Binding Mechanism and Shrinkage Prediction of Polypropylene, Polyethylene and Silica Particles in Isothermal Sintering Process

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Abstract

Combination between sintering process and automation system has proven to be a manufacturing process that can produce mechanical parts with high geometric complexity. However on the development of various materials, varying material in one part has not been studied by many researchers. In addition the uniformity of strength and shrinkage aspects are problems that have not been perfectly solved. Based on these reasons, research of binding mechanism and the shrinkage prediction of particles on sintering process are importantly conducted.

In this research, binding mechanisms were observed in isothermal sintering. Materials were used polypropylene, polyethylene and silica, while the occurring shrinkage prediction used polypropylene material. The images of in situ sintering process were observed by optical microscope camera with magnitude of 100x. Based on these images data, the equivalent diameter can be determined with projection area method and then, the shrinkage prediction can be calculated.

The results show that the mechanism of sintering of polypropylene and polyethylene is solid state sintering. During sintering, the expansion of dimension of polyethylene particle is greater than that of polypropylene. In sintering process between polyethylene and silica, the lower melting temperature particle (polyethylene) is as the driving binding among silica particles. From shrinkage prediction, the final stage of polyethylene sintering at temperature of 1100°C and holding time of 780 s yields shrinkage of 20.47%.

Keywords: binding mechanism, shrinkage, sintering, polyethylene, polypropylene

1. Introduction

From a technological viewpoint, dimensional control is one of the most important practical problems in liquid phase sintering. The occurring shrinkage prediction during sintering is important so that the final dimension of product can be controlled. Modelling of sintering process is a method to solve this problem. Shimosaka et al (2003) made a modelling for 2 type material. In their research, they presented that the sintering behaviour of 2 type material was not only influenced by solution property of both materials but also by sintering conditions.

The existing of solid solution is in grain boundary area with transport mechanism which consists of surface diffusion, volume diffusion, grain boundary diffusion, evaporation-condensation, and grain growth. Maximenko and Olevsky (2004) proposed a constant parameter which was called an effective coefficient diffusion that was aimed to ease in modelling of sintering. This constant was a combination between grain boundary diffusion and volume diffusion. With using this coefficient, the
modelling of sintering can be solved in 2 dimensions.

In sintering process, prediction of shrinkage phenomenon can be easily done by experiment. Hambrir and Jog (2000) presented that a specimen, which was resulted by low density green part, would reveal a specimen with higher shrinkage than a specimen which was resulted by higher density. This matter was used as a base research of Garino et al (1995) who conducted research about shrinkage un-uniformity in multi material sintering. With sintering of 2 pieces green-parts, which had different density, shrinkage un-uniformity can be revealed. Their experimental results showed that the occurring shrinkage was determined by the layer dimension of each green-part.

In this paper, with using an optical microscope, binding mechanism in liquid phase sintering between polyethylene and PVC particle was periodically observed. These images data were used to calculate the occurring shrinkage during sintering with projection area method.

2. Fundamental

Based on sintering mechanism, the sintering process is classified into 3 types of sintering, namely: vapour phase sintering, solid state sintering and liquid phase sintering. The transport mechanism of vapour phase sintering is driven by evaporation-condensation process, while solid state sintering and liquid phase sintering are diffusion and viscous flow respectively.

Solid state sintering (SSS)

Based on shrinkage phenomenon, the sintering process is a changing geometric interval in which the pore dimension decreases. According to its definition, sintering process consists of three stages, namely initial stage, intermediate stage and final stage. During the initial stage, point contact of particles usually increases and the relative density increases from 60 to 65 percent (Barsoum, 1997). In the point contact, cohesive necks grow in atomic solid state condition under melting point temperature and in bonding microstructure scale (German, 1994).

Intermediate stage is signed by continuing of joining particles to shape the continuous pore channels, the relative density increases from 65 to 90 percent and pore starts loosing from cylindrical channels. While in the final stage, continuous pore channels loose and change to individual pore (Barsoum, 1997). The stages of sintering process are presented in Figure 1 below:

![Figure 1](image1)

Figure 1. a) initial stage of sintering process that presents a point contact of accompany particles, b) the final of initial stage that signed by obtaining of neck growth, c) intermediate stage in which the particle contact continued and obtained continuous pore channels, d) final stage, pore was obtained on contact angle of accompany particle (Barsoum, 1997)

The simple modelling of binding mechanism between the same types of two particles is presented in Figure 2 below:

![Figure 2](image2)

Figure 2. Modelling binding mechanism of two particles which have the same dimensions in sintering process
In the end of intermediate stage possibly occurs the particle growth, therefore would reveal the particles with higher dimension but in the less number. In the final stage the particle growth with densification process slowly occurs.

