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Standardizing milling process parameters for the narrowest pigment particle size distribution with optimum energy consumption

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Abstract

Water-based ink, used in production of a new type of green packaging material has an efficient application in the flexible packaging industry to resolve the environmental issues related to volatile organic compound. To get the best possible application properties of dispersed pigment whose performance is mainly measured by the particle size distribution, it is essential to reduce the size of agglomerates. Concentrated dispersed material is manufactured by using a stirred bead mill, which is an energy-intensive process. The process of dispersion must be done efficiently and in the shortest possible time to draw out of the pigment its maximum color properties at the minimum cost. The grinding-energy efficiency is a significant parameter in bead milling as that affects the amount of energy used during grinding of pigment particles. The milling process needs to be optimized to reduce energy consumption. The objective of this study is to determine optimum dispersion process parameters to optimize energy consumption to achieve the narrowest pigment particle size distribution of rubine red pigments used in water-based ink. Experiments were conducted for fine grinding of organic rubine red pigment using a vertical bead mill. The experiments were conducted for varying sizes of grinding media from 0.5 mm to 1.0 mm, for two pigment loadings of 30 % and 37 %, and by extending milling time from 4 h to 6 h. The pigment particle size distribution and power consumption during each trial were measured to optimize process parameters with minimum energy consumption. Response surface design was performed to analyze data. Analysis of variance (ANOVA) techniques were used to check the significance of factors and the interaction of factors. The regression model for specific energy consumption was developed and tested; validation trial for dispersion process parameters concludes that 30 % pigment loading and mixed grinding media size provides narrowest pigment particle size distribution of 128 nm with minimum energy consumption of 1.67 kWh/t.

Keywords: pigment dispersion, bead milling, grinding efficiency, transparency, regression analysis

1. Introduction

The dispersion process of ink involves complete wetting of the pigment surface and then their uniform distribution in the application vehicle. Thus, the dispersion process includes the breakdown of pigment particles, agglomerates (with primary particles touching each other at the corner), and aggregates (with primary particles having surface to surface contact), into smaller particles and their distribution in a vehicle, leading to a colloidal suspension. A colloidal suspension is characterized by the behavior that the finely divided particles do not settle under their gravitational forces.

The dispersion of pigments in printing inks is important for several reasons as it improves the color strength of pigment, increases the transparency of pigment, affects rheological behavior and flow properties of the ink, provides more gloss and finally, it provides stability to ink throughout its shelf life (Pal and Fleming, 2006; Simpson, et al., 2015). For getting maximum benefits of pigment, it is recommended to get the maximum reduction of pigment size as possible to its primary particle size. The color strength of pigment depends on its exposed surface area, so the smaller the particles, the higher the surface area and thus stronger the color (Herbst and Hunger, 2007; Klein, 2010).

Concentrated dispersed material is manufactured by using a stirred bead mill. Stirred bead milling is however an energy-intensive process. Along with this, the high prices of raw materials make it essential to look the efficiency of the dispersion process (Weber and Langlois, 2010). The process of dispersion must be done efficiently and in the shortest possible time to draw out of the pigment its maximum color properties at the least cost (Klein, 2010).

Pigment dispersion process by using a bead mill is the major energy-consuming process during the manufacturing of water-based ink. Milling process parameters like milling time, size and density of grinding media (GM), pigment loading, and speed of machine are majorly affecting the pigment particle size distribution and energy consumption during the milling operation. Therefore, the milling process parameters need to be optimized to increase the milling efficiency to achieve the desired particle size distribution with minimum energy consumption. (Zheng, Harris and Somasundaran, 1996; Hamey, 2005; McDowell, 2006; Choi, Lee and Kim, 2009; Weber and Langlois, 2010; Inam, Ouattara and Frances, 2011; Schmidt, et al., 2012; Ohenoja, Illikainen and Niinimäki, 2013; Senthilkumar and Akilamudhan, 2014; Simpson, et al., 2015)

The researchers highlighted that the use of chemical additives during wet dispersion improves the surface and mechanical properties of individual particles such as surface energy and flow of medium which improves the efficiency of the grinding process. (Farrokhpay, 2004, p. 147; McDowell, 2006; Nsib, Ayed and Chevalier, 2006; Choi, Lee and Kim, 2009)

Kwade's model (Kwade, 2004) summarized that the milling efficiency of a stirred media mill is a product of stress intensity and stress frequency. The best milling efficiency is obtained by selecting GM size, the density of GM, and mill speed as these parameters approach to an optimum stress intensity and the maximum stress number (Choi, Lee and Kim, 2009; Weber and Langlois, 2010; Simpson, et al., 2015). During the pigment dispersion, specific energy is the amount of energy or work required to grind a pigment agglomerate to a desired particle size. The most common units of specific energy are J/kg or kWh/t of slurry. The amount of specific energy required to grind a particular solid to a desired particle size is a fixed value, only dependent on the efficiency of the equipment, milling time, density of GM, and mill base (McDowell, 2006). Various studies were conducted to analyze the influence of materials of GM of different specific weights, sizes of GM, stirred tip speeds, milling times, and solid concentrations on particle size and particle size distribution with optimum energy consumption. (Weber and Langlois, 2010; Ohenoja, Illikainen and Niinimäki, 2013; Simpson, et al., 2015)

Weber's rule predicts that when the particles are of 0.4 µm to 0.7 µm in size they provide the maximum scattering for wavelengths between 400 nm and 700 nm. Thus maximum scattering from pigment particles is observed if the size of the pigment is equal to its wavelength. Exactly opposite to this for maximum transparency is that the pigment particle should be as small as possible. (Herbst and Hunger, 2007)

Therefore to achieve maximum transparency and maximum color strength it is important to obtain the narrowest pigment particle size distribution during dispersion.

