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The impact of raw material properties and process conditions on the color of a powdered formulated detergent product

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Abstract

The appearance of detergent powder plays an important role in how the consumer perceives the powder will work for cleaning applications. In this study the influence of numerous formulation and processing conditions are investigated to understand causes for product discoloration. Product color was scrutinized using the L a*b* color space in particular L and b*. Particle size, method of introducing the optical brightener and the raw material grade in particular were found to have great importance in controlling the optical properties of the granules. As the particle size decreases the

light reflected to the observer appears brighter and bluer corresponding to increased whiteness (from L = 84.03 when particle size is above 841 μm to L = 90.59 when particle size is smaller than 250 μm), whilst reducing impurities will result in improved color definition. A key finding when investigating the optical brightener is that it should be dispersed rather than molecularly dissolved and sprayed on. This leads to improved distribution within the granules and this increased 'whiteness' when compared to pouring it in the agglomeration vessel. Subsequent spray on steps highlight that the surface contributions of the brightener on the granule surface influence whiteness more than those embedded inside. In addition the effect of drying temperature, mixer impeller speed on powder color are reported herein.

Keywords: Detergent powders; powder processing; powder discoloration, optical brighteners; characterization;

1. Introduction

Modern granular detergent products contain more than 20 - 25 different components each having a specific function. These different components can be widely classed into four categories; 1) surfactants, 2) builders, 3) bleaching agents and 4) auxiliary additives (Bajpai & Tyagi, 2007; Saouter & Hoof, 2002; Smulders & Sung, 2000). Detergent powder manufacturing processes in industry are conventionally produced by spray drying the majority of the ingredients with the addition of sensitive components at a later stage. For production of smaller volumes there has been a move away from spray-drying routes with focus towards so called 'non-tower' routes (Appel, 2000; Smulders & Rähse, 2000). This typically involves agglomeration by fluidization using either a) mechanical agitation for high density granules and b) air-fluidization to achieve a low density

product (Flore, Schoenherr, & Feise, 2009; Gao, Jain, Motheram, Gray, & Hussain, 2002; Hausman, 2004).

Wet granulation is a size enlargement process whereby a group of powder particles are bound together by a liquid binder (Ennis, 1997; Iveson, Litster, Hapgood, & Ennis, 2001). The objective of the granulation process is to improve the properties and characteristics of the powder such as flow, dustiness, composition and structure, strength, handling, and appearance (Capes & Darcovich; J. Litster & Ennis, 2013; Pietsch, 1997). In industry, granulators are typically classified into two categories: i) low-shear mixer granulators (e.g. tumbling drum and pans) and ii) high-shear mixer granulators (e.g. Lödige, Fukae, Drais, Ballestra, Eirich, Schugi) (Smulders & Rähse, 2000).

High-shear mixer granulators are typically employed in the pharmaceutical, agrochemical and detergent industries as they offer numerous advantages over other granulators such as; i) ability to process wet and sticky materials, ii) robust to processing conditions, iii) handling of viscous binders and iv) production of small (< 2 mm) high density granules (Reynolds, Le, & Nilpawar, 2007). The characteristics of the final granular product are determined by both the process operating conditions as well as the feed formulation characteristics as alluded to by Lister (J. D. Litster, 2003).

The color of the end product is a key criterion that dictates consumer perception with regards to how the product will perform during application (i.e. the whiter the appearance the better the product). The agglomeration process in detergents occurs through the mixing and neutralization of sulfonic acid (e.g. linear alkylbenzene sulfonic acid, HLAS) with sodium carbonate to produce granular anionic surfactants (e.g. linear sodium alkylbenzenesulfonate, NaLAS) (Dang, Lang, Li,

& Wei, 2018; Dang, Li, Fu, Lang, & Wei, 2018). The stoichiometric equation describing the reaction can be written as:

$$2HLAS + Na_2CO_3 \longrightarrow 2NaLAS + H_2O + CO_2 \quad (1)$$

This neutralization reaction is highly exothermic and although the heat of neutralization using sodium carbonate is not known, using sodium hydroxide as the base in liquid form results in an enthalpy change of 502 kJ/kg (De Groot, 1991). In dry neutralization with sodium carbonate the enthalpy change is expected to be lower as the reaction is dependent on particle surface area.

