Characterization of Flow-Induced Surface Defects in Injection Moulded Components – Case Studies

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ABSTRACT

Injection-moulded parts involving long flow lengths can exhibit visible flow defects characterized by alternating glossy and dull bands on the polymer surface, approximately perpendicular to the flow direction. These flow defects are sometimes referred to as "flow marks" or "tiger stripes" and are considered to have strong negative effect on the appearance of the component and thus on the perceived quality. The reasons for the appearance of these stripes are not entirely clear; two main hypotheses relate to a slip-stick phenomenon or to an unstable flow front during the mould filling (which however may be interrelated). The present work deals with an experimental characterization of the banded regions of automotive components manufactured in mineral-filled polypropylene-based materials. It is shown using a stereographic scanning electron microscopy technique that the dull band regions were more surface rough than the other type of region. Furthermore, it was found that the surface regions of the dull bands contained less filler than the corresponding glossy regions. Possible causes for this are discussed. No significant difference in chemical composition between the surfaces of dull and glossy bands was detected.

INTRODUCTION

Polypropylene (PP)-based materials are widely used for instance in the automotive industry, which uses e.g. elastomer-modified PP containing inorganic fillers, for car bumpers and other parts in order to obtain adequate or improved impact strength and other important mechanical properties. Generally, components of this type are manufactured by injection moulding. In this context, the appearance is a very important issue towards which the automotive industry puts significant efforts. Unfortunately injection-moulded thermoplastic details can exhibit surface defects which impair the appearance and hence the perceived quality impression. Especially when the injection moulding of the components involves rather long flow lengths, a certain type of "flow marks" or "tiger stripes" may appear[1-3].

Tiger stripes are characterized by alternating glossy and dull bands on the polymer surface, approximately perpendicular to the flow direction. Often they are opposite in phase, i.e. if one side of the moulded part has a dull area, the corresponding area on the opposite side will be glossy. The surface morphology of the glossy and dull bands has been reported in several studies to differ substantially. A number of possible explanations have been put forward accounting for the appearance of these defects. Some of these attribute the tiger stripes mainly to a snake-like flow front instability during the filling phase of the injection moulding cycle. This would also explain why the appearance of the stripes is reversed on opposite sides.

Another hypothesis is that a stick-slip flow during the mould filling causes the flow marks. A high shear stress accumulation close to the mould surface can in principle overcome the adhesion between the polymer and the mould surface leading to slip. These hypotheses as well as others are described in some more detail below. The present study focuses on the characterization of these dull and glossy areas in injection-moulded car components. The specimens used were from industrially manufactured components. Several surface analysis techniques were used, in particular scanning electron microscopy (SEM) applied to surfaces and cross sections.

Possible causes for flow marks/tiger stripes

Flow marks are often encountered with filled polymers and polymer blends[3,4,5], but defects of this kind have also been reported for neat polymers[6]. Higher melt and mould temperatures may reduce the tendency for flow mark formation[4,7] and an increase in the injection rate or speed appear to lead to more pronounced flow marks[3,4,8]. The mould and gate geometry can also affect the generation of flow marks/tiger stripes[4,7].

Most reports dealing with flow marks appear to associate these with a flow instability during the mould filling, i.e. a snake-like flow[9] of the melt front giving an asymmetric fountain flow[4,5,8,10,11] (Fig. 1).

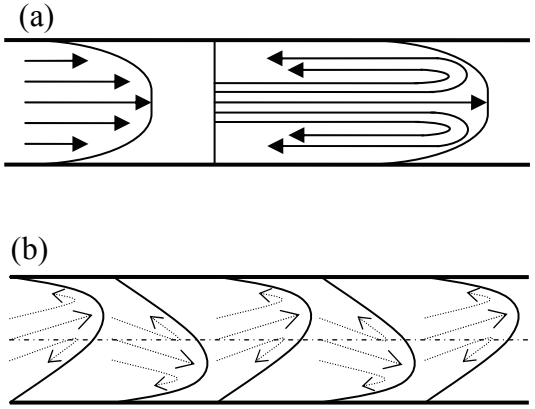


Figure 1. a) Stable symmetric fountain flow, b) Snake-like flow due to unstable fountain flow exhibiting an oscillating stagnation point.

