The Ideal Temperature Setting of Polymer Blends: 
Investigational Characterization Effects of Color Matching

J. Al Sadi, A. Hawary
Jadara University, Irbid, Jordan
Email: jamal.alsadi@ontariotechu.net, j.alsadi@jadara.edu.jo

Abstract. The goal of this work, to study the characteristics of a mixture of the two Lexan polycarbonates, having different (MFI) the characterizations were run in two stages. The first stage: The two-polycarbonate resins of different weight percentages and MFI were melt-blended using (Coperion) a Co-rotating twin-screw extruder (SB). This grade rheologically characterized using the rotational rheometer and MFI tester. The same material was included; the same composition was blended in steps of eleven in a Thermo Haake Mini Lab II twin-screw micro compounder (ML). Further experimental design dictated a sample without pigment and additive (WOP) consisting of only PC resin, a sample with PC resin mixed with pigment and additives (WP) to study the characterization of polycarbonate formulation at different temperatures were also analyzed. They were rheologically characterized using the rotational Rhoemeter, MFI tester, Particle size analyzer and Scanning electron Microscopy (SEM). This work was mainly focused on examining the viscosity at the mid (Centre) temperature =255°C. The focus was extended to the polymer grade to extract the impact of the rheological and dispersion characteristic at 255°C to study their correlations in the viscosity data and their bearing effects on color matching.

1. Introduction
Producing plastic with a marketable color requires the addition of one or more resins; however, achieving the correct color in the first attempt is a challenge. The color properties of the polymer are directly affected by formulation for various ratios of PC resins. The viscosity for a certain ratio may vary due to the effect of formulation such as (PCs weight percent blends, pigments, and additives), and processing parameters. A few studies have been carried out by various researchers regarding the effects of processing parameters by the dynamic mixing in a screw extrusion during polymers’ compounding [1, 2]. In order to control the mentioned points, a system of additives and resins can be formulated to modify viscosity and improve thermal stability to have better dispersion [3]. Researchers have studied the importance and effects of adding the pigment to the base resin; especially, it has been shown that incorporating additives into polymeric materials during production often unpredictably affects rheological and optical properties [4-6]. However, rheological properties are an essential link between the processing steps and the final performance of the product [7-8].
Polymer blending is an important field of polymer science and has been reviewed by many scientists. The PC/PBT blends are transparent in the melt state, and most transparent in the solid-state are partially miscible blends, as was shown by Sanchez et al. [8]. In their work, Liang and Gupta (2000) studied the rheological properties of a recycled PC blended with virgin PC; they stated that separated PC could be added to pure PC up to the 15% level without significantly varying the properties of pure PC [9]. Khan et al. developed in their study characterizing alloys of ABS and PC through blending these two materials [10]. Lee S. et al. studied PC/polyester blends’ rheological behavior and their phase behaviour [11]. However, they found that the blends do not follow the rule of mixtures which is common in all studies.
The particle sizes, fillers (pigments and additives), and processing parameters like temperature affect the rheology and the pigmented polymer's color. The significance of the processing variables was correlated with the rheological result [12]. Difficulties with the dispersion of pigments or obtaining a uniform blend, can be overcome by reducing the resin viscosity and extending the mixing time [13]. A system of additives and resins can be formulated to modify viscosity, increase mechanical prosperities, improve thermal stability, or improve wear performance [14]. Many researchers have studied the importance and effects of adding the pigment to the base resin, especially since it has been shown that incorporating additives into polymeric materials during production often affects rheological, mechanical, and optical properties unpredictably [15, 16,17]. Rheology, as a conception, applies to all types of materials (solid, semi-solid and fluid) like polymers and their composites. Rheological properties are an important link between the processing steps and the product's final performance [18, 19].

This study included additional literature reviews, investigation for the materials' rheological characterization, and Melt flow adjustment (MFA). Haake mixer and viscosity measurements [20, 21, 22, 23, 24]. More recent studies have indicated that the effective processing conditions were dependent on the materials used or blending different materials (resins, additives, pigments, and fillers) to improve the rheological characteristics [25, 26, 27, 28]. The effect of particle sizes, parameters like temperature, pressure, and amount of additive can be determined and used for improving processing conditions [29, 30].

