

# UNDERSTANDING HOMOGENEITY OF POWDER-POLYMER MIXTURES—EFFECT OF MIXING ON TUNGSTEN POWDER INJECTION MOLDING FEEDSTOCK

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## **ABSTRACT**

Binder-assisted processing techniques are widely used in the fabrication of metal and ceramic components from powders. Central to the success of the processing techniques is the ability to obtain homogeneous mixtures. The present study investigates the effect of mixing technique on the homogeneity of tungsten based feedstock. Experiments were conducted with as-received (agglomerated) and rod-milled (deagglomerated) tungsten powder mixed in wax-polypropylene binder system. Increase in the shear rate during mixing decreased the size of the agglomerate particles of as-received tungsten powder, enhancing the homogeneity of the feedstock. Higher solids loading, lower mixing torques and improved homogeneity were observed with deagglomerated tungsten powder, emphasizing the importance of particle characteristics and mixing procedures in powder injection molding process.

## **KEY WORDS**

powder injection molding, homogeneity, mixing

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## **INTRODUCTION**

The importance of particulate processing ranges across many fields and has a large economic impact in many segments of industry. Powder injection molding is a near-net shape processing technique that permits the manufacture of complex parts using ceramic or metallic powder. The end product is a result of compounding a thermoplastic binder and powder followed by injection molding, binder removal and sintering. Mixing of powder and binder is an important phase in this process, as the uniformity of the mixture influences the flow behavior of the feedstock and the sintered properties [1]. The focus of the present work is to evaluate the effect of mixing on the homogeneity of tungsten feedstock.

Factors that influence the feedstock homogeneity include processing variables such as speed and time of mixing, geometry of mixing blades, raw material feed rate, processing temperature and material variables such as particle characteristics, solids loading, binder composition and viscosity. Typical particle sizes encountered in powder injection molding (PIM) are in the range of 0.1  $\mu\text{m}$  to 15  $\mu\text{m}$ , which can interact with each other and form clusters, generally referred to as agglomerates.

The presence of agglomerates alters the flow behavior of the feedstock [2-4] and causes defects in the sintered body [5]. Obtaining a homogeneous feedstock involves a reduction in the scale and frequency of the agglomerates. Studies on agglomerate dispersion pertaining to PIM have generally been conducted on ceramic systems such as alumina and titania [6-9]. Agglomerates in these systems are a result of surface interactions and Van der Waal's forces. Deagglomeration of ceramic powder is caused by a combination of rupture and erosion during mixing [10]. Few studies have been conducted on the mixing of cohesionless metal powders [1,11]. In these cases, homogeneity is predominantly due to distributive mixing. However, there are no reported studies on the mixing behavior of tungsten. Agglomerates of this material are formed during the powder production stage and constitute particles that are welded or fused together. In practice, deagglomeration of tungsten is accomplished by mechanical attrition during the rod milling process.

It is assumed that the agglomerates are broken down by hydrodynamic forces during mixing, resulting in dispersion of tungsten powder in the polymer. The viscosity of the feedstock is influenced by the presence of undispersed agglomerates [3,4,12,13] and is considered a good indicator of feedstock homogeneity [2,14,15]. In this study homogeneity is qualitatively estimated by viscosity measurements.

## **EXPERIMENTAL PROCEDURE AND DISCUSSION**

The material used in this experiment was a feedstock of 97%W-2.1%Ni-0.9%Fe prepared using as-received and rod-milled tungsten powder. The powder characteristics are given in Table 1. The particle size distribution was measured using Horiba LA-920 laser scattering particle analyzer and the pycnometer density in a Micromeritics Pycnometer. The tap density was obtained using a Dual Autotap Quanta Chrome equipment and the apparent density followed the ASTM B 703 [16] standard test. The rod-milled powder was prepared by milling the as-received powder with tungsten rods in a plastic jar backfilled with argon. The powders morphology observed by scanning electronic microscopy (SEM), is given in Figures 1 (a) – (d).

The powders were weighed to the desired composition and blended in a Turbula mixer for 30 minutes. Wax-polypropylene was used as the binder system. The feedstock was evaluated for its critical solids loading in a Haake torque rheometer, which was also used as low intensity mixer. The feedstock mixing temperature used was 170 °C. The variation in mixing torque with solids loading is given in Figure 2. The as-received powder exhibits lower critical solids loading ( $\phi_c = 63\%$ ) and higher mixing torque for a given solids loading compared to the rod milled powder ( $\phi_c = 66\%$ ). The error bars in the graphs represent standard deviation in the torque during mixing. The presence of agglomerates in the as-received powder hinders effective packing [12] decreasing the critical solids loading content [3,4]. The viscosity of a suspension varies with the ratio of the solids loading to the maximum packing fraction and can be related as:

$$\eta_m = \eta_b (1 - \phi / \phi_c)^{-2} \quad (1)$$

where  $\eta_m$  is the mixture viscosity,  $\eta_b$  is the binder viscosity,  $\phi$  is the powder solid loading fraction and  $\phi_c$  is the critical powder solid loading fraction.

