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FULL PAPER

Bio-based polyethylene masterbatch preparation and investigation of the effect of process conditions on pigment dispersion

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Abstract

Although the importance of bio-polyethylene is increasing, academic research on the subject is still limited. The dispersion level of pigments and functional additives plays a crucial role in achieving the desired effect from polymer composites. This study investigates the effects of additives and process conditions on pigment dispersion in bio-polyethylene masterbatches using four different factors and three different parameters by the Taguchi method. To achieve this, the relative colour strengths of extruder-produced masterbatches were measured using a spectrophotometer, blown film samples were visually observed and filter pressure values were examined in a filter tester. The results were subjected to Taguchi analysis to determine which factors and parameters are important to the process and which can be neglected. The study identified the physical properties of the wax, extruder screw speed, barrel temperature and mixer time as the most effective factors on pigment dispersion, respectively. Micronised wax was found to be the most critical parameter among wax types, while mixer mixing time was found to be the least important. It was observed in all the experiments that the colour strength obtained from a constant pigment content in the masterbatches was improved by increasing the dispersion and reducing the number of agglomerated particles.

1 | INTRODUCTION

Masterbatch is a mixture of polymer carrier, pigment, wax and additives used to colour plastic objects. The granules contain a high percentage of pigment and are mixed with the main plastic carrier in varying proportions according to requirements. These mixtures are melted together under high temperature and pressure in machines called extruders. The dispersion varies depending on the parameters used at this stage. Some factors that affect the dispersion are mixing time, extruder profile temperature, feed rate, screw speed, screw design, shear stress and dispersants.[1–4]

Masterbatch dispersion is a critical parameter that is often overlooked in many studies. To achieve the highest colour strength that the pigment can provide in coloured masterbatches, or the most pronounced effect desired from that additive in masterbatches with different additives, a good dispersion study must be conducted. A poorly dispersed masterbatch will not yield the desired result in terms of colour strength or additive efficiency. Improper dispersion of pigment or additives can weaken

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the masterbatch matrix, causing mechanical instability. This can lead to an inadequate colour or effect, prompting an increase in the amount of pigment or additive to achieve the desired strength, which may impact the research or product design.[5–8]

A variety of waxes and additives have been developed for masterbatch dispersion. The most commonly used type is homopolymer polyethylene wax. However, other waxes, such as montan, copolymer, oxidised, polypropylene and Fischer–Tropsch, are also utilised. It is important to consider particle size and physical form, in addition to the chemical structure of the waxes. Homopolymer wax (HW) can have particle sizes of 500 μm or less, while micronised wax (MW) can have particle sizes of 15 μm or less. Liquid wax emulsions typically have particle sizes in the nanometre range. It is important to note that these particle sizes can affect the performance of the wax in various applications.[9–13]

Bio-based polyethylene (BIO-PE) is a renewable plastic that is produced without the use of fossil fuels. Unlike conventional polyethylene, which is derived from petroleum, BIO-PE is made from biomass such as sugar cane, corn or other plant-based sources. The sugars in the biomass feedstock are converted to ethylene and then polymerised to form plastic. It is important to note that BIO-PE is not biodegradable by nature. However, this material is more environmentally friendly and sustainable than petroleum-based polyethylene because it achieves negative carbon emissions during production.[14–19] This type of polymer is commercially marketed as green, and it is a type of polymer that is commercially available and has a growing number of applications in various industries, such as packaging, automobiles, agriculture and consumer goods. However, its use in masterbatch production for colouring plastics is limited, both commercially and academically.

This paper addresses the topic of dispersion, which has been largely overlooked in many scientific studies. It examines parameters that are often considered proprietary by masterbatch manufacturers and rarely studied in the scientific literature. The objective is to optimise dispersion and eliminate unnecessary parameters to save time and obtain more accurate data from scientific studies. The current study investigated the dispersion of CI Pigment Blue 15:1 (alpha-crystal modification of copper phthalocyanine), which exhibits excellent acid-alkali resistance, high light fastness and temperature resistance,[20,21] in a BIO-PE carrier developed as an environmentally sustainable solution.

A series of experiments was conducted to assess the impact of varying wax types and associated mixer and extruder parameters. The experiments were analysed for significance using the orthogonal matrix Taguchi method. The variables were calculated methodically, analysed statistically using Minitab 21.1.1 software and reported.

2 | EXPERIMENTAL

2.1 | Materials

Biopolyethylene resin from Braskem, with a minimum of 95% bio-based carbon content, was used to produce the masterbatch. The melt flow index of this polymer is 2.7 (190°C, 2.16 kg) g/10 min. CI Pigment Blue 15:1 Allo-ment Alpha Blue AGK (alpha-crystal modification of copper phthalocyanine), from Alliance Organics LLP, was used as the pigment. Luwax A homopolymer polyethylene wax from BASF (D_{50} = 500 μm), Ceretan MX 9815 micronised polyethylene wax from Münzing (D_{50} = 15 μm) and Hordamer PE 02 wax emulsion (WE) from BYK Additives & Instruments (D_{50} = 130 nm) were used for pigment dispersion.

