

Influence of polymer processing parameters and coloring agents on gloss and color of acrylonitrile-butadiene-styrene terpolymer moldings

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Abstract: The results of examination of the influence of polymer processing parameters and addition of coloring agents in the form of yellow organic pigment PY 191 and its masterbatches based on acrylonitrile-butadiene-styrene copolymer (ABS) or polystyrene (PS) on the color and gloss of injection molded parts have been presented. Gloss investigations, at the light incidence on the molding surface in the direction of polymer flow in the cavity and in the opposite direction at an angle of 60° and 20° were made (Figs. 3–6). Color tests were also carried out (Table 4). The obtained results showed that high mold temperature ($T_f = 80$ °C) and lower injection temperature ($T_w = 230$ °C) allow to obtain parts of higher gloss and smaller variation in their color.

Keywords: injection molding, coloring agents, gloss, color, acrylonitrile-butadiene-styrene copolymer.

Wpływ warunków przetwórstwa oraz środków barwiących na połysk i barwę wyprasek z terpolimeru akrylonitryl-butadien-styren

Streszczenie: Omówiono wpływ warunków przetwórstwa oraz dodatku środków barwiących w postaci żółtego pigmentu organicznego PY 191 oraz jego koncentratów na bazie kopolimeru akrylonitryl-butadien-styren (ABS) lub polistyrenu (PS) na barwę oraz połysk wyprasek. Wykonano badania połysku powierzchni wyprasek przy świetle padającym zgodnie z kierunkiem przepływu tworzywa w gnieździe formującym oraz w kierunku przeciwnym pod kątem 60° i 20° (rys. 3–6). Przeprowadzono również badania barwy porównując barwę wytworzonych wyprasek do wzorców wybranych ze wzornika NCS (z ang. *Natural Color System*) (tabela 4). Na podstawie otrzymanych wyników określono, że najwyższa z badanych temperatura formy ($T_f = 80$ °C) oraz niższa temperatura wtryskiwania ($T_w = 230$ °C) umożliwiają uzyskanie wyprasek charakteryzujących się większym połyskiem oraz mniejszym zróżnicowaniem barwy.

Słowa kluczowe: wtryskiwanie, środki barwiące, połysk, barwa, kopolimer akrylonitryl-butadien-styren.

Most plastics without the addition of coloring agents are white or similar thereto, or are transparent. Plastics can be colored both in mass and surface. The quality of colored plastic products depends mainly on the thickness, volume and method of dispensing a colorant and also conditions of their manufacturing process [1, 2]. Color is a subjective visual impression in the human brain caused by radiation incident on the receptors of the eye, the latter being the result of two phenomena: the absorption (color formation) and scattering (transparency). At the same time, objectively, the color is a feature by which

one can measure the properties of trans-reflective material, *i.e.*, the amount of light of a given wavelength reflected or absorbed by the material [2–4].

The color is not an absolute property, because it is dependent on many factors, *e.g.* the state of the tested samples surface or attitude examiner testing the color of the product [2–5]. In the case of polymeric products an additional factor for their color is the orientation of the polymer macromolecules, resulting primarily from the conditions of flow in channels of processing machines and tools. As a result of the macromolecules orientation the dichroism (*i.e.* bicolor state) can occur [1, 6]. This phenomenon relies on the absorption, at varying degree depending on the orientation of the macromolecules, of light incident on the sample. The amount of light absorbed depends on its polarization. Dichroism is a characteristic of materials of anisotropic structure [6]. The color is measured using de-

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pendent models, e.g. RGB, CMYK and independent ones for example CIE XYZ, CIE $L^*a^*b^*$ [3, 4, 7–14].

Increasingly, attention of polymeric products' manufacturers focuses not only on their color, but also on the gloss of products, which is one of the primary (besides color) marketing tools. Evidence of this shows even much greater demand for the products of a high-gloss, and about half the size lower than the mat ones. Gloss is the optical impression emerging from the reflection and scattering of light on the surface of solids and liquids, or directly above the surface. It can also be defined as the amount of light reflected at the same angle as the light falling on the test surface [5, 15]. Gloss is dependent on the refractive index and absorption of light, transparency and color of product, type of lighting, part surface (including the structure, roughness and the position of the tested plane) and the angle and distance of observation [1, 15], and is measured with glossmeters. The state of products surface determines their gloss. This surface state depends on processing conditions in which parts were produced, especially the mold temperature [16–19], and also on the surface roughness of mold cavities.

