle.
3. The molten particle, made spherical by surface tension, then solidifies rapidly as it travels through the air. The morphologies in figure 4 are a result of this rapid solidification process and likely involve multiple nucleation sites.

While an analogous mechanism has been proposed before, see for instance, [25] and references therein, we present, perhaps for the first time, detailed observations and calculations in support of it.

4. Results and supporting evidence

We now present experimental evidence and corresponding analytical calculations that lend support to the hypotheses forwarded in §3. We start from the origin of a single particle—as a chip emerging from the substrate—and follow its journey until it is collected.

(a) Temperature of removed material is lower than the melting point

The first stage involves interaction between abrasives on the wheel and a thin material layer on the substrate within the contact zone. Determining whether or not melting occurs in this zone requires the evaluation of local or ‘flash’ temperature due to the action of a single abrasive. Experimental measurement of flash temperatures in the contact zone is well known to be nearly impossible in
Figure 5. Schematic showing heat sources and heat partitioning during wheel-substrate interaction. On the global scale (left), heat \( Q_0 \) is generated at the contact, of which \( Q_w, Q_c, Q_s \) go into the wheel, chip and substrate, respectively. On the local scale (see inset, dashed line), the heat at an individual asperity \( q_0 \) is partitioned into \( q_w, q_c, q_s \) going into the substrate, wheel and chip, respectively. The model heat distributions for \( q_s, Q_s \) are also shown (right).

grinding, especially with the stochastic distributions of abrasive geometry and penetration depth [21]. This problem is further complicated by the near-complete absence of predictive theories for estimating temperature of the ejected chips—thermal heat partition problems in grinding almost always consider only the substrate (or workpiece) and wheel [21,33]. For the present case, prediction of the temperature rise in the ejected chip is crucial, while estimates of accompanying substrate temperatures provide necessary information for validating this prediction. We thus confirm our results by comparing these substrate temperature estimates with those obtained experimentally using IR imaging.

(i) Three-way heat partition in the contact zone

To evaluate the temperature rise in the chip as it exits the contact zone, we use the three-way heat partition model presented in our recent work [34], and originally conceptualized by Ju & colleagues [35]. Temperatures in both chip and substrate are determined using coupled global-local heat partitioning, see schematic in figure 5. The global scale represents gross interaction between the abrasive-laden wheel and substrate, while the local scale accounts for heat sources arising at individual abrasive-substrate contacts.

At the global scale, the wheel interacts with the substrate, generating heat \( Q_0 \) in the process, see figure 5. Part of this heat goes into the wheel \( (Q_w) \), part into the substrate \( (Q_s) \) and the surrounding air/coolant medium \( (Q_f) \), with the rest convected away by removed chips \( (Q_c) \). Since spherical particle formation occurs in dry grinding, and given the length/time scales involved, it is expected that heat convected to the surrounding is minimal [34]. Therefore, the determination of local chip temperature requires evaluating how \( Q_0 \) is partitioned into \( Q_w, Q_c, Q_s \). From conservation of energy, \( Q_0 = Q_w + Q_c + Q_s \), so that we define heat partition coefficients \( \epsilon_w, \epsilon_c, \epsilon_s \) as

\[
\epsilon_w = \frac{Q_w}{Q_0}, \quad \epsilon_c = \frac{Q_c}{Q_0}, \quad \epsilon_s = 1 - \epsilon_c - \epsilon_w. \tag{4.1}
\]

Assuming that \( Q_0 \) is known, the problem hence requires the determination of two unknowns \( \epsilon_w, \epsilon_c \). Note that the global heat \( Q_0 \) is generated non-uniformly over the wheel-workpiece contact
length $-L < x < L$, consequently each $\epsilon$ will be a spatially varying function. This will be explicitly accounted for later.

On the local scale, heat is actually generated via individual asperities interacting with the substrate. If the (known) ratio of nominal to real contact area is $\zeta$, then from energy balance, the local scale heat fluxes $q_0$, $q_w$, $q_c$ (shown in figure 5 (inset)) are related to the global values by

$$q_0 = \zeta Q_0 \quad q_w = \zeta Q_w \quad q_c = \zeta Q_c$$

with $Q_0$, $Q_w$, $Q_c$ denoting initial grain temperature. Here again, the coefficients $K_w$, $\alpha_w$ correspond to the grain thermal conductivity and diffusivity, respectively. Finally, for the chip temperature, we use

$$T_c = T_w \quad \text{and} \quad \tau_w = T_s + \tau_s,$$

where the relations are assumed to hold at each point of the corresponding intimate contact interface. Note that in the second relation, contributions from both local ($\tau_s$) and global ($T_s$) sources are taken into account for the substrate temperature. Details of this local scale heat fluxes $q_w$ are accounted for later.

