Inhibition of the surface levelling of thermosetting polyester powder coatings caused by surface tension gradients

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Previous theoretical and experimental work has shown that surface tension gradients in liquid layers create surface defects and inhibit the levelling of an uneven surface. In coatings deposited from thermosetting polyester powders, which are studied here, small amounts of a low molecular-weight acrylate are incorporated to act as a "flow agent." We find that this additive lowers the surface tension of the polymer melt and has a minor effect on the melt viscosity. A slower rate of levelling results from the decreased surface tension. We provide experimental evidence that lateral gradients in the surface tension of the polymer melt, resulting from the non-uniform distribution of the flow agent, inhibit the levelling of the surface. Specifically, the surface roughness of a powder coating is up to three times greater when a steep surface tension gradient is purposely created through powder blending. Surface tension gradients might also be responsible for the greater surface roughness (observed with atomic force microscopy on lateral length scales of 100 μm) that is found in coatings that contain flow agent. © *2002 Kluwer Academic Publishers*

1. Introduction
A well-established method of depositing a hard, glossy coating on a variety of substrates is through thermosetting polymer powder technology [1]. During the film formation process of such coatings, dry polymer particles undergo coalescence and levelling to create a smooth surface, while simultaneously crosslinking reactions build up a three-dimensional molecular network. Although an attractive technology from the perspective of its low energy use, minimal environmental impact, and good product quality [2], powder coating technology still has some drawbacks. A primary one is that the coating surface is sometimes subject to dimpling and undulations, referred to as an "orange peel" defect, which diminish its attractive appearance. Considerable effort has been expended to understand the origins of this problem.

One tactic to create smoother, defect-free surfaces is to incorporate levelling aids in the formulation [3]. This type of additive encourages the flattening out of surface undulations by *increasing* the surface tension to enhance the driving force for the process [4]. Another type of additive, known as a flow agent (often a low molecular weight polymer), is intended to serve the related purpose of eliminating any surface tension gradient by diffusing to and along the surface. Flow agents usually lower the surface tension. As noted by Wulf *et al.* [3], these terms are rather arbitrary, and the classification cannot usually be made unambiguously. Powder formulations usually have a balance of levelling aids and flow agents with the intention of their acting independently and at different stages during the film formation [4].

Quite separately from the work on powder coatings, there has been progress in understanding the levelling of solvent-borne paints. The relationship between gradients in surface tension and defects in paints has been recognised for many years [5, 6]. Most significantly, mathematical models have been developed by Schwartz and co-workers to take into account the role of surface

(a)

uniform γ

(b)

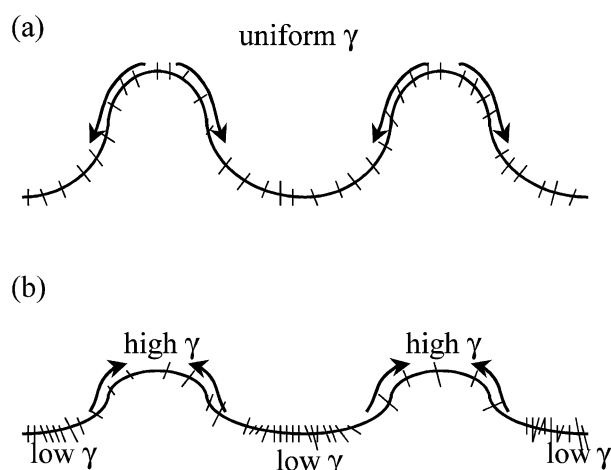high γ   high γ

low γ   low γ   low γ

Figure 1 Schematic illustration of how surface tension gradients can retard levelling in coatings. (a) Initially, there is an even distribution of surfactant and hence a uniform surface tension. The Laplace pressure (i.e., capillarity) drives lateral flow that carries surfactant from "hills" to "valleys." (b) The surface now has a surface tension gradient. Material flows from regions of lower surface tension (i.e., valleys) to regions of higher surface tension (hills).

tension gradients (STGs) in inhibiting the levelling of paint films [7, 8] and in leading to crater formation [9]. As a brief summary of this work, it has been found the shear stress imposed by the Laplace pressure on a curved surface causes surfactant molecules at that surface to be displaced from their equilibrium, uniform distribution. A gradient in the surface tension then results. Flow is then encouraged from regions of low surface energy to regions of higher surface energy. This flow can counteract the flow from the crest of surface undulations to the valleys. This phenomenon is represented schematically in Fig. 1. The degree of retardation of the flow is a function of the strength of the surfactant, R, which, in turn, depends on the decrease in surface tension induced by the surfactant, among other parameters [7].

STGs have been proposed as a possible cause of poor levelling in powder coatings [3, 10], but there has been limited experimental or theoretical work on this topic reported in the literature. We suggest that the basic concepts of the STG models should also apply to powder coatings, in which there is an analogy between flow agents at the surface of a polymer melt and surfactants at the surface of water.

The effects of STGs are expected to be particularly acute in the thermosetting polymer coatings studied here. In a thermoplastic melt, a retardation of the levelling is not an insurmountable problem, because if the time is sufficiently long, complete levelling will still occur. In a thermosetting melt, on the other hand, an exceedingly high viscosity develops over time, and the levelling process then comes to a halt. In this work, we provide experimental evidence that flow agents can contribute to STGs and result in a rougher surface of coatings made from thermosetting polymer powders.

2. Experimental details
2.1. Materials
The standard powder formulation consists of a polyester (PE) polymer (64% w/w), triglycidyl iso-

cyanurate (TGIC) as a curing agent (5% w/w), a flow agent (1% w/w), a titanium dioxide pigment (30% w/w, Kronos 2160), and a small fraction of benzoin, which functions as an anti-pinholing agent [11]. The PE is a carboxyl functional resin with an acid number of 30 mg KOH/g PE. The flow agent, which is sold under the tradename of Resiflow PV5 (Estron Chemical Co., USA), consists of a low molecular weight acrylic (M_w ranging between 6,000 and 13,000 g mol^{-1}).