Many researchers have studied the degradation pathway for the polymer. The degradation Photo-oxidative of weathered plastic constituents is due to the combined effect of UV light (295nm to 400nm), which is existing in the solar spectra, moisture, and heat [31]. Most polymers' primary degradation pathway is measured to be their reaction with atmospheric oxygen, consequential resulting in chain cleavage, crosslinking, and discoloration. Although photodegradation of polycarbonate (PC) resin extensively been measured [32-34], more investigation was continuing about the effect chemistry involved. When manufacturing plastics, e.g., bisphenol, a polycarbonate (PC), are used in outdoor applications, oxygen, sunlight, humidity, and can be the source of degradation of the polymer leading finally to mechanical failure and loss of optical and aesthetical properties [35]. The instrument settings were as follows: color space – CIE Lab; measurement mode; observer angle – 10° and D65 illuminant (light source) [36]. Taking the above results into account makes sense that the addition of colorant will minimize the viscosity, and the absorbance mechanisms here for degradation and yellowing will be decreased as was seen for samples subjected to an earlier study [37-38].

This work was mainly focused on examining the viscosity at the mid (Centre) - temperature =255°C. The focus was extended to the polymer grade to extract the impact of the rheological and dispersion characteristic at 255°C to study their correlations in the viscosity data and their bearing effects on color matching.

2. Experimental procedure and materials
This research investigated two PC resins grades referred to as R1 and R2 in the thesis, each having a different melt flow index (MFI). One had an MFI of 25, and the other had an MFI of 6.5g/10min, respectively, here forth. The resins are manufactured by General Electric (GE) and traded under the name of Lexan. Four different color pigments, black, white, red and yellow, were used. Also, three additives were used called F1, F2, and F3 in this work. One was a stabilizer, one a light stabilizer, and the third offers weather-resistant properties. Materials were extruded in an intermeshing, 25.5 mm, Coperion twin co-rotating screw extruder(SB). The total weight of the color additives (pigment and additive) was 0.86%. The two PC resins, R1 and R2, were used in a ratio of 30 and 70 wt. %, respectively. The additives and pigments were mixed with the resins at a 100:0.86 ratio and were blended by a super floater. [39-40] Table 1 shows the formulation used.
Table 1. The composition of compounding material (g3)

| Ingredients  | Material Name      | PPH |
|--------------|--------------------|-----|
| R1           | Bisphenol A (BPA)  | 30  |
| R2           | Bisphenol A (BPA)  | 70  |
| F1,F2,F3     | Additives          | 0.3 |
| W,B,R,Y      | Pigments           | 0.56|

To study the effects of the blending of two resins, a Thermo Haake Minilab II twin-screw micro compounder (ML) was used to prepare PC formulations with both resins individually as well as by varying the composition of the resins in steps of ten for a total of eleven blends. The concentration ratios between the two polycarbonate resins used in %R1/%R2 were 100%/0%, 90%/10%, 80%/20% and 0%/100%.

3. Results & Discussion

3.1 Rheological behaviour at 255°C.

The PC blends were rheologically characterized at 255°C using the Ares rotational rheometer. Results of complex viscosity at 255°C vs. frequency for the blends without additives (WOA) are presented in Figure 1.a. Blend 100% R1 exhibited the lowest viscosity with a solid-like behaviour at the low frequencies compared to blend 0% R2, while both presented shear thinning behaviour at the higher frequencies. With the addition of R2 into the R1 resin, a viscosity of the blends increased substantially. The dynamic responses of R1/R2 blends do not appear to follow the “rule of mixing,” which expects a variation of the parameter to be in proportion to the amounts of constituent elements of the blend. The increase in viscosity with the addition of R2 may be due to the solvation of the highly entangled structure of R2 molecules in R1 molecules. Similarly, Figure 1.b presents the viscosity of blends where the pigments and additives (WA) are added. Resin R2 (100%) shows a higher viscosity and exhibits a strong shear thinning effect while Resin R1 (100%) shows a lower viscosity in comparison to 100% R2 and the other blends. In general, the blends’ viscosity decreased in the presence of pigments and additives by approximately 10-20%.