Secondly, in order to apply the conditions in equation (4.3), we have to obtain the spatial temperature distribution along each of the contacting zones, as a function of the heat input (equivalently, $\epsilon_w$, $\epsilon_c$, $\epsilon_s$) using suitable temperature models. We use the following temperature models for the present case, see figure 5 (right). For the substrate on the global scale, the heat flux is assumed to be uniformly distributed in the form of a strip source of width equal to the contact zone width $L$. Consequently, the global substrate temperature is obtained as (see, for instance, [36])

$$T_s(x) = \int_{-L}^{+L} \frac{\epsilon_s(x')}{\pi K_s} Q_0(x') \exp \left( \frac{v_s(x - x')}{2\alpha_s} \right) \times \frac{1}{K_0} \left( \frac{v_s|x - x'|}{2\alpha_s} \right) \, dx',$$

where $v_s$, $\alpha_s$, $K_s$ are the substrate traverse speed, thermal diffusivity and thermal conductivity, respectively. $K_0(\rho)$ is the Bessel function of order 0 and $\epsilon_s$ is as defined in equation (4.1). Note that this solution is obtained by solving the steady-state heat equation for a continuously moving point source and using the principle of spatial superposition.

For the local scale, given the changing engagement geometry of the abrasive, a triangular source approximation (figure 5 (right)) is used. Additionally, the short engagement time implies that a steady-state approximation cannot be employed, thereby requiring a finite-time integral [36]:

$$\tau_s(x) = \int_{-L}^{+L} \frac{1 - h(x')}{|q_s(x')/2K_s - \epsilon_s(\sqrt{\pi v_w|x - x'|}) \cdot \text{erf} \left( \frac{b_g}{\sqrt{(4\epsilon_s/v_w)|x - x'|}} \right) - \text{erf} \left( \frac{x - x' - l_g}{\sqrt{(4\epsilon_s/v_w)|x - x'|}} \right) \right)} \, dx',$$

where $l_g$, $b_g$ are length and breadth of a single abrasive, while $v_w$ is wheel linear velocity, respectively. The other constants are the same as earlier and the function $h(x) = 1$ if $x \geq 0$ and $h(x) = 0$ otherwise.

For the local abrasive temperature $\tau_w$, a one-dimensional approximation is used to obtain (see also [34]):

$$\tau_w(x) = \tau_{w0} + \int_{-L}^{+L} \frac{h(x)q_w(x')}{K_w} \sqrt{\frac{\alpha_w}{\pi v_w|x - x'|}} \, dx',$$

with $\tau_{w0}$ denoting initial grain temperature. Here again, the coefficients $K_w$, $\alpha_w$ correspond to the grain thermal conductivity and diffusivity, respectively.
Figure 6. Analytical temperatures obtained using the three-way heat partitioning scheme. (a) Partition functions $\epsilon_c, \epsilon_w, \epsilon_s$ along the corresponding non-dimensionalized contact length $\xi$. The blue axis depicts variation of non-dimensional temperature $\theta$ (see definition in text) along $\xi$. (b) Two-dimensional temperature field showing variation into the depth of the substrate ($z$-direction). The maximum substrate temperature of approximately $400^\circ C$ occurs at the substrate surface.

a lumped estimate based on the contact geometry:

$$T_c(x) = \epsilon_c(x) \frac{q_c(x)}{A \rho_s C_s} \frac{2L^2}{\gamma (L + x)}$$

with $\rho_s, C_s$ are mass density and specific heat of the chip/substrate material, $\gamma$ is the imposed material removal rate per unit width.

Finally, we can use the temperature fields equations (4.4)–(4.7) in the two matching conditions of equation (4.3) to obtain expressions for the partition coefficients $\epsilon_c, \epsilon_w, \epsilon_s$. The end result is two equations, valid point-wise along the contact that must be solved numerically. Details of this calculation, including the discretization step and a typical solution scheme using Simpson’s one-third rule, are provided in [34].

(ii) Numerical estimates of chip temperature

The three-way heat partition described above can be used to obtain the temperature for the chip as it exits the contact zone. For this purpose, we have to solve for the partition functions in equation (4.1) using the relations in equation (4.3). In order to obtain values for the experiments described in §2, the following parameters were adapted from [35,37]: for the substrate (AISI 52100 steel), $K_s = 60.5 \text{ W/m-K}$, $\alpha_s = 1.8 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$. For the abrasive (alumina), $K_w = 36 \text{ W/m-K}$, $\alpha_w = 9.8 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$. The following nominal values were used: $Q_0 = 88 \text{ W mm}^{-2}$, $\zeta = 0.25\%$, $2l_g = 2h_g = 0.25 \text{ mm}$, $L = 1.25 \text{ mm}$.

The resulting partition functions are shown in figure 6a as a function of the non-dimensional distance $\xi = x/L$ within the contact zone. It is clear from this figure that, for the present experimental configuration, we should expect the substrate to take away most of the heat within the contact zone, see $\epsilon_s$ curve (dash-dot line) in figure 6a. By contrast, the fraction of heat carried away by the chip is quite small, $\epsilon_c \simeq 6\%$ on average. This is not surprising, and is the reason that nearly all analysis of grinding temperatures neglect heat lost to the chip [35,37]. These functions can then be used to evaluate the temperatures of the wheel, chip and substrate using equations (4.4)–(4.6).

