Abstract

The objective of this work is to study the variations of how independent processing parameters such as temperature, speed, and feed rate affect the dependent responses for consistent output colour (L*, a*, b*, dE*). In this study, the compounded material was processed on an intermeshing twin-screw extruder (TSE) and injection molded to evaluate their effect on the colour stability, rheology and dispersion of the polycarbonate resins. Focus was extended to the interaction of the speed, which correlates to the dispersion and colour changes.

Introduction

Mastery of this science includes physics, chemistry, and psychology. It is essential for the design and the control of most manufactured colored materials, including polymers. In the 21st century, colour science is more significant than ever. Producing plastic with a marketable colour requires the addition of one or more resins; however, achieving the correct colour in the first attempt is a challenge. Apart from pigment formulations, the colour properties of polymers are directly affected by the processing parameters for polymer compounding during the extrusion process. Data mining, in which historical data is analyzed to select formulations (PC compound grades, e.g., G-43), pigments (e.g., red pigment), and processing parameters (temperature, speed, and feed rate) may cause colour mismatches. Blending mixer, co-rotating intermeshing-twin screw extruder and injection moulding were employed. General factorial design, (BBD) and 3level were conducted in design of experiments [1-4]. ANOVA of the obtained experimental data was utilized to study the effects of these processing parameters. The regression model was generated based on the ANOVA to predict the optimal matching colour. Producing the right colour with minimal wastage has been a great challenge [5] to obtain a desired colour, vary with changes of the light source, the object and the observer [6]. The effects of screw geometry and operating conditions on dispersion have also been studied [7]. Controlling the particle size and dispersion of yellow pigment, PY62 directly affects properties such as film transparency, colour development, extruder pressure build and processing time [8]. The colour variation is due to degradation, temperature, particle size, and colorant [9]. Screw speeds have no apparent effect on the kneading block performance [10]. Shear forces that occur in the extruder is a good determinant of the level of mixing for different polymer [11]. High shear rates, processing temperatures, and processing pressures are involved in the manufacturing processes. The increasing speed (screw speed) will enhance the longitudinal distribution [12, 13]. Processing affects the rheological properties and dispersion ability of pigments in the resin [14]. Because pigments are metallic products, they do not exhibit exact and uniform size, or range of sizes [15]. Narrower particle size distributions are superior for Chroma. Scattering and absorption are functions of particle size. Control properties are linked to particle size and morphological structure. Characteristically, the narrow particle size distribution shows a cleaner effect and wider provides greater hiding power. Therefore, the shapes offer different levels of reflectance and opacity [16]. To obtain primary particle size, this is due to smaller particles in the higher surface area and consequently strengthens the colour [17, 18]. There are two types of reduction in particle size, shear and collision. The best quality of the dispersion degree depends on the characteristic of: pigment volume concentration, size, dwell time, rotation speed, energy input, and temperature [19, 20]. The dispersion degree depends on the amount of energy input (i.e., speed) and the residence time in the chamber and intensity of scattered light by a particle depends on molecular weight, size and shape [21].

The objective of this research is to investigate the effect of processing parameters, rheology and dispersion. Experiments were produced in terms of three different processing parameters (temp., feed rate, screw speed), and while keeping the other parameters fixed at (255°C, 25kg/hr, 750rpm). In our previous work [27-28] found that the obtained results of the variation in polycarbonate content and temperature significantly effect rheological properties and the obtained viscosity data was correlated to dispersion and colour changes [27-28]. In this study we used the same compound grade at three levels of design. Each of the runs was replicated three times to allow estimation of experimental error or trend analysis This work investigated how the variations of independence of processing parameters such as temperature, screw speed, and feed rate affect the dependent responses for consistent output colour (L*, a*, b*, dE*). Focus
extended on the speed, correlated to dispersion and colour changes to evaluate their effect on the colour stability, rheology and dispersion of the translucent compounded plastic grade of polycarbonate (PC) to achieve minimum deviation of colour differences.