The components were co-extruded and then pulverised to make particles with a wide size distribution, ranging from about 10 to 60 μm, according to examination by scanning electron microscopy. Modulated differential scanning calorimetry (TA Instruments model 2910) found that the glass transition temperature of the standard formulation is 55°C. The analysis also indicated that crosslinking of the polymer *started* near a temperature of 87°C, which is well below the recommended stoving temperature of 190°C, but the rate increased with increasing temperature.

2.2. Techniques
The viscosity of the polymer was measured as a function of time at the stoving temperature of 190°C using a dynamic rheometer with a parallel plate configuration (Rheometrics Scientific model SR5). The specimen was prepared by the compression of the powder in a die at room temperature to create a disc with a thickness of 1.5 mm and a diameter of 25 mm. In testing, the disc was loaded between the parallel plates of the rheometer. The powder was melted and equilibrated at 80°C with a gap spacing of 1 mm. Measurements were carried out in the stress-controlled mode with a strain frequency of 10 rad s^{-1}. The complex viscosity, η^*, was derived from the ratio of the complex modulus over the angular frequency.

A relative comparison of the surface tensions of the various formulations was obtained by contact angle measurements. Small amounts of powder were loaded onto clean, polished stainless steel plates heated in air on a hot stage (Linkam model TP 93, Leatherhead, UK) to a temperature of 190°C. The powder melted to form a shallow dome. A digital video camera was positioned to view across the plane of the substrate in order to record the shape of the polymer melt as a function of time. An equilibrium shape was usually obtained within a few minutes, before the completion of the crosslinking (according to independent viscosity measurements). The contact angle was determined through quantitative analysis of the images.

Coatings were deposited on steel substrates by spreading known quantities of powder uniformly across the surface by hand. The substrates were heated on a hot stage in air at 90°C/min to the stoving temperature of 190°C and held for 10 minutes before cooling at the same rate.

The film formation process was observed *in situ* using a reflected-light laser scanning confocal microscope (Zeiss LSM510). The key feature of confocal microscopy is that only light from the focal plane of the objective lens is detected. Samples for confocal

microscopy were prepared by spreading the powder onto a glass coverslip. The powder was then heated in air using a temperature-controlled stage (Linkam) mounted on the microscope. Heating was carried out at 10°C min^{-1} to a maximum temperature of 190°C. This heating rate allows time for each stage of the film formation process to be recorded in detail. Images were acquired using 543 nm laser light. A low magnification objective ($\times 5$ with a numerical aperture of 0.15) was used to give a large field of view (2 mm $\times$ 2 mm). Images were acquired at selected temperatures with an acquisition time for a single image being on the order 10 s.

Surface topographies of the coatings were determined at room temperature via atomic force microscopy (Digital Instruments, Nanoscope IIIa) in tapping mode using a large area (120 μm $\times$ 120 μm) scanner. A silicon cantilever, oscillating at a frequency of 300 kHz and having a spring constant of 42 N/m, was employed. Scan frequencies were typically 0.2 Hz and always less than 0.5 Hz. The topography over areas of up to 20 mm $\times$ 20 mm was determined with a contact surface profiler (KLA-Tencor P-11) using a contact force of 2 mg and a scanning rate of 400 μm s^{-1}.

3. Results and discussion

A simple preliminary experiment shows that it is essential to include the flow agent in the formulation in order to achieve a coating without craters and pinholes. Fig. 2 compares the appearance of coatings prepared with and without the addition of the flow agent. Both coatings were prepared by electrostatic spraying of the powders onto steel plates and then baking in an industrial oven
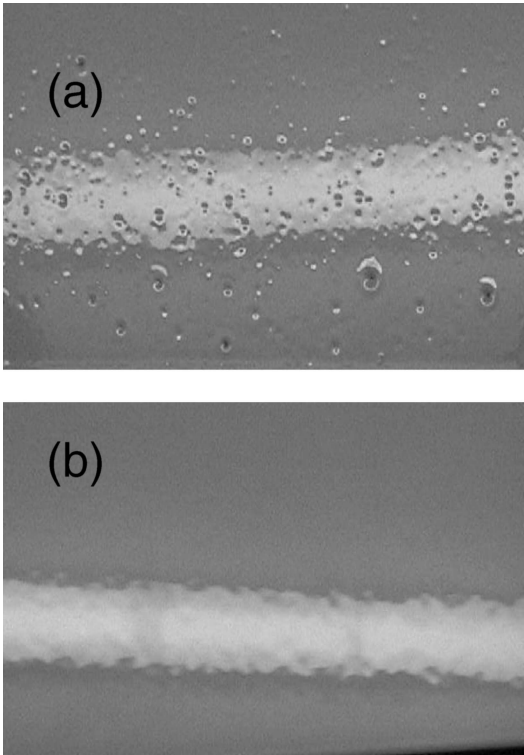
Figure 2 Photographs of coatings with (a) 0% flow agent and (b) 1% flow agent. The field-of-view is about 150 mm by 70 mm. The bright strip in each image is the reflection from a fluorescent ceiling lamp.
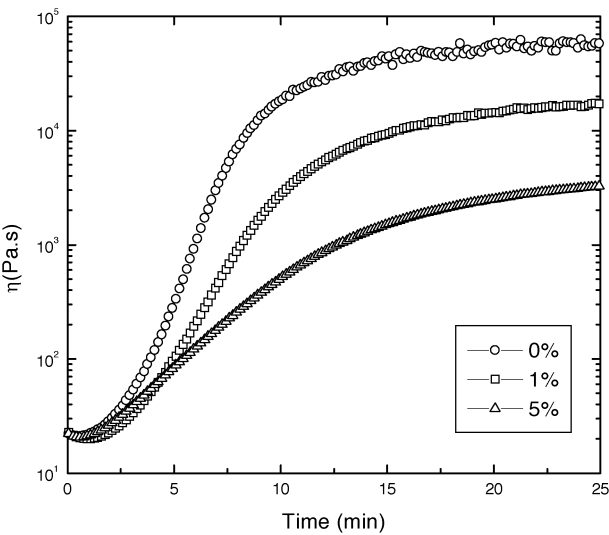
Figure 3 The time dependence of the complex viscosity at a temperature of 190°C for powder formulations with varying concentrations of flow agent: 0% (○); 1% (□); and 5% (△).