If the light is incident on the scattering centers, e.g. pigments or, in the case of semi-crystalline materials, spherulites, then there is a scattering of the beam as it was shown in Fig. 1. The amount of light reflected by the tested sample comprises the amount of light reflected from the surface and from the inner layer. Due to this one can observe the color of a sample given by the pigment [1].

Measurement and control of color and gloss of polymeric products play an important role in the evaluation of their quality and performance. The aim of this study was to investigate the effect of coloring agents in the form of a powder or masterbatches (coloring concentrates in the form of granules), and the conditions of the injection molding process on the color and gloss of injection molded parts made of ABS.

EXPERIMENTAL PART

Materials

A terpolymer of acrylonitrile-butadiene-styrene (ABS) with trade name Terluran® GP 35 Natur produced by BASF was chosen for injection molding. As a colorants a

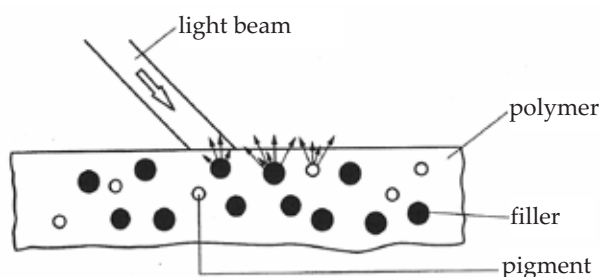


Fig. 1. Diagram of internally diffused light deflection (according [1])

yellow organic azo pigment PY 191, as well as the masterbatches containing 20 wt % of pigment on the carrier resins ABS Terluran GP 35 Life (MBABS) or PS Empera 622 N (MBPS) were used. Coloring agents were provided by Permedia SA Chemical Plant from Lublin (Poland). Pigment PY 191 has temperature stability in the range between 260 and 280 °C when used for ABS dyeing [8, 20, 21].

Sample preparation

Rectangular test samples of dimensions 150 × 23 × 4 mm were produced using the two-cavity mold mounted on the injection molding machine Krauss Maffei KM65-160 C4. The surface of cavities was polished and characterized by a roughness $R_a = 0.02 \mu\text{m}$. Samples were prepared with varying injection temperature T_w (230 or 250 °C) and variable mold temperature T_f (30, 50 or 80 °C). Other injection molding parameters were as follows: holding pressure 70 MPa, injection velocity 96 mm/s, injection time 0.8 s, holding time 10 s. The surface of moldings was without sinks and other defects. Injection molding parameters were determined based on data from the literature [22] and manufacturers' recommendations.

The granulate was not subjected to preliminary drying as the ABS does not exhibit any high water absorption (up to 1 %), and plastic bags were not damaged. The samples were made of undyed ABS or with addition of coloring agents. MBABS or MBPS were added in the amount 1 or 2 wt %. Two samples were colored using the additive of 1 or 2 wt % of pigment (PIGM).

The polymer was mixed with the pigment in a sealed zipper bag for 5 min, weighted with Sartorius CP225 with an accuracy of $\pm 0.01 \text{ mg}$ before and after mixing of the pigment with the polymer in order to determine the amount of pigment remaining in the package. Dosage level of each coloring agent was determined on the basis of manufacturer recommendations.

Methods of testing

Gloss measurements of molded parts surface were performed using a glossmeter Elcometer 406, accord-

Table 1. Values of the angle of the light incidence used in gloss examinations of different materials (according to [16])

Gloss range	Gloss value at 60° angle of the light incidence on the surface, GU	The angle at which the measurement should be done
Medium	10–70	60°
High	> 70	20°
Low	< 10	85°
Special applications		
Ceramics	–	45°
Paper, vinyl products	–	75°

ing to PN-EN ISO 2813:2001 standard. Preliminary gloss tests were carried out at the angle of light incidence of 60°. Depending on the results of surface gloss degree obtained, additional measurements at other angles of light incidence were done, according to rules listed in Table 1. Gloss of parts was evaluated on the basis of the value of gloss units (GU).