Such flow instability may in principle arise from several causes. Bulters and Schepens[1] associated it with an instability of the flow front due to a coupled elongation process and the numerical stability analysis performed by Grillet et al.[2] using a Phan-Thien-Tanner-type of constitutive equation indicated for some fluids a swirling flow in the vicinity of the flow front.

The associated uneven deformation distribution in the surface regions will yield differences in molecular orientation and surface deformation (also after solidification). This will be reflected in the surface appearance making it less uniform, i.e. flow marks or tiger stripes. Hamada and Tsunasawa[4] noted a difference in composition between the dull and glossy areas when injection moulding a blend of polycarbonate and acrylonitrile-butadiene-styrene copolymer (PC/ABS) which they attributed to rupture of the flow front. Differences in surface appearance and morphology can also be related to differences in break-up and relaxation of incorporated rubber particles due the swirling motion of the flow front[11].

Another phenomenon that is discussed in conjunction with tiger stripes is wall slip. Fluid flow in channels and similar are often modelled assuming zero-velocity at the boundaries of the solid channels. This means that a no-slip condition is at hand. However, above a certain (critical) shear stress, the fluid may slip. Hence, the wall velocity may be non-zero during a certain time period. During the slip, the stress in the surface regions decreases due to loss in friction and this allows the adhesion to the walls of the channel to build up again, i.e. the fluid sticks again. This behaviour is called *stick-slip flow* (Fig. 2). As a result, the surface of injection-moulded parts is deformed and can exhibit defects[3]. Fig. 2 is simplified, since in reality slip may occur on side one of the mould walls which in itself can give rise to a flow instability resembling that previously described[5,12]. Rather closely associated with slip is the mechanism

proposed by Heuzey et al.[6] by which the flow marks are associated with stretching and deformation of semi-solid material close to the mould surface.

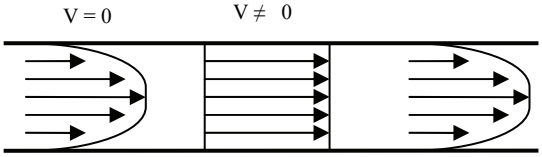


Figure 2. Schematic stick-slip flow.

Among other possible mechanisms, it has been suggested that the thermal contraction of the material close to the mould surface could be one reason for the formation of flow marks[13].

With regard to the relation between the rheological properties of the melt and the formation of flow marks, several studies have pointed to that high melt elasticity, given e. g. through recoverable shear strain or die swell measurements, reduces, at least partly, the tendency towards flow marks[8,10,12]. A more pronounced extensional strain hardening of the melts could also provide benefits in this respect[11]. Adding rubber particles of a high molecular weight can give the desired change in rheological behavior[1] (and indeed many of the materials used contain such a rubber phase) or adding a high molecular tail to the molecular weight distribution[10,12]. In elastomer-modified polypropylene, the difference in gloss between the bands can be associated with a difference in orientation/deformation of the rubber particles[9,11]. Hirano et al.[9] showed that this difference could be controlled by the intrinsic viscosity ratio between the rubber phase and the matrix which then in a sense constitutes a tool for controlling the formation of tiger stripes. Furthermore, a higher recoverable strain of the elastomeric phase could promote the formation of flow marks[11].

The reasons for the occurrence of flow marks considered here cannot be said to be completely clarified[5,7,11] and such a clarification is not within the scope of the present work either. In fact, there may be several possible causes, but whatever the cause is, the unstable flow is likely to be reflected in a variation in the surface topography and/or in the structural morphology of the surface regions. As already noted, this will cause an undesired appearance of the surface but also local variations in properties of the components which certainly are less wanted for products which are expected to have a long life time. When aiming at reducing problems associated with the flow marks, a structural characterization of the defects can be of significant value or even required. Such a characterization of industrially manufactured components is the topic of the present work. A forthcoming work will be focused on the rheological behaviour of the corresponding polymeric materials.