2.2 | Manufacturing

Taguchi orthogonal arrays are a method of designing experiments and can be used to analyse the influence of many factors simultaneously. In traditional full factorial experimental designs, all possible combinations are tested, which is very costly in terms of time and resources. Taguchi orthogonal arrays, on the other hand, enable analysing these effects by conducting the minimum number of experiments across several factors. The masterbatch production process involved developing the Taguchi L9 orthogonal array formulations using the Minitab program (Table 1). Four parameters were utilised at this stage: wax type, extruder speed, mixer time and extruder temperature profile. There were three different levels for each of these four parameters. At the end of the experiments, the signal-to-noise (S/N) ratio (smaller is better) (Equation 1) values were analysed, considering the filter pressure value (FPV). This analysis was conducted because a lower FPV is expected to result in improved dispersion values.

The S/N ratio is used to measure the robustness of a system's performance in the face of variability. This ratio evaluates the strength of the signal (desired performance) against noise (unwanted variability).

$$\frac{S}{N} = -10\log\left(\Sigma\frac{Y^2}{n}\right) \quad (1)$$

Here, "n" represents the number of measurements, and "Y" represents the value of each measurement. A

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TABLE 1 Taguchi L9 orthogonal array parameter table.

No.	Sample	Wax type	Extruder speed (rpm)	Mixer time (min)	Extruder temperature (°C)
1	BIO-PE/HW/01	Homopolymer	100	5	160
2	BIO-PE/HW/02	Homopolymer	200	10	200
3	BIO-PE/HW/03	Homopolymer	300	20	240
4	BIO-PE/MW/01	Micronised	100	10	240
5	BIO-PE/MW/02	Micronised	200	20	160
6	BIO-PE/MW/03	Micronised	300	5	200
7	BIO-PE/WE/01	Emulsion	100	20	200
8	BIO-PE/WE/02	Emulsion	200	5	240
9	BIO-PE/WE/03	Emulsion	300	10	160

Abbreviations: BIO-PE, bio-based polyethylene; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

TABLE 2 Table of masterbatch formulations.

No.	Sample	BIO-PE (wt%)	CI Pigment Blue 15:1 (wt%)	Homopolymer wax (wt%)	Micronised wax (wt%)	Wax emulsion (wt%)
1	BIO-PE/HW/01	60	25	15	-	-
2	BIO-PE/HW/02	60	25	15	-	-
3	BIO-PE/HW/03	60	25	15	-	-
4	BIO-PE/MW/01	60	25	-	15	-
5	BIO-PE/MW/02	60	25	-	15	-
6	BIO-PE/MW/03	60	25	-	15	-
7	BIO-PE/WE/01	60	25	-	-	15
8	BIO-PE/WE/02	60	25	-	-	15
9	BIO-PE/WE/03	60	25	-	-	15

Abbreviations: BIO-PE, bio-based polyethylene; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

higher S/N ratio indicates that the system works well without being affected by unwanted changes (ie, noise). Therefore, those levels with the highest S/N ratio were considered the most successful. During the production of masterbatch, the percentages of the formulation components were kept constant. This enabled observations of the effects of wax type and the process parameters (Table 2).

The BIO-PE was ground using a grinder to achieve a more uniform blend. The pigment, polymer powder and wax were added simultaneously to a laboratory turbo mixer (Labtech Engineering) and mixed at 1500 rpm for 5-20 minutes, depending on the parameters. The resulting mixture was then transferred to a laboratory extruder (Labtech Engineering) with a 20-mm diameter twin screw and an L/D ratio of 44:1. Temperature profiles ranging from 160 to 240°C were used, while the screw speed of the extruder was maintained between 100 and 300 rpm. The masterbatches were then cooled to room temperature and held in a dry air dryer at 80°C for 8 hours. To observe agglomerated particles, films with a thickness of 100 μm were produced using 4% masterbatch (Labtech Engineering).

Tensile test specimens were produced using an injection moulding machine (Haitian) with a clamp force of 600 kN, in compliance with ASTM D638 Type 1. The injection temperatures for each specimen were maintained at a constant range of 210-230°C. The samples were produced directly in the injection moulding machine without being mixed with any polymer. The

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purpose of this step was to observe the mechanical test results of the obtained compounds based on the production parameters.

2.3 | Characterisation

The Fourier Transform–infrared (FTIR) spectra of the masterbatches were obtained using an IR spectrometer (Bruker ALPHA-II) equipped with an attenuated total reflectance crystal. The spectra were collected from 4000 to 450 cm^{-1} with a data range of 1 cm^{-1} and a resolution of 4 cm^{-1}. A background scan was performed between each sample.