Color measurements of moldings were performed using the SP60 colorimeter by X-Rite at three measurement points on the surface of each sample, numbered sequentially in the direction of material flow in the cavity as it is shown in Fig. 2. The tests were made for undyed samples and for parts from ABS with the addition of coloring agents. The color was determined using the independent model CIE $L^*a^*b^*$, where L represents the luminance (brightness), a color in the range from green to red (magenta), and b color from blue to yellow. With independent models the differences between shades of color or ΔE can be assessed (the distance between compared colors), corresponding to the impression of the difference between colors, which has an observer [9–14]. To determine the differences in color ΔE between the measurement points of samples or between particular parts the following equations were used [9]:

$$\Delta L = L_1 - L_2 \quad (1)$$

$$\Delta a = a_1 - a_2 \quad (2)$$

$$\Delta b = b_1 - b_2 \quad (3)$$

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \quad (4)$$

where: $L_1, L_2, a_1, a_2, b_1, b_2$ – values that determine the color in two measurement points of the same sample or two different samples (the reference and tested sample).

RESULTS AND DISCUSSION

Gloss of injection molded parts

Gloss tests were carried out in two series. In the Series I the gloss was determined at the light incidence on the surface in the direction of polymer flow in the cavity, while in the Series II – in the opposite direction. The test results for Series I are shown in Figs. 3 (angle 60°) and 4 (angle 20°) but for Series II in Figs. 5 (angle 60°) and 6 (angle 20°).

The results presented in Fig. 3 show that almost all samples made of undyed material are characterized by a medium gloss. Their gloss values are lower than those obtained for samples made of ABS containing coloring agents. The smallest gloss values of undyed ABS parts (66.86 GU) were noticed for those produced at $T_f = 30^\circ\text{C}$ and $T_w = 230^\circ\text{C}$. Moreover, it may be noted that only undyed ABS parts produced at a high mold temperature ($T_f = 80^\circ\text{C}$) and low injection temperature ($T_w = 230^\circ\text{C}$)

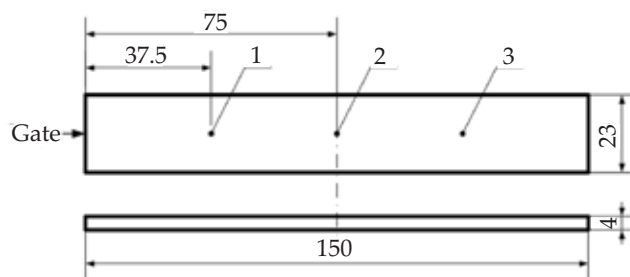


Fig. 2. Injection molded part with color measurement points

have higher gloss, its value is slightly higher than 70 GU.

Colored parts, regardless of the type and content of the coloring agent, obtained with using high $T_f = 80^\circ\text{C}$ have at the angle of light incidence of 60° (Fig. 3) the highest gloss values ranging from 71.40 GU for polymer colored with 1 wt % of MBABS to 77.46 GU in the case of 2 wt % content of this coloring agent. The lowest value (70.15 GU) was obtained for parts produced at $T_f = 30^\circ\text{C}$ and $T_w = 250^\circ\text{C}$, when 1 wt % of PIGM was used. In addition, parts produced at an average $T_f = 50^\circ\text{C}$ had low gloss values ranging from 70.46 to 72.76 GU. Because the values of gloss determined at the angle of light incidence of 60° in many cases were greater than 70 GU, additional tests were performed at the 20° angle of light incidence (Fig. 4). Measurements were performed for colored parts. In the case of undyed parts only those produced at $T_f = 80^\circ\text{C}$ and $T_w = 230^\circ\text{C}$ were subjected to further testing.

Measurements carried out at the angle of light incidence of 20° showed much larger variety of gloss values. The parts colored with MBPS have quite high gloss. The highest gloss was found for parts produced at $T_f = 80^\circ\text{C}$ and $T_w = 230^\circ\text{C}$, made of ABS containing 2 wt % of MBPS, for which the gloss value is 64.68 GU. Instead, the smallest gloss values (36.72 GU) were obtained for parts produced under the same molding conditions, but made of ABS with the addition of 2 wt % of MBABS. Moreover, it may be noted that for samples prepared at $T_f = 30^\circ\text{C}$ from ABS with the addition of 1 % and 2 wt % of masterbatch on ABS carrier, 2 wt % of masterbatch on PS carrier and 2 % of PIGM with the increase in T_w to 250 °C the gloss value is reduced.

