ORIGINAL ARTICLE

Efficiency of Laundry Polymers Containing Liquid Detergents for Hard Surface Cleaning

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Received: 29 March 2014 / Accepted: 8 August 2014 / Published online: 20 August 2014 © AOCS 2014

Abstract Three water soluble laundry polymers were employed for the first time in liquid detergent formulations for hard surface cleaning. The polymers included in the formulations were the sodium salt of maleic acid/olefin copolymer (P1, anionic), polyethyleneimine (P2, cationic), and polyethylene glycol-g-vinyl acetate (P3, nonionic). Commercially available surfactants $(C_{10}$ Guerbet alcohol alkoxylate (FAEO), caprylyl/decyl glucoside (APG), and the sodium salt of ethoxylated alkyl ether sulfate (SLES) were chosen to formulate bathroom, kitchen, and all-purpose cleaners, which provide the desired broad of pH range for hard surface applications. Their hard surface cleaning efficiencies were also compared with an amphoteric polymer (amino modified polycarboxylate, P4) as amphoteric polymers are the most suitable structures for hard surface cleaning. The standard test method and the cleaning device, the so-called cleaning robot, were used to investigate the primary cleaning performances and synergies of the chosen polymers in a hard surface cleaners system. Secondary cleaning performance tests, which indicate the effects of the hard surface cleaners on surface modification, were also performed. The results revealed that the formulations containing P3 and P4 gave the better cleaning performance for primary cleaning tests whereas only P4-containing formulations showed the significant results for secondary cleaning tests.

Keywords Hard surface cleaning · Polymer efficiency · Cleaning performance - Surfactant - Detergency

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Introduction

Detergents have the composition of chemical agents, which provide cleaning properties on defined surfaces. Cleaning action is progressed basically by two steps. The first step is the removal of soil from a certain surface such as with textiles or ceramics, and the second step is the dispersion of the soil in washing liquid.

Detergents are generally used for industrial/institutional and household cleaning. The formulations contain surfactants as a main ingredient, with hydrotropes, chelating agents, and acids or bases to adjust the pH as minor ingredients. Compositions are formulated by selecting specific ingredients to perform the desired benefits on a certain surface [[1\]](#page-10-0). Detergents can be classified into three subsegments according to their application fields. These are called hard surface cleaners, laundry detergents, and dishwashing detergents.

The term ''hard surface'' means that surfaces are mostly resistant to penetration by solid materials and generally do not absorb liquids that are found in both households and industrial places. Examples are tiles, stone, marbles, pots, ceramic and metallic sinks, and porcelain and metal kitchen appliances [\[2](#page-10-0)]. Hard surface cleaners are a class of cleaners formulated for application to soiled surfaces and removal therefrom without a rinsing process. The expected benefits for hard surface cleaners are the fast wetting, effective emulsification of the soil, and the prevention of significant streaks or fatty films on the surface after cleaning. Hard surface cleaners are mainly produced for household and industrial or institutional applications. Household hard surface cleaner formulations are generally mild on skin with pleasant odors and without or very less toxicity. On the other hand, institutional or industrial based ones tend to be more powerful and fast effective. Skin

mildness or irritation sensitivity generally is not required for these applications [[3](#page-10-0)].

Surfactants are the main component of the detergent formulations. They are used to wet the surface and decrease the surface tension to remove soil from the surface easily. The surfactants are classified according to their hydrophilic parts as anionic, nonionic, amphoteric, and cationic. Typical nonionic surfactants are based on polyoxyethylene, polyoxypropylene, or alkypolyglucosides. Alkylpolyglucosides are the surfactants with excellent cleaning properties and ecological properties [\[4](#page-10-0)]. They are produced from natural sources as glucose and fatty alco-hols [\[5](#page-11-0)], and they have been known for a long time [[6,](#page-11-0) [7](#page-11-0)]. They are commonly used in detergents [[8\]](#page-11-0) due to their compatibility with other types of surfactants, cleaning performances, foaming properties, and also skin compatibility. They are not only a major ingredient in the cleaners, but they are also used as a cosurfactant to enhance the efficiency [[9\]](#page-11-0). On the other hand, ethoxylated and alkoxylated alcohols are the most commonly used surfactants of the nonionic surfactant family [[10\]](#page-11-0). While the nonionic surfactants with long carbon (12–18) chains are mainly used for powder and liquid laundry detergents, short carbon (8–10) chains have types that are for hard surface cleaning. They are also classified as readily biodegradable surfactants [[11\]](#page-11-0). As for ionic surfactants, the cationic contain positively charged hydrophilic groups whereas anionic surfactants contain negatively charged hydrophilic groups such as sulfate, sulfonate or phosphates. Regarding amphoteric surfactants, they contain both an anionic and a cationic character such as the betaines. Surfactants and polymers are combined to give different effects such as, rheology modification, emulsification, structuring, and suspension properties. Even so, in many cases synergistic effects are achieved [[12\]](#page-11-0).

