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SOLVENT DEBINDING PROCESS FOR MIM COMPACTS**

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Application of RSM Technique to Optimize the Solvent Debinding Process for MIM Compacts

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ABSTRACT

Extraction of polyethylene glycol (PEG) from metal injection molding compact has been investigated at various powder loadings, leaching temperatures and times, referred as independent variables. Central composite rotatable design (CCRD) of experiments is used to study the effect of these independent variables on the brown body's strength, or a dependent variable. This paper reports the use of Response Surface Methodology (RSM) technique to optimize the processing factors for solvent debinding gas atomized 316L stainless steel (of average size 11.2 μm -fine and 19.6 μm -coarse) for maximum brown strength. For fine powder (11.2 μm), to achieve maximum brown strength of 5.6 MPa, the optimised values were as follow; powder loading 64% volume, debinding temperature of 60°C and debinding time at 2 hours. Meanwhile from the optimized values of powder loading (64% volume), debinding temperature (59°C) and debinding times (3 hours), the maximum brown strength for coarse powder obtained was 5 MPa. This protocol provides improved opportunities for solvent debinding techniques particularly to extract PEGs from the moulded components mathematically without experimentation.

Keywords: Solvent debinding, PEG, RSM.

1.0 INTRODUCTION

Metal injection molding (MIM) is a novel process, which combines plastic injection molding and conventional powder metallurgy technologies. MIM process comprises of four steps: mixing, injection molding, debinding and sintering. Among these processing steps, debinding is the critical step that determines the success of the whole process. Failure to remove most of the binder before sintering can result in component distortion, cracking and contamination (German & Bose 1997). The debinding process is often carried out either by thermal or solvent processing or by a combination of these processes. To shorten the overall debinding cycle, a major fraction of binder is removed by solvent extraction followed by thermal debinding. The green components are immersed in a suitable organic fluid, which dissolves the binder partially and this results in the formation of open pore channels in the body. Through these channels, the remaining binder fraction can be removed in a much shorter time.

The powder used in this study is 316L stainless steel, one of the most widely used materials for MIM research and industrial applications. Since 316L stainless steel is a highly alloyed material, the mechanical and corrosion properties are a concern (Ji et al. 2001). Therefore a high brown strength is important for optimization of the water leached specimens, so called brown body. Previous studies (Zu & Lin 1997, White & German 1993) have shown that the important factors of the debinding steps are: debinding temperature, debinding time and powder loading. An understanding of the effects of the debinding factors on the brown strength can be used to optimize the debinding process.

The traditional approach to experimental work is to vary one factor at a time, holding all factors fixed. This method does not produce satisfactory results in a wide range of experimental settings. In MIM process investigations, statistical design of experiment (DOE) is used quite extensively. Statistical DOE refers to the process of planning the experiment so that the appropriate data can be analysed by statistical methods, resulting in valid and objective conclusions (Noordin et al. 2004). Design and methods such as factorial design, response surface methodology (RSM) and Taguchi methods are now widely used in place of one factor at a time experimental approach which is time consuming and exorbitant in cost.

Zu and Lin (1997) used the Taguchi method to study the effects of debinding factors on the mechanical properties (strength and ductility) of injection molded tungsten heavy alloys. The Taguchi method was also used by Ji et al. 2001 in characterizing and optimizing the process factors for sintering water-atomized 316L stainless steel. A 2⁴ factorial design was used by Loh and German (1996) to establish the effects PIM parameters on the shrinkage properties of Fe-Ni powder system. Meanwhile, Park and Ahn (2004) determined the optimal conditions for injection molding process of a deflection yoke by DOE approach based on factorial design considering two way interactions and have been applied for both mold and process design.

In the present study, RSM was used to determine the optimum processing conditions (debinding temperature and time, powder loading) for maximum brown strength in brown bodies after PEG extraction. These conditions will serve as a preliminary basis for conducting further studies on solvent debinding of PEGs mathematically without any experimentation.

2.0 EXPERIMENTAL METHODS

Gas atomized stainless steel SS316L powder with median size (D_{50}) of 19.606 μm (coarse powder) and 11.2 μm (fine powder) is used in this study. The powder was then added with a binder system composed by a polymethyl methacrylate (PMMA) and polyethylene glycol (PEG) according to volumetric percentage as follow: 73% PEG and 25% PMMA, while 2% of stearic acid is added to facilitate the flowability of the feedstock during molding stage. The

green body is injected molded using a Battenfeld BA 250 CDC machine, and then immersed in a distilled water bath according to the process variables given: five different powder loadings, 61, 62, 63, 64 and 65% volume, debinding times ranging from 2 to 6 hours, and distilled water temperature at 59, 60, 62, 64 and 65°C. The brown strength is determined by implementing three points bending test according to MPIF 41 Standard.

