Effect of sodium dodecylbenzene sulfonate on the dispersion stability of ceramic glaze suspension

Suphapan Satchawan*,*******, Wimol Naksata*,*******, Chairoj Rattanakawin***, Sakdiphon Thiansem**,*******,
Preecha Panya****, Ponlayuth Sooksamiti*****, Peter J. Scales******, and Orn-anong Arqueropanyo*,*******,†

*Department of Chemistry, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand
**Department of Industrial Chemistry, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand
***Department of Mining Engineering, Faculty of Engineering, Chiang Mai University, Chiang Mai 50200, Thailand
****Department of Chemistry, Faculty of Science and Technology, Kamphaengphet Rajabhat University,
Kamphaengphet 6200, Thailand
*****The Office of Primary Industries and Mines Region 3, Chiang Mai 50200, Thailand
******Department of Chemical and Biomolecular Engineering, The Melbourne School of Engineering,
The University of Melbourne, Parkville Victoria 3010, Australia
*******Materials Science Research Center, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand
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Abstract−Sodium dodecylbenzene sulfonate (SDBS) was used to render the stability of ceramic glaze dispersion which is composed of limestone, feldspar, quartz, kaolin and ferric oxide. The measured zeta potential showed negative values for the systems in deionized water and 0.001 M $MgCl_2$ media at pH above 2, but a positive value was observed in 0.1 M $MgCl_2$ at pH higher than 6.7. Adsorption of SDBS in aqueous suspensions of ceramic glaze in deionized water and in 0.001 M $MgCl_2$, within the concentration range studied, followed both the Langmuir and Freundlich isotherms, but the Freundlich isotherm was more favored. Adsorption of SDBS in 0.1 M $MgCl_2$ corresponded to the Freundlich isotherm. From dispersion stability investigation, SDBS could render the suspension in deionized water and in 0.001 mM $MgCl_2$ more than in 0.1 mM $MgCl_2$.

Keywords: Ceramic Glaze Suspension, Sodium Dodecylbenzene Sulfonate, Zeta Potential, Langmuir Isotherm, Freundlich Isotherm

INTRODUCTION

Glazing is an important step of ceramic production. It is a coating that has been matured to the glassy state on a formed ceramic article, and the material or mixture from which the coating is made. Five raw materials--quartz, limestone, feldspar, kaolin and ferric oxide-- were employed in this research. These raw materials were dispersed in water. The problem of this system is the particle's aggregate and the quick settlement under the influence of gravity. This results in aggregates of dense sediment at the bottom of a container being hard to redisperse. These aggregates then cause flaws in the final sintered product, reducing the overall strength of the object. By controlling the inter-particle forces, it is possible to eliminate these aggregates. There are two methods of imparting stability to the particles to prevent or slow down their aggregation. The first is the charged groups on the surface of the particles. The surface groups can be positively or negatively charged depending on the pH condition. The second method of imparting stability is to adsorb surfactant onto the particle surface [1]. Dispersion may produce some additives that called 'dispersants' such as surfactant or polymer. Surfactant or polymer adsorbs on particles surface and stabilizes dispersion by increasing interparticle repulsive forces through electrical charging and sterically hindering the close approach of neighboring particles. Optimum amount of the added polyelectrolyte resulted in good dispersion. For stabilized systems it is important to know the exact location of the isoelectric point (IEP), the pH value at which the particles have zero zeta potential. At this pH value there are no repulsive forces and the particles will be heavily flocculated. This is an unstable state and the systems are designed such that the suspension pH is well away from the IEP. The further away the suspension pH from the IEP, the greater the surface charge and hence the greater the zeta potential [2]. Surfactants are widespread in the application in many industrial processes. An important property of surfactant is its tendency to adsorb onto solid surfaces. The surfactant is used in many fields to regulate the stability and flocculation properties of disperse systems [3-5]. If too little surfactant is added then some flocculation will persist, and if too much surfactant is added then destabilization may occur [6]. Another factor that affects the adsorption of surfactant from solution onto ceramic particles is the ionic strength of the suspension [7]. The adsorption of single surfactants at solid/liquid interactions is fundamentally important for many technical applications. However, some studies have used a mixture of organic and inorganic surfactant systems [8].