To measure the colour strength, the masterbatches were mixed with titanium dioxide (TiO_2) in a thermokinetic mixer (Gülnar) at a pigment-to-TiO_2 ratio of 1:10. Although the thermokinetic mixer has no heating element, its special blade design and 2000 rpm mixing speed can melt the mixture by increasing the barrel temperature to 190°C through the impact effect on the granules. The molten samples were then moulded in a press machine (Labtech Engineering). The relative colour strength (RCS) was determined by comparing the K/S ratio of the sample with that of a standard reference, as reported in Equation 2. The K/S value was determined using the Kubelka–Munk equation. Equation 3 defines K as the absorbance, S as the scattering coefficient and $R\lambda$ as the surface reflectance of the masterbatch plates to light of wavelength λ. All samples were produced five times and measured separately.

$$\mathrm{RCS} = \frac{\left(\frac{K}{S}\right)Sample}{\left(\frac{K}{S}\right)Reference} \times 100 \tag{2}$$

$$\frac{K}{S} = \frac{(1-R\lambda)^2}{2R\lambda} \tag{3}$$

To observe the distribution and dispersion of pigments in the masterbatch samples, films were produced using a blown film machine (Labtech Engineering). The masterbatches were added to the mixture at a rate of 4%. The resulting film samples were examined under strong fluorescent light.

The dispersion quality of the masterbatches was tested using a filter tester (Labtech Engineering) in accordance with the ISO 23900-5 standard. The test measures the pressure generated by agglomerated coarse particles in the material, with the result given in bar/g (Equation 4) as the FPV. Where $P1$ is the initial pressure of the melt, $P2$ is the maximum pressure reached and M is the total mass of pigment in the blend. Because the masterbatches contain 25% pigment, mixtures prepared with 50 g of masterbatch correspond to 12.5 g of pigment. The filter type used was a standard three-layer 15-μm filter. The higher the FPV value, the higher the number of coarse particles that are not dispersed in the matrix. This test provided an understanding of the effect of the parameters used on dispersion. Each test was repeated five times.

$$\mathrm{FPV} = \frac{P2 - P1}{M} \tag{4}$$

To measure the mechanical strength, dog bone samples of the masterbatches were produced on the injection moulding machine with a clamping force of 60 kN in a mould set according to the ASTM D638 Type 1 standard, without mixing with any raw material. Each sample was moulded at a speed of 60 rpm, a pressure of 80 bar and a temperature range of 210-230°C. The tensile strength and elongation at the break of the specimens were observed using a Devotrans tensile testing machine with a capacity of 10 kN. The specimens were tested at a speed of 50 mm/min according to the ASTM D638 standard and observed until failure. Each test was repeated five times, and specimens exceeding the standard deviation were retested.

Fractographic analysis was conducted on the ends of the broken dog bone samples resulting from the tensile test of the masterbatches using scanning electron microscopy (SEM). The samples were coated with gold to ensure conductivity, and images were taken at different magnifications on a Carl Zeiss 300VP scanning electron microscope by setting the acceleration voltage to 5 kV. The distribution and dispersion of pigment in the matrix were examined and compared with the results of the blown film and filter tests.

3 | RESULTS AND DISCUSSION

The first part observed in the FTIR analysis of the masterbatches is the strong asymmetric and symmetric stretching vibrations of the methylene group (CH_2) between 2917 and 2848 cm^{-1} in all polymeric samples. With the peak at 1643 cm^{-1} seen in the BIO-PE neat sample, it was possible to distinguish the characteristic vibrations of amines originating from Hindered Amine Light Stabilizers (HALS). However, this peak shifts in the masterbatch forms. The band at 1417 cm^{-1} is due to the CH_2 scissoring vibrations in polyethylene.[22,23] The sharp band at 719 cm^{-1} is due to CH_2 rocking vibrations. Furthermore, because of the phthalocyanine pigment in the masterbatch, a C=N bond at 1506 cm^{-1}, a C–H group

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bonded from an aromatic ring at 1460-754 cm^{-1}, a C–C bond from isoindole at 1417-1331 cm^{-1}, C–N bonding of isoindole at 1284-1167 cm^{-1}, C–H bonding at 1118-1090 cm^{-1}, Cu ligand vibration at 899-862 cm^{-1},[24] as well as metal–oxygen bonding at 569-502-430 cm^{-1}, were observed (Figure 1).

Phthalocyanine blue is one of the most widely used pigments in the plastics industry and one of the most challenging products in terms of dispersion and distribution in the polymer matrix. The colour strength of this pigment, and as a general approach in all pigment work, is increased by improving the dispersion.[25] This study demonstrates that decreasing particle size and homogeneous dispersion play an important role in increasing colour strength. Analysis of the colour strength results of the masterbatches (Figure 2) revealed that, as a general trend, samples containing HW yielded lower colour strengths than the other samples. Formulations containing MW and WE showed a significant increase in colour strength. It is assumed that the dispersion and distribution of pigment in the matrix are better in these formulations than in the formulations containing HW. The results indicate that there was no distinctive difference in masterbatch colour strength due to process conditions, while samples with smaller wax particle sizes resulted in a significant increase in colour strength.