Given the time and length scales involved, it is clear that the chip temperature is not directly experimentally measurable using IR thermography. Instead, we obtain the chip temperature by measuring the substrate/workpiece temperature rise and comparing it with the predictions of the three-way partition model. The corresponding spatial non-dimensionalized substrate temperature within the contact zone, defined as $\theta_s = K_s (T_s + r_s)/(Q_0 L)$ is shown in figure 6a, see blue solid curve. The temperature increases from the engagement point ($\xi = -1$) to the disengagement point ($\xi = +1$) with the wheel. The maximum temperature occurs inside the
The accompanying two-dimensional temperature field into the depth of the steel substrate (−z direction) is shown in figure 6b. Here the wheel contact zone is between 0–2.5 mm and the maximum temperature rise occurs on the surface within the contact zone, as described earlier in figure 6a. It is clear that the model computations negate the possibility of melting occurring on the substrate/workpiece surface since the maximum predicted temperature is approximately 400°C. Consequently, the corresponding temperature rise in the chip (equation (4.7)) is hence predicted to be much lower (≈650°C) than the melting point (approx. 1400°C) of steel. For reference, using larger, albeit less realistic, values for $Q_0 = 108$ and 150 W mm$^{-2}$ yields substrate temperatures of approximately 450°C and 600°C, respectively. The accompanying chip temperatures are again significantly lower than the melting point.

**(iii) Experimental temperature measurements**

These model predictions for the substrate temperature were compared with experimental measurements obtained using infrared thermography. We also simultaneously evaluated the substrate surface condition post-grinding to check for signs of heat-induced damage, see figure 7. The thermal field in figure 7 must be compared with that predicted by the theory (cf. figure 6). Despite minor differences in maximum temperature (400°C and 360°C), the spatial distribution in the two cases appears to be nearly identical. Furthermore, microscopic observations of the cross-section of the ground surface (inset to figure) show no signs of any consequent phase transformations (e.g. martensite formation), consistent with these temperature predictions. It is thus safe to conclude that the workpiece does not even reach its recrystallization temperature, let alone its melting point. Sub-surface temperature field measurements also corroborate this observation, see data presented in figure 7b.

**(b) Exothermic surface oxidation causes particle melting**

In line with the hypothesized mechanism ($\S$3), once formed and in the presence of ambient oxygen, the freshly generated particle surface is postulated to undergo rapid oxidation, leading to significant energy release. This energy is sufficient, under situations to be described below, to melt the particle and result in a spherical drop.
Figure 8. (a) Map showing spatial distribution of elemental oxygen within a thin layer of thickness $\delta$ around 1% of the sphere’s radius. (b) X-ray diffraction data for the bulk specimen (top row) and the collection of spherical particles (bottom row). Peaks corresponding to Fe$_3$O$_4$ are clearly visible in the bottom plot and are absent in the top.

(i) Remnant oxide layer on particle surface

We first present experimental evidence for the occurrence of oxidation using post mortem analysis of the final spherical particles. A single spherical particle was first sectioned using FIB milling and an EDS spectrum extracted from the cross-section, see figure 8. The presence of a thin oxide layer is clear from this data, with insignificant or complete absence of oxygen towards the particle’s centre. Typical weight percentages of oxygen (41.3%) and iron (58.7%) within this outer layer (a qualitative indication) are adequate for the formation of iron oxide Fe$_3$O$_4$. Further, the layer thickness is not more than 1–2 $\mu$m, indicating that only a thin surface layer of the ejected particle is subject to oxidation. Traces of diffused oxygen were also registered inside the sphere but their concentration was found to be insignificant in comparison to the surface.

Corresponding X-ray diffraction analysis is reproduced in figure 8b showing diffraction data for the bulk specimen (top) and spherical particles (bottom). The peaks were matched with ICDD/JCPDS data, XRD of the bulk specimen (before grinding) shows diffraction peaks at 44.9, 65.7 and 83.3°, which correspond to standard peaks of $\alpha$-Fe BCC phase (110), (200) and (211), respectively. On the other hand, XRD spectrum of grinding swarf has additional peaks at 30.1, 35.4, 43.05, 53.3, 57.5 and 62.5°, which can be attributed to Fe$_3$O$_4$ (220), (311), (400), (422), (511) and (440), respectively. These peaks were matched with ICDD data for Fe$_3$O$_4$ file no. JCPDS-19-629.

(ii) Analysis of the oxidation process

If we assume the initial temperature of a single chip leaving the grinding zone is $T_0 < T_m$ (melting point), considerations of oxidation kinetics and the ensuing temperature rise can be used to evaluate whether the chip will melt entirely, partially or not at all. In order to do this, we first note that oxidation occurs along a freshly generated surface post removal, which is typically the underside of the chip.