In many previous studies, the effect of pH, ionic strength and surfactant concentration on the adsorption of surfactants has been studied on the single mineral ceramic glaze components, namely,

silica [9-16], quartz [17,18], feldspar [19], kaolinite [20-27], and limestone [28-30]. The most numerous and in-depth investigations have been carried out on silica and kaolinite. In our previous work [3], the dispersion stability of a ceramic glaze mixture composed of limestone, feldspar, quartz and kaolin was investigated in the presence of NaCl. It was found that, at low adsorbed amounts of SDBS, the dispersion stability was consistently poor. If the amount of adsorbed SDBS exceeded about 20 μmol/m^2 at pH 7 and about 2 μmol/m^2 at pH 9, the dispersion stability increased dramatically for all ionic strengths. The suspensions were completely stabilized and no flocculation occurred in the presence of the maximum adsorbed amount of cetylpyridinium chloride (CPC) in 0.001 and 0.01 M NaCl for both pH 7 and 9. But the dispersion in 0.1 M NaCl could not be stabilized by addition of CPC, although the level of adsorption was greater than at lower salt concentrations. Bremmell et al. [10] found that adsorption and mobility data indicated that SDBS adsorbed at the silica solution interface, though without improving the flotation efficiency. Cetyltrimethylammonium bromide (CTAB) was found to adsorb on the silica particles as a result of electrostatic interaction, initially to neutralize the surface charge and destabilize the suspension. At higher concentration, adsorption reversed the particle charge and re-stabilized the suspension. Taffarel and Rubio [31] reported that the adsorption of SDBS did not occur on natural zeolites. The adsorption of SDBS occurred on zeolites, if the surface of zeolite was modified with CTAB (ZMS). The medium pH influenced the SDBS ion adsorption rate onto ZMS, and SDBS adsorbed by ZMS increased with the increase of medium pH. This adsorption followed Langmuir isotherm model. Somasundaran and Krishnakumar [32] studied adsorption of sodium dodecylsulfate (SDS) on alumina from aqueous solutions. They found that the S-F isotherm could be observed in four regions. The first region showed the slope of unity under constant ionic strength condition and was characterized by electrostatic interaction between the ionic surfactant and oppositely charged solid surface. The second region showed the rapid increasing of adsorption, which was ascribed to surfactant aggregation at surface by interactions between hydrocarbon chains. The slope of adsorption isotherm in the third region decreased more than the second. It was due to increasing of electrostatic to surfactant adsorption following interfacial charge reversal caused by the adsorption of charged species. The fourth region showed the plateau of adsorption isotherm, which was the maximum surface coverage by micelle formation in the bulk or monolayer coverage. Jian-Xiao et al. [33] investigated interaction between SDS micellar solutions and several metal salt solutions. They found that the system of SDS-Al$_2$(SO$_4$)$_3$ occurred towards precipitation, which was the main reason due to the effect of adsorption charge neutralization between Al^{3+} ion and SDS micelles. The result of SDS-FeCl$_3$ system showed that the charge neutralization effect was not the main mechanism of precipitation. The main cause of precipitation of this system was the bridge connection effect of Fe species of Fe(OH)$^{2+}$ ions. However, in SDS-CaCl$_2$ system crystal of Ca(DS)$_2$ occurred, which caused precipitation of this system. Moreover, the system of SDS-MgCl$_2$ could be observed visually that there was no turbidity and precipitation under the experimental condition, which might be because of the low affinity between Mg^{2+} ion and the surfactant.