The milling process needs to be optimized to reduce energy consumption. Energy consumption can be different as per the mill base properties. Energy consumption during the resin-free dispersion of the pigment to achieve narrow particle size distribution as well as the effect of the pigment particle size distribution on transparency are relatively unexplored.

In this study process parameters like milling time, GM size and pigment loading were optimized to achieve target pigment particle size distribution. During each trial, power consumption was measured to optimize process parameters with minimum energy consumption. The optimum value for the GM size, milling time, and pigment loading were determined to obtain the narrowest particle size distribution with the optimum stress intensity and lowest specific energy consumption.

2. Experimental procedure

2.1 Materials

Rubine red pigment (PR 57:1) manufactured by Sudarshan Chemical Industries Ltd. was used for the pigment dispersion process. Dispersing additive, polydimethylsiloxane based antifoaming agent, was provided by BYK. Constant percentages of a dispersing agent, and antifoaming agent were used during the resin-free dispersion process. De-ionized water was used as a solvent in the grinding experiments and used to adjust the pigment loading of the mill base. Grinding media of different size ranges were used, purchased from M/s Jyoti ceramics.

2.2 Premixing of material

To prepare a premixture, initially, a mixture of deionized water, dispersing agent, and the antifoaming agent was prepared and later slow addition of pigment was allowed under low shear rate by using a stirrer (make-REMI). Prepared premixture was allowed to soak for 24 h. The formulation used for mill base is provided in Table 1.

Table 1: Formulation of a mill base used during a resin-free dispersion of PR 57:1

Ingredient	Specification	Amount (%)	Purpose of ingredient
Pigment	PR 57:1	25 to 30	Coloring agent
Dispersing agent	Disperse BYK	7 to 8	Dispersion of pigment
Antifoaming agent	BYK 019	0.35 to 4	Rupture of the foam
Deionized water	DI Water	60 to 68	Acting as a vehicle

2.3 Variables for experiment

Experiments were conducted to fix the levels of few parameters such as the speed of the stirrer, the density of GM, range of GM size, and pigment loading. The significant parameters and their levels for the pigment particle size distribution were screened and considered as input parameters to optimize the energy consumption. Higher density of GM as 6.2 g·cm^{-3}, speed of the machine as 2 600 rpm, and GM size as 0.5 mm to 1 mm were determined to be the more efficient values for the reduction of the pigment particle size. Response surface design of experiments (DOE) was generated to evaluate the energy consumption for narrow particle size distribution. The experimental design consists of three factors, namely GM size, milling time, and pigment loading, and different values of each factor (called levels of the factors). In the design it includes three groups of sizes of GM (zirconox beads): 0.4–0.6 mm (named as 0.5 mm), 0.8–1.0 mm (named as 1 mm) and 50 : 50 ratio of both 0.4–0.6 mm and 0.8–1.0 mm (named as 0.75 mm), two pigment loading percentages: 30 % and 37 %, and three milling times: 4 h, 5 h and 6 h. Thus, the total trials in the design were 18. The detailed experimental design along with input parameters and their levels are indicated in Table 2.

2.4 Equipment

Most of the mills used for grinding of pigments are vertical and horizontal bead mills due to their high efficiencies, low energy consumption, and high unit output. The lab-scale vertical bead mill equipped with a stationary grinding chamber, a high-speed stirrer with cowl blade fixed on a drive shaft which rotates at 2 600 rpm (tip speed is 6 m·s^{-1}) was used for pigment dispersion. The capacity of the grinding chamber is 1.5 liter. After dispersion 600 mesh was used to separate GM from the dispersed material. For lab scale 500 g of premixture was prepared. To maintain the constant temperature of mill base as 25 °C to 27 °C during dispersion, the grinding vessel is equipped with a water jacket. The particle size distribution of dispersed samples was measured by Malvern particle size analyser (Zetasizer Nano s90 model) at M/s Sudarshan Chemical Industries. Two watt meters were connected to the power supply to measure active power during the dispersion of the material.

Initially power consumption for the unloaded mill (empty mill) is measured from reading observed in two watt meters W1 and W2. It was measured ten times for every ten minutes. The average value of active power provides the value of N_0. Active power was measured throughout the dispersion process and for each trial power consumption was calculated.

2.5 Calculations

Width of particle size distribution (WPSD), transparency, specific energy consumption (SEC) and stress intensity (SI) were calculated by using Equations [1] to [5].

Width of particle size distribution, which provides a size range of pigment particles which occupies a total 80 % volume of the dispersed material, was calculated by using Equation [1].

$$\text{WPSD} = D_{90} - D_{10} \qquad [1]$$

where D_{90} and D_{10} are the particle sizes in nanometres representing the 90 % and 10 % values in the cumulative volumetric size distribution (Ding, et al., 2013; Barth, Schilde and Kwade, 2013).