Most metals and alloys obey a parabolic law for oxidation kinetics, at least at temperatures below the melting point [38,39]. As a first approximation, we assume this law to be valid so that the thickness of the final oxide layer $l_{ox}$ may be related to the oxidation time $t_{ox}$ as $l_{ox}^2 = K_p t_{ox}$, where $K_p$ is a rate constant. The corresponding (constant) heat flux generated due to the oxidation process is computed as

$$Q = \frac{\rho \Delta H_{ox}}{t_{ox}} = \frac{\rho \Delta H}{\sqrt{K_p t_{ox}}},$$

where $\Delta H$ and $\rho$ are oxidation enthalpy and density, respectively.

The geometry for the heating problem is now postulated as follows. The length $L_c$ is taken in the uncut chip thickness direction $\eta$, see figure 9. It is further assumed that $l_{ox} \ll L_c$. We set the
time \( t = 0 \) to when the chip exits the deformation zone at temperature \( T_0 \) and comes in contact with air. Continuous heat flux input \( Q \) occurs at \( \eta = 0 \) due to the oxidation process on the freshly created surface, from \( t = 0 \) until \( t = t_{ox} \). We assume that the other end \( \eta = L_c \) is insulated and ignore any convective effects. For concreteness, the typical convective heat transfer coefficient is \( h \approx 50 \text{ W m}^{-2} \text{ K} \), which results in an outward heat flux of approximately \( 2500 \text{ W m}^{-2} \), assuming a temperature difference of 500 K. By contrast, heat generated during the oxidation process obtained from equation (4.8) for typical values of \( \Delta H \) and \( K_p \) is approximately \( 1.5 \times 10^5 \text{ W m}^{-2} \), and is nearly 20 times larger [40]. In the interest of being able to obtain a closed-form solution, we believe that this serves as a good first approximation.

Melting, if it occurs in the chip, must happen in two stages: firstly, the temperature of the solid chip is increased from \( T_0 \) to \( T_m \), the latter occurring first at the surface \( \eta = 0 \) at some time \( t_m \) to be determined. Secondly, a melting front begins to propagate from \( \eta = 0 \) at \( t = t_m \) towards \( \eta = L_c \) with continued heat flux \( Q \) input. This second process is fairly complex and its analysis requires accounting for latent heat release due to phase change. This is done using the heat integral method introduced first by Goodman [41,42]. As shown in figure 9, if the melting front \( s(t) \) reaches \( \eta = L_c \) at time \( t_L < t_{ox} \), the entire chip will melt, leading to the formation of a spherical particle. On the other hand, if \( t_m < t_{ox} < t_L \), then only partial melting of the chip will occur before the heat source is extinguished. Finally, if \( t_m > t_{ox} \), a melting front will not start, resulting in unmelted chips.

Using this framework, we first compute the temperature at \( \eta = 0 \) using an analytical one-dimensional transient finite domain heat conduction solution for the geometry and boundary conditions outlined in figure 9 and the heat flux \( Q \) given by equation (4.8) using an integral transform method, see Ch. 3 (p. 112) of [43]:

\[
T(\eta = 0, t = t_m) = T_m = T_0 + \frac{Q L_c}{K} \left( \frac{a t_m}{L_c^2} + \frac{1}{3} - 2 \sum_{n=1}^{\infty} \exp\left(\frac{-n^2 \pi^2 t_m}{L_c^2}\right) \frac{n^2 \pi^2}{(n\pi)^2} \right),
\]

from which \( t_m \) is determined by inversion as

\[
t_m \approx \frac{L_c^2}{a} \left( \frac{K(T_m - T_0)}{Q L_c} - \frac{1}{3} \right).
\]

This expression is obtained by neglecting the infinite series in equation (4.9) after numerical evaluation.

The surface of the chip subject to oxidation (\( \eta = 0 \)) hence starts melting at \( t = t_m \). We define a non-dimensional parameter \( c = l_{ox}/L_c \) and set an upper bound on its value as follows (recalling that \( c < 1 \) always by assumption). Considering a chip of thickness \( L_c \) with cross-section area \( A \) that would eventually form a sphere of radius \( r \) (cf. figure 8), we have from volume conservation

Figure 9. Schematic showing geometry of a chip subjected to oxidation on the freshly generated surface \( \eta = 0 \). The melting front location is denoted by \( s(t) \).
Table 1. Pairs of values for c and minimum T₀ for which the inequality equation (4.12) is satisfied. For all the values shown here, at least partial melting will occur. Initial T₀ significantly larger than the minimum values in the second row can lead to complete melting (see text for details).