3.0 EXPERIMENTAL DESIGN AND STATISTICAL ANALYSIS

Based on the preliminary experimental results, the levels chosen for the independent variables, debinding temperature X_1 , debinding time X_2 and powder loading X_3 were shown in Table 1. Central composite rotatable design (CCRD) of experiments was used, where the values of independent variables were coded as the variables, x in the range of +1.682 and -1.682 levels. CCRD involves outlining the composition of the experimental process conditions around a central combination and then subsequently used to develop the regression models. For three independent variables process ($k = 3$), the design consists of factorial design points with eight runs (2^k , $k = 3$), three centre points, and six ($2k$) axial or star points. The central combination for the experimental design was as follows: debinding temperature, $T = 62^\circ\text{C}$, debinding time, $t = 4$ hours and powder loading, $P = 63\%$ volume.

Table 1 Real and coded values of independent variables in the experimental design

| Debinding temperature ($^\circ\text{C}$) | | Debinding time (h) | | Powder loading (% vol.) | |
|--|--------------------|--------------------|--------------------|-------------------------|--------------------|
| Real value, X_1 | Coded value, x_1 | Real value, X_2 | Coded value, x_2 | Real value, X_3 | Coded value, x_3 |
| 59 | -1.682 | 2 | -1.682 | 61 | -1.682 |
| 60 | -1 | 3 | -1 | 62 | -1 |
| 62 | 0 | 4 | 0 | 63 | 0 |
| 64 | 1 | 5 | 1 | 64 | 1 |
| 65 | 1.682 | 6 | 1.682 | 65 | 1.682 |

For each combination of the independent variables in the experimental design, the dependent parameter brown strength y_1 was determined. Duplication for each combination was done at 6 times. To study the effect of independent variables on y_1 (dependent variable), also for optimization of dependent variable and to check the sufficiency of the experimental design, a non linear regression equation (equation 1) was developed between dependent and independent variable for both coarse and fine powders.

$$y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_{11}x_1^2 + b_{12}x_1x_2 + b_{13}x_1x_3 + b_{22}x_2^2 + b_{23}x_2x_3 + b_{33}x_3^2 \quad (1)$$

where $b_0, b_1, b_2, b_3, b_{11}...$ regression coefficients and x_1, x_2 and x_3 are the coded values of the independent variables. The analysis of variance (ANOVA) was performed to determine the lack of fit and the effect of linear, quadratic and interaction terms on dependent variables y_1 . Response surfaces were generated by MINITAB (version 14) statistical analysis software.

4.0 RESULTS AND DISCUSSION

4.1 EFFECT OF INDEPENDENT VARIABLES ON BROWN STRENGTH USING COARSE POWDER

Table 2 presents the estimated regression coefficients of all factors and their respective values on brown strength of compacts. From the table, it shows that the insignificant factor that affected brown strength was square coefficient (temperature \times temperature) as it was the only factor with a P -value above 0.05. Thus, this factor was excluded to perform the regression analysis since it has little effect on the modal (have a high P -value). Based on this analysis, the regression coefficients for the estimation of brown strength for coarse powder, S_a were found (equation 2).

$$S_a = 4.46 - 0.15x_1 - 0.23x_2 + 0.45x_3 + 0.26x_2^2 - 0.69x_3^2 \quad (2)$$

Where S_a is the brown strength, x_1 , x_2 and x_3 are the coded values of the independent variables, debinding temperature (X_1), debinding time (X_2) and powder loading (X_3) respectively.

The regression equation indicates that the brown strength was negatively correlated with temperature and time and positively correlated with powder loading. Therefore, an increase in debinding temperature and time decreased the brown strength, whereas in increase in powder loading increased the brown strength. From equation 2 also, it was observed that the magnitude of coefficients for powder loading was larger than the coefficients for debinding temperature and time, which indicated that the powder loading had much significant effect on the brown strength than other two independent variables.