Transparency was determined by ASTM D344-11(2016) method for visual assessment (ASTM, 2016), and instrumental ASTM D2805-11(2018) standard method (ASTM, 2018) used to calculate opacity. The BYK opacity chart (Figure 1) was used to calculate opacity by using Equation [2]. A uniform ink drawdown by using bar coater no. 1 was applied on a black/white contrast chart. After air-drying the drawdown, a spectropho-

Table 2: Variables and their levels used during optimization of resin-free dispersion process for PR 57:1

Variables	Level 1	Level 2	Level 3
Zirconox beads	0.4 mm to 0.6 mm (0.5 mm)	0.8 mm to 1.0 mm (1 mm)	Mixture of both (50 : 50), (0.75 mm)
Pigment loading	30 %	37 %	–
Milling time	4 h	5 h	6 h

tometer (make: Gretag Macbeth, model: Eye-One Pro) was used to measure *Y* (tristimulus value) against white and *Y* against black of contrast chart. Transparency was calculated by using Equation [3].

Figure 1: Byko-chart Opacity 3B (BYK, n.d.)

$$\text{Opacity (\%)} = \left[\frac{Y\,(\text{against black})}{Y\,(\text{against white})}\right] \times 100 \qquad [2]$$

where *Y* is a tristimulus value.

$$\text{Transparency (\%)} = 100 - \text{Opacity (\%)} \qquad [3]$$

where opacity is calculated from Equation [2].

Specific energy consumption was determined by using two wattmeter method, which was used to measure the power throughout the dispersion process time. Power data was measured after every 10 min from both wattmeters to calculate power consumption for each design of the experiment. The power was recorded during grinding and SEC was calculated (Equation [4]) as the power input integrated over the grinding time t_G and divided by the mass of slurry (*m*),

$$\text{SEC} = \int (N - N_0) \cdot dt_G \; / \; m \qquad [4]$$

where SEC (kWh/t) is specific energy consumption, *N* (kW) is necessary electrical power running the filled mill with beads and slurry, N_0 (kW) is necessary electrical power running the empty mill, $(N - N_0)$ (kW) is active electrical power, and *m* (t) is a mass of slurry.

The stress-energy or stress intensity of GM describes the maximum kinetic energy of two colliding grinding beads. Optimum stress intensity and maximum energy utilization will happen when the stress intensity is just sufficient to break the particle. The stress intensity of the GM (SI_{GM}) *was* calculated using Equation [5].

$$SI_{GM} = d_{GM}^3 \cdot \rho_{GM} \cdot v_T^2 \qquad [5]$$

where SI_{GM} (Nm) is stress intensity of the GM, *d* (m) is bead diameter, ρ (kg/m^3) is specific weight of GM, and v_T (m/s) is stirrer tip speed.

2.6 Analysis of the impact of WPSD on transparency of water-based ink

The effect of WPSD on the transparency of water-based ink was analyzed. When there is one categorical independent variable and one quantitative dependent variable, one-way analysis of variance (ANOVA) is preferred statistical technique. It tells us if the dependent variable changes according to the level of the independent variable (Vik, 2014).

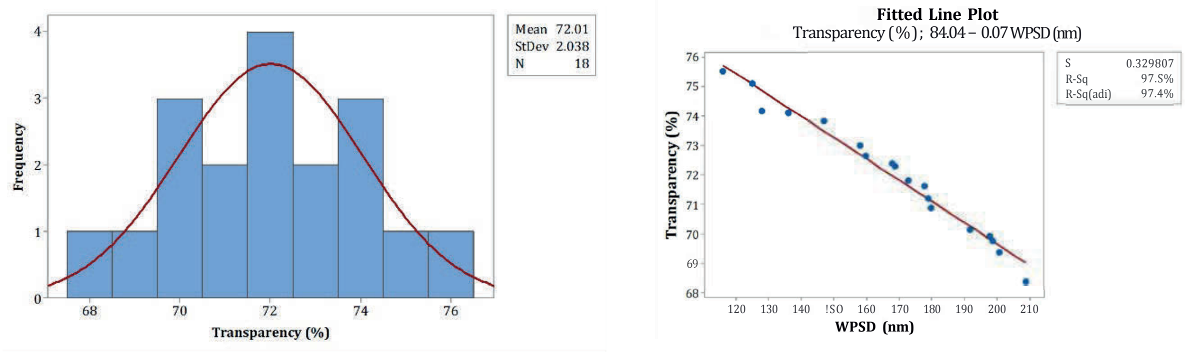
In this experiment WPSD is considered as independent variable and transparency is dependent variable. For the analysis, WPSD is the only factor considered; so, one-way ANOVA is used to predict the effect of WPSD on transparency of water-based ink. Above model with one predictor is referred to as simple linear regression model.

3. Results and discussion

3.1 Effect of WPSD on transparency of water-based ink

Dispersion experiment based on different pigment loading, GM size, and milling time provides varying widths of WPSD, which were measured by using particle size analyzer. Figure 2a presents the normal distribution of data measured for transparency of various formulations. Figure 2b describes the negative linear relation between WPSD and transparency of water-based ink. Narrower WPSD provides higher transparency. This is attributed to the fact that narrowest WPSD provides more surface area to interact with light and improves the absorbency of pigment. The amount of light scattering which influences the transparency depends strongly on the particle size of the pigment. Very small particle scatters very little light. Scattering increases with increasing particle size until the particles are about the same size of the wavelength of light and then it decreases for still larger particles. Thus, less scattering power of smaller particles, results in a higher degree of transparency, which is very important in four color conventional printing processes to achieve the higher strength of colors.