EXPERIMENTAL

Materials

The materials used for injection moulding the different components were based on a number of grades of elastomer-modified polypropylene (PP) containing talc as filler. The talc content was typically around 15 weight-%, but the content of elastomeric particles was, according to the available information, quite low (considerable lower than that used by Hirano et al.[9] and Patham et al.[11]). The melt flow indices of the used PP-grades were between 11 and 16 g/10 min (230°C/2,16 kg) and their densities were typically around 1000 kg/m^3.

Characterization techniques

Several techniques have been employed in order to characterize the structure and properties of the components.

The *gloss* of the dull and the glossy surface areas was measured with a ZGM 1020 Zehntner glossmeter. The analyzed area was 75 mm^2 and the measurements were performed at an incidence/reflection angle of 60^o. The accuracy was ≤ 1 gloss unit (GU) within the range 0 – 100 GU.

A Carl Zeiss DSM 940 A digital scanning electron microscope (SEM) was used to examine the *surface structure* as well as *cross-sections* of the different specimens. A secondary electron detector was used to obtain micrographs at different magnifications. The examined surfaces were initially coated with an approximately 5 nm thick gold layer using a Sputter Coater S150B, BOC Edwards, UK. Cross-sections of the specimens were obtained by fracturing along the flow direction in liquid nitrogen. In some cases the cross-sections were etched in order to partially remove amorphous polypropylene and enhance the visibility of the filler distribution. The etchant consisted of a mixture of potassium permanganate, concentrated sulphuric acid, orthophosphoric acid and water. The fracture surfaces were etched for 2 hours and then immersed in a solution of concentrated sulphuric acid, water and hydrogen peroxide and cooled down close to 0 ^{o}C. The etched sections were finally rinsed with water, methanol and acetone. The etching procedure is described in more detail by Shahin et al.[14].

Stereo-pairs of SEM images, i.e. two micrographs of the same specimen taken at two different viewing angles, can be combined in order to obtain a three-dimensional (3D) image (or model) of the analyzed area[15]. In order to combine the stereo-pairs, the software MeX 5.0 from Alicona Imaging GmbH was used. The 3D micrographs give the possibility to reproduce the surface topography and then calculate a measure of the surface roughness. In particular, the *root-mean-square roughness* S_q (Eq. 1) has been evaluated in order to characterize the surface topography in the height direction over the analyzed area:

$$S_q = \sqrt{\frac{1}{A}\int_A z^2(x,y)\cdot dx \cdot dy} \qquad (1)$$

where $z(x,y)$ is the surface height measured at the coordinates (x,y) of area A. The stereo technique is described in detail by Ariño[16].

In order to ascertain any differences in *chemical composition* between the banded surface regions Fourier transform infrared spectroscopy, FTIR, (Perkin-Elmer System 2000 with a TGS detector performing ATR technique with a zinc-selenide crystal) was used. Electron spectroscopy for chemical analysis (ESCA) was employed for the same purpose. The instrument used was a Perkin Elmer PHI 5500, USA, with a Al(K_α) X-ray source with energy of 1486.6 eV. The accelerating voltage was kept at 14 kV and the take-off angle was 45^{o}.

RESULTS AND COMMENTS

Gloss measurements

An automotive tunnel console (interior part) injection moulded in a PP-based material was evaluated with regard to its visual appearance. The surface exposed towards the driver was textured (Fig. 3), whereas the non-exposed surface had a glossier and smooth appearance. This component exhibited clear tiger stripes, especially on the non-exposed, non-textured surface.

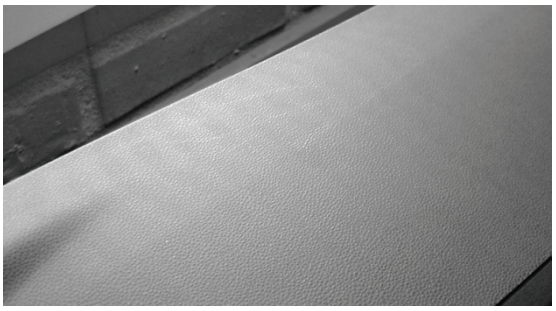


Figure 3. Detail of an automotive tunnel console exhibiting tiger stripes on the textured surface.