Organic polymers are the performance ingredients of the detergent formulations to give some benefits for specific claims. They have been an important part of the detergents for many years [[13\]](#page-11-0). They provide different properties depending on the application field of the detergents. These tailor-made designed polymers can be synthesized by considering the parameters, such as polymer composition, different charged-monomers, side chain character and functionality, molecular weight, which can affect the formulation stability and solubility, and also polymer structure as being linear, branched or grafted. While they are used as incrustation inhibitors, soil dispersants, soil release or dye transfer inhibitors for laundry detergents, they are also employed as rheology modifiers or surface modifiers for hard surface cleaners. For example, polycarboxylates are generally used for incrustation inhibition combined with zeolite in laundry formulations. They are mainly composed of acrylic acid

homopolymers or acrylic acid/maleic acid copolymers [\[14](#page-11-0)]. They reduce incrustation by being dependent on two primary actions [[14,](#page-11-0) [15\]](#page-11-0). The first one is dispersing the big and growing crystals, and the second one is particle dispersion, which prevents deposition. On the other hand, polyvinylpyrrolidone (PVP) is commonly employed to provide dye transfer inhibition for laundry detergents. Copolymers of vinylpyrrolidone/vinylimidazole monomers also show effective dye transfer inhibition [\[16](#page-11-0)]. Regarding the soil release effect, the most widely used polymers are based on polyethylene terephthalate, which can be modified with polyethylene glycol or sulfonates. Alkylene oxide/vinyl ester copolymers have also a soil release effect [\[17](#page-11-0), [18\]](#page-11-0).

Polymers for hard surface cleaners are generally used to adjust the rheology and viscosity as well as disperse soil or modify surface like improving gloss retention. Polymer structure plays a very important role to modify the surface. As hard surfaces are generally negatively charged, polymers with amphoteric structure show effective results. While the cationic part adheres on the surface to stay longer, anionic and nonionic parts affect the hydrophilicity of the hard surface $[19]$ $[19]$. Use of various amphoteric polymers in hard surface cleaning formulations is very well known. Amphoteric polymers, such as copolymers of diallydimethylammonium chloride (DADMAC) and acrylic acid are known to give benefits and claims to be hard surface cleaners. One of the benefits of amphoteric polymers is to impart hydrophilic properties to hard surfaces. This hydrophilicity of the surface reduces the occurrence of water spots and provide anti-streak properties. In addition, they are also known to provide additional benefits such as quick drying [\[20](#page-11-0)].

The motivation of the present study is to investigate the soil removal performances of the polymers, which are not employed for hard surface cleaners, but commercially available for laundry applications and also have a cost effective market price compared to amphoteric polymers like amino-modified polycarboxylates. Three different applications of the hard surfaces were employed to see the effect of different pH values since the market products include bathroom, all-purpose, and kitchen cleaners as a majority. Acidic (pH value: 2.0–3.0) for bathroom, neutral (pH value: 6.5–7) for all-purpose, and alkaline (pH value: 9.5–10) for the kitchen were used to formulate the cleaners. The formulations for primary cleaning performances were applied to an oily soil on PVC surfaces. Primary cleaning performances were investigated by using a cleaning device. Regarding the application of the secondary cleaning performance tests, quick drying and a streak-free effect (gloss retention) were examined for only all-purpose cleaners on black ceramic tiles.