The regression result (Table 3) tells that particle size is significant for transparency because of their low *p*-values. Particle size distribution account for 97.5 % of the variance of transparency. For each 1 % increase in the amount of transparency, the percentage of WPSD is expected to decrease by 0.07 %.



a) b)

Figure 2: (a) Histogram of transparency, and (b) negative linear relation between transparency and WPSD

Table 3: ANOVA, regression analysis and regression equation for transparency
Regression analysis: transparency (%) versus WPSD (nm)
Analysis of variance for transparency

Source	DF	Adj. SS	Adj. MS	*F*-value	*p*-value
Model	1	68.9	68.9	633.4	0
Pure error	16	1.7	0.1		
Total	17	70.6			

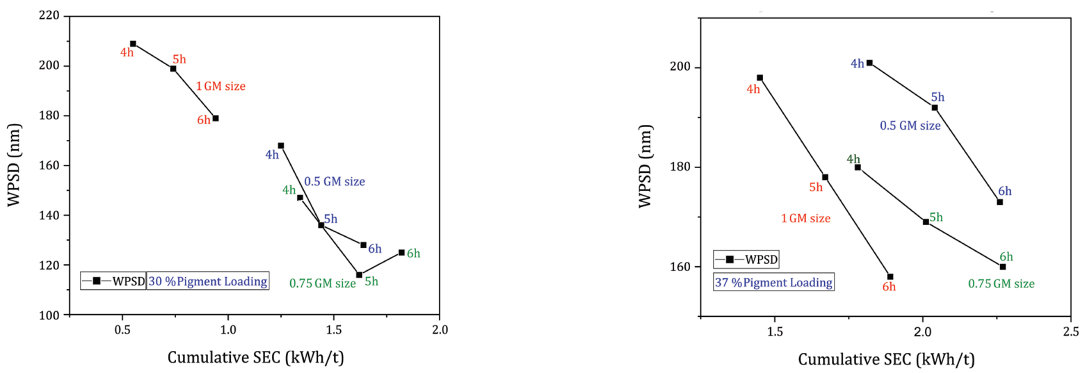
R-sq = 97.5 % *R*-sq(adj.) = 97.4 %
Regression equation:
Transparency (%) = 84.04 – 0.07 WPSD (nm)

3.2 Effect of GM sizes on WPSD and SEC

The effect of three different GM sizes on WPSD and cumulative SEC were analyzed. The GM used for the experiment was zirconium oxide with different levels of varying bead diameter named as 0.5 mm, 1 mm, and 0.75 mm.

Figure 3 indicates the effect of GM sizes on WPSD and cumulative SEC for two different pigment loading percentages. It is seen from Figure 3a larger GM size provides wider WPSD than smaller GM size. It is also observed that with longer milling time the WPSD is reduced.

Referring to Figure 2b narrow WPSD provides maximum transparency. Hence narrow WPSD is recommended which will provide higher strength for dispersed pigment. The pigment dispersion process to achieve narrow WPSD is least expensive when the grinding process works for 37 % (Figure 3b) it is for largest GM at the minimum of the SEC. It is observed from Table 4 that minimum specific energy was consumed to achieve narrowest WPSD for 30 % pigment



a) b)

Figure 3: Effect of GM size on WPSD and cumulative SEC for three different milling times at two different pigment loadings: (a) for 30 % pigment loading, and (b) for 37 % pigment loading

loading and when milling continued up to 5 h with 0.75 mm GM size. But if milling extended for 6 h it increases the WPSD with increased SEC. The 0.5 mm GM size also provides moderately narrow WPSD but is slightly higher than 0.75 mm GM size with low energy consumption. Higher GM size consumes less energy but provides wider WPSD which reduces the overall performance of pigment. Smaller GM size of the same density as larger GM size dramatically increases the number of grinding beads per liter of the mill working volume. Along with this the kinetic energy of the grinding beads is increased by the ratio of the specific weight of the bead material by equal stirrer tip speed in the mill. Both parameters, reduction in bead size and using beads with higher specific weight, improve the milling efficiency because the increased surface area of beads achieves more contact points where impact, shear and compression forces takes place between the beads and pigment particles; along with this higher kinetic energy generated helps to reduce particle size effectively. Due to increased surface area of GM, utilization of free material was observed, which increases the viscosity of mill base and needs more energy at a specified tip speed for the momentum of the mill base.

Large-sized grinding beads have different motion paths and speeds compared to a smaller one due to their mass difference. Single GM size provides monotonous movement among the material. The 0.75 mm GM size, as it is the mixture of both 0.4–0.6 mm and 0.8–1.0 mm, provides rise to two different momentums as a result of the different mass of the beads, which breaks the monotonous movement between them and increases the irregular movements. This will help to break more effectively the pigment particles and provides higher rate for reduction of WPSD value in comparison with a single size of GM, hence it reduces the dispersion time and indirectly reduces the energy consumption. Thus the 0.75 mm GM size optimized the energy consumption to provide narrowest WPSD.

3.3 Effect of pigment loading on WPSD and SEC

The effect of different pigment loading for three different GM sizes on the WPSD is presented as a function of a cumulative SEC as shown in Figure 4.