![Figure 1a](image1.png)

![Figure 1b](image2.png)

Figure 1. Complex viscosity at a temperature of 255oC: (a) without additives and pigments, and (b) with additives and pigments [36]
The change in viscosity at the higher end of the shear rate range occurred due to shear thinning – the slippage of chains over each other and molecular alignment. The increase in temperature led to a decrease in viscosity because of the mobility of polymer molecular chains increases with temperature.

**Table 2.** Effect of processing parameters on color in terms of tristimulus values.

| Parameters          | °C   | kg/hr | rpm  | dE*  |
|---------------------|------|-------|------|------|
| Temperature (°C)    |      |       |      |      |
| 230                 | 25   | 750   | 1.096|
| 255                 | 25   | 750   | 0.346|
| 280                 | 25   | 750   | 0.303|
| Feed Rate (kg/hr)   |      |       |      |      |
| 255                 | 20   | 750   | 0.44 |
| 255                 | 25   | 750   | 0.346|
| 255                 | 30   | 750   | 0.320|
| Screw Speed (rpm)   |      |       |      |      |
| 255                 | 25   | 700   | 0.633|
| 255                 | 25   | 750   | 0.347|
| 255                 | 25   | 800   | 0.623|

**Figure 2.** Complex viscosity for R1 30% blends at 255°C

According to Figure 2, the viscosity value is less at 255°C than unity at temperatures of 230 °C, indicative of lower viscosity values and greater wettability. Initially, the composites were rheologically characterized using an Ares-G2 rheometer, in three different dynamic modes at 230, 255, and 280°C. Figure 2 shows the complex viscosity behaviour vs. frequency, measured at a frequency of 10Hz. a. Complex viscosity exhibited mostly constant behaviour in the range from 0.1 up to a strain magnitude of 100%. As expected, complex viscosity decreased as the temperature increased. The relationship between viscosity and strain was constant at lower strains and all temperatures but started to show shear thinning behaviour at higher strains. Also, as the temperature increased, shear-thinning began at the higher strains [39;40]

**3.2 Rheological behaviour of PC blends (Micro-TSE) – ML**

The rheological properties of polycarbonate blends were characterized without (WOA) and with pigments and additives (WA) while changing the blend proportions in discrete equal intervals. The PC blends were rheologically characterized at 255°C, as shown in Figure 1, using the rotational rheometer. The blending of two polycarbonate resins (R1, R2) was studied by modifying the formulations of the pigments and additives in controlled steps; This resulted in eleven batches named ML. A decrease in viscosity due to the presence of additives was observed. An increase in temperature at 255°C decreases the viscosity and improved the surface tension of polymer blends, thereby increasing the wetting properties of pigments. The increase in R2 in the mixture of the two-polycarbonate formulations significantly decreased the melt flow index; the melt flow index for R1 was 28.22 gm/10min, and that of R2 was 8.10 gm/10min. [39]
3.3 Rheological behaviour of compounded plastic (Coperion TSE) – SB
Grades were prepared using SB extruder and injection molding. Output samples were measured by oscillatory rheometry at three levels of temperature, 230, 255, 280°C. The temperature was found to affect the color difference. It was also determined that SB’s viscosity was different from that of ML for the same-compounded plastic.
Lower viscosity improves pigment wetting, lowers color difference and ultimately results in higher dispersion. Thus, the higher pigment dispersion occurred at the point of 255°C or greater. Compounding plastic at 255°C showed that the material exhibited a Newtonian plateau at low frequencies. At a high frequency or shear rate, the viscosity curve exhibited a much more significant decrease and exhibited non-Newtonian behaviour at or above 255°C. Viscosity at 255°C was quite adequate for wetting and deagglomeration of particles. Therefore, 255°C may be regarded as a rheological threshold composition and a good sign for higher colorant dispersion and lower color difference (dE*). In general, as frequency increased, the storage and loss modulus increased, and viscosity decreased. Thus, the reduced viscosity seems to exhibit a correlation with greater pigment dispersion leading to a reduced color deviation. As presented in Table 2 and Figure 2. [36; 40]. The blending and mixing process is paramount to the screw design. Therefore, the processing screw design of SB significantly results in a higher viscosity than ML. Compounded plastic's experimental characterization will enable us to perform the design of the proper experiments with consistent parameters. [36-40]

3.4 Effect of temperature on viscosity and color values
Table 2 illustrates the effect of temperature on the color difference (dE*) at 230, 255, and 280°C, for the samples produced at 750 rpm and a feed rate of 25 kg/hr. It indicates a reduced value of color difference as temperature increased. The best color difference value (dE*) occurred at higher temperatures 255 and 280°C, with dE*=0.3.