at 190°C for 15 minutes. Although surface undulations with a wavelength on the order of one to two mm are seen in a coating based on the standard formulation, the surface of the coating without flow agent has more severe macroscopic defects. Subsequent experiments consider the influence of flow agents on shorter lateral length scales.

As surface levelling is driven by the surface tension and opposed by the polymer viscosity, we next consider the influence of the flow agent on these two properties. Fig. 3 shows how the viscosity changes over time for three different powders containing 0, 1 and 5% w/w flow agent. The viscosity at time $t = 0$ represents the viscosity when the powder has melted but no crosslinking has occurred. For all three powders, this viscosity is initially 20 Pa s. The flow agent has no observable effect on the *initial* melt viscosity. In thermoplastic epoxy melts, it has likewise been found elsewhere [3] that flow agents do not alter the viscosity. The viscosity for all three powders approaches a plateau value (listed in Table I) after about 20 minutes. The slowing down in the rate of the viscosity increase indicates the completion of crosslinking reactions. The highest plateau value (*ca.* 6×10^4 Pa s) is obtained in the powder without flow agent, whereas a much lower plateau is reached (*ca.* 3×10^3 Pa s) when there is 5% w/w flow agent. The flow agent does not appear to affect the initial viscosity of the melt but it influences the crosslinking density that develops. The rate of crosslinking, as indicated by the rate of increase in viscosity is slowest with 5% w/w flow agent.

TABLE I Effect of flow agent on polyester melt viscosity and equilibrium contact angle on steel at 190°C

Flow agent concentration (% w/w)	Plateau value of viscosity (10^3 Pa s)	Equilibrium contact angle ($^\circ$)
0	60	28 ± 2
1	20	20 ± 2
5	3	19 ± 2

Table I also lists the equilibrium contact angles on steel for these same three formulations at 190°C. The contact angle of the powder without flow agent is about eight degrees higher than with the formulation containing 1% w/w flow agent. Increasing the concentration of flow agent to 5% w/w causes no change in the contact angle within the uncertainty of the measurement. The melt surface obtained the contact angle within a few minutes of the powder melting, and the angle did not change significantly over time.

Contact angles are clearly reduced as a result of the addition of flow agent. This result can be used to an indication of a change in the surface energy of the melt with the addition of flow agent. Assuming that the Young–Dupré equation [12] holds for this system and that the flow agent does not affect the melt/substrate interfacial energy, we attribute the differences in contact angle to differences in the melt's surface tension. Using a literature value [13] for the surface tension of a polyester melt without flow agent at a temperature of 190°C, $\gamma = 33$ mN m^{-1}, we *estimate* that the surface tension is reduced by $\Gamma \approx 2$ mN m^{-1} through the addi-

tion of 1% w/w flow agent. (An upper limit to the value of Γ is 7 mN m^{-1}, because the surface tension of the neat flow agent is 26 mN m^{-1}.) According to standard theories of levelling [14, 15], a lower surface tension will increase the characteristic time for the decay of a surface wave. From that consideration alone, one would expect the addition of flow agent to slow down the rate of levelling.

We also note that the surface tension of the powder melts is the same at the two concentrations of flow agent (1% w/w and 5%). In work reported elsewhere [3], accurate measurements of the surface tension of melts of epoxy resins were obtained using axisymmetric drop shape analysis. It was similarly found that the surface tension was reduced by a poly(acrylate) levelling aid, but Γ was the same for two different concentrations (0.1 wt% and 1.0 wt%). In both sets of experiments, a small amount of additive is sufficient to saturate the surface. Additional additive does not reduce the surface tension any further.

