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Improvement of flow of an iron-copper-graphite powder mix through additions of nanoparticles

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ABSTRACT

Nanoparticles of carbon black, silica and ferrous oxide were added to FC-0208 powder metallurgy blends prepared using ethylene bis stearamide as lubricant to investigate their influence on improving flowability of this formulation that tends to be cohesive. This study consisted in preparing multiple premixes having different quantities of nanoparticles of different types and evaluating their impact on flowability and apparent density according to MPIF standards. The important variables for the distribution of nanoparticles on the surface of the host particles were the sequence of addition of all of the constituents, the quantity of nanoparticles and the mixing time. It was possible to identify a condition presenting good flowability with a flow time of 34.9 s/50 g and an apparent density of 2.78 g/cm³.

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1. Introduction

The powder metallurgy (PM) process refers to the densification of a predetermined quantity of metal powders and additives to form green compacts that are subsequently sintered. By far, the most popular consolidation technique in PM is uniaxial compaction where a die cavity is filled by gravity with a given quantity of powder, which is compacted by precise kinematics of punches and the die itself. Powder flow during die fill is an important aspect of the consolidation process as it significantly influences apparent density of the powder mix, fill uniformity, density gradients within green compacts and ultimately mechanical properties of sintered components.

One of the most popular metallurgical systems used in the PM industry is based on the Fe-Cu-C system. More specifically, alloy FC-0208 according to Metal Powder Industries Federation Standard 35 [1]. This alloy is obtained by typically admixing 2 wt-% of copper and 0.8 wt-% of graphite to an iron powder. Die compaction requires lubrication to assist densification as well as lowering the shearing forces generated during ejection. Typically, there are two lubrication approaches that are used in press-and-sinter powder metallurgy: admixed lubrication and die-wall lubrication. In the case of the former, lubricant under the form of particles is admixed with the base powder and its additives to obtain a homogeneous dispersion of lubricant particles throughout the powder mix [2]. Die-wall lubrication refers to the application of a coat of lubricant at the surface of the compaction tooling prior to die fill [3–4]. When talking about particle flow, it is important to make the distinc-

when talking about particle flow, it is important to make the distinction between adhesion and friction. Adhesion stems from the forces present when there is a contact between two surfaces. Friction relates to the force that limits or prevents tangential movement of particles. In the case of PM premix flow, we are seeking conditions that minimize both friction and adhesion. Flow of particulate materials can be sensitive to the presence of inter-particle forces that may sometimes exceed the gravitational force and therefore prevent particle movement. The importance of these inter-particle forces is influenced by particles size, their morphology, their chemistry and relative humidity in their surroundings. The inter-particle forces that are most frequently encountered are Van der Waals, hydrogen bridges, capillary and electrostatic. In the case of Van der Waals forces, Hamaker stated that the force of









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adhesion between two base particles having a diameter D_1 and D_2 , where $D_1 \neq D_2$, is given by:

$$F_{VdW} = AD/(12z_o^2) \tag{1}$$

Where:

- D $D_1D_2/(D_1 + D_2);$
- A Hamaker constant, which is a property of the material. In the case of dissimilar materials A is replaced with $A_{12} = \sqrt{(A_{11} A_{22})}$;
- Z_0 separation distance between the two particles μ m.

Thus, introduction of a smaller intermediate particle of diameter d between the two larger ones increases the separation distance leading to a decrease of the Van der Waals force (Ref. Fig. 1a) Thus, Eq. (1) becomes:

$$F'_{VdW} = AD/12(2z_0 + d)^2$$
 (2)

Adding the forces between the two original particles and the intermediate one we obtain:

$$F''_{VdW} = A(D'_1 + D'_2) / (12z_o^2)$$
(3)

Which leads to:

$$\begin{split} F^{\text{Total}}{}_{VdW} &= F'_{VdW} + F''_{VdW} = AD/12(2z_o + d)^2 \\ &\quad + A(D'_1 + D'_2)/(12{z_o}^2) \end{split} \tag{4}$$

Eq. (4) and Fig. 1b show that Van der Waals inter-particular forces decrease with an increase of the diameter of the intermediate particle



Fig. 1. a) Schematic representation of the arrangement of a smaller intermediate particle "d" lodged between two larger particles and b) the effect of the diameter of the intermediate particle on the Van der Waals Forces. $D_1 = 100 \,\mu\text{m}$, $D_2 = 80 \,\mu\text{m}$, $Z_0 = 5 \,\text{\AA}$ and the Hamaker constant for iron is 300z] [6].

up to a critical value. Therefore, it is possible to minimize Van der Waals forces between base particles by adding intermediate particles that have a diameter equal to this critical diameter. This diameter can be calculated providing that the Hamaker coefficients are known. Results by Meyer and Zimmermann [5] indicate that the propensity of nanoparticles to reduce Van der Waals forces is practically independent of their chemistry. Nevertheless, their method of production, which may influence their size and their tendency to form agglomerates, may bring about differences of efficiency. Another important factor related to the utilization of intermediate particles to lower the Van der Waals forces between larger particles is the spatial distribution of the former. Indeed, if the covering of intermediate particles on the surface of the larger ones is not continuous, the number of contacts between larger particles will increase, thus increasing the Van der Waals forces and the cohesiveness of the blend. Similarly, the potential agglomeration of intermediate particles will have the same effect as an improper covering.

Hydrogen bridges may form when given chemical species are found at the surface of particles in intimate contact. This type of bond takes place typically between proton-donor and proton-receiver molecules [7]. Hydrogen bridges may form bonds that are 10 times stronger than Van der Waals cohesive forces. Veregin [8] established a relationship to evaluate the force of hydrogen bridges that form at the surface of starch particles:

$$F_{\rm H} = 2\pi L \rho_{\rm H} f_{\rm H} r \tag{5}$$

Where:

FF

 $\begin{array}{ll} r & radius \ of \ curvature \ of \ particles. \\ 2L & length \ of \ hydrogen \ bridge. \\ \rho_{H} & surface \ density \ of \ hydrogen \ bridges. \end{array}$

 f_H force of one hydrogen bridge.

Thus, using intermediate particles, it is possible to increase the distance between the base particles and minimize the formation of hydrogen bridges. Similarly, the presence of intermediate particles can reduce the number of OH groups at the surface of base particles [5,9,10].

One of the most popular admixed lubricants used in PM is ethylene bis stearamide (EBS) ($C_{38}H_{76}N_2O_2$). Fig. 2 shows a schematic representation of the EBS molecule. It can be seen that this molecule contains sites for potential hydrogen bridges. Thus, it can be assumed that an addition of small particles with a chemistry that minimizes the formation of hydrogen bridges could significantly improve powder flow. Therefore, in the case at hand, it is interesting to consider hydrophobic silica fumes as a potential flow-enhancing additive. Indeed, the latter has much less SIANOL groups at its surface than hydrophilic silica, making it less prone to the formation of hydrogen bridges.

Capillary forces constitute another factor that may influence negatively flow of particulate materials. These forces stem from the presence of humidity within the atmosphere surrounding neighbouring particles. According to McFarlane [11] and Fisher [12], the capillary force exerted on two spherical particles of identical diameter is given by Eq. (6).

$$H = 2\pi\gamma_{\rm L} R \cos\Theta \tag{6}$$



Fig. 2. Schematic representation of a molecule of ethylene bis stearamide (EBS).

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