3.5 Evaluation of pigment dispersion
From analyzing pigment dispersion using the particle size analyzer (PSA) in the wet test, the raw pigment size was broken to lower size values, measured down to approximately 0.1-0.2 µm. Scanning electron microscopy (SEM) produced images to evaluate the pigment dispersion of raw pigment materials. The average pigment size measured approximately 0.1-0.2 µm. Lastly, the thin slices' pigment size results from the injection-moulded chips, imaged with SEM, recorded average particle sizes of 0.2 µm. The obtained measurement test typically agreed with the optimized measurement of the optimal minimum color difference. Furthermore, it could be used as a powerful tool to optimize compounding materials for the color matching process.

3.6 Viscosity and particle size distribution
Further investigation and correlation between viscosity and dispersion were carried out for different screw extruders at a temperature of 255°C. Figure 3 shows the viscosity (a) and particle size distribution (PSD (b)) measurements for the blends processed in Minilab (ML) and (SB). The blend processed in ML shows a lower viscosity than the one processed in SB. For the ML blend, a larger number of particles (69%) had a small particle size, compared to 50% for SB. This could be attributed to the lower viscosity of the melt, which caused greater wetting, leading to the molecular bonds to break to produce a more significant number of particles. The presence of additives, blending, extrusion and injection molding, high pressure, shear rate and shear heat could also cause the particles to break into smaller sizes. The compounds produced with SB showed a higher viscosity, which could be due to the smaller particle size distributed in the resin matrix. The most favourable result in terms of color was achieved when the blend exhibited higher viscous behaviour. Smaller particle sizes present a higher total surface area and thus give rise to a higher viscosity; therefore, a higher shear heating would not affect the color mismatch and degradation.
3.7 Morphological dispersion analysis

Figure 4 illustrates the compound grade's SEM micrograph for temperature processing parameters, with a magnification range of 500X-3000X. The pigment particles here came from the 30%-R1 sample. Agglomeration of the pigment occurred at lower processing parameters, e.g. 230°C. The total color difference, $dE^*$, increased at lower processing parameters, and higher agglomerations than that of the sample were produced at higher parameters.

At the higher temperature range (255°C and 280°C), the samples were exhibited a lower color difference. According to these results, the sample processed at 255°C showed better-dispersed pigment than that processed at 230°C. Agglomeration can, in principle, take place in zones with low shear. By increasing the temperature to 255°C, deagglomeration occurred in the zones with high shear. By raising the temperature to mid-255°C, the color differences started to decrease. This can be attributed to the relatively low overall shear forces through the entire mixing zones, which separate the composition's pigment particles.

The color difference values ($dE^*$) decreased significantly with the increase in temperature. It showed a consistent higher peak distribution. Equally, large shear forces and frictional heat were possibly generated that may have affected pigment's heat stability and caused damage to the other components of the blend. Consequently, failure in appearance, physical properties, or degradation is possible. Viscosity plays a crucial role in determining pigment wetting, pigment agglomeration, pigment flow and dispersion, and, ultimately, the color shifts. Particle size and color differences are reduced, and a higher peak distribution occurs with increasing the temperature. However, the color difference was reduced at the center level. In general, the color differences were reduced at the center level of the three processing parameters at (i.e. 255°C, 750 rpm, 25 kg/hr), and color output was improved.
4. Conclusion
At the centre level of temperature (255°C). Materials of different formulations processed with different screw configurations were attributes for several applications: It was found that at the centre level of temperature (255°C), smaller sized particles with better dispersion were observed. Scattering efficiency increased rapidly as the average particle size decreased to approximately 0.2 µm, and the optimal number of particles increased. It was also observed that agglomeration occurred in zones of high pigment size. The particle size analyzer (PSA) and imaged with SEM were used in the test to study pigment dispersion. It was found that raw pigment sizes were approximately 0.1–0.2 µm (particle diameter). Thin slices of chips were also imaged with SEM. The obtained measurements were exhibited the minimum color difference. A correlation between viscosity and dispersion was supported for the two screw extruders at 255°C. The blend exhibited higher viscous behaviour in SB than ML. Smaller particle sizes present a higher total surface area and thus give rise to a higher viscosity; therefore, a higher shear heating would not affect the color and degradation. The particle size distribution (PSD) measurements for the blends processed in Minilab (ML), Coperion extruder (SB), and the (SEM) micrograph of the compound grade were used to test the pigment dispersion, the pigment size was decreased to approximately 0.2 µm, deagglomeration occurred in the zones with high shear, and exhibited the minimum color difference (dE*)=0.3). The color values (dE*) decreased significantly by raising the temperature to mid-255°C and color output was improved.