**Materials**

R1 was 25g/10min and that for R2 was 6.5g/10min. In addition it contained four different colour pigments (black, white, red, yellow) and three additives (a stabilizer, light stabilizer, and weather resistant). The composition for one compounded grade (R1 30%- R2 70%) is shown in Table 1. The materials were extruded in an intermeshing 25.5 mm, 27 kW twin co-rotating screw extruder (TSE). The experiment was designed with the three processing parameters (temperature, speed, and flow rate). The additives were mixed with the resins at a 100:0.86 ratio and were batch blended by super floater in ratio per weight to assure higher consistency.

Table 1: Composition of compounding material for PC.

<table>
<thead>
<tr>
<th>S.No</th>
<th>Ingredients</th>
<th>Material Name</th>
<th>PPH</th>
<th>weight</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>R1</td>
<td>Bisphenol A (BPA)</td>
<td>30</td>
<td>4.95</td>
<td>gm</td>
</tr>
<tr>
<td>2</td>
<td>R2</td>
<td>Bisphenol A (BPA)</td>
<td>70</td>
<td>10.05</td>
<td>gm</td>
</tr>
<tr>
<td>3</td>
<td>F1</td>
<td>Weather resistant(L)</td>
<td>0.035</td>
<td>0.00525</td>
<td>ml</td>
</tr>
<tr>
<td>4</td>
<td>F2</td>
<td>Stabilizer (Liquid)</td>
<td>0.065</td>
<td>0.00975</td>
<td>ml</td>
</tr>
<tr>
<td>5</td>
<td>F3</td>
<td>Light Stabilizer</td>
<td>0.2</td>
<td>0.03</td>
<td>gm</td>
</tr>
<tr>
<td>6</td>
<td>White</td>
<td>White Pigment</td>
<td>0.278</td>
<td>0.041625</td>
<td>gm</td>
</tr>
<tr>
<td>7</td>
<td>Black</td>
<td>Black Pigment</td>
<td>0.036</td>
<td>0.0054</td>
<td>gm</td>
</tr>
<tr>
<td>8</td>
<td>Red</td>
<td>Red Pigment</td>
<td>0.175</td>
<td>0.02625</td>
<td>gm</td>
</tr>
<tr>
<td>9</td>
<td>Yellow</td>
<td>Yellow Pigment</td>
<td>0.071</td>
<td>0.01065</td>
<td>gm</td>
</tr>
</tbody>
</table>

**Experimental Setup**

Producing a specific colour plastic using extrusion compounding requires proper operating conditions. Changes in the operating conditions will affect color. In the present work, the operating conditions were varied in a controlled manner to study their effects on color. Three parameters including temperature, speed, and feed rate were varied individually to three different levels, while keeping all other parameters fixed.

The experiments were set up as follows: The recommended processing temperatures were 230°C, 255°C and 280°C with a speed of (750 rpm) and flow rate of (25 kg/hr) fixed. A similar procedure was used for both the speed and flow rate. The following are recommended: flow rate was 20 kg/hr, 25 kg/hr, and 30 kg/hr, with a constant speed (750 rpm) and temperature (255°C). Lastly, focus extended on the recommended processing speeds of 700 rpm, 750 rpm and 800 rpm with a flow rate of (25 kg/hr) and temperature (255°C) fixed, [1-4].

Extrusion was performed on co-rotating intermeshing-TSE to assure a uniform melt mixing. Three parameters were varied individually to three different levels. While keeping the other parameters constant, one parameter was varied; the others remained fixed at feed rate, screw speed, and temperature, while studying their effects on colour shifts. Experimentation was focus on varying the speed parameter; the others remained fixed at feedrate and temperature. Upon exiting the die, the extrudate was quenched in cold water, dried using air and then converted in to pellets using a pelletizer. These pellets were then molded by using injection molding into three rectangular colour chips of (3x2x0.1") size. These were processed at 85 ton, at about 1000 PSI, and at 280 °C. The specimens were then dried in the lab at room temperature. They were measured at 3 different spots. The colour difference (dE*) tristimulus values of these coupons were measured and characterized by spectrophotometer, using the CIELAB Colour Space system which is widely used in the industry. It provides 3 tristimulus colour readings L*, a*, and b* values [22], target values (Tg): CIE (L*, a*, b*) = (68.5, 1.43, 15.7).