Binding mechanism of this particle (Figure 2) is driven by transport mechanism which is categorized into two types, namely surface transport and bulk transport. The surface transport consists of surface diffusion and evaporation-condensation, while the bulk transport consists of volume diffusion, grain boundary diffusion, plastic flow and viscous flow. Surface transport yields the neck growth without shrinkage.

The driving force of sintering process is a form of surface energy in which the magnitude per unit volume increases with decreasing of particle diameter. During the sintering process, mass transport occurs from particle to the neck. According to the balancing law, mass transport aims to decrease the surface energy of particle which is run by increasing of particle surface area, such that, during the sintering process, surface energy decreases.

**Liquid phase sintering (LPS)**

In LPS system, the liquid may provide for rapid transport and therefore rapid sintering if certain criteria are met (Lenel, 1980). The liquid must form a film surrounding the solid phase. Thus, wetting is the first requirement. Secondly, the liquid must have solubility for the solid. Finally, the diffusive transport for the solid atoms dissolved in the liquid should be high enough to ensure rapid sintering (Huppmann, 1975). In the liquid phase sintering, the densification rate is much faster than in solid state sintering, and times as short as 15 min at the maximum temperature can be successful in producing full density (German, 1996).

On liquid phase sintering, shrinkage gradients in a compact sintered powder traditionally attributed to gradient in green density. Reduced shrinkage during sintering at the bottom of the compacts is attributed to the friction between the compacts and the substrate material (Gurland, 1962).

Densification and shape distortion during liquid phase sintering depend on the driving force and the resistance to viscous deformation (German, 1996). Liu et al (1999) presented that the capillary force was used the driving forces of densification and surface tension and gravitational were used the driving force of shape distortion.

**Equivalent diameter**

Practically, particle powders have shape variations. For irregular shape powders, calculations of their volume commonly use an equivalent volume, while it is calculated by using equivalent diameter. One of the methods, which can be used to determine an equivalent diameter, is projected area method. If the particle is assumed in ball geometry, the equivalent diameter can be calculated with Equation 1.

\[ D_A = (4A/\pi)^{1/2} \]  

(1)

So that the equivalent volume of particle is calculated with:

\[ V = \frac{4}{3} \pi D^3 \]  

(2)

![Figure 3. Projected image of an irregular particle.](image)

**3. Methodology**

**Properties of polypropylene and polyethylene**

This research used polypropylene, polyethylene and silica sand as sintering materials. These Polypropylene and polyethylene are made by PT. Tri Polya Indonesia. Their properties are presented in Table 1.
Observation of binding mechanism and shrinkage prediction

Observations of binding mechanisms consist of single material and multi material. In single material, sintering process is conducted between polypropylene and polypropylene particles; while in multi material the sintering is carried out between polypropylene and polyethylene, and silica sand with polyethylene particles. The nominal diameter of particle is around 2 mm. Observation of binding mechanism is conducted on isothermal condition. The set-up of this experiment is presented in Figure 4.

4. Result and Discussion

Binding mechanism of single material

Binding mechanism of single material used polypropylene materials and the sintering process was conducted at 105°C. The binding mechanisms during sintering between two polypropylene particles are presented in Figure 5:

According to Figure 5 above, the occurring binding mechanism is a form of solid state sintering. During sintering, each particle periodically experienced the same dimensional changing.

Binding mechanism of multi material

According to Table 1, the particles of polypropylene and polyethylene have a different of melting point of 35°C. This possibly makes a binding mechanism with solid state sintering. Figure 6 shows a binding mechanism between polypropylene and polyethylene at sintering temperature of 105°C.