During the blown film tests, film samples containing 4% masterbatch underwent examination under a fluorescent light source to observe pigment distribution and dispersion (Figure 3). The colour strengths of the films were consistent with the values in the Colour Strength graph. The samples with HW have the lowest colour strength, indicating poor pigment distribution due to the presence of coarse agglomerated pigment residues in the film samples. By contrast, the other samples exhibit stronger dark tones and better pigment distribution. Upon visual inspection, although the particles in the samples with HW are coarser than those in the samples made with WE, they have a similar particle density. It was determined that BIO-PE/MW/02 exhibited the highest colour strength and particle count. This sample also gave one of the highest colour strengths in the spectrophotometer results. Among the emulsion wax samples, BIO-PE/WE/03 showed a similar colour strength. It has the best dispersion and distribution in blown film samples compared with samples made with WE.

Upon analysing the filter tests (Figure 4), it was observed that the results were consistent with those from the previous tests. The samples containing HW exhibited the highest filter value. Similar to the blown film samples, these samples contained numerous particles in the matrix. The high concentration of these particles resulted in quicker clogging of the filters and a greater increase in pressure compared with the other samples. The masterbatches containing WE exhibited good dispersion and high colour strength, but still contained more coarse particles compared with the samples with MW. However, the number of coarse particles was lower than that for the samples with HW, resulting in less clogging of the filters. The best results were obtained with masterbatch

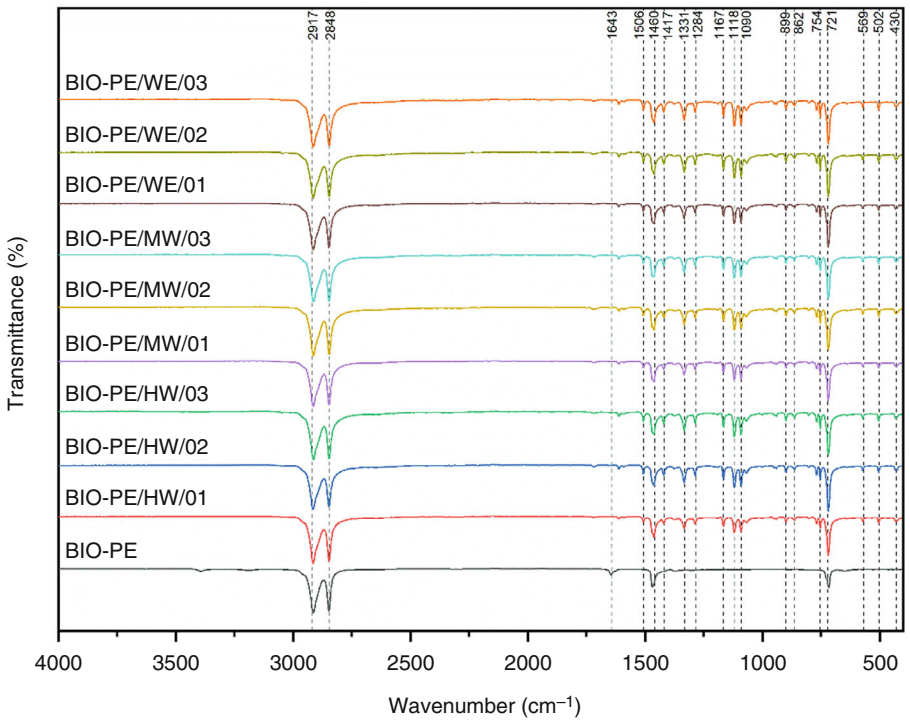


FIGURE 1 FTIR spectra of masterbatch samples. BIO-PE, bio-based polyethylene; FTIR, Fourier Transform–infrared; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

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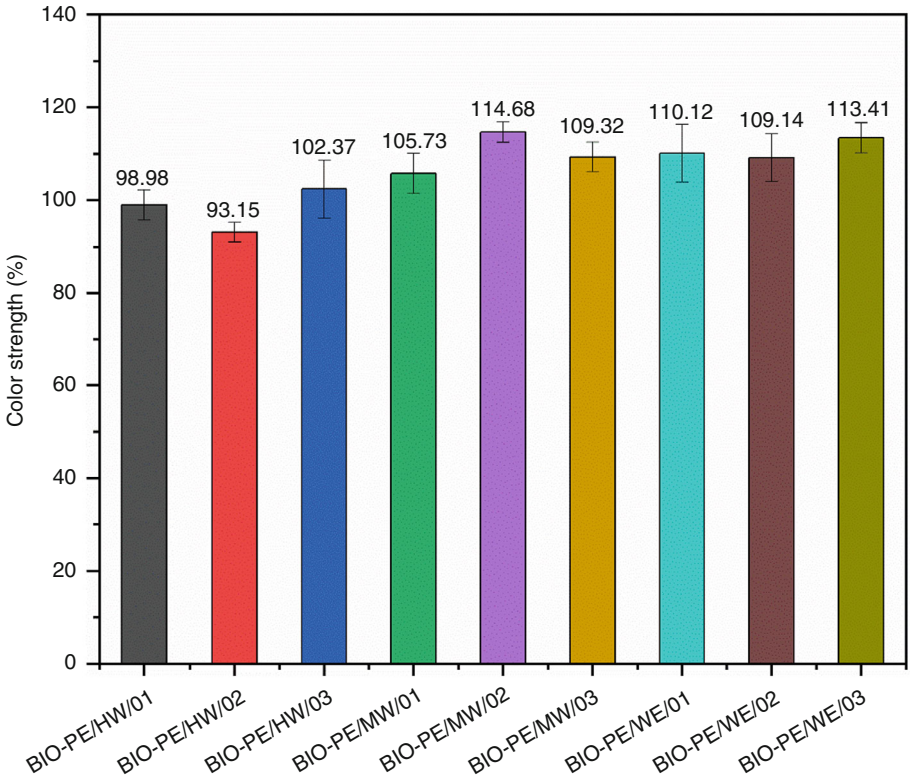