5. References
[1] Syang-Peng, Rwei. Polymer Engineering and Science, 2001
[2] Wong, A and Lam, Y, J Polym Res 15:11–19, 2008
[3] Mishra, A, Kumar. Scientific Journal & Industrial Research 67 (2008).
[4] Dealy, J, and Wissburn, KF. In Melt Rheology and its Role in Plastics Processing: Van Nostrand New York: Kluwer Academic Publishers, 1990.
[5] Abdel-Goad, M, and Potschke, P. "Rheological characterization of melt-processed polycarbonate multiwalled carbon nanotube.," J. 2-6. 2005.
[6] Mange Sana, N, Chi kuku, RS, AN. "Journal of the South African Institute of Mining & Metallurgy / Saimm, 108, no. 4 (2008): 237-243.
[7] Zaeh, M and Hess, R. "Impact of stabilizers on the color of pigmented polymer articles Geneva." Switzerland, Additives Geneva, 2002.
[8] Sanchez, P. M. Remiro, Nazabal, J. J. Appl. Polym. Sci., 50, 995-1005 (1993)
[9] Liang, R. F. and. Gupta, R.K, 2000, Society of Plastic Engineering ANTEC, 2903 2907.
[10] M. Khan, "Rheological and mechanical properties of ABS/PC blends," Korea-Australia Rheology Journal, vol. 17, no. 1, pp. 1-7, 2005.
[11] Lee S, Mather P, Pearson DMMK. Journal of applied polymer science 59, 243-250, 1996
[12] Fatoni, Rois. Product Design of Wheat Straw Polypropylene Composite. Waterloo: University of Waterloo, 2012
[13] D. Meade, Introduction to colorant selection and Application Technology, in Coloring of Plastics Fundamentals, 2nd ed., R. Charvat, Ed., 2004.
[14] B. Mulholland, "Effect of Additives on the Color & Appearance of Plastics," ANTEC, Ticona Engineering Polymers, 2007.
[15] J. Birmingham, "Volatility of Titanium Dioxide Pigments in Polyethylene Film Extrusions," ANTEC, p. 3290–3294, 1995.
[16] E. Jaffe, C. Campbell, S. Hendi and F. Babler, "Rheologically Effective Organic Pigments," J. Coating Technol, p. 47–54, 1994.
[17] D. Williams and M. Bevis, "The Effects of Recycled Plastics and Compound Additives on the Properties of Injection Polypropylene Sci, p. 2834–2842, 1980.
[18] A. Mishra and Kumar, Scientific Journal & Industrial Research, vol. 67, 2008.
[19] D. J and K. Wissburn, In Melt Rheology and its Role in Plastics Processing: Theory and Applications Van Nostrand Reinhold, New York: Kluwer, 1990.
[20] K. H, H. Lee and J. Wook, "Rheological properties of branched polycarbonate prepared by," Korea-Australia Rheology Journal, vol. 19, no. 1, pp. 1-5, 2007.
[21] P. R. Hornsby, "Rheology, Compounding and Processing of Filled Thermoplastics," Mineral Fillers in Thermoplastics I, pp. 155-217, 1999.
[22] A.-G. M and P. Pötschke, "Rheological characterization of melt-processed polycarbonate multiwalled carbon nanotube composites," Fluid Mech, pp. 2-6, 2005.
[23] C. K and W. Cook, "Structure-Property Relationships of Blends of Polycarbonate," Polymer engineering and science, vol. 43, no. 11, pp. 1727-1739, 2003.
[24] B. V and P. Kitchloo, "Rheological characterization of polycarbonate resins and its applications," Society of Plastics Engineers, pp. 103-106, 2011.
[25] H. S, K. Tapan and S. Chattopadhyay, "Dynamic and capillary rheology of LDPE-EVA–based thermoplastic elastomer," vol. 31, no. 3, p. 377–391, 2010.