| c   | 0.01 | 0.02 | 0.03 | 0.04 | 0.05 | 0.06 | 0.07 | 0.08 | 0.09 | 0.10 |
|-----|------|------|------|------|------|------|------|------|------|------|
| min T₀ (°C) | 1288 | 1176 | 1064 | 953  | 841  | 729  | 617  | 505  | 293  | 282  |

\[ A L_c = 4\pi r^3/3, \text{ so that for an oxide layer of thickness } \delta, \]
\[ A l_{ox} = 4\pi r^2 \delta \Rightarrow c = \frac{l_{ox}}{L_c} \sim \frac{3\delta}{r}. \]  

Equation (4.12) provides an estimate for the minimum initial chip temperature T₀ required for final melting to start occurring. Following Goodman and Shea [42], we obtain s(t) using a perturbation expansion in the small parameter \( \kappa = QL_c/2K\Delta T < 1 \) to get \( t_L \).

The inequality in equation (4.12) represents conditions under which melting will begin (at \( \eta = 0 \)). As mentioned earlier, we further check if the chip will melt completely (partially) if \( t_L < t_{ox} \) \((t_L > t_{ox})\). For concreteness, we choose particular values for c and determine the corresponding T₀ such that equation (4.12) is satisfied. Corresponding numerical data are shown in Table 1. It is clear that for \( c = 0.1 \), any chip with initial T₀ > 400°C would start melting. Correspondingly, complete melting, i.e. \( t_L < t_{ox} \), would require an initial T₀ in excess of 850°C. The values used to evaluate these expressions are summarized in electronic supplementary material.

(iii) Explaining the occurrence of melted/unmelted and partially melted particles

Based on the analysis presented above, a complete ‘oxidation-melting phase diagram’ may be constructed to depict regions of no/incomplete and full melting of the chip in T₀ versus L_c space, see Figure 10. The vertical demarcation line shows the bound determined by \( c^2 L_c^2/K_p = t_{ox} < t_f \) where the typical particle flight time \( t_f \simeq 600 \text{ ms} \), see also [44]. This diagram shows unmelted (grey, \( t_m > t_{ox} > t_f \)), partially melted (orange, \( t_m < t_{ox} < t_f \)) and fully melted (blue, \( t_m < t_L < t_{ox} \)) chips in various hues corresponding to different values of the parameter c. For a given value of c, determined by oxidation duration/kinetics and chip dimension, the inequality in equation (4.12) determines the region where melting (either partial or full) of the chips will occur. As explained earlier, complete melting will occur beyond a certain T₀ for which \( t_L < t_{ox} \). We term this change from partial to complete melting as occurring at a critical initial temperature \( T_0^* \), which is also dependent on c. The maroon dotted line in Figure 10 depicts the functional dependence of \( T_0^* \) on c. For the largest value c = 0.12, nearly all the chips exiting the contact zone with uncut chip thickness below approximately 20 µm will melt completely provided \( T_0 > T_0^* \sim 650°C \). Chips exiting with lower initial temperature will partially melt. Analogous interpretations may be made at lower values of c, as shown by the dash-dot (c = 0.08) and dotted (c = 0.10) lines.

Corresponding to these predictions, experimental images of typical melted/unmelted/partially melted particles are reproduced in Figure 11. The fully melted sphere in (a) has a radius \( r = 20 \mu m \). Assuming typical chip length of 200 µm and width 20 µm, we obtain \( L_c \sim 9 \mu m \) from
volume conservation. Consequently, assuming c = 0.12, this chip must have left the contact zone at a temperature anywhere above approximately 650°C, corresponding to the blue region in the phase diagram of figure 10. This is the equivalent of the completely spherical particles observed in Hooke’s original drawings, see figure 1a. The partially melted particle shown in figure 11b has a much larger size so that it must have formed with a likely initial temperature of $T_0 \leq 650$°C if $c = 0.12$. Analogous partially melted ‘slivers’ are also seen in figure 1a. Finally, unmelted chips with length $L_c > \sim 30 \mu m$ are shown in (c); these most likely have exited the abrasion contact zone with $T_0 < 500$°C or are so large that $t_m > 600$ ms so that even significant surface oxidation is unable to cause any melting.

(c) Rapid solidification results in characteristic microstructures

Post-oxidation and, when appropriate, complete melting, the ejected particle takes the form of a spherical molten drop at temperatures greater than 1800 K. Subsequently, it undergoes convective cooling and solidification over a timescale of approximately 100 ms to form a spherical particle in the collection bin. This constitutes the final stage in its evolution; the accompanying cooling rate of approximately $10^4$ Ks$^{-1}$ is typical of rapid solidification processes, resulting in characteristic features.

Recall that three distinct microstructures are observed in the powder particles—cellular, dendritic and mixed, cf. figure 3. The cooling rate is the key parameter that determines which of these structures actually occurs in a drop of a given size. In the present problem, the cooling

Figure 10. Phase diagram depicting conditions on $T_0$ and $L_c$ for which full (blue), partial (orange) and no (grey for $c = 0.08$) melting occurs in the chip exiting the deformation zone.