An increase in the factor  $\phi / \phi_c$ , due to the presence of agglomerates, in the present case, increases the viscosity of the feedstock. The measured torque is proportional to the viscosity of the feedstock.

The hydrodynamic force ( $F_h$ ), that is, the driving force for agglomerate rupture, given as,

$$F_h = \eta \times \gamma^* \quad (2)$$

where  $\gamma^*$  is the shear rate, increases with an increase in the viscosity of the feedstock and shear rate during mixing. Hence, experiments were conducted to observe the effect of change in the viscosity of the feedstock on the morphology of the agglomerates. The viscosity of feedstock, mixed at the same torque rheometer conditions (180 RPM for two hours), increased with solids loading, as shown in Figure 3. The morphology of the powder was observed using a small sample obtained from feedstock that was debound in a retort furnace at 600 °C in hydrogen for one hour. No discernable change in the particle morphology was observed as evident from Figures 4 (a) and (b). The powder shows significant agglomeration indicating negligible effect of torque rheometer mixer on deagglomeration. It was concluded that the hydrodynamic force due to the low intensity torque mixer was insufficient to induce agglomerate rupture.

High shear rate mixing conditions were realized using Readco continuous mixer. In some cases, feedstock discharged from the mixture was re-introduced into the mixing chamber, retrieving sample in each iteration. Viscosity variation of the feedstock, prepared with as-received and rod-milled powder respectively, with shear rate under different conditions, is shown in Figure 5. It can be seen that the viscosity of feedstock composed of as-received powder decreased with additional mixing iterations. It is also evident that for a given solids volume fraction, an increase in the mixing intensity, represented by shear rate x mixing time, decreased the viscosity and enhanced the flow stability, as given in Figure 6. This decrease in viscosity is attributed to the reduction in agglomerate size. As stated previously, decrease in the scale and frequency of agglomerates enhances the packing fraction. An increase in the mixing intensity makes the viscosity of feedstock composed of as-received agglomerated tungsten powder approach that of deagglomerated, rod-milled tungsten powder. The observed decrease in the viscosity is an indicator of enhanced homogeneity [14]. The effect of shear rate during mixing is also apparent from Figure 7, which shows the viscosity variation with shear rate for 60 volume percent tungsten

feedstock mixed in low intensity torque rheometer and high intensity continuous mixer. The change in the particle morphology is evident from Figure 8, which shows a marked difference in the scale of the agglomerates compared to the as-received powder shown in Figure 1 (a).

Particle size distribution was measured to quantify the change in the agglomerate size due to different mixing conditions. Table 2 summarizes the findings of these studies, where the feedstock was repeatedly mixed in the Readco continuous mixer and powder samples were debound, as described previously, for analysis. The results indicate that there is a difference in particle size distribution, with a decrease in the mean particle size as mixing increase. However, the extent of particle size reduction is not substantial, contrary to the morphological observations given in Figures 1, 4 and 8. The quantification of particle size reduction is being investigated [17].

Table 1 - Characterization of as-received and rod milled tungsten powder.

	Apparent density (g/cm <sup>3</sup> )	Tap Density (g/cm <sup>3</sup> )	Helium Pycnometer Density (g/cm <sup>3</sup> )	Particle Size (μm)		
				D <sub>10</sub>	D <sub>50</sub>	D <sub>90</sub>
As Received Powder	5.36	7.17	19.23	D <sub>10</sub>	D <sub>50</sub>	D <sub>90</sub>
				8.17	16.54	35.42
Rod Milled	7.62	9.9.0	19.23	4.08	8.89	14.82
Iron	2.43	4.63	7.80	2.9	5.9	9.9
Nickel	2.30	3.26	8.98	4.0	9.9	23.5

Table 2 – Feedstock size distribution at high shear rate conditions. Powder was obtained using a small sample from feedstock that was debound in a retort furnace at 600 °C in hydrogen for one hour.