Sulfonic acid is a reactive binder, therefore it is important to maximize the neutralization reaction as residual sulfonic acid can lead to powder discoloration (as the acid is brown in color), degradation of any perfumes added, and weaker powder granules (Boerefijn, Dontula, & Kohlus). The process of neutralization is time dependent and is dictated by the binder distribution and the wetting of the acid with the carbonate particles (Chaudhury, et al., 2015; Germanà, Simons, & Bonsall, 2008), thus influencing granule color.

The neutralization reaction is also influenced by the reaction temperature and impeller tip speed as illustrated by Schöngut et al. (Schöngut, Smrčka, & Štěpánek, 2013). They investigated the amount of CO_2 gas produced during the reaction to determine the conversion rate. It was found that the overall conversion increased with temperature (due to the high exothermic reaction) causing a reduction in the binder viscosity. This led to improved binder distribution and thus an increase in product yield which was also observed by Salman *et al*. (Balashanmugam, Cheong, Alam, Hounslow, & Salman, 2016). Increasing the impeller tip speed five-fold had little influence on the conversion rate. Shear stresses can also lead to structural changes in the neutralized

surfactant product. McKeown et al. highlighted that NaLAS can form multi-lamellar vesicles (MLVs) (multi-layered 3-dimensional geometry) when moderate shear values are used and do not form at high shear levels (McKeown, Mackley, & Moggridge, 2003). It was also noted that when MLV's were formed, NaLAS would become more elastic and the color was noted to become whiter in appearance. However this color change was simply observed and no follow up testing was done to ascertain the cause. This was later expanded to investigate the influence of temperature and shear on the formation of MLV's as a function of added electrolyte (Brooks & Moggridge, 2006).

Particle size also plays an important role in the product color perceived by the human eye. This is referred to as Rayleigh scattering which states that scattering of light is proportional to $1/\lambda$ and that the relative scattering of light decreases with increasing wavelength (Tilley, 2010). In essence, smaller particles will preferentially scatter light of shorter wavelengths (i.e. blue and violet). This phenomenon can be seen experimentally in a number of different research areas. One of the most common fields where particle size is a concern is pigment design and production. In a study conducted by Li et al. (Li, Zhang, Yang, & Guo, 2011). It was found that porcelain powder with and without yellow pigment appeared bluer as the pigment size decreased. This observation was matched by Symons and Dexter (Symons & Dexter, 1991), who investigated the influence of particle size on the color of flour.

Modern detergent formulations include fluorescent whitening agents (FWA's also known as optical brighteners), as they reduce the yellow hue appearance of properly washed and bleached white laundry. These organic compounds convert a portion of the invisible ultraviolet light into longer wavelength visible blue light making the overall appearance look whiter in color (Smulders & Sung, 2000). They can undergo structural transition and degradation during improper storage

and reacting with other compounds during product processing. This reduces their functionality within the product and can lead to product discoloration (Blanco, Jiménez, & Valverde, 2001; Dyer, Cornellison, Bringans, Maurdev, & Millington, 2008; Gold; Wong-Wah-Chung, Mailhot, & Bolte, 2001). Furthermore, another factor to consider during processing is that many of the commercially available optical brighteners are a deep yellow in color.

As highlighted above, investigations into detergent powder discoloration is limited and not well documented in the public domain. Therefore there is a need to understand the influence of process parameters and raw material properties on the final product color, to ensure that the product matches the aesthetics demanded by the consumers. In this study, the potential sources that influence product discoloration during the neutralization reaction within a lab-scale granulator will be investigated using a spectrophotometer. The influence of drying temperature, particle size, raw material quality, and raw material processing are reported therein.