FIGURE 2 Colour strengths of masterbatch samples. BIO-PE, bio-based polyethylene; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

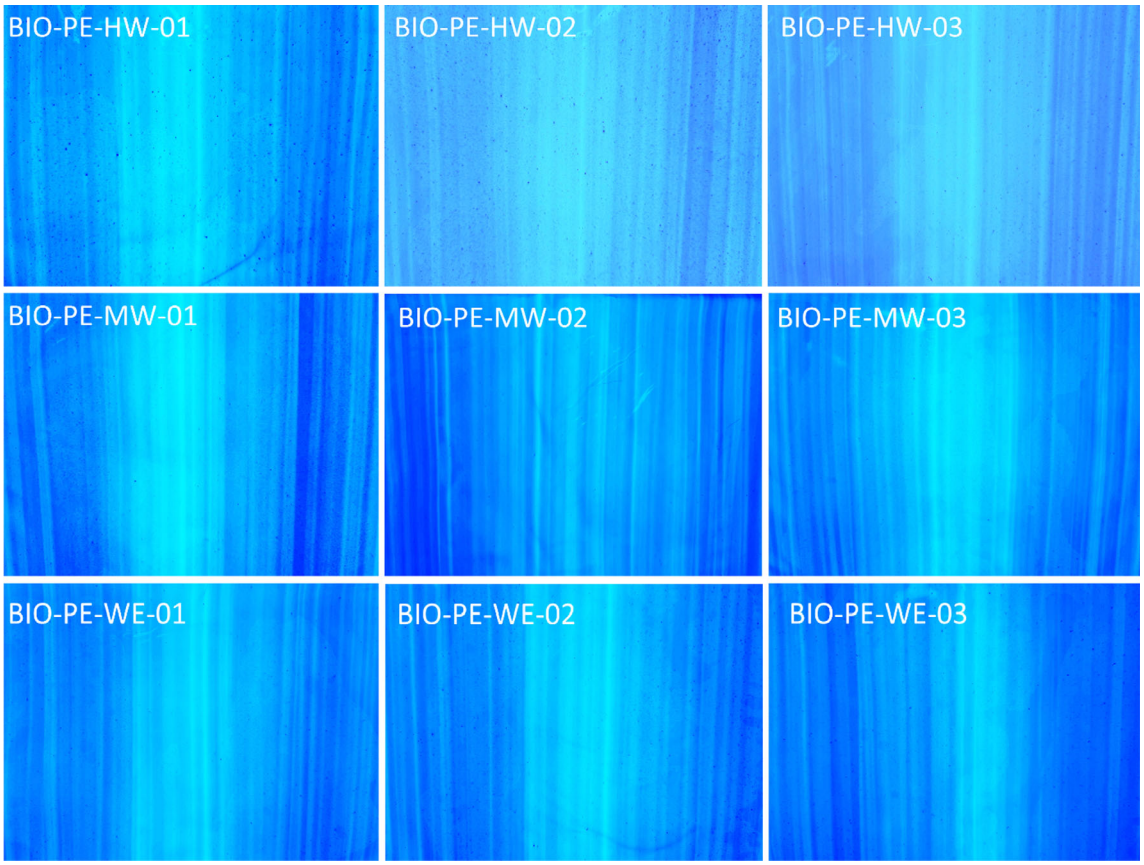


FIGURE 3 Blown film samples containing 4% masterbatches under a fluorescent light source. BIO-PE, bio-based polyethylene; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