	D <sub>10</sub> μm	D <sub>50</sub> μm	D <sub>90</sub> μm
<b>As-received feedstock–Torque Rheometer (180 RPM/ 120min), φ: 60%</b>	6.7	14.0	82.2
<b>As-received feedstock–Torque Rheometer (180 RPM/ 120min), φ: 63%</b>	7.2	15.4	75.5
<b>As-received feedstock (first iteration) Readco continuous mixer, φ: 60%</b>	6.0	13.8	83.0
<b>As-received feedstock (sixth iteration) Readco continuous mixer, φ: 63%</b>	6.0	13.4	73.0

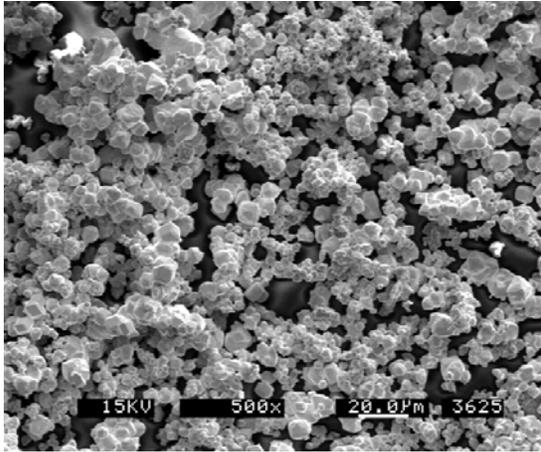


Figure 1(a) - Morphology of as-received tungsten powder observed by SEM, revealing the agglomerate structure.

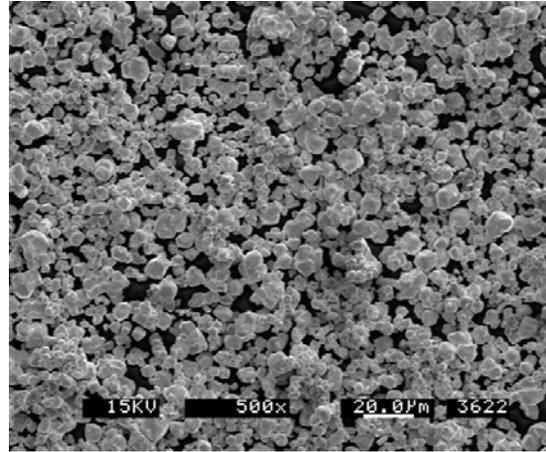


Figure 1(b) - Morphology of rod-milled tungsten powder observed by SEM, revealing the presence of a deagglomerated powder.

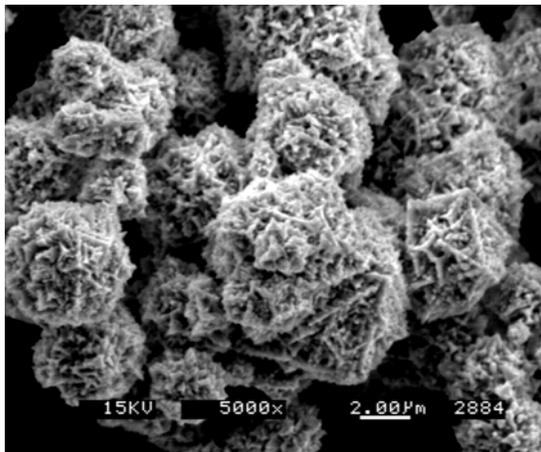


Figure 1 (c) - Morphology of nickel powder observed by SEM.

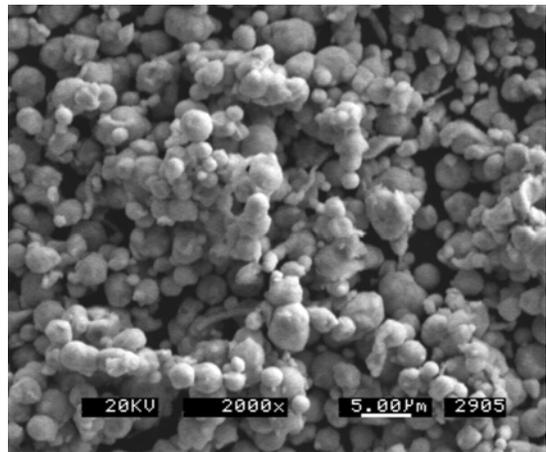


Figure 1 (d) - Morphology of iron powder observed by SEM.