2. Materials and Methodology

2.1. Materials: The powders and liquids (binders) used for the production of the granular detergent product were used as received without further purification. The bulk of the raw materials used in this were of commercial grade and were kindly supplied by Hosokawa Micron BV, The Netherlands. To investigate the influence of material purity, research grade alternatives for some of the raw materials (Linear Alkylbenzene Benzene Sulfonic Acid (HLAS), sodium silicate, sodium carbonate and sodium sulfate) were additionally purchased from Sigma Aldrich. The full list of raw materials and the typical formulations used in this study are outlined in section S1 within the supplementary information with a sample breakdown in section S2.

2.2. Methodology

2.2.1. Granulation: A food processor (Cookworks SG-500) was used as a laboratory-scale high shear mixer in these studies. The vertical axis granulator consisted of two vertically offset, curved steel blades connected to a plastic shaft. Later, a food blender (Tefal Blendiforce with Tripl'Ax impellers food blender) and a coffee grinder (Wahl) were used to compare color performance of prepared granules. To granulate, the powder components were pre-blended together in the mixer for 1 minute. In this pre-blend, sodium carbonate and sodium sulfate were the major components (60-85%) whilst Tinopal and CMC accounted for 1% of the total mass. To this pre-blend, the liquid components were added one at a time using a pipette and once all the liquids were added the mixer was turned on maximum speed for 10 s. HLAS and Sodium silicate accounted for 70 - 80% of the total liquid added. The resulting product (~120 g) was then dried using two methods a) overnight drying (16 – 18 hours) using a convection oven (Memmert) at different temperatures and at ambient humidity and b) using a lab scale fluidized bed dryer (Retsch TG100, temp range 40 - 120°C) for a drying time of 10 mins. In order to standardize the samples for color measurements, the moisture content of the powders was measured using a Rotronic water activity measurement probe. Once the powder samples had an equilibrium moisture content of less than 5% they were subjected to color analysis as wetted samples can manipulate the color values obtained using a color spectrophotometer.

2.2.2. Granule color characterization: A Macbeth Color-Eye 7000A spectrophotometer was used to quantify the color properties of granules produced that had an equilibrium moisture content of less than 5%. Approximately 10 g of detergent powder was placed into a clean, oil and fingerprint free petri-dish. It was important to ensure that the powder level completely covered the aperture of the device when placed vertically against the spectrophotometer. The sample was

against the aperture by means of a spring and a cantilevered arm. The samples were illuminated using a pulsed Xenon source and included the influence of UV light since optical brighteners were used in the formulation. The samples were measured six times and averaged to obtain color properties in the L and b* axis within the L a*b* color space. Where L is lightness index, a axis extends from green (-a) to red (+a) and the b axis from blue (-b) to yellow (+b) (HunterLab, 2015).

3. Results and Discussion

3.1. Drying temperature

Drying of the granular product is an important unit operation in the agglomeration process as it conditions the product by controlling the moisture content and thus stabilization of the granule structure (Boerefijn, et al.; Räsänen, Rantanen, Mannermaa, Yliruusi, & Vuorela, 2003). The influence of drying temperature on the 'whiteness' of the product was firstly investigated using a convection oven. Images of the dried samples and their optical micrographs of granules dried at different temperatures between 70°C and 170°C are presented in Figure 1.

From the images presented in Figure 1a it can be seen that at the elevated temperature of 170 °C the powder has a significantly 'yellower' appearance. The sample also contains distinct smaller particles which are dark brown in appearance. This is confirmed upon closer examination of the granules as presented in Figure 1b. The first type of discoloration observed were irregular brown particles similar in size to the 'whiter' detergent powder (Figure 1bi and zoom of region of interest Figure 1bii). These particles do not seem to be comprised of smaller particles forming a large agglomerate, instead they appear as a continuous solid brown particles with the presence of white speckles on the surface indicating partial neutralization. The solid nature of the particle indicates that the partially unreacted HLAS transformed from a liquid into a solid due to the following mechanisms; a) drying due to the exothermic nature of the reaction, b) drying occurring within the

convection oven, and/or c) change in material composition (e.g. increased viscosity) due to binder-solid interactions (Germanà, et al., 2008).