The colour chips were pressed into a 25mm disk shape mould by hot a presser to prepare 25mm dia and 1.2 mm thick chips for rheological characterization measurement. For the microscopy dispersion test, tin sheets were prepared either by molded colour chips by hot presses or microtomated into thin slices.

**Results & Discussion**

**Effect of Processing Parameters on dE**

**Effect of Temperature and Feed rate**

In our earlier work, the effect of processing parameters on colour of tristimulus values (L*, a*, b*) were characterized by using three processing parameters (General Trends) (GT) [3]. An increase in temperature and feed rate decreases the colour differences (dE*), see Figures 1 and 2. The colour difference (dE) approaches to 0.3 as temperature range approaches 255°C and stays such until 280°C [27].

![Figure 1. Effect of Temperature on dE*](image-url)
Figure 2. Effect of feed rate on dE*.

Figure 2. Illustrates the effect of feed rate on the colour difference (dE*) at 25kg/hr and 30kg, the minimum colour difference (dE*) also approaches to 0.3

Figure 3. Shows the effect of the screw speed. The colour difference initially decreases and increases as the speed exceeds above 750 rpm.

Effect of Speed

The three figures indicate, level two exhibits the minimum dE* value. (dE*)starts to exhibit improvement at the central point for the three processes. Moreover, the deviation in (dE*) increases with an increased speed, compared to the other two processes. This behavior may be due to shear rate having basically the same effect on speed as it does with temperature and feed rate.

Figure 3. Effect of Screw Speed on dE*.

The speed increases from 700 rpm to 800 rpm, while the feed rate at 25Kg/hr and temperature at 255°C are fixed. The colour values of dL* and db* are lower than the target values at three levels; except the da* colour difference value is slightly positive at 750rpm (See Fig.4). Fundamentally, this is an indication that the red inorganic pigment was better dispersed in the center point than in the 700 and 800 rpm samples.

Colour differences are (dL*,da*, db*, dE*, dC*): dL* is in negative for darker, da* is in positive for more red, and db* is in negative for bluer. DE or dE* is the total colour difference. The lower colour difference is at 750 rpm; dC* is negative for chromaticity. The negative dC* and db* values indicate the chromaticity of a bluer hue originate from incomplete dispersion. The transparent polycarbonate plastics do not scatter light; consequently to achieve a certain opacity level, white pigment is added to create scattering. Therefore, it affects the apparent colour strength [23, 24, and 25]. The negative of chromacity may be due to the low chroma in the compounded plastic grade of the translucent polycarbonate resins.

At a low speed (e.g., 700 rpm), the extruded material has shown a lower shear heat or lower shear rate. Agglomeration can, in principle, take place in zones with low shear.

With increasing the speed to 750 rpm, deagglomeration occurs in zones of high shear. Raising the screw speed higher than 750 rpm, the colour differences are decreased because of the relatively low overall shear forces through the entire mixing zones, which separate the pigment particles immediately from the composition.

In addition, directly after raising the speed to 800rpm, extruded resin material was increased with shear heat transition and the PC composites were confined to higher shear rates at the higher speed of 800 rpm.

The difference in the state of the pigment dispersion was proven also by measurements of the colour differences between the 700rpm, 750rpm and 800rpm samples relative to the most commonly used system in plastics which is CIE 1976 (L* a* b*) colour space or CIELAB, which allows the specification of colour in terms of a three-dimensional space.

The data are represented in Fig. 4. The total colour difference dE* increases at 700 and 800, but decreases at the center point 750rpm. The negative dC*, da* and db* values indicate the green-blue hue, originating from incomplete dispersion of the pigments. Pigment particles in sample 750rpm were better dispersed than the other two samples. Again, differences in colour measurement are in good agreement with the image and particle size distribution analysis results; (see Figures 4-9).