Images of powder coatings, obtained using confocal microscopy during film formation, are shown in Fig. 4.
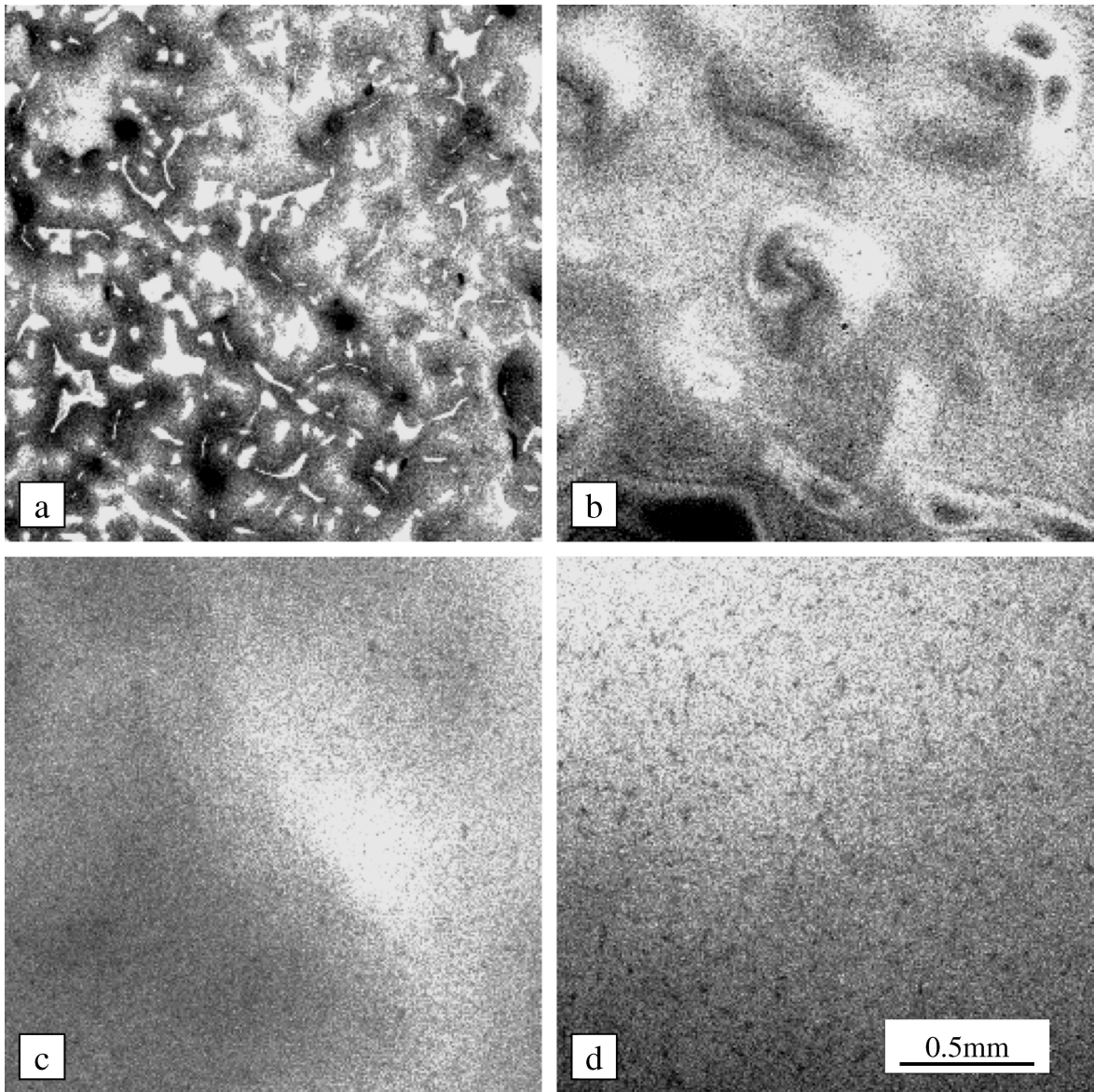

Figure 4 Confocal microscopy of the same surface of the standard formulation (1% flow agent) at different temperatures while heating at 10°C/min.: (a) 100°C; (b) 110°C; (c) 130°C; (d) 190°C.

In reflected light mode, contrast arises principally from differences in refractive index. The TiO_2 particles in the powder have a very high refractive index, and so they reflect light strongly. The particulate nature of the TiO_2 leads to the grainy appearance of the images. The level of this graininess is not reduced with slower scanning or increased image averaging, as would be expected if it was arising from image noise. A second source of contrast in the images is topography. Because light does not penetrate deeply into the opaque sample surface, if the focal plane lies beneath the sample surface no light will be seen. Likewise, if the focal plane lies *above* the sample surface no light is seen. For the low magnification objective used in this work the focal plane has a depth of 60 μm. Preliminary experiments showed that the dark and bright areas of the form shown in Fig. 4a arise from surface topography. Thus, in this instance, the confocal microscopy is providing qualitative information on how the surface morphology evolves over lateral length scales up to 2 mm.

The surface undulations have completely decayed when the melt reaches a temperature of 130°C in the formulation with 1% flow agent when heating at a rate of 10°C/min (Fig. 4c). Measurements of viscosity when heating at the same rate found that the viscosity at this temperature is *ca*. 300 Pa s. By comparison, in a formulation without flow agent, a smooth surface on a 2 mm length scale was obtained at a temperature of 110°C, as shown in Fig. 5. This temperature corresponds to a viscosity of 2×10^3 Pa s. The faster levelling observed in this formulation is attributed to its surface tension being higher by an estimated 2 mN/m.