The purpose of this work is to study adsorption of anionic surfactant, SDBS, in ceramic glaze suspension in the presence of ferric oxide at various factors such as surfactant concentration and ionic strength in dispersion. It was reported that electrolyte such as calcium chloride has the ability to form a stable suspension [34]. The amount in the range 0.025-0.1% could increase the viscosity of glaze suspension. In the absence of NaCl, brighter glaze effects were observed during the firing [35]. MgCl$_2$ was used as one of the craze-inhibiting compound for treatment of fired glazed ceramic ware [36]. Therefore, in this work, 0.001% and 0.1% of MgCl$_2$ was used as electrolyte instead of NaCl in the previous studied. Ferric oxide (Fe$_2$O$_3$) is commonly used as a colorant in its red iron oxide form. Iron is also another tricky colorant because of its ability to yield different colors under different circumstances. At low percentages (0.5-1%) and in the presence of potassium, iron will become light blue or light blue-green in reduction (as is seen in traditional celadon). In the presence of barium, iron may become yellow green. When used in combination with calcium, red iron oxide can become pale yellow or amber in oxidation or green in reduction. Langmuir and Freundlich adsorption isotherms will be employed to investigate the adsorption behavior. Dispersion stability of ceramic glaze upon addition of SDBS was also investigated. Novel knowledge in application of SDBS in ceramic glaze in the presence of ferric oxide and MgCl$_2$ to improve the colloidal processing of ceramics in the ceramic industry will be obtained.

MATERIALS AND METHODS

1. Preparation of Ceramic Glaze

Feldspar and Quartz were from Tak Province supplied by Mineral Assay and Services Co., Ltd., Nakornpathom, Thailand. Limestone, supplied by Mineral Assay and Services Co. Ltd., was from Saraburi Province, while kaolin, supplied by Cernic International Co. Ltd., was from Ranong Province. Feldspar and Quartz were ground in the ball mill for about 23 h and 41 h, respectively. All materials were passed through a 300 μm sieve. Each component was dried at 110 $^{\circ}$C for 12 h and mixed at a ratio as shown in Table 1 [3]. After mixing, ferric oxide was added into the glaze and mixed well in the ball mill again. The content of ferric oxide in the glaze was 1% by weight. The particle size (μm) of the ceramic glaze mixture measured by a particle size analyzer laser (Malvern, Masterizer) was 17.5 μm and the surface area measured by a Quantachrome autosorb automated (version 2.19) was 4.89 m^2/g. The pH of ceramic glaze suspension was around 8.0-8.5.

2. Preparation of Solutions

All solutions were prepared with analytical grade chemicals and deionized water. Stock solutions of 1 M MgCl$_2$ (Fluka, molecular mass=203.30) and 25.00 mM SDBS (Fluka, molecular mass=348.48) were prepared in deionized water. The 0.1 M and 0.001 M MgCl$_2$ solutions were prepared by dilution. The stock solution of SDBS was used to prepare dilute solutions in three media: deionized water,

Table 1. Compositions of ceramic glaze

Material	Formula	%Weight
Quartz	SiO$_2$	20.0
Feldspar	KNaO·Al$_2$O$_3$·6SiO$_2$	25.0
Kaolin	Al$_2$O$_3$·2SiO$_2$·2H$_2$O	27.0
Limestone	CaCO$_3$	28.0

0.001 M MgCl₂ and 0.1 M MgCl₂ for further investigation.

3. Zeta Potential Measurement

The ceramic glaze mixture for zeta potential measurement as a fuction of pH was prepared at 1%w/v in three media: deionized water, 0.001 M MgCl₂ and 0.1 M MgCl₂ The suspension was shaken overnight before being transferred to a zetameter, and the zeta potential was measured from high to low pH.

4. Adsorption Study of SDBS in Ceramic Glaze Suspension

All batch adsorption studies were carried out by addition of 0.1 g of ceramic glaze mixture into Erlenmeyer flasks containing 50 mL of SDBS solution in deionized water or 0.001 M MgCl₂ or 0.1 M MgCl₂ at various concentrations. The flasks were then immersed in water bath shaker at 25 °C and the pH was adjusted to pH 8 by using 0.01 M KOH. The suspensions were shaken with constant agitation rate of 125 rpm shaking water bath for 2 h, then centrifuged for 15 minutes. The concentrations of SDBS in the clear supernatant were determined by measuring UV absorbance at 223 nm using UV-Vis spectrophotometer and the adsorbed amount of SDBS per unit mass of ceramic glaze was calculated.