Experimental

Materials

All materials used in this study were obtained from the stated suppliers and used without any further treatment. Alkoxylated guerbet alcohol (FAEO, nonionic), caprylyl/ decyl glucoside (APG, nonionic), and sodium salt of ethoxylated alkyl ether sulfate (SLES, anionic) were obtained from BASF and employed as the surfactants in the formulations. Their specifications are given in Table 1. Four commercially available polymers from BASF were used for hard surface cleaners. Among these polymers, only the amphoteric one is used for hard surface cleaners as commercially available. The application area of other polymers is not for hard surface cleaners. The chemical structures of the polymers are depicted in Scheme 1, whereas their technical specifications and charge densities at different pH values are presented in Tables [2](#page-4-0), and [3](#page-4-0), respectively. Isopropyl alcohol and propylene glycol n -butyl ether were the solvents in the formulations and obtained from Merck. Citric acid (Merck), methanesulfonic acid (BASF), and sodium hydroxide (Merck) were employed to adjust the pH of the liquid formulations. Methylglycinediacetic acid (BASF) was used as the complexing agent.

Soil Mixture Ingredients

Primary cleaning test soil Industrieverband Putz und Pflegemittel (IPP) 83/21 was a mixture of 40.0 % Nytex 801 (Nynas; processing oil, naphthenic), 36.0 % Benzine 80/110, 17.0 % Myritol 318 (Trigylcerid), and 7.0 % Carbon black (Degussa Cl 77266).

Cleaning Surfaces

(i) Primary cleaning performance test: White Polyvinylchloride (PVC) strips (35 \times 42 cm) were used as the hard surfaces in the robot cleaning system.

Scheme 1 Structures of the COOH polymers used in the formulations ċоон Polyethyleneimine **Maleic/Olefin Copolymer** $(P2)$ $(P1)$ \sqrt{m} ≈o C. нó Poly(ethylene glycol)-graft-poly(vinyl acetate) $(P3)$ CH₃CI

> Acrylic acid/Methacrylamido propyl trimethyl ammonium chloride/Ethyl acrylate Terpolymer $(P4)$

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(ii) Secondary cleaning performance test: Smooth and black ceramic tiles (15 \times 15 cm) were employed to investigate the surface modification properties.

Instruments

The pH values of the test formulations were measured both in preparing and during the stability steps.

- (i) For the primary cleaning performance test, a cleaning device was the cleaning system for primary cleaning tests.
- (ii) For the secondary cleaning performance test, a glossmeter was used to analyze the light reflection values for secondary cleaning performances on black smooth ceramic tiles.

Preparation of Liquid Detergent Formulations

Three different types of hard surface cleaners (all-purpose, bathroom, and kitchen) were studied. Each cleaning application was treated by twelve formulations. In total, thirty-six different cleaners were formulated. To prepare each cleaner formulation, we first introduced water and then added the remaining components in the given order at room temperature and with homogenization. Besides, the pH value, which is an important parameter, was adjusted by citric acid or sodium hydroxide to get a desired pH value for the application. Correspondingly, the physical stability of the formulations was tested (1) under room temperature, (2) at 40 °C in the incubator, and (3) in the fridge (4 °C). All the prepared formulations were stable under these conditions for three months. The test formulations for allpurpose, bathroom, and kitchen cleaners are given in Tables [4,](#page-5-0) [5](#page-6-0), [6](#page-7-0), respectively.

Test Method Description

(i) Primary cleaning performance test principle (soil preparation and application).

The primary cleaning system had two steps to perform the tests. They included (1) the soiling and (2) cleaning and rinsing the surfaces. The evaluation of the results was done by a camera system. The results were calculated as the differentiation of before and after cleaning surfaces and given as a percentage.

The soil mixture (IPP) was prepared as follows: mineral oil, benzin, Myritol 318, and Carbon black were slowly stirred until having a homogenous mixture. Then the final stirring was done 30 min with Ultra-Turrax $^{\circ}$ at level 4-5.

After this homogenization process, the soil mixture was stirred constantly to swell in an erlenmeyer on a magnetic stirrer plate for 21 days. Before using the soil, it was homogenized again for 30 min with Ultra-Turrax[©] at level 4–5. For the remaining mixture, the storage was kept under constant stirring on a magnetic stirrer in erlenmeyer at room temperature. After 14 days of storage, the viscosity was relatively stable again. The finished soil mixture was applied on to PVC strips. Application of the soil was done by brush. IPP soil was brushed, and the stain was dried on the white PVC strips. The soil was then treated with a sponge with a defined amount (20 ml) of detergent under a defined pressure (2 bars). The amount of soil was 0.16 ml per strip. Before the cleaning system was started, 60 min was required to dry the soil. In this study, the test solutions were dosed at 20 ml for each cleaning process. For each cleaner the evaluation was done five times. A standard test solution was used at the beginning, middle, and end during the cleaning process.