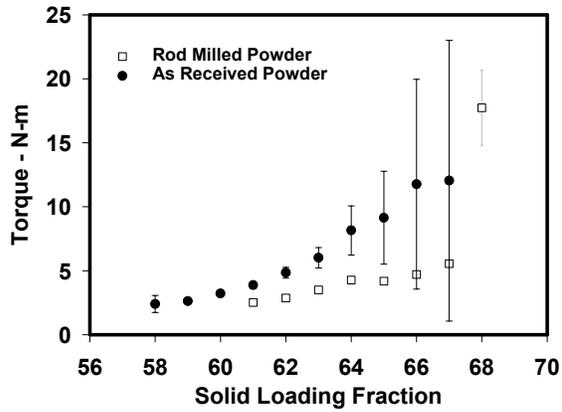


Figure 2 - Mixing torque variation with solids loading for feedstock composed of rod-milled and as-received powder. The critical solids loading for as-received powder is estimated at 63% and 66% for rod-milled powder.

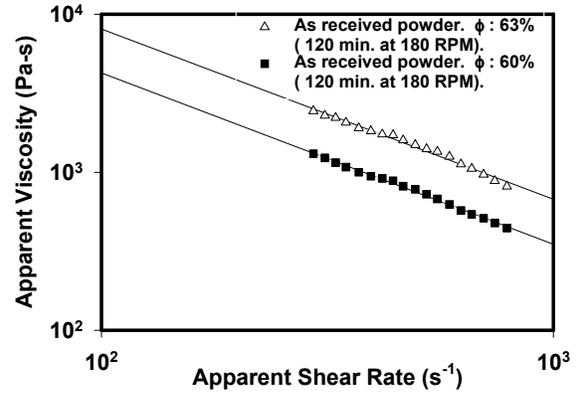


Figure 3 - Variation in the apparent viscosity with shear rate for feedstock of 60 and 63% solids loading. The mixing experiments were conducted in torque rheometer at 180 RPM during 120 min.

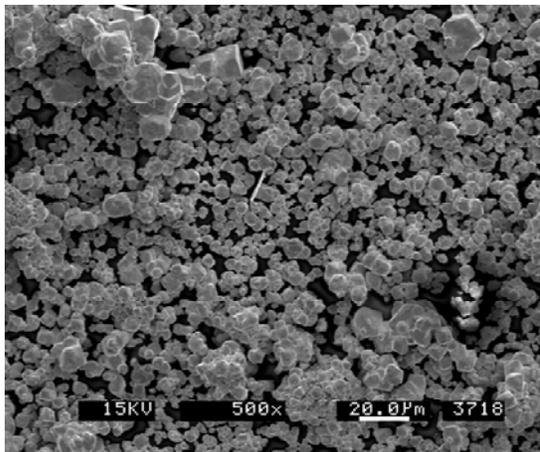


Figure 4 (a) - Particle morphology of the feedstock ( $\phi = 60\%$ ) after mixing at 180 RPM for 120 min. in a torque rheometer.

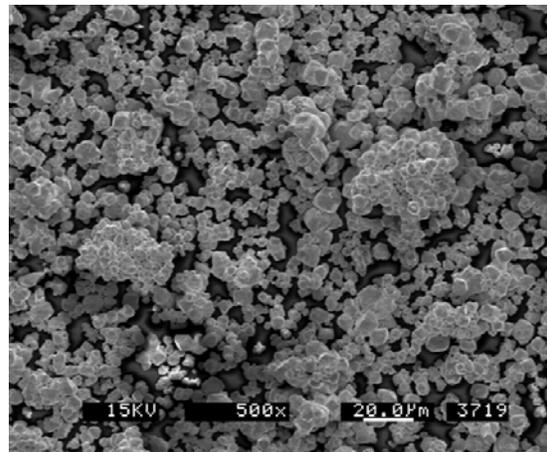


Figure 4 (b) - Particle morphology of the feedstock ( $\phi = 63\%$ ) after mixing at 180 RPM for 120 min. in a torque rheometer.

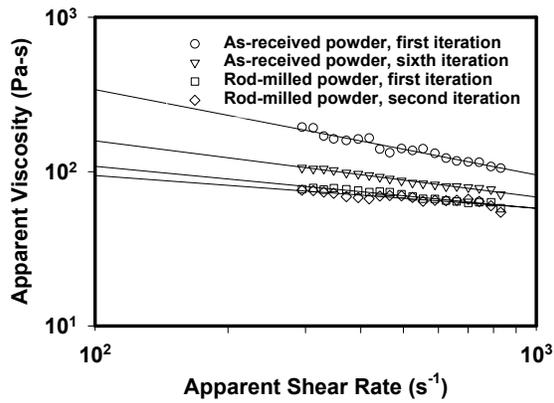


Figure 5 - Apparent viscosity versus apparent shear rate obtained for a 60% solid loading feedstock mixed in continuous mixer.