Narrow WPSD is observed for the lowest solid concentration i.e. 30 % with minimum energy consumption. This result is applicable for 0.5 mm and 0.75 mm GM size when used for dispersion. But for the 1 mm GM size (Figure 4c), narrow WPSD is observed at higher pigment loading (37 %) with moderately less energy consumption. This is attributed to the fact that 1 mm GM size i.e. higher GM size reduces the number of grinding beads required in a specified volume.

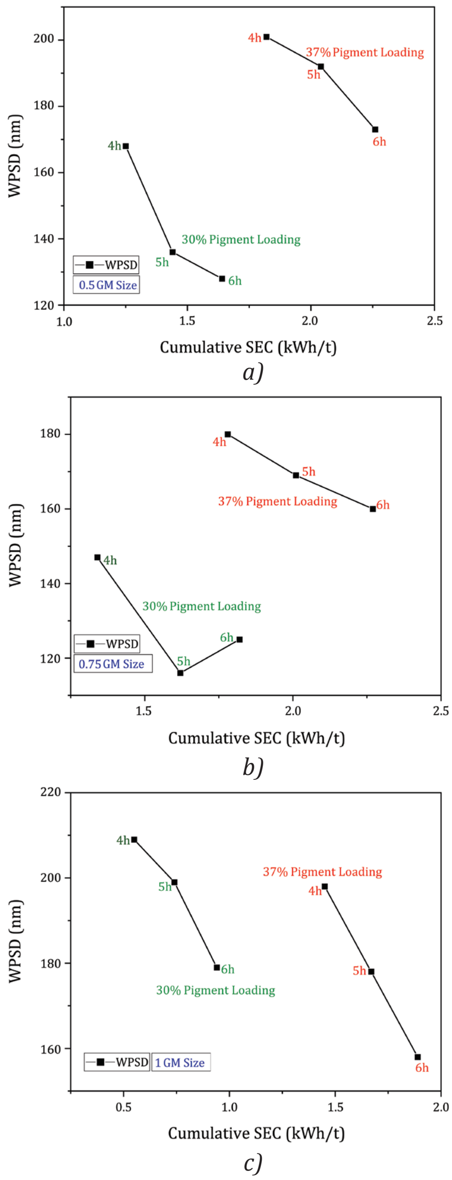


Figure 4: Effect of two different pigment loadings on WPSD and cumulative SEC at three different milling times with constant GM size: (a) for 0.5 mm GM size, (b) for 0.75 mm GM size, and (c) for 1 mm GM size

Along with this it also reduces the surface area of grinding beads, which reduces the ratio of the surface area with material volume. More free moving material is present, which provides unused collisions of the grinding beads inside the mill. When pigment loading is increased it consumes the free moving material and provides the required viscosity to mill base which utilizes the maximum collisions of grinding beads to reduce the particle size. Thus, when 1 mm GM size i.e higher GM size and 37 % i.e. high solid percentage are used for dispersion it provides narrow WPSD with moderately less energy consumption. For the 0.5 mm GM size (smaller GM size) as well as the 0.75 mm GM size (mixed GM size), ratio of surface area to the material is very high, which reduces free-flowing material; so when pigment loading is 30 % it provides sufficient viscosity to mill base to utilize the maximum collision of GM, which helps to reduce the particle size distribution. But for higher pigment loading (37 %) viscosity gain is observed, which restricts the free motion of grinding beads and ultimately reduces the frequency of collisions; hence wider WPSD is observed even though high energy is consumed during dispersion. The 0.75 mm GM size provides narrowest WPSD after 5 h of milling time with minimum energy consumption.

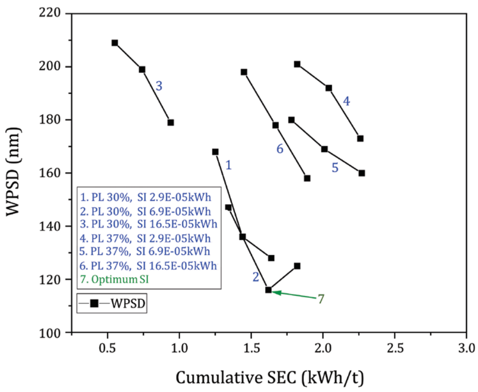


Figure 5: Stress intensity with optimum (marked by the green arrow) of GM with respect to WPSD and cumulative SEC

The stress intensity of GM corresponds to the maximum kinetic energy of two colliding grinding beads at the stress events. When the maximum utilization of kinetic energy takes place at stress events to break the pigment particles then it is an optimum stress intensity. Stress intensity for different solid percentages of dispersion remains constant as it is assumed that the velocity of the mill base is equivalent to a tip speed of stirrer. Figure 5 shows the WPSD at different SEC as a function of stress intensity. It can be seen from Figure 5 that the lower the stress-energy, the lower the particle size. When energy consumption is considered, the optimal stress-energy is seen to be around $6.9{\cdot}10^{-5}$ kWh (written as 6.9E-05 kWh in the legend in Figure 5) to the low energy consumption with narrowest WPSD. The stress-energy $2.9{\cdot}10^{-5}$ kWh (2.9E-05 kWh) can also be considered as optimal stress energy as it also provides moderately narrow WPSD with slightly higher energy consumption than $6.9{\cdot}10^{-5}$ kWh stress intensity. Thus, the most efficient grinding is observed at the smallest GM size, but the optimum condition is observed at mixed GM size.