5. Dispersion Stability Study

Dispersion stability of ceramic glaze suspension was investigated by measuring relative absorbance at 850 nm by spectrophotometer. The experiment carried out the same procedure as adsorption study except that 0.5 g of ceramic glaze powder and 25 mL of the deionized water or 0.001 M MgCl₂ or 0.1 M MgCl₂ were employed and without centrifugation after being shaken. Hence, after shaking at 25 °C for 2 h, the ceramic glaze suspension was poured into a cell and then the absorbance was recorded for a period of time.

RESULTS AND DISCUSSION

1. Zeta Potential of Ceramic Glaze Mixture

The zeta potentials of ceramic glaze mixture in deionized water media, 0.001 M MgCl₂ and 0.1 M MgCl₂ measured by a Zeta Size are presented in Fig. 1. The isoelectric point (IEP) values of ceramic glaze mixture in deionized water and in 0.001 M MgCl₂, if extrapolated, were probably less than 2, whereas the IEP value in 0.1 M MgCl₂ was about 6.7.

The zeta potential indicated the increase in negative surface charge for the system in deionized water and 0.001 M MgCl₂ media when pH was higher than 2, whereas the increase in positive surface charge for the system in 0.1 M MgCl₂ was observed at pH above 6.7. This might be considered as generation of variable charge mechanism.
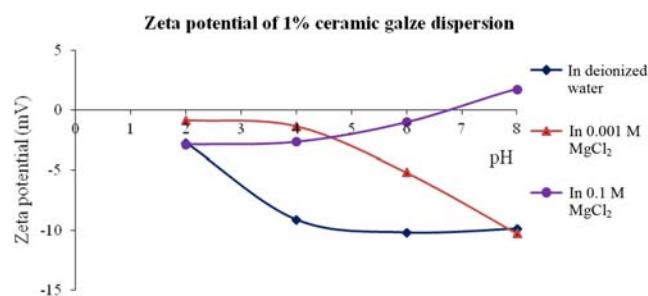

Fig. 1. Zeta potential values versus pH values of 1% w/v of ceramic glaze mixture in distilled water, in 0.001 M MgCl₂, in 0.1 M MgCl₂.
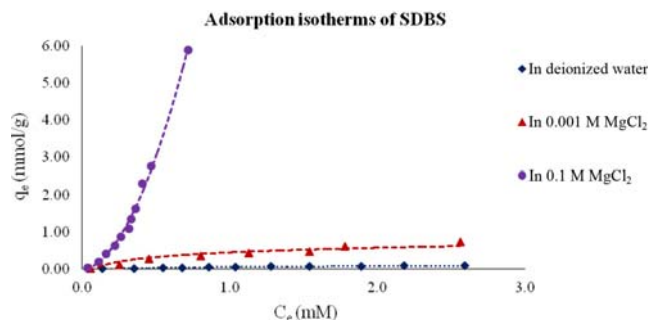
Fig. 2. Adsorption isotherms of SDBS on ceramic glaze in three media.

The net surface positive charge of ceramic glaze in deionized water media and 0.001 M MgCl₂ media increased with decreasing the suspension pH value due to protonation of surface hydroxyl group on ceramic glaze mixture.

2. Adsorption of SDBS on Ceramic Glaze Suspension

Adsorption isotherms of SDBS on ceramic glaze in all three media are shown in Fig. 2. These isotherms relate the uptake per unit weight, q_e, to the equilibrium concentration in the bulk fluid phase C_e. In water and in 0.001 M MgCl₂, the amounts of SDBS uptake increased gradually with increasing the SDBS concentration. On the other hand, in 0.1 M MgCl₂, the amount of adsorbed SDBS increased sharply with increasing SDBS concentration.