Comparison of the micrographs in Figs 4 and 5 also reveals some differences in the appearance of the coatings of different formulations. Small dark spots are seen at the surface of the formulation containing 1% flow agent (Fig. 4d), which are not seen when at the same temperature when there is no flow agent (Fig. 4c). When the concentration of flow agent is higher (5%), these spots emerge at a temperature as low as 130°C. We
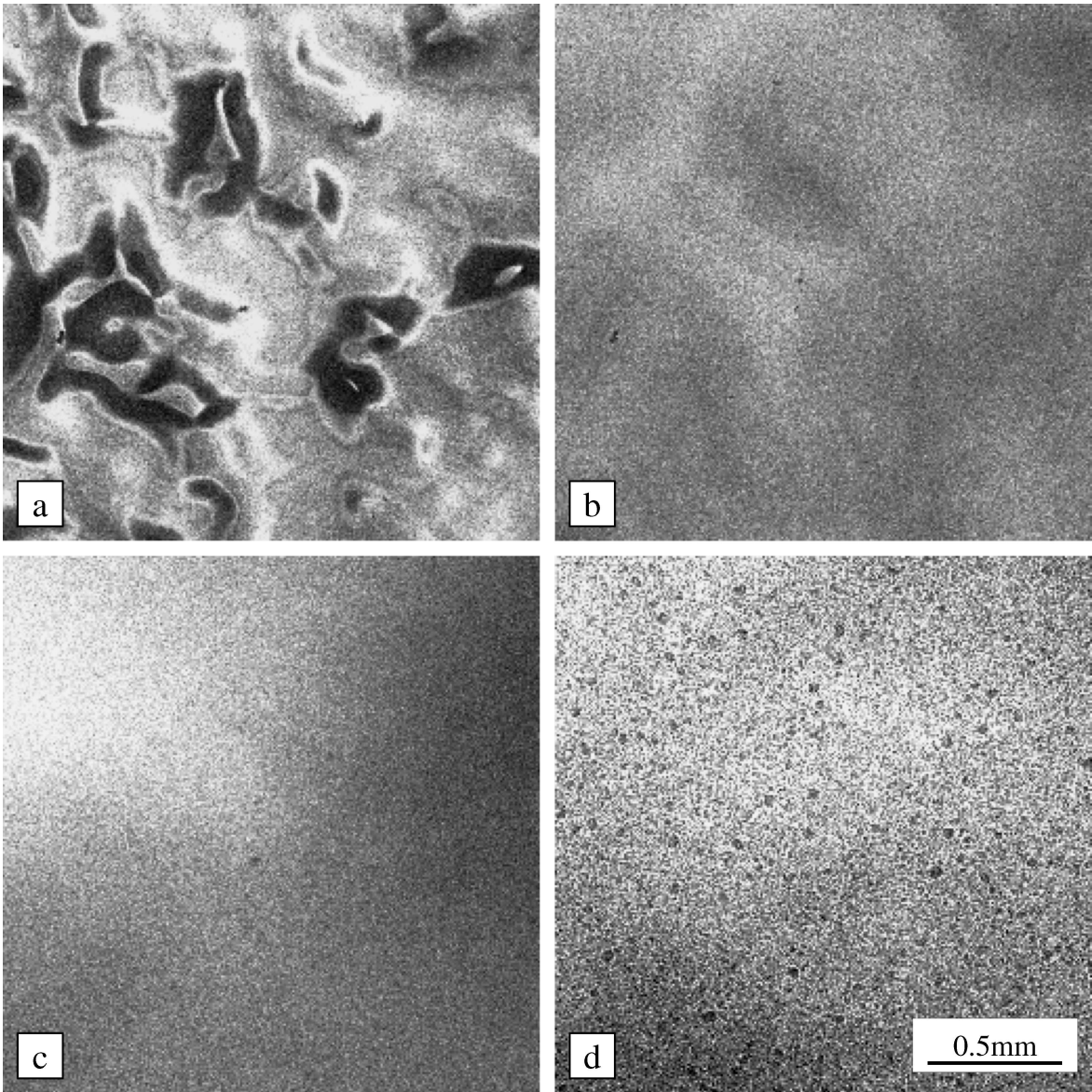

Figure 5 Confocal microscopy of the same surface of a powder formulation containing 0% flow agent at different temperature while heating at 10°C/min.: (a) 100°C; (b) 110°C; (c) 190°C for 10 minutes. (d) Surface of a coating with 5% flow agent at a temperature of 130°C.

interpret these spots as clusters of the flow agent. As they are not seen at lower temperatures, it seems that they migrate to the surface during the thermal treatment.

It is relevant now to consider the theory relating to surface tension gradients. Schwartz and co-workers [7] defined the "strength" of the surfactant, R, as

$$R = \frac{3\Gamma}{\gamma h^2 k^2}$$

where h is the coating thickness (*ca.* 1.6×10^{-4} m in coatings studied with AFM) and k is a wave vector given as $2\pi/\lambda$, with λ being the wavelength of the surface undulation. For the system studied here, and using estimated values of Γ and γ from above, R ranges from 6.5×10^{-4} to 4.5 as λ varies from 6×10^{-5} m (the size of the largest particles) to 5×10^{-3} m (a macroscopic length scale) when the concentration of flow agent is 1%. Simulations have predicted that with intermediate values of R, on the order of 0.1, levelling is maximally retarded, provided there is no diffusion of surfactant to reverse the STG. The length scale over which the effects of the flow agent on levelling is predicted from theory to be dominant, λ, is thus between about 0.5 and 1 mm.

Subsequent experiments therefore were designed to examine the influence of surface tension non-uniformity on the levelling of lateral features with a characteristic length scale of $\lambda \approx 0.5$ mm. It is unfavourable for a polymer melt to spread on a surface with a lower γ, whereas it is favourable for it to spread on a surface with a higher γ. It was hypothesised therefore that a powder without flow agent (and a higher γ) should not spread readily on the surface of a powder with flow agent. This hypothesis was tested in a simple experiment in which 160 μm layers were deposited from two powders: 0% flow agent and 5% flow agent. On top of this base, clusters of the opposite powder

were deposited. The lateral size of the larger clusters was approximately 0.5 mm. The two specimens were then heated under the recommended stoving conditions of 190°C for 10 min.

Fig. 6 shows a comparison of the surface topography of the "0% on 5%" and the "5% on 0%" coatings. Both surfaces exhibit a broad undulation associated with the curvature of the substrate. The surface of the coating with 0% flow agent on the 5% flow agent base exhibits additional "hill-like" features attributed to poor levelling of the deposited clusters. In the reverse situation, these features are not observed, which indicates that good levelling was achieved. The powder with 0% flow agent has a surface energy that is estimated to be higher by 2 mN/m. The results suggest that this small difference is sufficient to impede levelling.

Other experiments were conducted to determine if the presence of flow agent influenced the surface topography observed on a scale of μms in the final coating. Fig. 7 shows representative AFM images of coatings with 0%, 1%, and 5% flow agent. The cross-sectional traces give an indication of the differing amplitudes and wavelengths of the surface waves. The RMS roughness values obtained from this analysis for each of the powder compositions are given in Table II. On a lateral

TABLE II Root-mean squared roughness values for coatings from various powder formulations obtained from AFM (120 μm × 120 μm area)