3.4 Statistical analysis of SEC

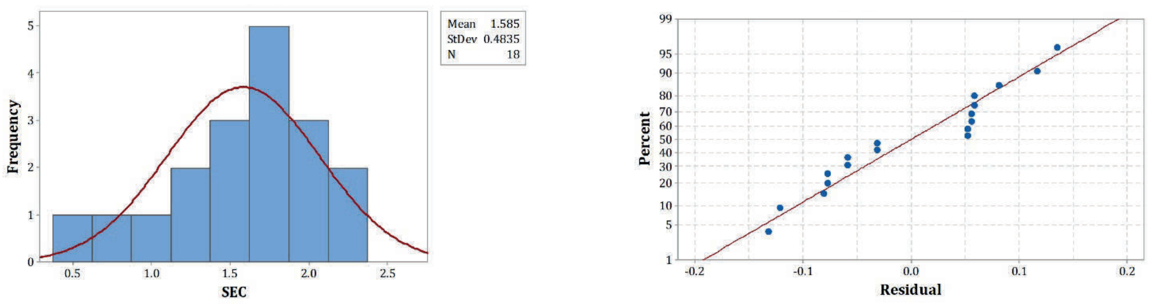
Histogram of the residuals (Figure 6a) is an exploratory tool to show general characteristics of the residuals including typical values, spread, and shape. It shows the normal distribution of samples. Normal probability plot (Figure 6b) of residuals shows that the points in this plot form a straight line, which means that the residuals are normally distributed.

3.5 Main effect plot and interaction plot

Main effect plot (Figure 7a) explains the significance of GM size, milling time, and pigment loading on specific energy consumption. The 0.75 mm GM size, higher pigment loading i.e. 37 % and higher milling time provide higher specific energy.

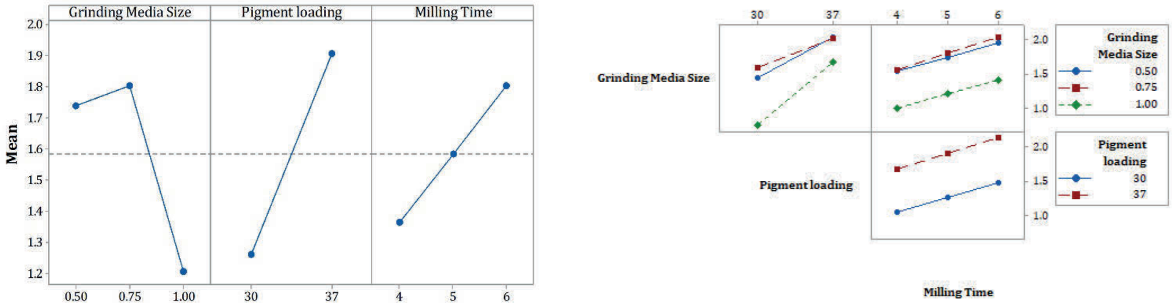
In the interaction plots (Figure 7b), the upper-left one consists of non-parallel lines representing there is an interaction between GM size and pigment loading.

The ANOVA table (Table 4) summarizes the linear terms, the squared term and the interaction. Higher *F*-value of linear terms compared to square term and interaction term, indicates that the model is majorly explained by the linear terms. The *p*-values from the ANOVA table, which are below α value of 0.05, indicate that all the main factors such as GM size, pigment loading and milling time are significant for energy consumption. The higher *F*-statistics values for pigment loading, GM size and milling time indicate these are the most significant factors that influence energy consumption. The inter-



a) *b)*

Figure 6: (a) Histogram of SEC residuals, and (b) normal probability plot of residuals for SEC response



a) *b)*

Figure 7: (a) Main effect plot of SEC and (b) interaction plot of SEC, both for different GM sizes (in mm), pigment loadings (in %) and milling times (in h)

Table 4: Response surface regression model for energy consumption
Response surface regression: energy consumption versus GM size, pigment load, milling time
Stepwise selection of terms α to enter = 0.15, α to remove = 0.15
ANOVA for energy consumption

Source	**DF**	**Adj. SS**	**Adj. MS**	***F*-value**	***p*-value**
Model	5	3.86	0.77	79.56	0.00
Linear	3	3.33	1.11	114.57	0.00
GM size	1	0.86	0.86	88.68	0.00
Pigment loading	1	1.90	1.90	195.50	0.00
Milling time	1	0.58	0.58	59.53	0.00
Square					
GM size * GM size	1	0.44	0.44	45.69	0.00
2-way interaction					
GM size * Pigment loading	1	0.08	0.08	8.39	0.01
Pure error	12	0.12	0.01		
Total	17	3.97			