(ii) Secondary cleaning performance test (surface modification properties) principle and application.

Surface modification performances were studied forthe all-purpose cleaners by two experiments: quick drying (water repellency) and the gloss retention test.

Quick Drying Test Principle

The test formulation was applied by wiping with a tissue on a black ceramic tile. After drying of the test solution, the ceramic tile was rinsed with water and the drying time was measured with a stop watch. The ceramic tile was cleaned in a dishwashing machine with a 55 \degree C program with the standard cleaning powder. After drying, the surface was cleaned successively with isopropanol and ethyl acetate. Under warm running tap water, the surface of the tile was cleaned with a sponge, and the process was repeated under deionized water again. Finally, the draining of deionized water was visually checked to exclude any pretreatment before starting the test. Twenty-five drops of the test solution in five rows with five drops per row were applied via a 3 ml disposable pipette and distributed with a folded paper towel. The distribution was done by wiping without pressure in ten rows from top to bottom and then jumped back in ten rows from the bottom to top. After drying of the test solution, the surface was rinsed by a stream of tap water. The stream was moved twice at the top of the tile from the left to the right border and back in order to create a complete water film on the surface. After complete rinsing of the tile, a stop watch was used to measure drying speed of the water from the surface of the tile.

Table 3 Charge densities of the polymers at different pH values

Polymer	pH at 3 (meg/g)	pH at 7 (meg/g)	pH at 10
P1	-1.7	-3.1	-3.4 meq/g
P ₂	$+19.5$	$+2.2$	unstable
P ₃	$-\leq 0.5$	$+0.7$	0.0 meq/g
P4	$+1.9$	$+0.7$	$+\leq 0.5$ meg/g

Gloss Retention Test Principle

The application of the test formulation onto a black ceramic tile for the gloss retention performance test was the same as that of the quick drying performance test. The light reflection (20[°] light angle) was measured at five different points on a clean, dry, and untreated black ceramic tile by using a BYK Gardner glossmeter. After drying, the light reflection was measured once again, possibly at the same points as initial measurements on clean black tiles.

Results and Discussion

Primary Cleaning Performance Test

For the first time, cleaning efficiency of three commercially available polymers were investigated in three different applications of hard surface cleaners with two different test methods. Three different types of hard surface cleaners were studied. The formulation of the hard surface cleaners contained two nonionic (FAEO and APG) surfactants and one anionic surfactant (SLES). All employed surfactants were stable in each formulation for all applications. Water solubility of the surfactants was good enough to formulate stable formulations. But, APG was the most soluble surfactant in water. The polymers were also stable in all detergent formulations except the cationic one (P2). It was not stable in an alkaline media to formulate kitchen cleaner and also with SLES due to its cationic nature. Therefore, the cleaning efficiency of P2 was investigated for bathroom and all-purpose cleaners. Triple vertical bar graphs in Figs. [1](#page-8-0), [2](#page-8-0) and [3](#page-8-0) gives percentage soil removal results obtained from robot cleaning system for all formulations. Each bar in the triple vertical bar graph represents the type of the surfactants used in the formulations. The figures, for the first time, also include the results of the formulations possessing an amphoteric polymer (P4) and no polymer to compare their surface cleaning efficiencies with those of P1, P2, and P3 used in hard surface cleaners.

Figure [1](#page-8-0) presents the results of robot cleaning for allpurpose cleaner formulations. The results show that the formulations containing the surfactant, FAEO, gave a better cleaning performance. On the other hand, SLESbased formulations did not show any increase of performance in the presence of polymers. The formulation without polymer removed 61.73 % of the soil from the surface whereas a P4-based formulation increased the soil removal up to 76.35 %. In the presence of P3, a slightly better result (79.29 %) for the soil removal was obtained. Regarding P1 and P2, there was no cleaning improvement compared to the formulation without polymer. It can be said that both anionic and cationic polymers are not worth using in all-purpose cleaner formulations to increase cleaning performance. Consequently, P3 is comparable to the commonly used P4, when it is formulated with the surfactant FAEO.

The experiments for the kitchen cleaner formulations also showed comparable results as seen in Fig. [2](#page-8-0). As P2 was not stable for the alkaline formulation type, there is no cleaning performance result for the cationic polymer included formulations. The kitchen cleaner formulations containing FAEO gave better cleaning results as in the allpurpose cleaner formulations.