Table 2 Estimated regression coefficients for brown strength of coarse powder

| Term | Coef. | T-value | P-value |
|------------------------------------|-------|---------|---------|
| Constant | 4.46 | 35.93 | 0.00 |
| Temperature | -0.15 | -1.98 | 0.05 |
| Time | -0.23 | -2.98 | 0.00 |
| Powder loading | 0.45 | 5.68 | 0.00 |
| Temperature × temperature | 0.04 | 0.50 | 0.62 |
| Time × time | 0.26 | 3.34 | 0.00 |
| Powder loading × Powder loading | -0.69 | -8.73 | 0.00 |

Table 3 ANOVA for the regression equations of coarse powder

| Source | d.f | Seq. SS | Adj. SS | Adj. MS | F-ratio | P-value |
|-------------------|-----|---------|---------|---------|---------|---------|
| Regression | 6 | 69.686 | 69.686 | 11.614 | 25.06 | 0 |
| Linear | 3 | 25.826 | 20.912 | 6.971 | 15.04 | 0 |
| Square | 3 | 43.860 | 43.860 | 14.620 | 31.54 | 0 |
| Residual error | 104 | 48.205 | 48.205 | 0.464 | | |
| Lack-of-fit | 8 | 3.122 | 3.122 | 0.390 | 0.83 | 0.578 |
| Pure error | 96 | 45.084 | 45.084 | 0.470 | | |
| Total | 110 | 117.891 | | | | |

The analysis of variance (ANOVA) for the non-linear regression equation (equation 2) showed that the linear and square terms were significant at 5% level. The lack of fit was found not to be significant, which indicated that the number of experimental combinations formed in the design of experiment was enough to find out the effect of independent variables on the brown strength.

To study the effect of these independent variables on brown strength of molded parts, Figure 1 shows the response surface plots of brown strength for various combinations of debinding temperature, time and powder loading.

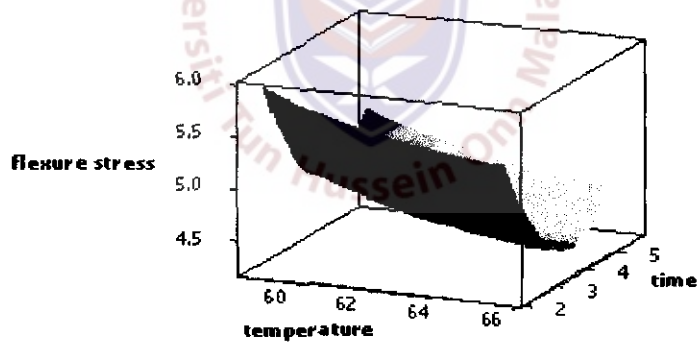
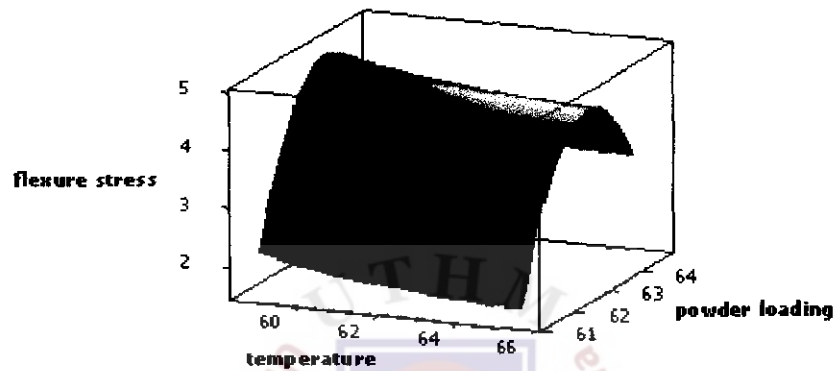
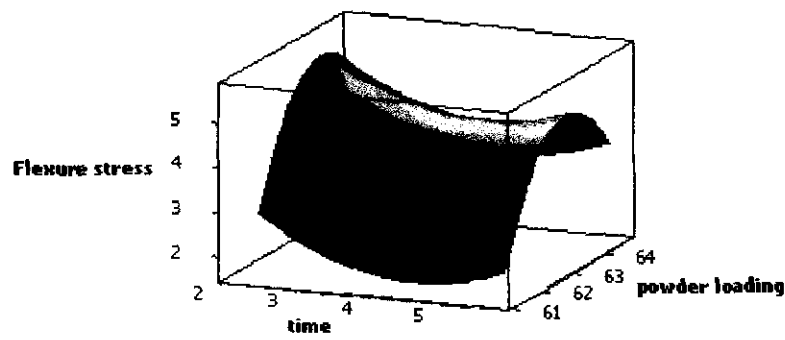


Figure 1 Response surface plots of brown strength using coarse powder

The brown strength reached the highest value in the middle level of powder loading, and did not increase thereafter and there before. Lowest value of brown strength was detected in the smallest value of powder loading (e.g. 61% volume). Similar trend was observed between the debinding temperature and time effect on brown strength. Figure 1 also showed that the brown strength value could be improved at lower debinding temperature, if the debinding time is sufficiently short. However the value could then be reduced at higher debinding temperature and longer time. After some moment of time, the value increased up to some value. The maximum brown strength was achieved in the combination of debinding temperature between 59 to 62 °C and debinding time between 2.5 to 5.5 hours, while powder

loading must exceed 64.5 % volume. Therefore, the molded parts should be injected molded at least at powder loading of 64.5 % volume for maximum value of brown strength.