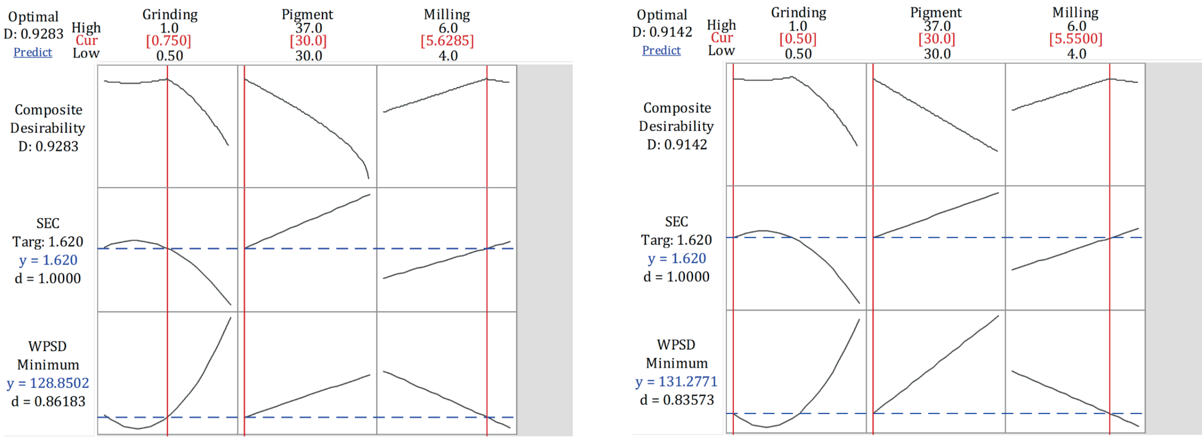
R-sq = 97.07 % *R*-sq(pred.) = 95.85 % *R*-sq(adj.) = 93.87 %
Estimated regression coefficients for energy consumption:
Energy consumption = −2.22 + 3.76 GM Size + 0.02 Pigment loading + 0.21 Milling time − 5.33 GM Size * GM size + 0.09 GM size * Pigment loading

action of GM size and pigment loading is significant for energy consumption at a 95 % confidence level. Table 4 shows a higher percentage of *R*-sq., indicating that 97.07 % of the variability could be explained by the model at a 95 % confidence level. The adjusted *R*-sq of 93.87 % indicates a significant improvement of the model by using three parameters. The predicted *R*-sq of 95.85 % indicates that the model predicts new observations nearly as well as it fits the existing data. The regression equation describes the statistical relationship between dispersion process factors and energy consumption. Table 4 provides the regression equation that helps to predict new observations for desired particle size distribution with energy consumption.

3.6 Optimization from regression model

The optimization plot (Figure 8) shows the effect of each factor ie. GM size, pigment loading, and milling time on the cumulative energy consumption to achieve the desired particle size. The vertical red lines on the graph represent the current factor settings. The numbers displayed at the top of a column in a square bracket show the current factor level settings. The horizontal dash lines and numbers represent the responses of WPSD and energy consumption for the current factor level. Minitab software calculates the narrowest WPSD and minimum energy consumption for two hold values of GM size, namely 0.75 mm and 0.5 mm.

Figure 8 explains that when the same amount of energy consumption is considered it is observed that 0.75 mm GM size will provide narrowest WPSD compared to 0.5 mm GM size. Hence 0.75 mm GM size, 30 % pigment loading, and milling time of 5.63 h are the optimized parameters which will provide narrowest WPSD with minimum energy consumption as shown in the Figure 8.



a) *b)*

Figure 8: Optimization plot from the regression model for the smallest WPSD and minimum SEC: (a) hold value 0.75 mm GM size, and (b) hold value 0.5 mm GM size

Table 5: Multiple response prediction for 0.75 mm hold value of GM size
Response optimization: SEC, WPSD

Optimized value

Variable	**Setting**
GM size	0.75 mm
Pigment loading	30 %
Milling time	5.63 h

Solution

Solution	**GM size**	**Pigment loading**	**Milling time**	**WPSD fit**	**Energy consumption fit**	**Composite desrability**
1	0.75 mm	30 %	5.63 h	1.62	128.85	0.93

Solution

Response	**Fit**	**SE fit**	**95 % CI**	**95 % PI**
SEC (kWh/t)	1.62	0.0497	(1.51, 1.73)	(1.38, 1.86)
WPSD (nm)	128.85	5.2600	(117.39, 140.31)	(103.44, 154.26)

Additionally, as presented in Table 5, summarizing the optimization results the confidence interval (CI) indicates the 95 % confidence that the mean of the WPSD at these settings is between 117.39 nm and 140.31 nm, and the SEC is between 1.51 kWh/t and 1.73 kWh/t. The prediction interval (PI) indicates 95 % confidence that WPSD of a single new observation will fall between 103.44 nm and 154.26 nm and SEC will be between 1.37 kWh/t and 1.86 kWh/t.

Table 6: Response predicted values for optimum energy consumption

Variable	**Setting**	**WPSD (nm)**	**SEC (kWh/t)**
GM size (mm)	0.75		
Pigment loading (%)	30.00	128	1.67
Milling time (h)	5.63		

3.7 Validation trial

A validation trial was conducted for the optimized setting (Table 6). The WPSD of 128 nm was achieved with 1.67 kWh/t SEC. These values are close to those estimated using the model (see Table 5).

4. Conclusion

Width of particle size distribution (WPSD) shows negative linear relation with transparency of water-based ink. Narrower WPSD provides higher transparency. In the bead milling process size of grinding media (GM), the pigment–binder ratio, and milling time have a significant effect on the milling rate and efficiency of the milling process. Smaller GM size provides narrow WPSD with minimum energy consumption. Mixed GM size provides a slightly higher rate of WPSD reduction with the lowest energy consumption. Percentage of pigment loading should be sufficient enough for the maximum utilization of collisions of grinding beads which affects the rate of particle size reduction and energy consumption. The model showed more than 96.57 % predictability. Validation trial for dispersion process parameters concludes that 30 % pigment loading, mixed GM in wide range of sizes (named 0.75 mm, and milling time of 5.63 h) provide narrowest pigment particle size distribution of 128 nm with minimum energy consumption of 1.67 kWh/t.