In Series II the gloss measurements were conducted at the light incidence on the sample surface in the direction opposite to the polymer flow in the cavity (Figs. 5 and 6). It can be seen that in most cases the gloss values are lower than those obtained in tests carried out at the light incidence on the part surface in the flow direction.

Analyzing the results of the gloss measurements shown in Fig. 5, it can be found that the undyed molded ABS parts have lower gloss than colored ones, similarly as it was stated before, for above described experiments (Fig. 3). Gloss values of undyed parts are between 53.50 and 68.74 GU, so they are in the range in which gloss is defined as medium. The lowest gloss features undyed parts produced using $T_f = 30^\circ\text{C}$ and $T_w = 230^\circ\text{C}$. Smaller gloss values were also obtained for parts pro-

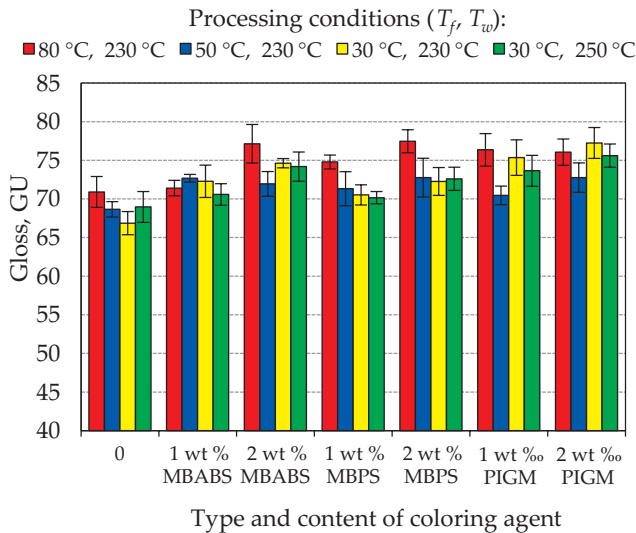


Fig. 3. Gloss values of ABS injection molded parts in a function of coloring agents; gloss examined at the light incidence on molding surface in the direction of the polymer flow in the cavity, 60° angle of the light incidence

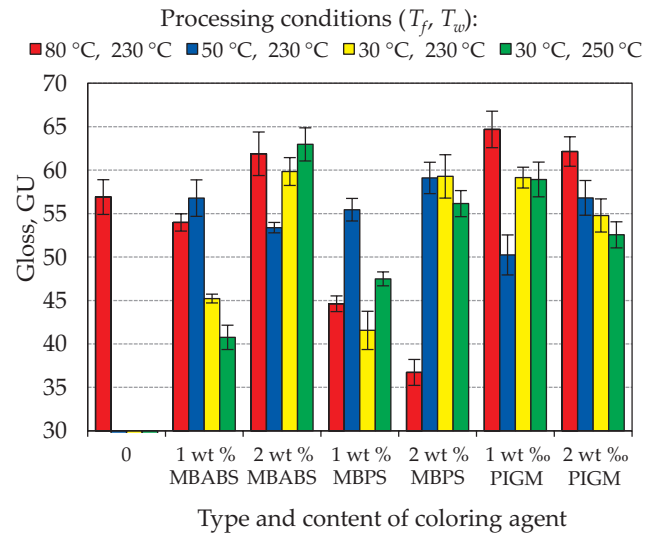


Fig. 4. Gloss values of ABS injection molded parts in a function of coloring agents; gloss examined at the light incidence on the molding surface in the direction of the polymer flow in the cavity, 20° angle of the light incidence