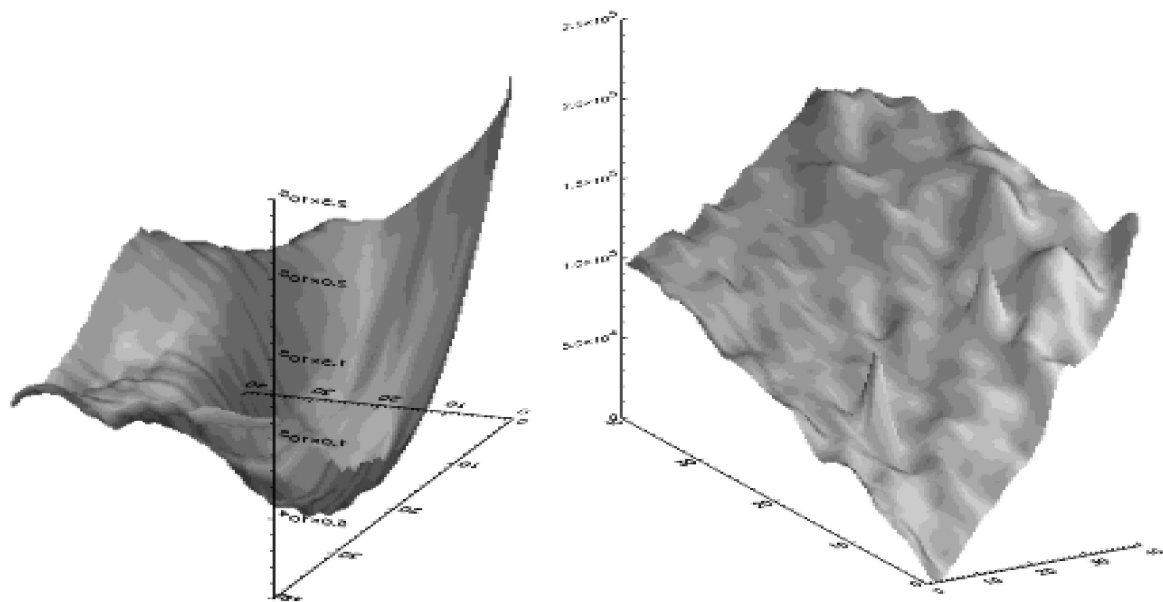| Powder formulation	Coating thickness (μm)	RMS roughness (nm)
0% w/w flow agent	161	11.8
1% w/w flow agent	157	23.0
5% w/w flow agent	159	25.6
1:1 blend of powders with 0% and 1% flow agent	161	31.8
1:1 blend of powders with 0% and 5% flow agent	157	35.9

Figure 6 Comparison of surface topography as determined with surface profilometry for coatings heated at 190°C for 15 min. (a) Powder particles with 5% flow agent were spread in clusters on a base of 0% w/w flow agent. (b) Powder particles with 0% flow agent were spread in clusters on a base of 5% w/w flow agent. Image areas are 3 mm × 3 mm. The vertical scale is the same for both images.
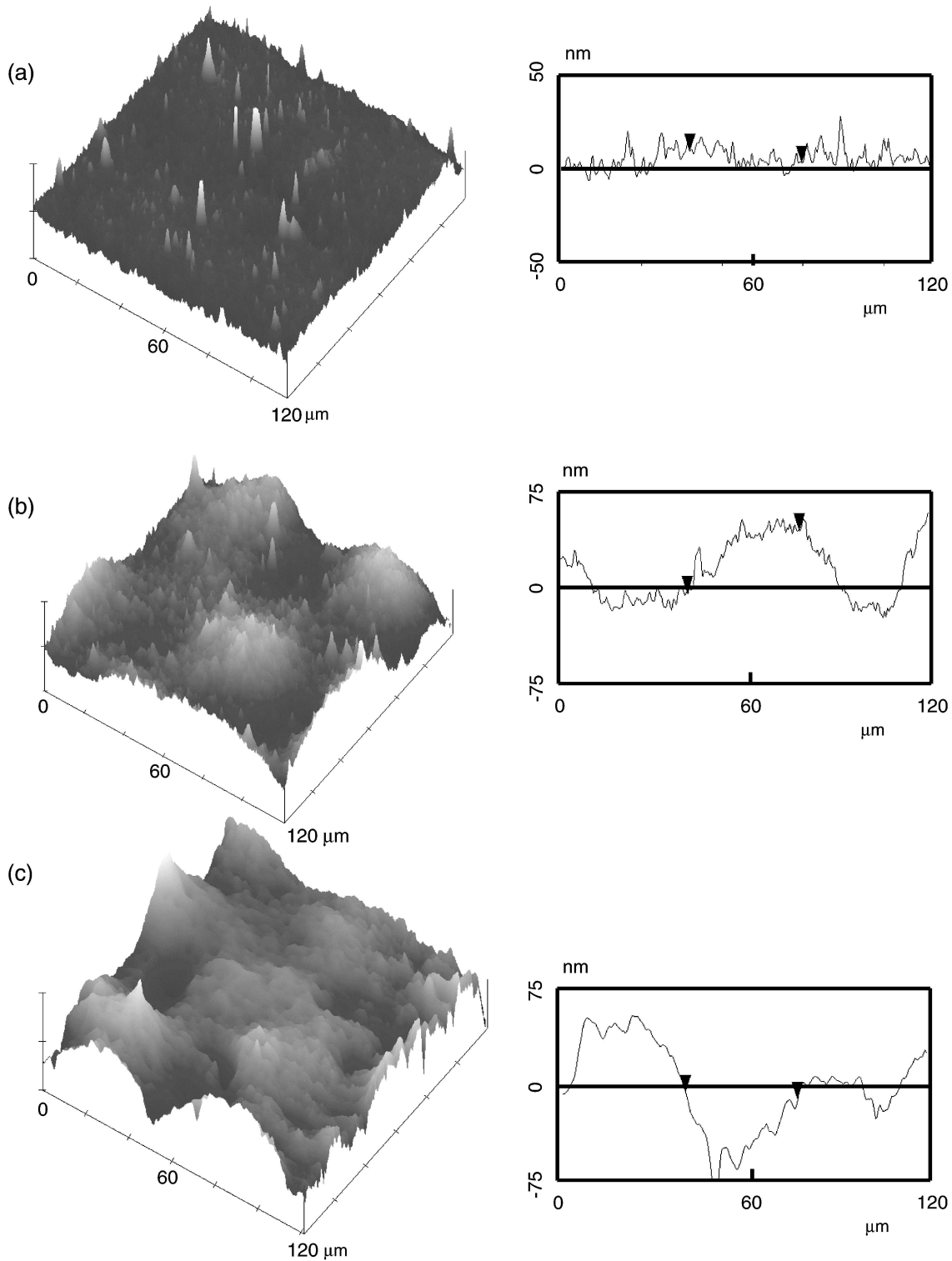
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Figure 7 AFM height images of coatings deposited from powders containing (a) 0%, (b) 1% and (c) 5% flow agent.