In this research the regression model was developed for a specific type of pigment. Research can be continued in a direction that would include energy efficiency and optimization of pigment particle size distribution for the grinding of other process colors as well as special color pigments. Also, the optimization of percentage of dispersing additive for resin-free dispersion for all process color pigments is recommended to develop cost-competitive stable pigment dispersion for water-based ink.

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References

ASTM, 2016. *ASTM D344-11(2016) Standard test method for relative hiding power of paints by the visual evaluation of brushouts*. West Conshohocken, PA, USA: ASTM International.

ASTM, 2018. *ASTM D2805-11(2018) Standard test method for hiding power of paints by reflectometry.* West Conshohocken, PA, USA: ASTM International.

Barth, N., Schilde, C. and Kwade, A., 2013. Influence of particle size distribution on micromechanical properties of thin nanoparticulate coatings. *Physics Procedia*, 40, pp. 9–18. https://doi.org/10.1016/j.phpro.2012.12.002.

BYK, n.d. *Opacity charts.* [online] Available at: <https://www.byk-instruments.com/si/en/Physical-Properties/Test-Charts/Opacity-Charts/c/p-5916> [Accessed December 2020].

Choi, H., Lee, W. and Kim, S., 2009. Effect of grinding aids on the kinetics of fine grinding energy consumed of calcite powders by a stirred ball mill. *Advanced Powder Technology*, 20(4), pp. 350–354. https://doi.org/10.1016/j.apt.2009.01.002.

Ding, B., Zhang, X., Zhou, D.L. and Miao, C.W., 2013. Effects of water-borne polyurethane on calcium silicate hydrate and toughness of properties of cementitious composites. *Advanced Materials Research*, 619, pp. 545–552. https://doi.org/10.4028/www.scientific.net/AMR.619.545.

Farrokhpay, S., 2004. *Interaction of polymeric dispersants with titania pigment particles*. PhD thesis. University of South Australia. https://doi.org/10.13140/RG.2.2.24446.59200.

Hamey, R.G., 2005. *Production of organic pigment nanoparticles by stirred media milling*. Master thesis. University of Florida.

Herbst, W. and Hunger, K., 2007. *Industrial organic pigments: production, properties applications*. 3rd ed. Weinheim, Germany: WILEY-VCH.

Inam, M.A., Ouattara, S. and Frances, C., 2011. Effects of concentration of dispersions on particle sizing during production of fine particles in wet grinding process. *Powder Technology*, 208(2), pp. 329–336. https://doi.org/10.1016/j.powtec.2010.08.025.

Klein, L.C., 2010. Solgel coatings. In: A.A. Tracton, ed. *Coatings Materials and Surface Coatings*. 3rd ed. Boca Raton, FL, USA: CRC Press..

Kwade, A., 2004. Mill selection and process optimization using a physical grinding model. *International Journal of Mineral Processing*, 74(Supplement), pp. S93–S101. https://doi.org/10.1016/j.minpro.2004.07.027.

McDowell, R.I., 2006. *Particle size reduction of phthalocyanine blue pigment.* Master thesis. University of Louisville. https://doi.org/10.18297/etd/946.

Nsib, F., Ayed, N. and Chevalier, Y., 2006. Selection of dispersants for the dispersion of C.I. Pigment Violet 23 in organic medium. *Dyes and Pigments*, 74(1), pp. 133–140. https://doi.org/10.1016/j.dyepig.2006.01.047.

Ohenoja, K., Illikainen, M. and Niinimäki, J., 2013. Effect of operational parameters and stress energies on the particle size distribution of TiO_2 pigment in stirred media milling. *Powder Technology*, 234, pp. 91–96. https://doi.org/10.1016/j.powtec.2012.09.038.

Pal, L. and Fleming, P.D., 2006. The study of ink pigment dispersion parameters. *The Hilltop Review,* 2(1), pp. 61–70.

Schmidt, J., Plata, M., Tröger, S. and Peukert, W., 2012. Production of polymer particles below 5 µm. *Powder Technology*, 228, pp. 84–90. https://doi.org/10.1016/j.powtec.2012.04.064.

Senthilkumar, K. and Akilamudhan, P., 2014. Experimental studies on effect of grinding additives in size reduction process, *International Journal of ChemTech Research*, 6(9), pp. 4428–4433.

Simpson, A.B.G., Byrne, J.A., McLaughlin, J.A.D. and Strawhorne, M., 2015. Effect of solids concentration on particle size distribution of deagglomerated barium titanate in stirred media mills. *Chemical Engineering Research and Design*, 93, pp. 287–292. https://doi.org/10.1016/j.cherd.2014.04.006.

Vik, P., 2014. *Regression, ANOVA, and the general linear model: a statistics primer*. Thousand Oaks, CA, USA: SAGE Publications.

Weber, U. and Langlois, D., 2010. The effect of grinding media performance on milling and operational behaviour. *Journal of the Southern African Institute of Mining and Metallurgy*, 110(3), pp. 147–152.

Zheng, J., Harris, C.C. and Somasundaran, P., 1996. A study on grinding and energy input in stirred media mills. *Powder Technology*, 86(2), pp. 171–178. https://doi.org/10.1016/0032-5910(95)03051-4.