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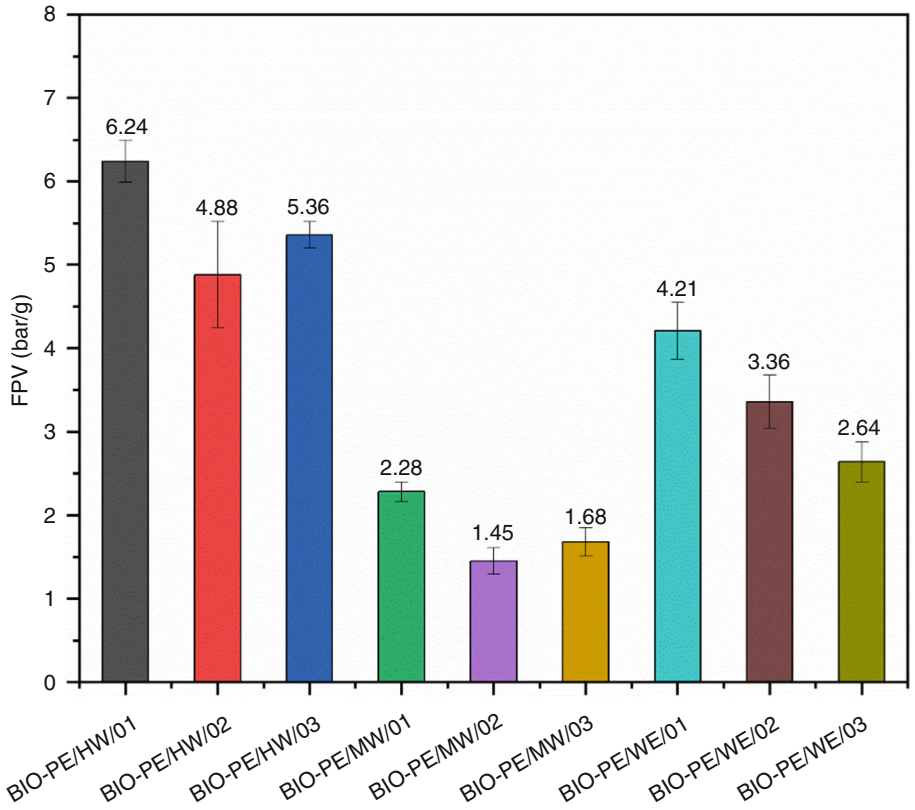


FIGURE 4 FPVs resulting from filter testing of masterbatches. BIO-PE, bio-based polyethylene; FPV, filter pressure value; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

samples made with MW, as observed from the colour strength values and the film samples. As in previous steps, the type of wax used was found to be more significant than the process conditions.

Buccella et al[26] conducted a similar study investigating the relationship between relative colour strength and filter pressure tests using halogenated copper phthalocyanine pigment in polyamide-based masterbatch. The study found that the colour strength of the masterbatches increased proportionally as the samples were extruded more than once, while the filter results decreased continuously as the samples were extruded more than once. It was concluded that dispersion improved with each new extrusion. The results of our study are consistent with the increase in tinting strength due to dispersion. All of the samples made with MW displayed better filter results than those for the other samples, with the best result being obtained from BIO-PE/MW/02.

Upon examination of the mechanical properties, it was observed that the Young's moduli of all samples exhibited an increasing trend when compared with the raw polymer. The addition of fillers, such as pigments and minerals, to the matrix, resulted in a composite that is stronger, stiffer and less ductile. These findings are consistent with those from previous studies.[27–33]

The best results were obtained in samples containing MW, which are the formulations with the best dispersion. MW strengthened the matrix more than the other additives. However, when the fracture occurred, all samples showed similar tensile strength values. From raw polymer, even better results were obtained than for masterbatch samples. All samples were very poor in terms of elongation at break percentage compared with the crude polymer. While the tensile specimens of the raw polymer showed 115.03% elongation, the tensile specimens from the masterbatches failed as soon as they entered the plastic zone. However, samples containing MW showed slightly more brittleness than the others. This is agreement with the increase in Young's modulus values and shows that the material loses its elasticity at the same rate as its stiffness increases.

Ersoy and Köse[34] obtained various composites by mixing calcium carbonate ($CaCO_3$) into a polypropylene matrix after surface modification with coupling agents. In the FPV tests performed on these composites, similar to our study, the raw polymer showed better results than the composites in both tensile strength and elongation at break. Among the composites, calcite, coated with isopropyl tristearyl titanate, showed the best results for FPV and mechanical tests compared with its counterparts. The authors concluded that the uncoated calcite had a high energy surface, which resulted in poor dispersion in the polymer matrix due to low energy levels. This caused intense agglomeration, leading to the worst FPV values

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due to incompatibility. Similarly, in our study, the mechanical values of the samples made with HW were poor, indicating bad dispersion due to incompatibility between polymer–pigment bonds (Table 3).

SEM analysis of the ends of the fractured parts in the mechanical tests showed that all parts were brittle, in compliance with the values obtained in the tensile tests (Figure 5). In the samples that contained HW and WE in

TABLE 3 Mechanical test results of masterbatches.