The second type of discoloration is due to the presence of small brown spots on the white granules (Figure 1biii) indicating highly localized regions of unreacted solid HLAS binder. The high temperature will also serve to 'bake' any organic matter present (as the sample remains static during the drying process) and thus leads to a product with undesired color properties. These visual observations are matched by the spectrophotometer as shown in Figure 2, which report a low L value i.e. 'darker' and a large b* value indicating a yellowish color. As the drying temperature decreases the detergent powder became noticeably 'whiter' in appearance corresponding to a shift towards negative b* (bluer) and an increase in the L values (brighter). In addition, the standard deviation associated with the data show that the trend observed is of significance.

The samples in a convection oven remain static over the course of the drying period and this has a profound effect on the heat transfer rate. Fluidized bed dryers have a high heat transfer rate as the material is continually fluidized, therefore a lab-scale version was used to determine if this drying method led to an increased color performance. The b* and L values for samples dried with the fluidized bed dryer between 60 and 120 °C are presented in Figure 3.

The samples produced after drying with a fluidized bed dryer for 10 mins follow a similar trend observed with a convection oven between the temperature ranges of 120 to 80°C. As the temperature decreases, the b* value also decreases with an increase in the L values observed. This potentially suggests that at 120°C, the temperature causes a 'scorching or a self-heating' effect of HLAS and any organic matter present within the granules leading to product discoloration. This matches observations reported elsewhere (Beever & Crowhurst, 1989; X. Chen, Lake, & Jebson, 1993; X. D. Chen, 2001; Chong, Shaw, & Chen, 1996). As the temperature decreases to 80°C, the

magnitude of this effect reduces and thus a whiter product is observed. It is interesting to note that below 80°C, the product again undergoes some discoloration highlighted by an increase in b* and a reduction in L. This could be likely due to an increase in the moisture content as the drying temperature decreases, which can alter the reflectance wavelengths observed by the spectrophotometer leading to a change in the measured b* (Blum, 1997). Furthermore as the moisture levels increase, less light is reflected off the sample and thus the sample will appear darker (Lekner & Dorf, 1988). Since the b* and L values between the two drying techniques were within a similar range, the influence of other parameters were based on samples dried with a convection oven at 70°C.

3.2. Particle size

Initial color measurements with the convection oven dried samples presented significant variation in the measured L and b* values. Upon investigation it was found that particle segregation (i.e. smaller particles settling to the bottom of the petri-dish as the sample was presented vertically to the spectrophotometer) was causing this variation. This led to color measurements that showed that the sample (larger granules) were apparently darker and yellower in color. To negate this segregation effect all subsequent samples were shaken to ensure homogeneous distribution of the different sizes to allow repeatability in the color measurements. This interesting observation was further investigated to fully explore the impact of particle size on color.

Samples were sieved into four different size classes (above 841 μm, 500 – 841 μm, 250 – 500 μm and below 250 μm) and the resulting color measurements were compares with the unsieved sample. The resulting L and b* values obtained are presented in Table 1 with digital photographs of the samples presented in Figure 4.

As the particle size decreases the measured b* value increases in negative magnitude indicating a whiter sample, whilst the L value increases by almost 6 units indicating an increase in sample lightness. As the particles become smaller in size they scatter the light at shorter wavelengths i.e. blue and violet and as such the -b* value will increase in magnitude (Li, et al., 2011; Symons & Dexter, 1991). In addition more of the incident light is reflected back to the detector resulting in the sample appearing lighter (MacDougall, 2002). The standard deviation for L and b* shows that the trend observed is of significance. The bulk of the sample has a particle size of between 250 – 841 microns and therefore the L and b* values of the unsieved sample lies in-between the values measured at these size fractions.

3.3. Agglomeration vessel

McKeown and Hibare reported that changes in the shear profiles within the mixer can greatly influence the final properties of the formed detergent granule (Hibare & Acharya, 2014; McKeown, et al., 2003). To determine the effect of shear on granule color, studies were performed comparing the performance of the food processor (10000 rpm) with a blender (20000 rpm) and a coffee grinder (in excess of 20000 rpm). The hypothesis was that the increased impeller speed would aid in improving the binder distribution within the granules.