Figure 11. SEM image of the chip corresponding to (a) full melting (blue region), (b) partial melting (orange region) and (c) no melting (grey region) of the phase diagram.
The rate for the drop to form a powder particle is approximately $10^5 \degree \text{C/s}$. At these high solidification rates, high carbon steel is likely to form body centred tetragonal (BCT) martensitic structure [45]. Evidence for this is not discernible from the XRD analysis of figure 8b since both BCC and BCT structures show practically overlapping peaks at $2\theta = 44.7\degree$ in the XRD spectrum [46].

In order to investigate the structure of the crystal lattice, we performed TEM analysis on a sample section retrieved from the cross-section of a typical spherical particle, see figure 12a. The particle cross-sections were obtained in a manner similar to that in figures 4 and 8 using FIB milling. TEM selected area diffraction (SAD) and high resolution-TEM (HRTEM) data were taken at different distances from the outer surface of the particle and towards the centre, see figure 12. The obtained SAD patterns were indexed using zone axis $\overline{113}$. The corresponding lattice parameters were calculated from SAED patterns to be $a = 2.54 \text{Å}$, $c = 2.58 \text{Å}$, which give $c/a$ ratio of 1.02. These values are consistent with lattice parameters of BCT martensitic phase in high carbon steel [47]. Additionally, HRTEM analysis of the sample showed the $d$-spacing of 0.243 nm. These results, taken together, appear to strongly point at the occurrence of iron in the BCT martensitic phase, as is to be expected from the rapid solidification hypothesis.

It now remains to explain why each of the solidification morphologies of figure 3 occur. Contrary to conventional diffusion-controlled solidification processes, the conditions under which planar–cellular–dendrite morphology transitions occur during rapid solidification remain an area of active investigation [48]. Even the simplest analysis of a perfectly stable spherical solidification front presents significant difficulty [49,50]. The primary complication arises from the unknown location of the solid–liquid interface during solidification, which must be determined as part of the solution—the so-called Stefan problem in conduction heat transfer (see, for instance, p. 282 of [43]). A secondary complication is the singular nature of the geometry. Unlike conventional slab casting, a drop has a finite volume and if solidification proceeds inwards from the outer surface, the solution becomes singular as the front approaches the drop centre [51]. Consequently, even a stable interface growth problem remains challenging to solve analytically; potential
interface instabilities leading to cellular/dendritic growth remain, at present, intractable except by very approximate methods \cite{52,53}.

A third, more serious, complication and one that has not even been considered by the works cited above is the possibility of multiple nucleation sites. Under rapid solidification, alloys, such as the steel used here, inevitably solidify by the formation of several nuclei on the surface \cite{54,55}. In fact, additional images of similar morphologies suggest the cells in figure 3 actually constitute individual nucleation sites as opposed to surface features arising from a growing unstable spherical front. Given the seemingly insurmountable complexities of the thermal field surrounding multiple nuclei, we resort to a kinematic model to explain some of the void features in the cellular patterns.

We begin by considering a two-dimensional circle with multiple nucleation centres located on its periphery, see figure 13. Generalization to the three-dimensional case is straightforward and is discussed below. Consider a two-dimensional circular drop of radius \(R_0\) with \(n\) nucleation sites along its circumference. On the microscopic scale (greater than1 \(\mu m\)), crystals centred at each of these points grow with circular symmetry and first meet at a point midway between the two nucleation centres, termed \(N_1, N_2\) in the figure. Further growth proceeds along the perpendicular bisector of \(N_1N_2\)—if the growth rate ratio between the two crystals is \(\xi\), then this line of intersection of the two crystals will be distant \(\xi\) from one of the nuclei, yet still perpendicular to \(N_1N_2\). Taking the final crystal radius to be \(r\), with \(r < R_0\) being distinct from the drop radius due to solidification shrinkage, then the shape of the void is determined using elementary geometry as shown in figure 13.

The only changes to this calculation for the case of a three-dimensional sphere with arbitrary nucleation sites are as follows. Firstly, since growth is now spherically symmetric, the line of intersection between adjacent nuclei will become instead a plane passing through the sphere centre and perpendicular to the line \(N_1N_2\). The intersection of this line with the drop surface will be a great circle. Consequently, the original line \(AA'\) in figure 13a must be interpreted as the planar projection of a circle on the surface. Secondly, if the nucleation points are now moved closer to each other on the surface of the sphere, the calculation remains the same with the final void shape being determined only by \(r, R_0\) and the locations \(N_1, N_2\). These considerations apply to each pair of nucleation sites.