The gloss levels of the dull and the glossy areas (on the non-exposed backside) were 9.8 and 12.2 GU, respectively. These results are average values based on fourteen individual measurements on different bands along the console and the standard deviation

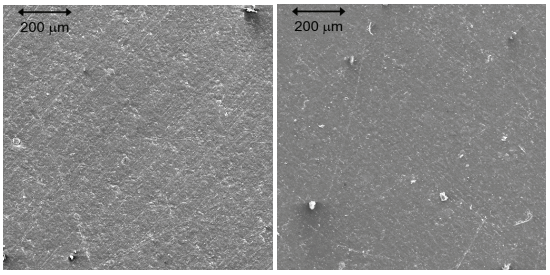
was 0.6 GU for dull bands and 1.3 GU for glossy areas. The difference in gloss between the bands was readily visible and can be considered as large. The magnitude of the gloss difference (2.4 GU) is also in good agreement (although on the high side) with that reported by Hirano et al.[9] for injection-moulded plaques of elastomer-modified PP. The dull bands also exhibited a lighter appearance than the glossy ones.

Surface appearance with SEM and surface roughness

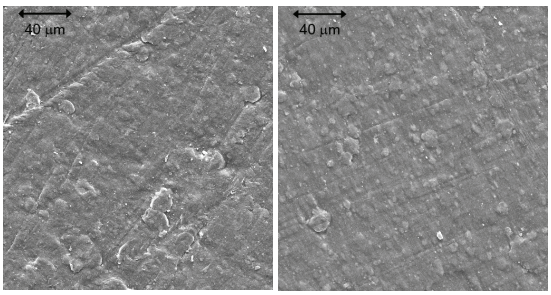
The surface of the non-exposed side of the tunnel console was investigated by SEM. Micrographs of samples from both the dull and the glossy bands were captured at magnifications from 50x to 2000x. Fig. 4 shows some representative micrographs of the two bands. At magnifications between 100x and 500x the difference in their surface appearance/roughness was quite clear. The glossy bands were visibly flatter than the dull ones, which is to be expected[7,11]. At magnifications lower than 100x and higher than 500x, the surfaces appeared to be quite similar, i.e. any difference in surface roughness could not be discerned.

The appearance exemplified by Fig. 4 was quite typical of all components exhibiting tiger stripes used in the present study.

A car bumper exhibiting tiger stripes and the tunnel console mentioned before were also analyzed using the stereo-pair technique described in the Experimental section. A comparison between the dull and glossy bands of the car bumper showed that at 100x magnification the S_q-value of the dull bands (S_q=59 µm) was about 20 % larger than that of the glossy bands (S_q=47 µm). A similar difference was observed also at a higher magnification (1000x); the dull areas having a S_q-value of 6.4 µm, whereas the corresponding value for the glossy regions was 4.8 µm. (A higher magnification in effect filters surface features of greater lateral dimensions, thus giving a lower value of the surface roughness.) The surface roughness of the dull and glossy areas of the tunnel console exhibited qualitatively a similar difference.



(a)



(b)

Figure 4. Scanning electron micrographs of the banded regions of the tunnel console: a) dull and glossy band 100x respectively; b) dull and glossy band 500x respectively.

Fractured cross-sections

The car bumper and the tunnel console were further analyzed using SEM. Samples from both components were cut from the flow marks and both surface regions and cross-sections were investigated. The samples were etched as described in order to enhance the visibility of the filler particles and other features.