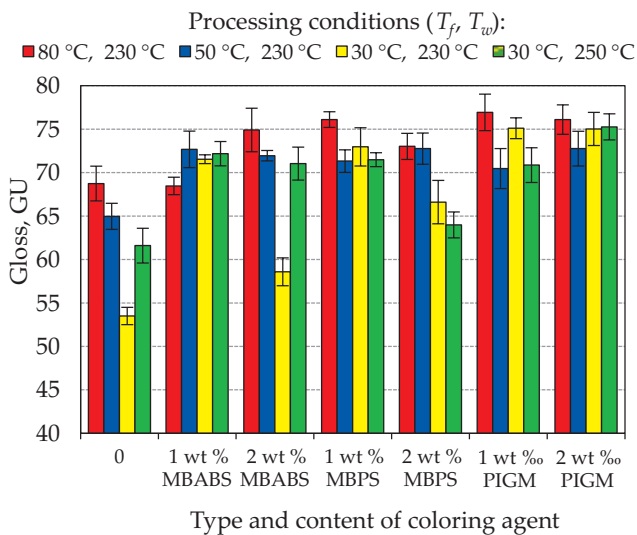


Fig. 5. Gloss values of ABS injection molded parts in a function of coloring agents, gloss examined at the light incidence on the molding surface in the direction opposite to the polymer flow in the cavity, 60° angle of the light incidence

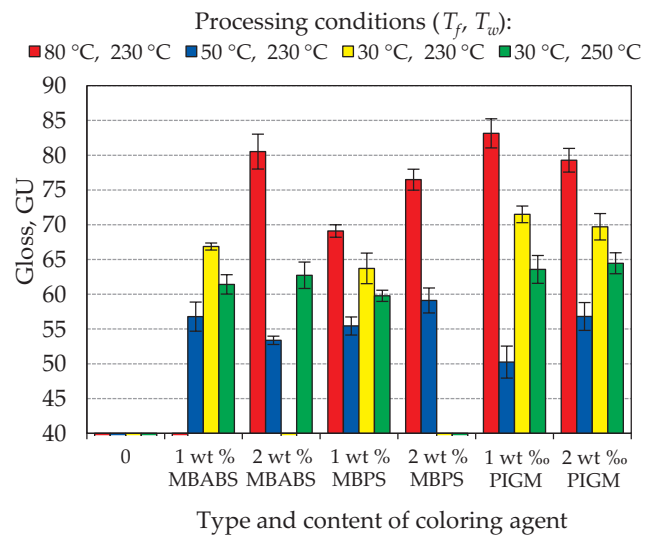


Fig. 6. Gloss values of ABS injection molded parts in a function of coloring agents; gloss examined at the light incidence on the molding surface in the direction opposite to the polymer flow in the cavity, 20° angle of the light incidence

duced under the same processing conditions, but made of ABS containing 1 wt % of MBABS.

In most cases, both for the undyed material and the material with the addition of coloring agents, the highest gloss characterized parts produced at high $T_f = 80$ °C and low $T_w = 230$ °C. The highest gloss value (76.92 GU) was measured for parts made of material dyed with 2 wt % of masterbatch on PS carrier. For samples made of ABS with the addition of coloring agents, obtained at $T_f = 50$ °C and $T_w = 230$ °C, small differences in gloss, varying from 72.68 to 72.76 GU were stated.

Next tests, performed at the angle of light incidence of 20°, were carried out only for parts, for which the gloss

was higher than 70 GU (in accordance with the recommendations presented in Table 1). The results are shown in Fig. 6. It can be noted that the smaller number of moldings can be characterized by the high gloss. The highest gloss, as before, was determined for parts formed at high $T_f = 80$ °C and low $T_w = 230$ °C, but the lowest values were obtained in the case of $T_f = 50$ °C and $T_w = 230$ °C.

Considering the effect of injection temperature on the gloss of injection molded parts produced at low $T_f = 30$ °C, it can be concluded that for majority of samples, especially made of ABS containing 1 wt % MBABS, 2 wt % of MBPS, 1 and 2 wt % of PIGM, the increase of T_w causes the decrease in parts gloss.

Color of moldings

Certain ranges of ΔE and the corresponding differences in color are listed in Table 2. In view of the fact that in experiments the color differences between the measuring points of tested samples ranged between 0.01 and 0.29, these values were averaged for further calculations. ΔE values were calculated by comparing the results obtained for the particular sample with the results determined for the model color. Samples with varying content of masterbatch on ABS, PS carrier, and the pigment were compared with the model color selected from NCS (Natural Color System) [23]. Samples made of ABS without the addition of coloring agent were compared with the model NCS S2002-G, samples obtained with the addition of 1 wt % of masterbatch (MBABS or MBPS), or 1 wt ‰ of PIGM were compared with the model NCS S2050-Y10R, and in the case of parts colored using 2 wt % of the masterbatch or 2 wt ‰ of PIGM the model NCS S1060-Y 20R was used. The results of measurements are shown in Table 3, wherein areas with determined ΔE values are marked with different shades of blue, in accordance with ΔE ranges given in Table 2.