(i.e., in-plane) length scale of 120 μm, the smoothest surface is found in the formulation without flow agent, which has an RMS roughness of 11.8 nm. In the two compositions containing flow agent, the roughness is more than twice this value. The wavelength of the observed surface roughness (in the plane of the film) is *ca.* 60 μm. This length scale is comparable to the size of the larger powder particles, indicating that the observed roughness might originate from the contours of individual particles or small clusters. The value of R corresponding to this λ and (using experimental values for Γ, γ and h) is ca. 6.5×10^{-4}. Simulations [7] pre-

dict that with this extremely low value of R, there is negligible retardation of surface levelling.

For the specimens shown in Fig. 7, the expected characteristic time for levelling, τ, can be calculated [14, 15]. With $h = 1.6 \times 10^{-4}$ m and $\lambda = 6 \times 10^{-5}$ m, which is consistent with experimental observation, we are in the limit $kh \to \infty$, and τ is given as $\eta\lambda/\gamma$. With $\gamma = 3.3 \times 10^{-2}$ N m^{-1} and an average η taken to be 10^2 Pa s, τ is predicted to be only 0.2 s. It is well established that shorter wavelengths of surface undulations will be the first to decay, whereas longer wavelengths decay more slowly. It is therefore remarkable that
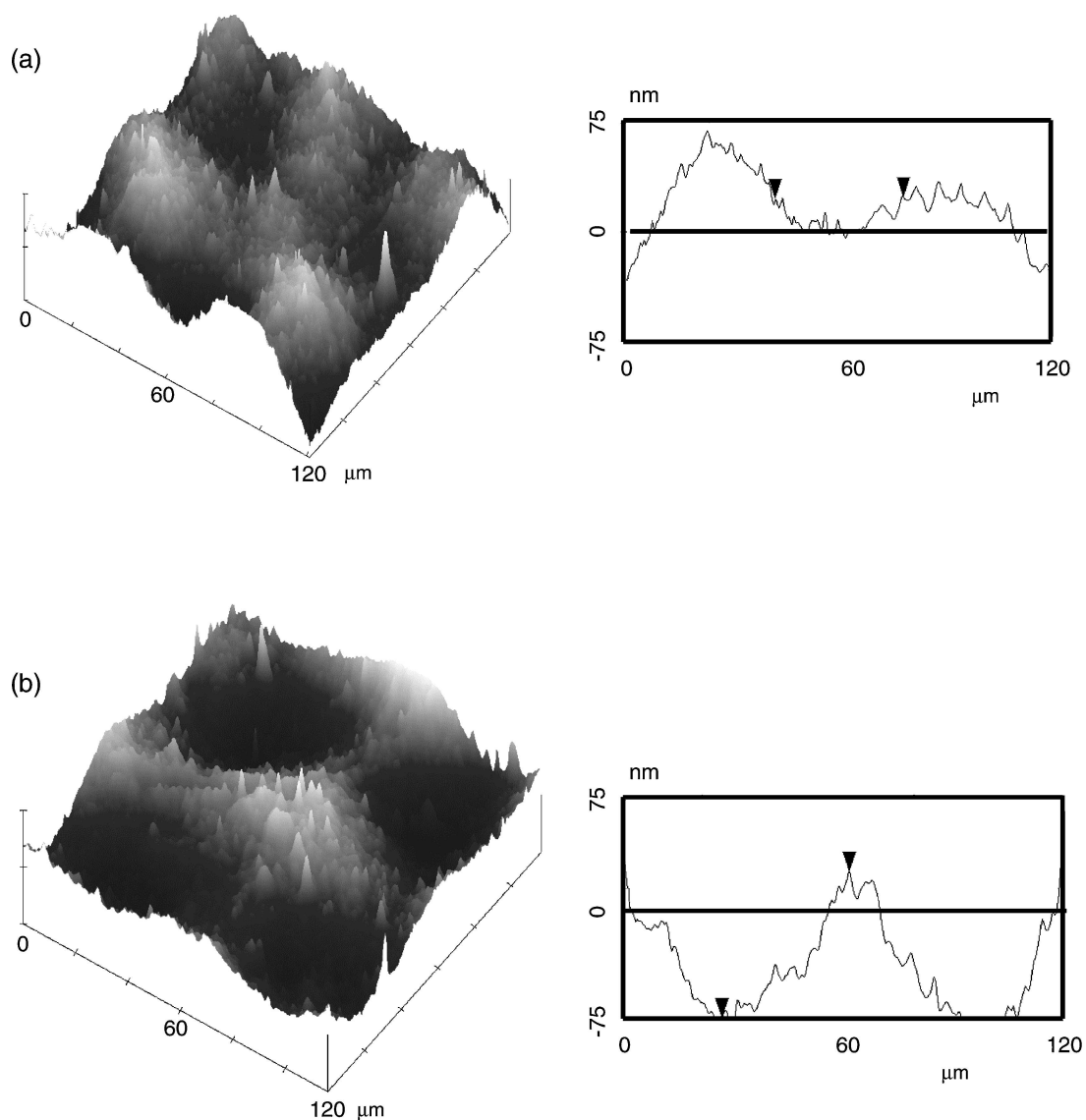
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Figure 8 AFM height images of coatings deposited from blends of powders: (a) 0% and 1% flow agent; and (b) 0% and 5% flow agent.

significant roughness on the relatively short length scale of the particles ($\sim$60 μm) exists on the surface of the coatings. Although surface features on this length scale are not directly noticeable by eye, they can diminish the glossiness [16].