Among the laundry polymers, only P3 increased the cleaning performance when the formulations included APG. However, both P4 and P3 increased the cleaning performance in this liquid alkaline formulation type. The effect of P3 was again slightly better than the commonly used P4. Both of them increased the cleaning performance of kitchen cleaner formulations approximately 17 %. Although there was not much of a comparable cleaning

cleaner formulations Table 4 All-purpose cleaner formulations more

Table 6 Kitchen cleaner formulations 11 atio \tilde{f} ÷ V itoh $\ddot{}$

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Fig. 1 Cleaning performance for all-purpose cleaner

formulations

Fig. 3 Cleaning performance for bathroom cleaner formulations

property with SLES-based formulations, the cleaning performance increased in the presence of P3 and P4.

The cleaning performance of the bathroom cleaners, which have pH values between 2 and 3 are given in Fig. 3. Considering the results shown in Figs. 1 and 2, there was not significant improvement compared to the other two applications for the formulations containing FAEO. Furthermore, the slight increase in the cleaning performance was attributed to the presence of P3 and P4. While the cleaning performance of P3 was 68.84% , P4 showed 67.24% for the cleaning

Fig. 5 Gloss retention performance for APG-based allpurpose cleaner formulations

Fig. 6 Gloss retention performance for FAEO allpurpose cleaner formulations

performance. This is again a remarkable result in that P3 is doing slightly better cleaning than P4. The formulation based on SLES with P1 is decreasing the efficiency, indicating the negative effect of P1.

Secondary Cleaning Performance Test

Considering the surface modification tests, two experiments were conducted for all-purpose cleaner detergents.

Quick Drying Performances

The results obtained in this part indicate how polymers modify the ceramic surface to show water repellency. The triple horizontal bar graph in Fig. 4 represents the results of the quick drying performance of all-purpose cleaners with the different surfactants on vertical ceramic surfaces. The x-axis shows time in minutes for drying.

The results in Fig. [4](#page-9-0) clearly show that there is an extremely hydrophobic effect for P4, which resulted in very fast drying with each surfactant formulation. It took approximately only 9 s to repel water from the surface. Although P3 gave a similar primary cleaning performance compared to P4, it was not as effective as P4 for the quick drying performance. Among the laundry polymers, although P2 showed the best water repelling performance, it is definitely not comparable to P4. Within these three surfactant systems, all the polymers showed quicker drying performance with SLES. On the other hand, the type of the surfactants didn't make any difference on the performance for P4-included formulations.

Gloss Retention Performances

The gloss retention was also measured to study secondary cleaning performance of all-purpose cleaner formulations. The double vertical bar graphs in Figs. [5](#page-9-0), [6](#page-9-0) and 7 show the gloss values on black ceramic tiles before and after the treatment of the formulations. This can be explained as the better formulation has the similar or the same gloss value as before treatment. The results in Fig. [5](#page-9-0) demonstrate the clear effect of P4 with 82.6 gloss units and P3 with 81.3 gloss units. Although the presence of P2 and P1 decreased gloss units, not many streaks were observed by the naked eye.

As seen in Fig. [6,](#page-9-0) there is a dramatic decrease with the formulation which doesn't contain any polymer (46 %). With the addition of P4, the negative effect of FAEO was reduced. The P2-based formulation was the worst when compared to others. Although P1 and P3 didn't show the same decrease in gloss unit like P2, many streaks were also observed on the black ceramic tile with P1 and P3.

The gloss retention effects of the polymers with SLES are given in Fig. 7. These experiments showed that P4 increased the gloss efficiency of SLES-based formulation as in other surfactant-based ones. P1 and P3 didn't have any efficiency to improve gloss behaviour of the formulation. The streaks appeared on the surface of the black ceramic tile when treated with these formulations. The gloss retention performances of P2 with SLES couldn't be carried out since they have different electrical charges.

Conclusions

The present study evaluated the efficiency of three water soluble laundry polymers with different charges on hard surface cleaning performances. Standard test methods were applied both for primary and secondary cleaning properties. The experiments were done for different applications depending on different pH ranges. The results of a nonionic polymer (P3) for primary cleaning performance were comparable to the amphoteric polymer (P4), which is commercially used in hard surface cleaner compositions. Moreover, for secondary cleaning performances, P3 showed also good results compared to P4. By considering the market price of these polymers, P3 could be modified and used as a surface modifier polymer for hard surface cleaning agents.

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