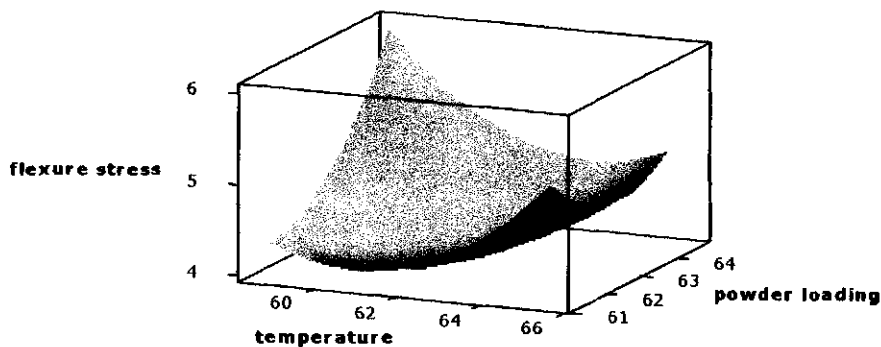
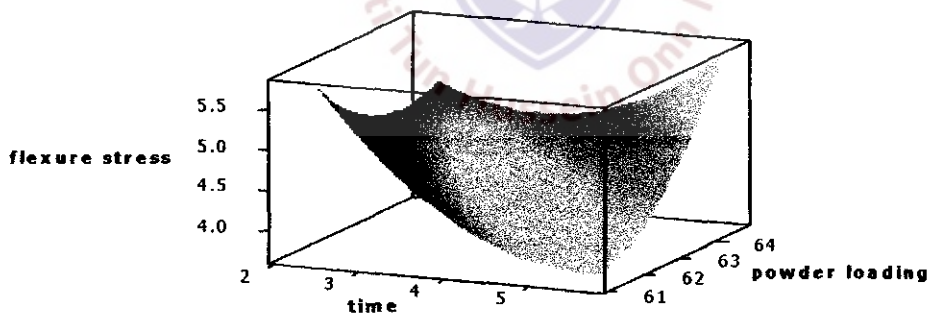
4.2 EFFECT OF INDEPENDENT VARIABLES ON BROWN STRENGTH USING FINE POWDER

From equation 3, it was clear that the brown strength was negatively correlated with debinding time and positively correlated with the powder loading, i.e., an increase in powder loading increased the brown strength. The analysis of variance (ANOVA-Table 4) for non-linear regression equation showed that the model was significant at 5% level. But the number of experimental combinations formed in the design of experiment was not enough to find out the effect of independent variables on the brown strength. This can be seen through the P value of lack of fit model which is smaller than 0.05, that indicates its significance to this model.

$$S_b = 4.14 - 0.21x_2 + 0.21x_3 + 0.21x_1^2 + 0.2x_2^2 + 0.14x_3^2 + 0.01x_2x_3 \quad (3)$$

Table 4 ANOVA for the regression equations of fine powder

| Source | d.f | Seq. SS | Adj. SS | Adj. MS | F-ratio | P-value |
|----------------|-----|---------|---------|---------|---------|---------|
| Regression | 8 | 15.82 | 15.81 | 1.98 | 6.43 | 0 |
| Linear | 3 | 5.37 | 5.76 | 1.92 | 6.24 | 0.001 |
| Square | 3 | 6.87 | 6.72 | 2.24 | 7.28 | 0 |
| Interaction | 2 | 3.58 | 3.58 | 1.79 | 5.81 | 0.004 |
| Residual error | 94 | 28.91 | 28.91 | 0.31 | | |
| Lack-of-fit | 6 | 6.11 | 6.11 | 1.02 | 3.93 | 0.002 |
| Pure error | 88 | 22.79 | 22.79 | 0.26 | | |
| Total | 102 | 44.72 | | | | |