Sample	Young's modulus (GPa)	Tensile strength (MPa)	Elongation at break (%)
BIO-PE	0.22 ± 0.04	10.87 ± 0.05	115.03 ± 6.49
BIO-PE/HW/01	0.35 ± 0.05	7.27 ± 0.77	6.51 ± 1.94
BIO-PE/HW/02	0.33 ± 0.04	7.62 ± 0.31	6.34 ± 1.08
BIO-PE/HW/03	0.37 ± 0.02	7.90 ± 0.25	6.40 ± 0.80
BIO-PE/MW/01	0.41 ± 0.08	8.49 ± 0.55	4.11 ± 1.11
BIO-PE/MW/02	0.57 ± 0.04	10.23 ± 0.79	4.10 ± 0.75
BIO-PE/MW/03	0.54 ± 0.07	9.72 ± 0.37	4.35 ± 1.44
BIO-PE/WE/01	0.36 ± 0.04	8.66 ± 0.50	4.48 ± 1.03
BIO-PE/WE/02	0.39 ± 0.09	9.25 ± 0.06	4.75 ± 1.27
BIO-PE/WE/03	0.36 ± 0.06	8.63 ± 0.24	4.30 ± 0.70

Abbreviations: BIO-PE, bio-based polyethylene; HW, homopolymer wax; MW, micronised wax; WE, wax emulsion.

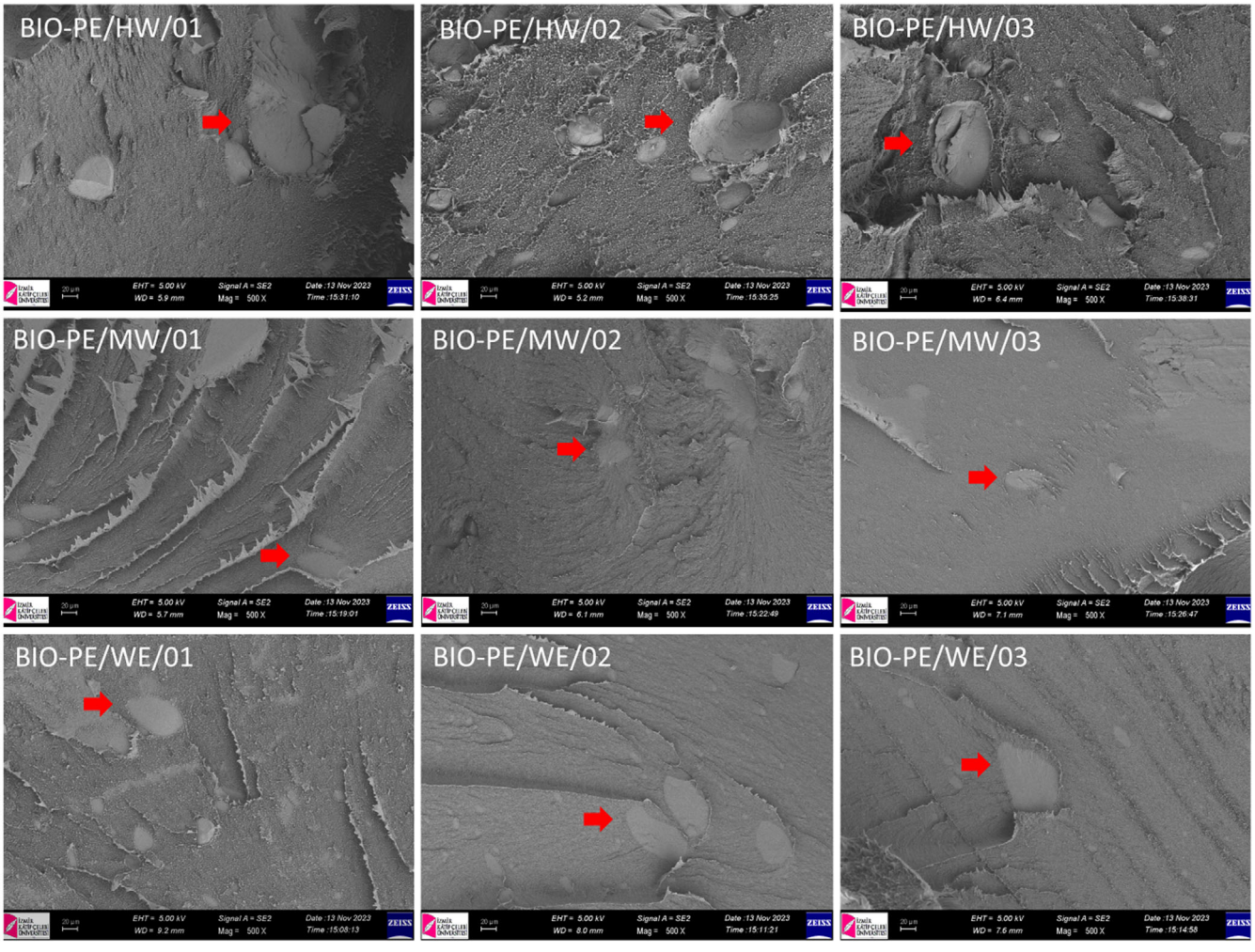


FIGURE 5 SEM analysis of fractographic samples of masterbatches. BIO-PE, bio-based polyethylene; FPV, filter pressure value; HW, homopolymer wax; MW, micronised wax; SEM, scanning electron microscopy; WE, wax emulsion.