Figure 4. CIELAB colour differences for speed
Rheological behavior

Rheological measurement was conducted by rotational rheometer at 230°C, 255°C, and 280°C. The main factor influencing the pigment dispersing in polymer is the viscosity. For pigment wetting rapidly, the viscosity must be low, however for rapid deagglomeration, it should be high. The applied temperatures for extrusions of the polycarbonate compound are 230, 255, and 280 °C. As temperature increases, complex viscosity ($\eta^*$) decreases. Figure 5 shows when frequency increases, it causes a drop in the melt viscosity (shear thinning). As temp increases, the onset of shear thinning occurs at higher frequency. The relationship between the shear stress $\tau$ and shear rate $\dot{\gamma}$ is described as follows.

$$\tau = \eta \dot{\gamma} \quad (1)$$

$$\dot{\gamma} = \frac{mxDxN}{60s} \quad (2)$$

Wetting is the more important factor for better dispersion, since the shear forces, created during extrusion, must be transferred onto the pigments, as a result, to increase the efficiency of deagglomerates [26].

The low viscosity is produced by high temperature; therefore, both pigment wetting and the colloidal stability of the dispersed pigment are improved.

![Figure 5. Complex viscosity of PC composite.](image)

Evaluate Dispersion at Variant Speed

Extrusion was carried out at a screw speed of 700rpm, at center point (750rpm), and 800 rpm, for which a screw speed of 750rpm was used and found significant lower colour difference. The results of the particle size and dispersion analyses are shown in Figures 6-9. The dispersions were characterized with digital optical microscope (DOM) analysis in combination with image scanning electron microscope (SEM) and MCT scanner.

Figure 6 shows dispersions of 30/70 wt % of PC compound prepared at 700, 750, and 800 rpm, at fix 255°C and 25 kg/hr. With the increase of screw speed, the peak of the distribution becomes narrow except the lower and higher speed, the average size of particles are 0.96, 0.79, and 0.88μm for 700, 750, 800rpm respectively.

At 750rpm higher peaks are shown at 52.43% in comparison to 42.8% at 700rpm and 44.98% at 800rpm.

![Figure 6. Pigment size distributions for Speed](image)

For optical microscopy a tin sheet was processed either by hot presses or rotary microtomed. Colour chips were used to get the images into DOM at magnification (1000X-5000X), analyses are shown in Figures 7.

Figure 8 illustrates the characterization of the pigment size distribution for screw speed. SEM-Jeol5500 is a high-resolution microscopy scan system. The samples were covered with a thin conductive colloidal graphite coater, used to characterize the compounding material, having a magnification of 1000X

Figure 9 shows the particle size distribution (PSD) graphs for the samples used are in good agreement with the input in the process. The image was characterized on a high resolution 3D X-ray detector; 10 mega (4000 x 2300) pixels. It shows a consistent distribution and a spherical pigment shape. The micro tomography measurements for the center point treatment chip were carried out by using CT scanner, at a speed of 750 rpm.

![Figure 7: (a) DOM micrograph at (a) 700rpm and (b) 800rpm](image)
The interaction relationship between tristimulus colour values (dL*, da*, db*, dE*) and processing parameters showed that the minimum color difference throughout the experiment was 68.42 for L*, 1.47 for a*, 15.35 for b*, and 0.34 respectively. The colour difference values (dE*) decrease significantly with the increase of the temperature and feed rates. Therefore, it shows a consistent higher peak distribution. Steadily, increasing the speed to a medium range (i.e., center point at 750 rpm) shows a significant minimum colour difference; therefore, it shows a consistent higher peak distribution and a spherical pigment shape. At a higher screw speed, these processing parameters can generate large shear forces and a frictional heat state that may arise and affect the heat stability of pigment and damage other components of the polymer matrix. The high velocities raise the material temperature; the failures of appearance, physical properties, or degradation are possible.

Agglomeration can in principle, take place in zones with low shear; deagglomeration occurs in zones of high shear. This spectacle is employed to increase pigment wetting, reduce particle size or prevent pigment agglomeration, improve the rheological behaviour of flow, dispersion process, and a higher peak distribution occurs, ultimately reducing the colour mismatch differences.

Conclusion

Figure 8: SEM micrograph of Polycarbonate grade compound at (a) 700rpm (b) 750rpm (C) 800rpm

Figure 9: Pigment distribution (a) particle shape,(b) Distribution , Process at center point using - μCT Scanner.
References


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