The linear form of Langmuir isotherm can be expressed by

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \frac{C_e}{q_m} \tag{1}$$

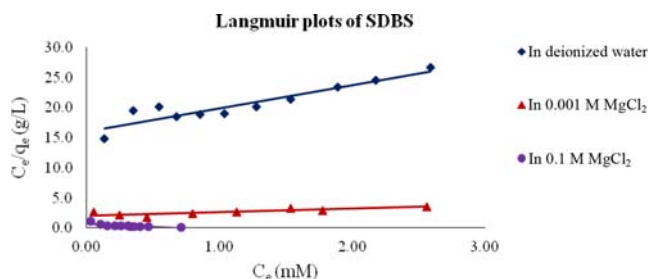where q_m is the maximal substance amount of adsorbate, the unit

Fig. 3. Linear plots of Langmuir isotherm of SDBS on ceramic glaze in three media.

Table 2. Comparison of parameters of Langmuir and Freundlich isotherm models of SDBS in three media

Model/Isotherm constants/R^2		SDBS in		
		Deionized water	0.001 M MgCl₂	0.1 M MgCl₂
Langmuir	q_m (mmol/g)	0.28	1.30	-
	b (L/mmol)	2.2	0.46	-
	R^2	0.9216	0.8755	-
Freundlich	K_f	0.31	0.39	9.65
	n	1.2	1.4	0.57
	R^2	0.9909	0.9573	0.9957

Fig. 4. Linear plots of Freundlich isotherm of SDBS on ceramic glaze in three media.

Fig. 5. Diagram illustrating the adsorption of DBS⁻ on the surface of ceramic glaze in the presence of MgCl₂.

of q_m is the same as q_e, and b is the adsorption constant. The constant b is related to the energy of adsorption and increases as the strength of the adsorption bond increases.

The plots of C_e versus C_e/q_e, shown in Fig. 3, correspond to the Langmuir model only in deionized water and in 0.001 M MgCl₂ and, thus, the values of q_m and b can be obtained from the slope and the intercept of the plots (Table 2). On the other hand, the adsorption of SDBS on ceramic glaze suspension in 0.1 M MgCl₂ within the concentration range studied did not follow the Langmuir isotherm.

The Freundlich model assumes heterogeneous surface and multilayer adsorption. The linear form of Freundlich isotherm is expressed by the following empirical equation:

$$\log q_e = \log K_f + \frac{1}{n}\log C_e \qquad (2)$$

where K_f and n are empirical constants depend on the nature of adsorbate and adsorbent.

From the Freundlich plots of $\log C_e$ versus $\log q_e$ shown in Fig. 4, the values of n and K_f were approximately calculated from the slope and the intercept of the plot, respectively, and summarized in Table 2.

The adsorption behavior of SDBS in deionized water and in 0.001 M MgCl₂, within the concentration range studied, followed both the Langmuir isotherm and the Freundlich isotherm (Figs. 3-4), but the Freundlich isotherm was more favored. Even though the Langmuir binding equilibrium constant (b) of SDBS in deionized water was more than in 0.001 M MgCl₂ indicating higher affinity for SDBS adsorption; the q_m value in deionized water was much less than that in 0.001 M MgCl₂. Also, the q_m value of SDBS on the glaze in the presence of ferric oxide and MgCl₂ was much more than in NaCl in our previous work [3]. The Freundlich constants, K_f, in these two media were less than one, which indicated low adsorption capacity, and 1/n values less than one indicated normal adsorption. Adsorption of SDBS in 0.1 M MgCl₂ did not agree with the Langmuir isotherm, but it followed the Freundlich isotherm with the constant K_f more than one representing high adsorption capacity, and 1/n also more than one indicated cooperative adsorption.