[26] S. E, A. Hossein and Behravesh, "Rheological Investigation of Wood-Polypropylene Composites", Journal of Polymers and the Environment, pp. 998-1006, 2012.
[27] A. S, W. Leelapornpisit, A. Derdouri and P. Carreau, "Morphology and Properties of Microinjected Polycarbonate/MWCNT Nanocomposites," in 66th Annual Technical Conference of the Society of Plastics Engineers, 2008.
[28] N. Mangesana, R. Chikuku and A. Mainza,"The effect of particle sizes and solids concentration on the rheology of silica sand-based suspensions Journal of the South African Institute of Mining & Metallurgy / SAIMM, vol. 108, no. 4, pp. 237-243, 2008
[29] M. Zaeh and R. Hess, "Impact of stabilizers on the color of pigmented polymer articles Geneva," Swissman, Clariant Business Unit Additives - Geneve, 2002
[30] J. E. Pickett. In-Service Life Prediction: Challenging the Status Quo; J. W. Martin; R. A. Ryntz; R. A. Dickie, Eds., 2005; pp 93-106.
[31] A. Factor; W. V. Ligon; R. J. May. Macromolecules 1987, 20, 2461-2468.
[32] P. Hrdlovic. Polymer News, 2004, 29, 187 -193.
[33] J.S. Humphrey Jr; A. R. Shultz; D. B. G. Jaquiss. Macromolecules, 1973, 6, 305-314.
[34] H. Zweifel. Stabilization of Polymeric Materials; Springer, 1998.
[35] ASTM Standard D2244, 1993 (2000), “Standard Test Method for Calculation of Color Differences,” ASTM International, West Conshohocken, PA
[36] J. Alsadi., Color Mismatch in Compounding of Plastics: Processing Issues and Rheological Effects (Doctor of Philosophy in Mechanical Engineering. The Faculty of Engineering and Applied Science University of Ontario Institute of Technology). Ontario, Canada (2015).
[37] J. Alsadi, U. Saeed, S. Ahmad, G. Rizvi and D. Ross., Processing issues of color mismatch: Rheological characterization of polycarbonate blends, Polymer Engineering & Science, Volume 55, Issue 9, September 2015, Pages: 1994–2001, Version of Record online: 8 DEC 2014.
[38] J. Alsadi, Investigation of the effects of Formulation, process parameters, Dispersions, and Rheology on using combined Modelling and experimental Simulations, published in Materials Today: Proceedings and published on April 2, 2019, ELSEVIER. Paper May 22, 2019. Volume 13, Part 3, 2019, Pages 530-540
[39] J, Al Sadi, Systematic review: Impact of Processing Parameters on Dispersion of Polycarbonate Composites, and Pigment Characterized by Different techniques, submitted on April 3, 2020, to Materials today: Proceedings Elsevier, Published on June 10, 2020. Volume 27,issue 4, pp -3254-3264,DOI information: https://doi.org/10.1016/j.matpr.2020.05.027
[40] J. Alsadi, “Materials Processing Issues and Experimental Study for Rheology and Dispersion of Polycarbonate Grades: Twin Screws design.” the manuscript has been accepted to the Second International Conference on Industrial, Systems & Manufacturing Engineering (ISME’19) arranged by, Jordan Engineers Association (JEA) and German Jordanian University (GJU), accepted on October 17, 2019, Presented on November 11-13, 2019

Acknowledgments
The author would like to extend his gratitude for the support provided by Engineer Nour Al Sadi at Ontario technical university, Canada for excellent assistance.