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TABLE 4 Signal-to-noise ratio response table for masterbatch filter pressure values.

Level	Wax type	Screw speed	Mixture time	Temperature
1	−14.752	−12.902	−10.312	−9.188
2	−6.017	−9.174	−10.839	−10.253
3	−10.482	−9.174	−10.099	−11.809
Delta	8.735	3.728	0.741	2.621
Rank	1	2	4	3

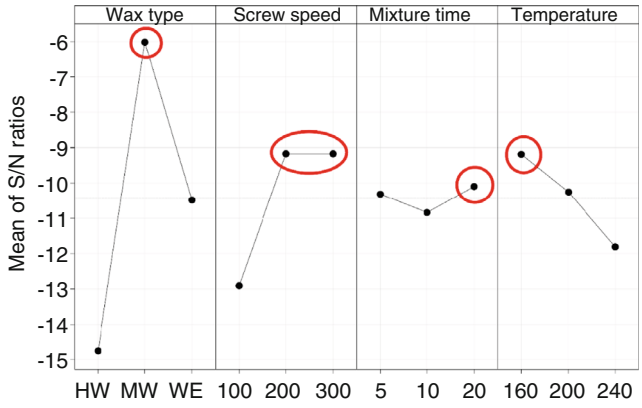


FIGURE 6 S/N ratio graphs for FPVs of masterbatches. HW, homopolymer wax; FPV, filter pressure value; MW, micronised wax; S/N, signal-to-noise; WE, wax emulsion.

the matrix, numerous particles above 20 μm were observed. These particles were caught in 15-μm filters in the filter test and caused high FPV results. The samples containing MW differ in particle density and size compared with the other samples, which is consistent with previous tests. Depending on the success of dispersion and distribution, the number and size of particles decreased, resulting in improved colour strength and stiffness in the samples containing MW.

The S/N ratio was calculated for each factor level combination with the objective of assessing the effect of variability on the FPVs. The S/N ratio is a measure used to identify the robustness of a process, intending to minimise variability and maximise performance. The ratios were then employed to construct the response table (Table 4), which facilitated the identification of the most significant factors influencing the FPV. Higher S/N ratios indicate superior performance with reduced variability, whereas lower S/N ratios indicate inferior performance with elevated variability. The examination of these S/N ratios revealed that wax type had the most pronounced impact on FPV, followed by screw speed, temperature and mixing time.

Examining the S/N ratios plot (Figure 6) reveals the levels at which factors have the greatest effect on dispersion. The results indicate that MW is the most effective type of wax, followed by emulsion waxes. HW, on the other hand, gave the poorest results. These results are consistent with previous tests.

In terms of screw speed, there is no significant difference between 200 and 300 rpm and both options can be applied depending on the type of pigment used. Higher screw speeds can be considered for sensitive pigments that should not remain in the barrel for extended periods, while lower screw speeds can be utilised in situations where temperature exposure time is beneficial for dispersion.

Although the graph shows that 20 minutes yields better results in terms of mixing time, the rates are quite close to each other. Therefore, considering that mixing time is the least effective factor on dispersion, it was concluded that the lowest mixing time during the process is preferred in terms of time savings.

Upon analysing the temperature factor, it was observed that the most effective dispersion occurred at 160°C. Mixtures processed at lower temperatures in the extruder were subjected to more shear stress. Therefore, it can be inferred that phthalocyanine blue, a challenging pigment to disperse that requires higher energy, yielded better results. Based on the results of all the tests, it was concluded that the BIO-PE/MW/02 sample, produced with MW, a screw speed of 200 rpm and a mixing time of 20 minutes, consistently performed the best. This indicates that the formulations prepared by the Taguchi method produced consistent results.

4 | CONCLUSION

This study examined the issue of dispersion, which directly affects the effectiveness of additives used in academic research on thermoplastic composites. For this purpose, the dispersion of copper phthalocyanine pigment in a BIO-PE masterbatch was studied using a Taguchi orthogonal array. The masterbatches were tested for colour strength, blown film, FPVs and mechanical properties.

Based on the findings, it is evident that colour strength increased as a result of improved dispersion. This result was consistently observed by spectrophotometer examination of relative colour strength, visual

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inspection of blown film samples and filter pressure tests. The masterbatches exhibited changes in their mechanical properties, becoming stiffer, stronger and more brittle, depending on the degree of dispersion. In the production of samples using the Taguchi L9 orthogonal array, wax type was found to be the most effective parameter in achieving dispersion. Specifically, MW was the most effective type, and the physical properties of the wax played a significant role in achieving dispersion. Additionally, screw speed was identified as the most effective process condition parameter.

This study can be extended using many other parameters that can affect dispersion. The findings can also be used for different pigment or filler ratios. In this way, dispersion can be improved depending on different additive types and the highest efficiency can be obtained from the additives used in academic studies.

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