Fig. 5 provides a typical illustration of the general observation. There was in most cases a noticeable difference in filler distribution between the dull and glossy bands. The figure includes two SEM micrographs at 500x of etched cross-sections from the tunnel console; in the light bands (dull bands) the concentration of filler

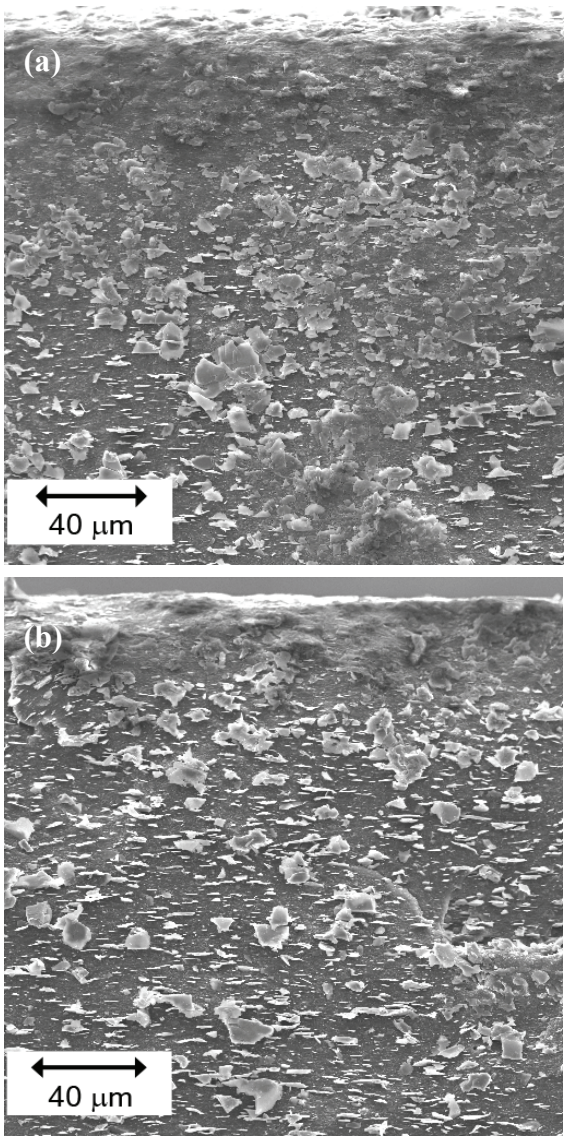


Figure 5. Scanning electron micrographs (500x) of etched cross-sections of the banded regions where a) dull band and b) glossy band.

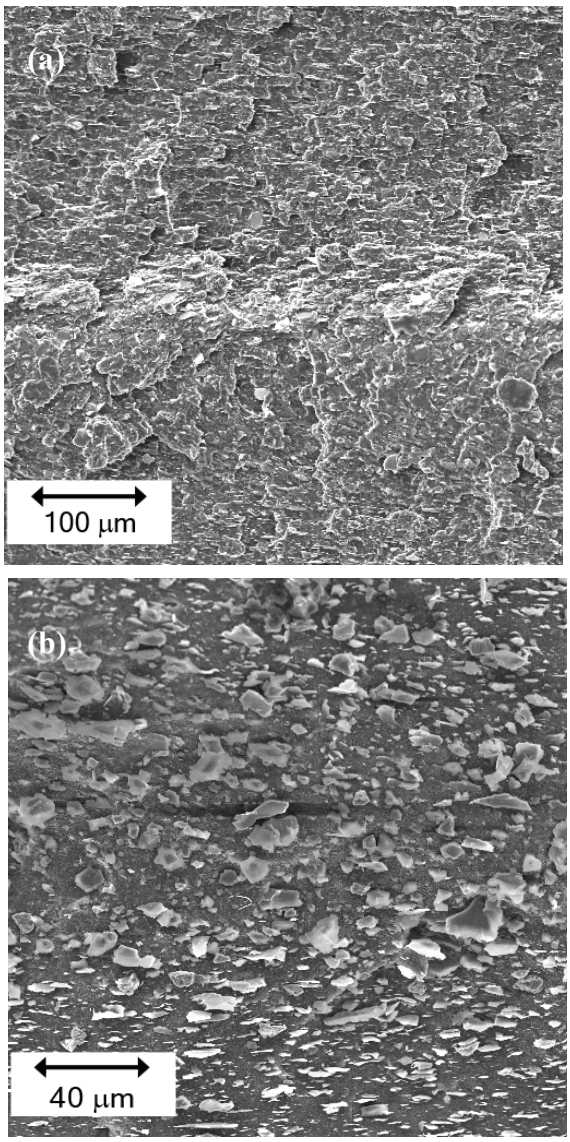


Figure 6. Scanning electron micrographs of the central area of an injection moulded tunnel console where a) is not etched sample 200x and b) etched sample 500x.