Two such simulated patterns for three-dimensional arbitrarily located nucleation sites are shown in the top row of figure 13a,b, and correspond to 3 and 4 nuclei, respectively. The final shape of the three-sided and four-sided voids is clear from the figure. These may be contrasted with corresponding experimental images showing analogous voids. Furthermore, the size of these voids can be predicted by suitably choosing the ratio \(r/R_0\), according to the expected density.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure13.png}
\caption{Kinematic nucleation model for describing voids showing (a) three-site (top numerical, bottom expt.) and (b) four-site solutions (top numerical, bottom expt.). \(AA'\) shows the perpendicular bisector of the line joining two nucleation sites \(N_1, N_2\). In three dimensions, this line represents a circle on the surface of a sphere.}
\end{figure}
difference between the liquid and solid phases. Voids with more sides were seldom observed in any of the powder particles.

These elementary considerations suggest that each facet on the surface observed in figure 3 is likely a separate grain and not a result of a cellular growth instability. The transition to dendritic growth could appear with each of these growing nuclei, and, given their near-equal growth rate, likely occurs simultaneously. This could explain the other two microstructures seen in figure 3. Unfortunately, a more quantitative description is beyond the scope of the present work and we hope to present a more complete analysis in a future publication.

5. Discussion

Our results have clearly shown that the standard melting-solidification hypothesis—that particles are in the molten state as they exit the wheel-workpiece contact zone—requires the inclusion of an additional oxidation step to explain the experimental observations. Given the difficulty in accurately measuring chip temperatures, we obtained bounds using a detailed three-body heat partition method. Our calculations strongly suggest that the chip does not reach anywhere near the melting temperature even under extreme cases, i.e. when $Q_0$ is 150 W mm$^{-2}$. Consequently, melting must occur post-chip ejection from the contact zone. Detailed evaluation of the oxidation process and related energetics showed that exothermic oxidation is the most likely cause for particle melting, while also elucidating conditions under which partial/full or no melting occurs. This was confirmed by cross-section analysis, which showed the occurrence of a thin oxide layer on the surface of the particles. Subsequently, the molten drop assumes a spherical shape and undergoes rapid solidification to form a single spherical particle. We now discuss some additional features of the process that couldn’t be explored in the present manuscript.

(a) Other consequences of oxidation during particle formation

As we have already noted in the introduction, the spectral properties of grinding sparks are commonly used to identify scrap metals [26]. The basis for this ‘spark test’ is the characteristic spark spectra and patterns that result when certain elements constituting an alloy are subjected to oxidation. Sparks patterns from carrier lines, carbon bursts and tongues have been observed in various metals and alloys [56]. In Fe-based alloys, frequency and intensity of bursts are related to the carbon content. Non-carbide forming elements such as nickel, cobalt and manganese suppress spark burst whereas carbide-forming elements such as molybdenum, chromium and tungsten change the colour of spark patterns [27]. Nickel-based superalloys and titanium alloys also have their characteristic spark patterns.

In steels, the formation of CO$_2$ due to oxidation of Fe$_3$C (cementite) may produce internal pressure leading to exploding particles. The reaction can be given as: Fe$_3$C + 3O$_2$ = Fe$_3$O$_4$ + CO$_2$ [27]. We observed evidence for this in our experiments as well, one such hollow particle with an apparent burst surface (see electronic supplementary material, figure S1). It is quite likely that this feature is a result of simultaneous carbon and iron oxidation leading to the sudden expulsion of CO$_2$ from inside the particle.

More recent studies within the spark test community have focused on temperature measurement during spark formation by tracking their evolution during abrasive cutting. For instance, the study in [44] employed colour-ratio pyrometry combined with particle streak tracing velocimetry and image processing to obtain non-intrusive temperature estimates. These were reported in the range from 1500 to 1700°C implying oxidation and melting of these particles, consistent with the mechanism outlined in §4b.

(b) Thermal analysis of surface patterns on solidified spheres

The three primary patterns that occur on the surface of the spherical particles (cf. figure 3) are typical of solidification structures observed during casting [57]. However, the precise thermal
conditions governing the development of each of these structures remains as yet unclear. The situation is complicated by multiple factors. Firstly, given the rapid timescales for solidification, the possibility of multiple simultaneous nucleation sites implies that the temperature field in the sphere is most likely not radially symmetric. This then necessitates the use of a fully numerical solution capable of solving multiple, coupled moving boundary Stefan problems in a confined geometry. Furthermore, the solution must also be able to handle rapid cooling rates $10^5 \degree C/s$ and non-equilibrium effects such as the formation of BCT metastable phases. To the best of our knowledge, such a comprehensive framework does not exist at present. The geometric model used in §4c circumvented this restriction by considering only the kinematics of multi-nucleus growth. This may be extended using a probabilistic description for the nucleation process [55] to provide additional analytical estimates. However, as we alluded to in §4c, it is quite likely that a more detailed evaluation of something as basic as the competition between multiple-nucleation sites and the spherical–cellular front transition is still in order.