In deionized water, without MgCl₂ electrolyte, DBS⁻ ions may adsorb by electrostatic attraction to positive charge surface sites present on the ceramic glaze, i.e., limestone and kaolin. Since only a few adsorption sites have positive charge, therefore, a very small amount DBS⁻ was adsorbed. At low SDBS concentrations, the surfactant adsorbed via electrostatic attraction between its negative head group of a single particle and the positive charged surface sites. At higher concentrations both electrostatic and particularly hydrophobic interactions are favored. The former occurs via the additional charged
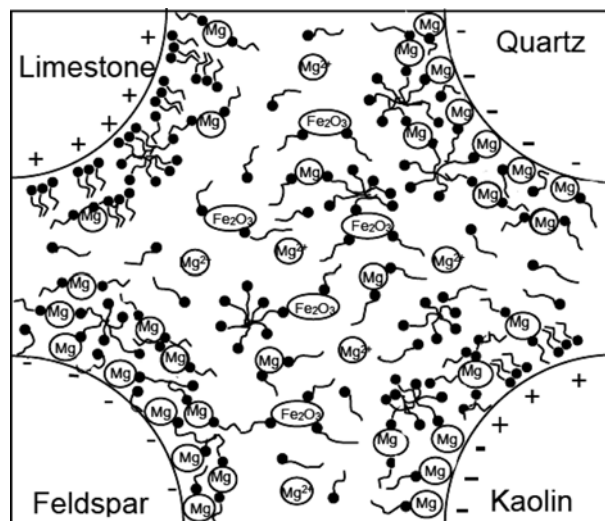
surface sites. The latter occurs via the already adsorbed surfactant, which has exposed hydrocarbon chains that provide nuclei for further adsorption via lateral interactions between adjacent chains and the subsequent formation of admicellar structures. If the concentration is above the CMC, there is expected to be direct adsorption of micelles to the surface. Therefore, the increase of adsorbed amount of surfactant with increasing concentration corresponds to the Freundlich isotherm rather than the Langmuir isotherm. In the presence of MgCl₂ electrolyte, the Freundlich constant K_f increased with increasing MgCl₂ concentration, indicating the increase in the adsorption capacity. The amount of adsorbed DBS⁻ increased significantly in the presence of high concentration of MgCl₂. It may be suggested that the magnesium ions were firstly adsorbed on the negative sites of feldspar, quartz and kaolin surfaces, which was confirmed by the positive zeta potential at pH 8, and behaved as a binder between the glaze and DBS⁻ ions. Therefore, DBS⁻ ions could be adsorbed as the second layer on the glaze via the adsorbed magnesium ions resulted in the high increase of adsorbed DBS⁻ on the surface as shown in Fig. 5.

3. Dispersion Stability of Ceramic Glaze

The relative absorbance is directly proportional to the stability of dispersion. The decrease of the relative absorbance indicates the decrease in dispersion stability, in other words, an increase in dispersion instability. The relative absorbance values at 850 nm of 2%w/v ceramic glaze suspensions in the absence and presence of SDBS at concentrations of 0.6, 2.0 and 3.5 mM in the three media as a function of time are measured and shown in Figs. 6-8. Without SDBS, the dispersion stability increased slightly with increasing the MgCl₂ concentration. Less than 10% of the suspension remained after one hour. In the presence of SDBS, the stability of suspension increased with increasing the concentration of SDBS, especially in deionized water and 0.001 M MgCl₂ media much higher relative absorbance was investigated.