particles in the surface region was lower than in the glossy bands. No clear evidence evidence of elastomeric particles could be discerned, probably due its low content.

Phase separation of fillers etc along the radius of pipes in Poiseuille flow is a well known effect[17]. In particulate suspensions, an increase in the particle concentrations towards the centre of the channel, which represent the region of highest local velocity and lowest shear stress, takes place. This was also evident in the case of the injection-moulded components studied here (see Fig. 6). A higher particle concentration could be observed in the central areas of the specimens. The flaky particles also appeared to be less oriented in the flow direction, which corresponds to the lower stress in the central part of the flow regime.

ESCA and FTIR analysis

ESCA and FTIR were used in order to characterize the chemical composition of the surfaces of the specimens. However, none of

these techniques pointed to any significant differences in composition between the dull and the glossy areas of the analyzed automotive components. As an illustration, the ESCA spectra shown in Fig. 7 from the dull and glossy regions of the tunnel console samples appear to be very similar. The carbon and oxygen peaks are clearly evident, which is not unexpected, and, in both bands, some small amounts of magnesium and silicon are noted. This is likely to be associated with the talc filler.

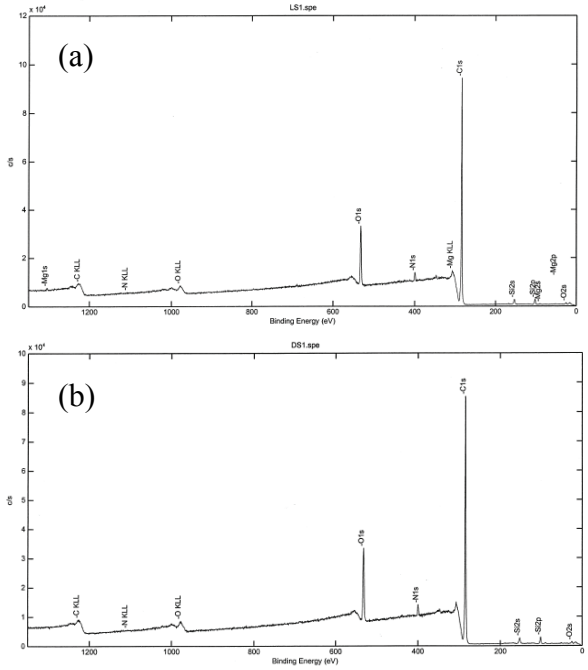


Figure 7. ESCA spectra (binding energy, eV) from the surfaces of the dull (a) and the glossy, (b) bands of the car tunnel console.

Fig. 8 shows the C(1s)-peaks separated into the components C1 – C4[18] for the dull and the glossy bands. No substantial differences between the spectra can be noted, although the C2-component associated with C-O bonds were slightly more developed in case of the glossy/darker bands. This can perhaps be interpreted in terms of some oxidation of the material, but this difference is not regarded as significant.

The FTIR-measurements provided similar information, i.e. significant amounts of carbon and oxygen, but no clear differences in composition between the different bands. This is in agreement with the observations of Mathieu et al.[5] who did not detect any noticeable differences in composition between the bands in the case of an ethylene-propylene copolymer.