The dominant wavelength of surface roughness that is typically observed in powder coatings is on the order of one to three mm [17, 18], because waves on shorter length scales decay at a faster rate under the action of capillarity alone [15]. If the surface tension remained uniform, then the roughness observed at the surfaces in Fig. 7 is thus predicted to diminish over a time much shorter than the time of film formation, even taking into account the changing viscosity in the thermosetting melt. Indeed, it is usually on longer lateral length scales, with λ up to several mm, where powder coating surfaces are known to exhibit surface roughness. It is not obvious if this short-scale roughness can be attributed to STGs stemming from the non-uniform distribution of flow agent. Nevertheless, the addition of flow agent leads to greater surface roughness over these short lateral length scales.

This effect was explored further in subsequent experiments. Under the assumption that diffusion of the flow agent is slow on the time scale of the levelling, surface tension gradients can be created by blending powders with differing concentrations of flow agent. Fig. 8 shows AFM height images of such coatings formed under standard conditions from a blend of two powders. These blended coatings have a higher RMS roughness (as given in Table II) than coatings made from either of their constituent powders. The blend of 0% and 5% powders (Fig. 8b) has the highest roughness of 35.9 nm. The three-dimensional image reveals four distinct "valleys" on the surface of this coating, each surrounding a central "hill." One interpretation of this image is that the central hill was formed by a polymer with a higher surface tension (0% flow agent). Flow of material from areas with lower surface tension (i.e., containing flow agent) to areas of higher surface tension (i.e., without flow agent) is expected. Shear forces are thus generated that carry material laterally. In the case shown in Fig. 8b, material was transported to a "hill" to oppose the counterflow under the Laplace pressure. The case of the mixed powders is expected to create very steep surface tension gradients that would otherwise not be encountered in typical powder systems. Nevertheless, the experiments illustrate

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TABLE III Comparison of surface roughness of spray-deposited coatings obtained after different heat treatments (20 mm × 20 mm area)

Heat treatment	Dominant wavelength(s) of surface roughness (mm)	Mean height of surface roughness (μm)	Standard deviation on height of roughness (μm)
85°C for 15 min.	0.38 and 0.52	35.0	4.6
190°C for 10 min.	3.62	3.57	0.59
85°C for 15 min. + 190°C for 10 min.	2.35	2.70	0.46

that rougher surfaces result from surfaces with steeper STGs.

Additional experiments determined the relationship between the stoving conditions and the resulting coating morphology. The stoving conditions could influence the distribution of flow agents, which, in turn, should have an impact on levelling. The standard formulation (with 1% w/w flow agent) was spray-deposited onto steel plates. Coatings were prepared using standard stoving conditions (190°C for 10 min.) in an oven and compared to coatings prepared via a two-stage process. This latter process consisted of a 15-min. hold at 85°C (during which negligible crosslinking is known to occur according to rheology studies) followed by the standard stoving at 190°C. The mean peak-to-valley distance and the mean wavelength of roughness in the lateral direction were determined using a three-dimensional surface profiler. Table III lists the results.

After heating for 15 min at 85°C, the coating surfaces have a high peak-to-valley roughness (4.6 μm) over lateral length scales corresponding to about ten particle diameters. This level of roughness is attributed to non-uniformities in the initial packing of particles. If this same heat treatment is followed by the standard stoving conditions in a two-stage process, the surface roughness decreases to a mean value of only 2.70 μm. By comparison, the standard conditions on their own result in a rougher coating (3.57 μm). Although the low-temperature treatment does not produce a smooth coating on its own, when it precedes the standard stoving, it creates a smoother film. One explanation is that the low-temperature treatment eliminates any extremes in surface roughness and provides a "head start" to the levelling process at higher temperatures. Another possibility, however, is that the low-temperature treatment enables the flow agent to distribute itself more uniformly at the coating surface. STGs are thereby minimised, so that levelling during the standard stoving is not impeded. In any case, these experiments point to a means of achieving a smoother finish through the adjustment of the heat treatment.

4. Conclusions

The flow agent studied here, a low molecular-weight acrylate, is essential to eliminating defects, such as dimples and craters, on longer length scales in a polyester powder coating. The inclusion of small amounts of flow agent in the powder formulation decreases the rate at which viscosity increases during stoving. More importantly, the flow agent lowers the surface tension of the polymer melt. One effect of the lower surface tension is that levelling is slower in coatings that contain flow agent, according to confocal microscopy analysis. Another effect is that melts of powders containing flow

agent spread out completely on a melt without flow agent, whereas the reverse is not true, because it is not thermodynamically favourable.

A non-uniform distribution of the flow agent is expected to create lateral gradients in the surface tension. In turn, these gradients will inhibit the levelling of the coating surface. Experiments support these ideas. In a coating in which surface tension gradients are purposely created by the blending of powders with and without flow agent, the surface roughness is higher than in coatings formed from either of the component powders. To achieve a level coating surface, it is essential that the flow agent is uniformly distributed in the powder formulation.

Even with an *initial* uniform distribution of flow agent, however, levelling might be inhibited since the flow agent acts as a weak surfactant. We suggest that when a curved surface of a coating undergoes shear stress resulting from capillarity, the flow agent might be displaced from its equilibrium uniform distribution. A counterflow is created to suppress the surface tension gradient, and levelling is therefore inhibited. This phenomenon might explain why a coating without flow agent is smoother (on short lateral length scales) in comparison to a coating containing flow agent.

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