Table 2. The ranges of ΔE values (according to PN-EN ISO 2813:2001)

The ranges of ΔE values:	
$0 < \Delta E < 1$ – normal, invisible color variations	
$1 < \Delta E < 2$ – small variations, recognizable only by an experienced observer	
$2 < \Delta E < 3.5$ – medium variations, recognizable by inexperienced observer	
$3.5 < \Delta E < 5$ – distinct color variations	
$\Delta E > 5$ – large color variations	

Table 3. ΔE values for samples made of ABS without and with coloring agents obtained at various injection (T_w) and mold temperature (T_f)

		Content of coloring agent						
		ABS	1 wt % MBABS	1 wt % MBPS	1 wt ‰ PIGM	2 wt % MBABS	2 wt % MBPS	2 wt ‰ PIGM
Processing conditions	$T_f = 80\text{ }^\circ\text{C}$ $T_w = 230\text{ }^\circ\text{C}$	1.91	0.89	0.85	3.27	0.75	1.28	1.89
	$T_f = 50\text{ }^\circ\text{C}$ $T_w = 230\text{ }^\circ\text{C}$	7.14	8.65	6.85	4.22	1.55	5.84	9.88
	$T_f = 30\text{ }^\circ\text{C}$ $T_w = 230\text{ }^\circ\text{C}$	1.52	3.98	3.06	3.52	2.24	7.33	8.97
	$T_f = 30\text{ }^\circ\text{C}$ $T_w = 250\text{ }^\circ\text{C}$	1.59	2.95	2.28	3.44	1.10	5.20	8.51
Standard NCS	S2002-G	S2050-Y10R			S1060-Y20R			

For most of the examined cases, the ΔE value is higher than 2, meaning that the difference in color between samples and models are visible to the inexperienced observer. At the same time about 36 % of ΔE values are lower than 2, which indicates that only about one third of compared samples is characterized by the difference visible for an experienced observer or these differences mean invisible color variations.

However, the color of samples is significantly dependent on injection molding conditions, especially T_f . In the case of undyed ABS samples compared to the color model NCS S2002-G it was found that only parts produced at an average $T_f = 50\text{ }^\circ\text{C}$ and low value of $T_w = 230\text{ }^\circ\text{C}$ are characterized by a substantial change in color ($\Delta E = 7.14$) in compare to the model. Instead, parts made at extreme T_f (30 and 80 $^\circ\text{C}$) and T_w (230 and 250 $^\circ\text{C}$) have a slight color change compared to the model ($\Delta E < 2$), visible only for the experienced observer.

On the example of molded parts made of ABS colored with 1 wt % of masterbatch or 1 wt ‰ of PIGM produced at $T_w = 230\text{ }^\circ\text{C}$ and $T_f = 80\text{ }^\circ\text{C}$, compared to the model NCS S2050-Y10R, it can be seen that the samples colored with 1 wt % of masterbatch (MBABS or MBPS) are characterized by normal, invisible color deviation – the value of ΔE is respectively 0.89 and 0.85. In contrast, the samples made of ABS with 1 ‰ of PIGM have medium deviation of color, visible even for the inexperienced observer. In the case of moldings produced from ABS with the same amount of coloring agents, but obtained at a lower mold temperature (50 $^\circ\text{C}$) the difference in color ΔE is much higher and is respectively 8.65, 6.85 and 4.22, while remaining within the ranges described as distinct or large color deviation. Thus the increase in T_f contributes to reduction of the difference in the color of parts.

A similar tendency can be observed in the case of ABS samples colored with 2 wt % of masterbatch on both carrier resins and 2 wt ‰ of PIGM compared to the model

NCS S1060-Y20R. The lowest ΔE value was reported for samples produced from ABS dyed with 2 wt % of MBABS, using high $T_f = 80$ °C. These are normal, invisible color deviations. For samples colored with 2 wt % of MBPS, produced under the same conditions, this value is larger and amounts to 1.28. Together with the decrease in T_f to 50 °C it was found, in all cases, that the value of ΔE increased. Only during comparison of the model and samples colored with MBABS the color difference was 1.56 and is defined as a very small deviation in the color. However, in other cases, the ΔE color deviations are higher than 5, which can be classified as large deviations in the color.