Food processor was used as the predominant method for laboratory scale detergent agglomeration in this study. Using a blender offered a different geometry as well as aggressive agitation with the main deterrent being that the blender was unable to remove powder that would build up on the sides of the vessel. Furthermore the increased shear could potentially lead to better binder and raw material distribution during the granule formation. The coffee grinder offered even more shear but had the drawback of significantly reduced particle sizes and the potential for poor

wetting with the binder due the blade design. Color data and associated statistical error from the resultant samples are presented in Figure 5.

The trend from the color measurements indicate that by increasing the shear rate results in increasing negative b* whilst the L value also increases leading to apparent increase in product whiteness. To determine whether this was an artefact of particle size as a result of increasing shear, the samples were sieved into different size fractions for further analysis. The data presented in Table 2 shows that by changing the agglomerator vessel and hence the shear profile the particle size distribution also changes and becomes smaller. As discussed previously the reduction in particle size will lead to a white appearance of the product. Although the powder appears whiter, further exploration is needed in future studies to investigate if the distribution and composition of the granules also improve.

3.4. Optical brightener dispersion method

Optical brighteners are colorless to weakly colored organic compounds (Grčić, Papić, Mesec, Koprivanac, & Vujević, 2014; Siegrist, Eckhardt, Kaschig, & Schmidt, 2000). They form a key component in many household detergent products to whiten laundry by re-emitting invisible ultraviolet into visible blue wavelengths. Their presence within the granule can also affect the granule color especially if they are not homogeneously distributed. In this study 80 ±40 μm powdered Tinopal® CBS-X was used as an optical brightener that is a based on a styryl derivative and yellow-green in color. The method of dispersing Tinopal within the granulation process was investigated to understand the effect on the granule color.

In Figure 4a the granular product shows visible signs of green speckling on the surface of the granules and is most likely due to poor distribution of the Tinopal optical brightener within the

granules. In this case Tinopal was introduced into the granulation process as a powder component as part of the pre-powder blend. Therefore the ability to disperse Tinopal within a liquid component for improved homogeneity within the granules was investigated.

Preliminary studies (S4 in supplementary information) were carried out to determine which of the liquid components could be used as candidate systems to disperse the Tinopal powder. From this study it was found that water, Neonol and the Sokolan polymer solution could be used as a potential delivery vehicle. The subsequent results of product color measurements using these liquids as delivery vehicles is presented in Table 3 along with the statistical error.

It can be seen from Table 3 that irrespective of the method used to disperse the Tinopal the L value remains consistent. This is because the purpose of the optical brightener is to increase the amount of light scattered at the shorter (blue) wavelength and hence has little influence on lightness. Examining b* it can be seen that the greatest discoloration in the granular product occurs when Tinopal is dispersed in water. Tinopal is highly soluble in water (25 g/L) and forms a yellow-green solution, which is then imparted onto the resultant granules (S4 in supplementary information).

When Tinopal is dispersed in the Sokolan solution the b* value decreases to ~1.5 indicating improvements into the product color. The Sokolan solution also contains water (~60 – 80%) and this lower water content may aid in lowering the solubility of Tinopal to yield a low b* value. Dispersing Tinopal in Neonol produces granules with the largest negative b* value (-4.75) indicating the whitest sample. This is due Neonol containing trace amounts of water (0.5 wt%) and therefore Tinopal forms a pseudo-stable dispersion in for a short period of time before settling out due to the density difference (Neonol density = 953 kg/m^3 whereas Tinopal has a density = 1400 kg/m^3). The importance of Tinopal in the formulation was highlighted as granules formed in the

absence of it were off-spec and yellow in color. These results indicate that Tinopal is an integral ingredient in forming a white looking granule and it is critical that Tinopal forms a dispersion rather than dissolving in the liquid of choice to ensure good distribution and improved granule color properties.