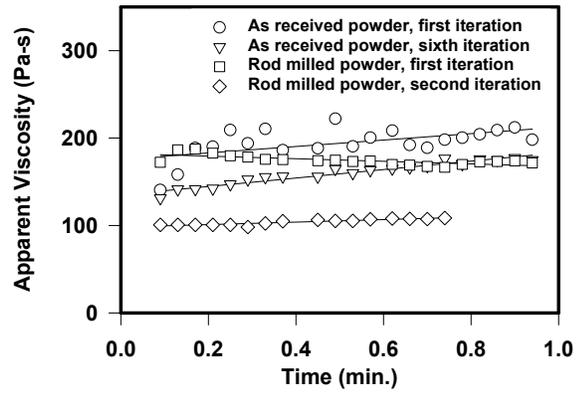


Figure 6 - Apparent viscosity versus time for 60% solid loading feedstock obtained at constant shear rate ( $294.53 \text{ s}^{-1}$ ) at Readco continuous mixer.

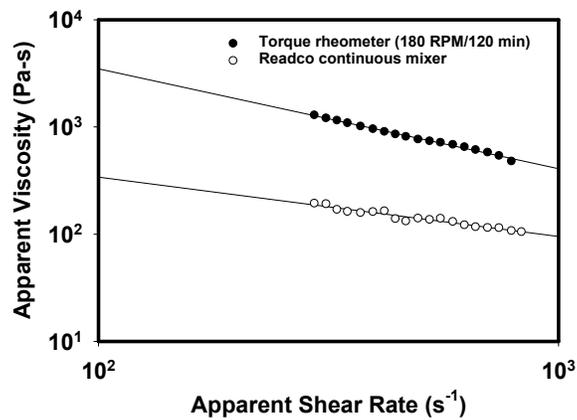


Figure 7 - Apparent viscosity versus apparent shear rate for a 60% solid loading feedstock, composed of as-received powder, mixed in the torque rheometer and Readco continuous mixer respectively.

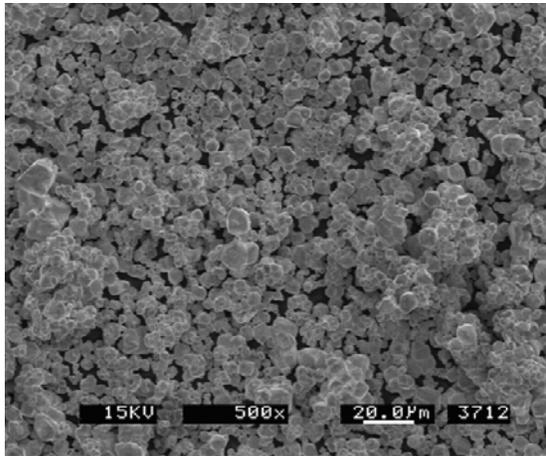


Figure 8 (a) – Powder morphology obtained from the debound feedstock ( $\phi = 60\%$ ) after first iteration in Readco continuous mixer exhibiting reduction of agglomerate size.

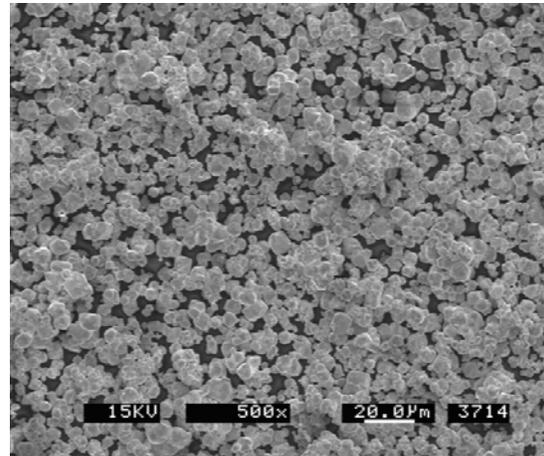


Figure 8 (b) - Significant reduction of feedstock agglomerate size after sixth iteration at Readco continuous mixer, compared with the same condition of Figure 8 (a)

## **CONCLUSIONS**

Experiments were conducted with as-received, agglomerated tungsten powder and rod milled tungsten powder to study the effect of mixing on the homogeneity of the powder injection molding feedstock. An increase in the mixing shear intensity increases the hydrodynamic force and contributes to the agglomerate size reduction. Deagglomeration during mixing improves the packing efficiency of the particles promotes homogeneity. Viscosity is a good indicator of the homogeneity of the feedstock. An increase in the homogeneity of the feedstock is reflected as a decrease in its viscosity. A perfectly homogeneous mixture will have the least viscosity. It is observed that with an increase in the intensity of mixing, the viscosity of the feedstock composed of as-received powder approaches that of the deagglomerated, rod milled powder.

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