Finally, the timescale for solidification also mandates the use of non-equilibrium phase diagrams to account for the formation of meta-stable phases [48,54]. This makes the problem significantly more complex and not particularly amenable to analytical treatment at present. Consequently, standard interface instability analyses [58–60] as well as paraboloidal tip solutions [61] are no longer strictly valid and cannot, for instance, explain morphology transitions like the ones seen in figure 3. We are presently evaluating a modified diffuse interface description and hope to present detailed investigations in a future manuscript.

### (c) Melting-resolidification versus abrasion-burnishing for spherical particle formation

At the outset, mention was made of two parallel mechanisms for the formation of spherical particles in abrasion, cf. figure 1. It is imperative that we now comment on when each of these mechanisms are operative. Firstly, it is clear that for the melting–resolidification mechanism, either local melting (as with large-scale, extreme pressure contacts) or an additional reaction mechanism (as with oxidation) is necessary to bring material into the molten state. Furthermore, for the melting-resolidification route, perfectly spherical shapes of the intermediate liquid drops, such as the ones shown in figure 2, are governed by the so-called Bond number (Bo) and Weber number (We) defined as

$$Bo = \left( \frac{R}{\lambda_c} \right)^2 \quad \text{and} \quad We = \frac{\rho v^2 R}{\gamma},$$

where $R$ is the drop radius and $\lambda_c = \sqrt{\gamma/\rho g}$ is the capillary length for the metal. Perfect spherical shape is possible only when $Bo, We \ll 1$ so that surface tension effects dominate over gravity or any other convective effects. For the particles observed and reported in this work, $R \sim 50 \mu m$ and $\lambda_c \sim 0.4 cm$ so that $Bo \sim 10^{-4} \ll 1$ and $We \sim 0.08 \ll 1$. When such a condition is satisfied, perfect sphericity results and we can be certain that the melting-resolidification mechanism is operative.

Secondly, in situations involving lubricated contacts (e.g. rolling fatigue), the possibility of either large local temperature or exposure to oxidation is remote. In these cases, it is almost certain that any spherical particles must form via the repeated mechanical action alone. While such action may, at first, appear far fetched, it is quite often observed in nature—the perfect rounding of pebbles in a river bed is one common instance. Here, the river flow imparts some directionality to the process so that ellipsoidal shapes result, yet in a completely isotropic situation, one can as well postulate the formation of nearly spherical final shapes. However, an explicit demonstration of this process, via suitable in situ studies, has hitherto not been undertaken, to the best of our knowledge.

Finally, we would like to comment on our use of the term ‘abrasion’ in general. The analyses and results presented in §4 do not take into account the precise mechanisms involved in the chip formation. While there has been significant debate on the mechanics of chip formation in grinding and abrasive processes in general [62], our analysis begins at the point where a chip does form. As a result, all of the inherent stochasticity involved in the process (e.g. various wear mechanisms,
potential chip fracture, plastic deformation etc.) are encoded in the single parameter $Q_0$. The thermal analysis presented here is hence applicable to a range of processes, including perhaps even cutting with circular saws.

(d) **Spherical abrasion particles as a source of powders**

In addition to its historical significance, the formation of spherical particles in the steel system is also of contemporary practical interest. The perfect spherical shape of the particles suggests their use as a starting point for powder-based metal additive manufacturing processes. Our preliminary investigations in this direction have yielded very promising results [63]. While requiring additional reduction steps to minimize oxygen content, this process is expected to be significantly more energy- and cost-efficient as a route for commercial powder production. If successful, it will perhaps link Robert Hooke and his monumental treatise with one of the most active areas of research in contemporary materials science, and perhaps not for the last time.

6. Conclusion

The formation of spherical powder particles in the abrasion of steels poses an interesting scientific problem that can be explained using the modified melting-solidification hypothesis presented in this manuscript. Heat partition models were used to obtain estimates for the exiting chip temperature and it was found that they were unlikely to reach anywhere near the melting point. Based on the consideration of oxidation kinetics and EDS analysis of the surface of the final spherical particles, we postulated the need for an intermediate oxidation route. Detailed investigation of the oxidation and attendant melting dynamics helped establish conditions under which completely melted (glittering spots) or partially melted (slivers) should occur. Finally, TEM analysis of the spherical particle surface provided evidence for the occurrence of rapidly solidified BCT structures post-solidification. Finally, we postulated a simple kinematic description of surface morphology using purely geometric arguments. The complexity of the rapid solidification process includes multiple nucleation sites, metastable phase formation and growth and morphological instability. We hope to present detailed investigations of these problems in a forthcoming manuscript.

**Data accessibility.** The data are provided in electronic supplementary material [64].

**Authors’ contributions.** H.S.D.: conceptualization, investigation, visualization, writing—original draft; P.R.P.: investigation, methodology, resources, validation, writing—original draft, writing—review and editing; P.S.: investigation, methodology, writing—original draft, writing—review and editing; K.V.: conceptualization, funding acquisition, resources, supervision, validation, writing—original draft, writing—review and editing. All authors gave final approval for publication and agreed to be held accountable for the work performed therein.

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