To determine if Tinopal distribution within the granules could be further improved, subsequent experiments investigated whether spraying the Tinopal-Neonol dispersions offered better performance compared to pouring it in within the agglomerator. The Tinopal-Neonol dispersions were sprayed using a household flower spray bottle. The resultant color data for the granules produced are presented in Table 4.

When the Tinopal dispersion is sprayed on during the agglomeration process the granules show an improvement in both the lightness and b* values compared to just pouring on. This is because when the dispersion is simply poured in, it creates a localized concentration of the brightener (at the point where it is added) that needs to distribute throughout the vessel within through shear. In comparison when using a spray nozzle, the dispersion is in an atomized state that allows for an even distribution onto the granule surface as the agglomeration process occurs. Furthermore, when the sample is dried and subsequently sprayed on again with the Tinopal dispersion, there are further improvements in the color performance. This highlights that it is key to have a good distribution of the Tinopal on the surface of the granules. The spectrophotometer measures the surface reflectance, therefore having the optical brightener embedded deep within the granule will have little or no influence on the color properties. It should also be noted that care should be taken that the brightener is evenly distributed on the surface as agglomerates will cause green speckling as shown earlier in Figure 4a.

3.5. Raw material purity

The presence of impurities within the starting raw materials can potentially lead to great variations in the optical properties of detergent product color (Johansson & Somasundaran, 2007). As highlighted earlier HLAS, sodium carbonate, sodium sulfate and sodium silicate contribute the highest percentage by mass in the manufacture of detergent powder. Therefore these raw materials were investigated further to determine if their purity (commercial vs. research grade) had any measured effect on granule color. The measured L and b* values of detergent powder as a function of purity grade along with statistical error are outlined in Table 5.

It is evident from Table 5 that changing the raw material source from commercial grade to research reagent grades leads to significant changes in the measured b* value, whilst there is little/no change measured in L. The greatest changes in b* occurs with HLAS and Sodium silicate and it is likely that these two components also have the greatest influence in improving b* when all four raw materials are substituted for research grades. Since the purity of the commercial grade materials are not known, it is difficult to deduct which impurities in HLAS and sodium silicate contribute to the granule discoloration without further chemical analysis.

It should be noted that there were visual differences when comparing the two grades for both materials as shown in Figure 6. For HLAS, the research grade was lighter in color and more transparent than its commercial grade equivalent, indicating different feedstock purities and/or different manufacturing routes resulting in difference in color (Foster, 1996). The difference in turbidity in the sodium silicate grades is also very stark. The research grade appears clear whilst the commercial grade appears cloudy and heterogeneous in color. The turbid appearance is due to the precipitation of silicates and presence of impurities. Furthermore the density of these precipitates will affect the color distribution within the liquid thus appearing heterogeneous.

Although the improvements with research grade sodium carbonate and sodium sulfate are much smaller, it does highlight that raw material quality dictate the color quality of the resulting product influencing consumer perception of product quality.

Conclusions

In this article we report the influence of formulation and processing conditions on the optical properties of detergent powder formed within a high-shear mixer. It is evident that particle size plays an important role on how white the sample appears especially if a sample undergoes segregation or if high shear rates are used during the granulation process. Another key finding is the importance of drying temperature as this can lead to discoloration of any unreacted organic compounds still present within the granule.

Additionally we show that the formulation itself also has a bearing on the perceived color of the final product. Improving the purity grade of raw materials as well as selecting the optimal vehicle to deliver key components such as optical brighteners, can lead to powders that appear both light and blue i.e. increasing whiteness, which is important for consumer acceptance of the final product. The biggest impact on improved color performance occurs when the optical brightener is in a dispersed state and sprayed on as opposed to being poured in. Further spray-on steps (after the granules have dried) illustrate that increasing the concentration at the granule surface vs. being embedded within the granule leads to large improvements in product 'whiteness'.

Supporting Information. S1. Materials and associated properties including typical formulation, S2. Breakdown of the samples used for color analysis S3. Dispersion of Tinopal study, S4. Impact of Tinopal dispersed in water on powder color.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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