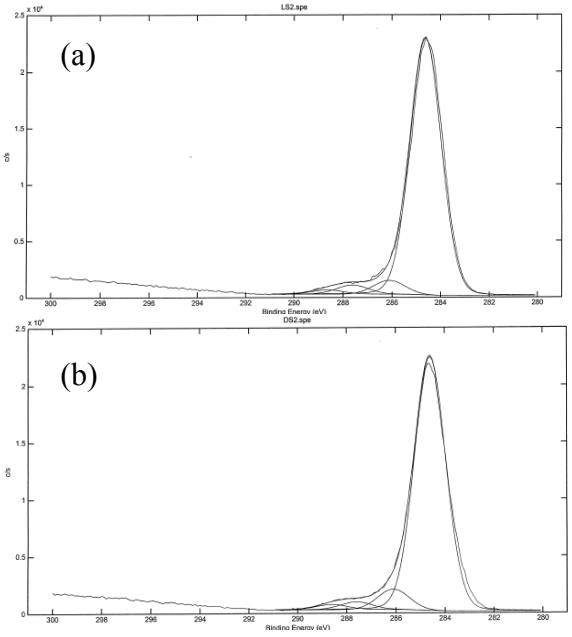


Figure 8. ESCA spectra (C(1s)) for the dull (a) and the glossy bands, (b) of the tunnel console.

DISCUSSION

As shown in Fig. 5, it is clear that, in the inspected samples, the filler content close to the surface of the dull bands was lower than in the glossy bands, likely due to a difference in deformation/stress field in the surface regions. This may be associated with a possible stick-slip phenomenon or a difference in deformation history associated with an instable flow front. The course of events is in the following depicted in terms of slippage, but a wiggling motion of the flow front could lead to a similar result. As a starting point, it may be assumed that the dull regions corresponds to a sticking to the wall of the mould (or less deformation of the surface regions due to an upward motion of the wiggling flow front) As mentioned earlier, the surface regions will always contain less filler than the interior of the flow channel during the flow[17].

Fig. 9 schematically illustrates that when a part of the melt sticks to the mould surface, the flow or deformation, which could be of a semi-solid nature, just behind of the sticking region will tend to accumulate more of the low-filler content material in the sticking part of the melt. At the same time the slipping surface regions downstream (or deforming regions due to a downward motion of the flow front) experience a tensile deformation which gives a thinning of the low-filler content surface regions in that region. Both phenomena contribute to a thicker region of low filler content close to the surface of the parts that "sticks" to the walls of the mould cavity.

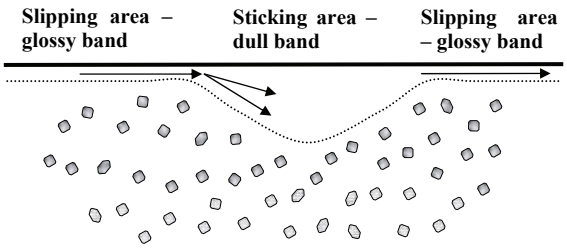


Figure 9. Schematic illustration of the possible mechanisms that govern the distribution of the filler particles in the surface regions of injection-moulded plastics showing tiger stripes.

The other clear difference between the glossy and the dull areas was revealed by their surface roughness, i.e. the glossy regions were smoother. If these areas correspond to the slipping zone (or a more deformed layer due to the motion of the flow front), this would imply that the somewhat rough mould surface is not replicated to the same extent as in the case of the dull regions. On the other hand, it may be argued that a slipping and stretching might result in surface deformations leading to a rougher surface, especially if this involves a cohesive failure of the surface layers close to the mould wall. In such a case one would expect the slip to give a dull appearance. It may also be remarked that the somewhat lighter appearance of the dull regions thus is not to be related to a higher content of filler particles. Instead, the rougher surface will give a higher fraction of diffusely reflected light giving a lighter color[19,20].

It is known that the lighter (dull) bands become more visible with time. To some extent this may be due to contamination preferably adhering to the somewhat rougher surface. It may also be speculated on the possibility of a difference in crystallinity in the surface regions between the lighter and darker bands. If the crystallinity is lower in the light band regions, it may be easier for stabilizers etc to migrate towards the surface in these areas. Such depletion would possibly enhance a local degradation leading to a pronounced banded appearance. More experimental work is however required in order to substantiate this point.

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