The value of T_w also influences the ΔE difference in sample color. Tests carried out using various T_w values (230 and 250 °C) and constant T_f (30 °C) showed that, for all ways of coloring, lower ΔE values were obtained when high $T_w = 250$ °C was applied. Based on these results it can be assumed that the high $T_f = 80$ °C and low $T_f = 230$ °C or low $T_f = 30$ °C and high $T_w = 250$ °C, is conducive to obtaining samples characterized by a color that most closely matches the model. This is especially evident for samples dyed with 2 wt % of MBABS.

Comparing the ΔE values obtained for samples dyed with the addition of different content and form of coloring agents it may be noted that the injection conditions have a significant effect on their color. The smallest differences in parts color occur when the high T_f and low T_w are used.

CONCLUSIONS

On the basis of experiments the influence of type and content of coloring agents in the form of pigment and masterbatches on different carrier resins on properties of ABS injection molded parts, rated by their gloss and color has been shown.

The studies have shown differences in the gloss values of moldings according to the way of light incidence on their surface. In the case of measurements performed upon light incidence in the direction of polymer flow in the mold cavity the addition of coloring agents, both in masterbatch form, and pigment powder influences the gloss significantly. All moldings made of ABS with the addition of coloring agents have high gloss. This may be due to the crystalline structure of the pigment (also present in large quantities in masterbatch). Instead, almost all not dyed moldings are characterized by the medium gloss. In the case of measurements made at the light incidence on the tested surface in the direction opposite to the polymer flow in the mold cavity it was found that some share of dyed moldings has lower gloss than those tested at the light incidence in the direction of polymer flow in the cavity. Gloss values, measured at the angle of light incidence of 60° in the direction of polymer flow, for dyed moldings are in the range between 70.15 and 77.46 GU, wherein the gloss is evaluated as high. Gloss of

these moldings, tested at the angle of light incidence of 60° in the opposite direction, varies in the range between 58.58 and 76.92 GU. The gloss measured at the angle of light incidence of 20°, in the direction of material flow in the cavity, is in the range between 36.72 and 64.68 GU, while in the opposite direction in the range between 50.24 and 83.14 GU. Differentiation of moldings gloss depending on the angle and direction of the light incidence on the surface can be important, when assessing their performance.

Processing conditions: T_f and T_w have significant effect on color of moldings. The calculated value ΔE allowed comparison of the color of moldings made under different injection molding conditions and with different content of coloring agents. For example, in the case of color model NCS S2050-Y10R compared to ABS moldings dyed with 1 wt % of MBABS or MBPS, produced using low $T_w = 230$ °C and a high $T_f = 80$ °C, the differences in color are invisible ($\Delta E = 0.89$ and 0.85). Meanwhile, for moldings with the same content of coloring agents, produced at lower T_f of 50 °C and 30 °C, the difference in color is already quite distinct. ABS moldings dyed with pigment, regardless of injection molding conditions, are characterized by the highest difference in color in compare to the model.

Uneven color of moldings produced in different injection molding conditions may be associated with the orientation of macromolecules in their surface layer formed during the melt flow and cooling in the mold. High T_w facilitates the polymer flow in the cavity and promotes greater macromolecules orientation in the flow direction as it was shown in previous studies [24]. When using low T_w and T_f the polymer flow in the cavity is somewhat inhibited by the material solidified on the cavity walls, resulting in non-uniform, along the flow path, orientation of macromolecules in the parts surface layer and the variation in their color. So, it can be assumed that the phenomenon of dichroism here occurs, which means variation in color depending on the orientation of macromolecules in the surface layer. Furthermore, in the case of moldings dyed with pigment the differences in color may be additionally caused by inadequate pigment dispersion in the polymer and also by the absence of additives.

It can be concluded that the high $T_f = 80$ °C and low $T_w = 230$ °C applied during processing of ABS with the addition of masterbatches (providing more effective dyeing due to better pigment dispersion in the polymer and the presence of waxes and different additives in the concentrate, e.g. a special interface lubricating effect) allow to obtain moldings characterized by a smaller differentiation in color and also having a high degree of gloss.

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