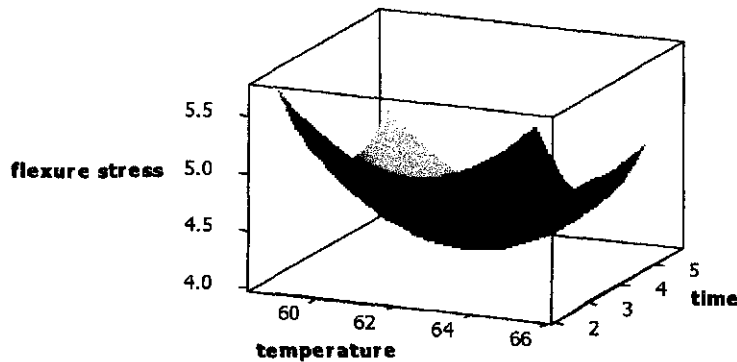


Figure 2 Response surface plots of brown strength using fine powder

Figure 2 shows the surface plot of brown strength using fine powder at constant debinding time of 4 hours at various values of powder loading and debinding temperature. The brown strength could be improved at higher powder loading, if the debinding time is sufficiently high. Figure 2 also showed that the combination of low debinding temperature and high powder loading produced high brown strength. The effect of independent variables on brown strength was different to the effect observed for coarse powder. However the minimum value of brown strength was observed in the combination of debinding temperature of between 62 to 64°C and the debinding time of between 3 to 4 hours. Therefore, the samples made from fine powders should be immersed in hot water not at the mentioned debinding temperature and time for maximum brown strength recovery.

4.3 OPTIMIZATION OF INDEPENDENT VARIABLES

Optimization of independent variables was carried out for maximum brown strength. The second order regression equation (equation 2 and 3) were solved using MINITAB optimization solver and the results were given in Table 5.

Table 5 Solvent debinding optimum conditions for maximum brown strength

| Particle size (µm) | Temperature, X_1 (°C) | Time, X_2 (hours) | Powder loading, X_3 (% volume) | Brown strength (MPa) | Desirability, D |
|--------------------|-------------------------|---------------------|----------------------------------|----------------------|-------------------|
| 19.61 | 59 | 3 | 64 | 5.0 | 1.00 |
| 11.2 | 60 | 2 | 64 | 5.6 | 1.00 |

Table 3 showed that the optimum combination for maximum brown strength of 5 MPa for coarse powder and 5.1 MPa for fine powder were within the range of levels of independent variables. The combination achieved maximum level of desirability, i.e. 1, which indicates that they are close to the desired values of responses.

4.4 OBSERVATION OF LEACHED SPECIMENS USING SCANNING ELECTRON MICROSCOPY

In the initial stage of water leaching, PEGs are extracted from the specimen surface or from near-surface regions. As the process of extraction proceeds deeper and deeper into the body, the pores extend from the sample surface into its interior. This provides more paths for water to enter into the body and diffusion of the PEGs out of the body. A typical micrograph which reveals the formation of pore channels in a leached sample is shown in Figure 2 these channels allowed more rapid removal of the remaining binder, without cracking, blistering or swelling during subsequent thermal debinding. After removal of the PEGs, the residual PMMA was found sufficient to retain the powder particles in position, so that the component retained its shape and could be handled without damage or distortion in subsequent processing. Most of the pores are considered to be interconnected and thus the leached body is permeable to monomer liberated by pyrolysis of the PMMA. The formation of these open pore channels was similar to the effect observed between coarse and fine powders.

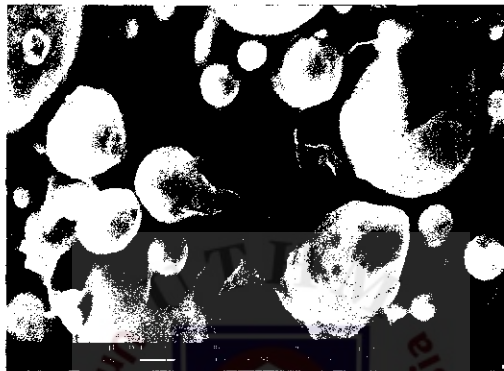


Figure 3 Scanning electron micrograph of water leached specimen, illustrating the formation of open pore channels.

5.0 CONCLUSIONS

RSM was used to identify the best combination of processing parameters (independent variables) that could maximize the brown strength. Central composite rotatable design (CCRD) of experiments is used to study the effect of these independent variables on the brown body's strength. ANOVA showed that all three debinding factors: debinding temperature, debinding time and powder loading affected the brown strength significantly. The optimal set of processing parameters for coarse powder were found to be powder loading (64% volume), debinding temperature (59°C) and debinding times (3 hours), to achieve the maximum brown strength of 5 MPa. Meanwhile for fine powder, the combinations are as follow: powder loading 64% volume, debinding temperature of 60°C and debinding time at 2 hours to achieve maximum brown strength of 5.6 MPa.

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