Table 1. Properties of polypropylene and polyethylene

<table>
<thead>
<tr>
<th></th>
<th>Polypropylene</th>
<th>Polyethylene</th>
<th>Silica*)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical formula</td>
<td>(C₃H₆)ₓ</td>
<td>(C₂H₄)ₓ</td>
<td>99% SiO₂</td>
</tr>
<tr>
<td>Density</td>
<td>0.85 gr/cm³</td>
<td>0.93 gr/cm³</td>
<td>2.65 gr/cm³</td>
</tr>
<tr>
<td>Melting point</td>
<td>173°C</td>
<td>138°C</td>
<td>1830°C</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>1200 to 11600 psi</td>
<td>1400 to 3010 psi</td>
<td>55 MPa</td>
</tr>
</tbody>
</table>

*) www.azom.com
Figure 6. Binding mechanism of polypropylene and polyethylene (dark) at sintering temperature of 105°C.

Figure 6 shows that the expansion of dimension of polyethylene is faster than that of polypropylene. This is caused by the melting point of polyethylene is lower than that of polypropylene.

If two types of particles which have big different of melting point are sintered, the binding mechanism is as liquid phase sintering. Figure 7 shows a binding mechanism between silica sand and polyethylene.

Polyethylene has a melting temperature of 130°C, this is lower than that of silica sand. From Figure 7, the viscous flow of polyethylene bound two silica sand particles around it.

Figure 7. Binding mechanism between silica (white) and polyethylene (dark) at sintering temperature of 95°C.

Figure 8. Transport mechanism in liquid phase sintering between silica sand and polyethylene, a) initial condition, b) final stage

Shrinkage prediction

According to the image data in Figures 6, 7 and 8, shrinkage predictions could produce errors of interpretations. During sintering, the particle shape was not in ball shape and it was glued to the base plate, so that the diameter of image (projection view) was greater than the actual shape.

Figure 9. Transformation of particle shape during heating process for particle size of around 2mm and un-polishing base plate surface. a) Ball particle shape before heating process, b) half ball particle shape after heating process.

Using a small particle and improvement of surface condition of base plate are possibly conducted to solve this problem. An observation of particle size of 200 µm and polishing of surface plate, the image data from sintering process at 110°C is showed in Figure 10:
Figure 10. Binding mechanism of polyethylene and polyethylene at sintering temperature of 110°C

From the images data in Figure 10, the equivalent diameter can be calculated with projected area method. The calculation results of equivalent diameter and shrinkage prediction are presented in Table 2.

Table 2. Calculations of equivalent diameter and shrinkage prediction of polyethylene on sintering process at 110°C.

<table>
<thead>
<tr>
<th>Holding Time (Minutes)</th>
<th>Projected area (mm²)</th>
<th>Equivalent diameter (mm)</th>
<th>Volume (mm³)</th>
<th>ΔV/V₀</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1853.23</td>
<td>48.69</td>
<td>480239.60</td>
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</tr>
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<td>7</td>
<td>1804.77</td>
<td>47.95</td>
<td>461526.68</td>
<td>-3.90</td>
</tr>
<tr>
<td>8</td>
<td>1743.15</td>
<td>47.12</td>
<td>431927.72</td>
<td>-8.78</td>
</tr>
<tr>
<td>9</td>
<td>1614.00</td>
<td>45.34</td>
<td>390310.65</td>
<td>-18.72</td>
</tr>
<tr>
<td>10</td>
<td>1605.66</td>
<td>45.23</td>
<td>397297.13</td>
<td>-19.35</td>
</tr>
<tr>
<td>11</td>
<td>1600.50</td>
<td>45.15</td>
<td>395431.69</td>
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<tr>
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<td>46.10</td>
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<tr>
<td>13</td>
<td>1590.74</td>
<td>45.02</td>
<td>381911.47</td>
<td>-20.47</td>
</tr>
</tbody>
</table>

5. Conclusions

In liquid phase sintering, viscous flow of a lower melting temperature particle is as the driving binding among the higher melting temperature particles.

In this case, particle size and surface condition of the base plate affect the result of equivalent diameter calculation. Shrinkage prediction with image calculation of equivalent diameter method can be accurately conducted if the particles size and surface condition of the base plate are small enough (200 μm) and polished respectively.

The final stage of polyethylene sintering at temperature of 110°C and holding time of 780 s yields shrinkage of 20.47%.

Reference