In the presence of 2.0 mM and 3.5 mM of SDBS, the glaze suspension after one hour in deionized water and in 0.001 M MgCl₂ was more than 86% remaining, while that in 0.1 M MgCl₂ medium less than 12% of the suspension remained. This should be in agree-
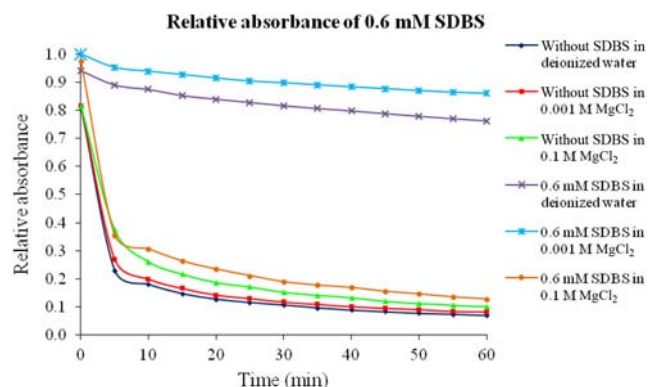
Fig. 6. Relative absorbance versus time of ceramic glaze suspension in the absence and presence of 0.6 mM SDBS in three media.
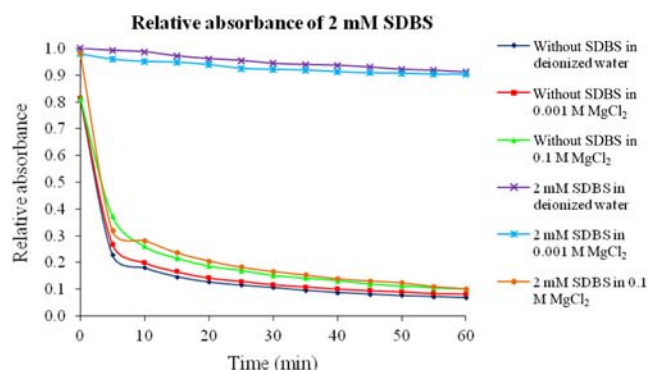

Fig. 7. Relative absorbance versus time of ceramic glaze suspension in the absence and presence of 2.0 mM SDBS in three media.
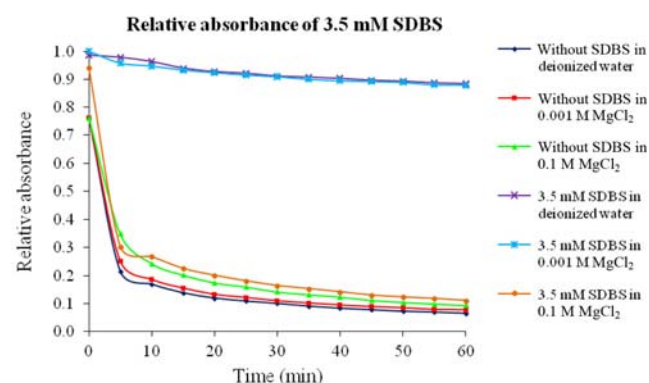

Fig. 8. Relative absorbance versus time of ceramic glaze suspension in the absence and presence of 3.5 mM SDBS in three media.

ment with the high adsorbed amount of SDBS in 0.1 M MgCl$_2$ medium that the DBS$^-$ ions might be adsorbed via the adsorbed Mg ions and hence neutralizing the positive charge on the particles which resulted in sedimentation.

CONCLUSIONS

The negative zeta potential of 1%w/v ceramic glaze suspension

in deionized water and 0.001 M MgCl$_2$ media at pH 2-8 indicated a more negative charge on the particle surface, while that in 0.1 M MgCl$_2$ the zeta potential became positive at pH above 6.7, indicating less negative charge or the more positive charge on the surface. Adsorption experimental work of SDBS on 0.2% ceramic glaze suspension was performed in a batch system at 25 °C, pH 8.0, adsorption contact times of 2 hours and agitate rate at 125 rpm. As a function of SDBS concentration, the adsorbed amount increased continuously with increasing the SDBS concentration. The adsorbed amount of SDBS on ceramic glaze from the same initial concentration in the three media increased from deionized water<0.001 M MgCl$_2$<0.001 M MgCl$_2$ media. It indicated that SDBS in deionized water and 0.001 M MgCl$_2$ media could be able to stabilize the dispersion of particles in the dilute suspension for more than an hour. The stability of suspension increased with increasing the concentration of SDBS from 0.6 mM to 3.5 mM.

ACKNOWLEDGEMENTS

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