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Boosting the impact of cinnamaldehyde-contained polymer microemulsion on the corrosion of C1018 alloy in an acidic medium

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Abstract

The corrosion and related limitations are the detriment of steel equipment installed in the wells which limits the useful life of the equipment and reuse their use in future wells. In this work a new type of environmentally friendly acid corrosion inhibitor composition based on the cinnamaldehyde-contained poly(methyl methacrylate) nanocolloid with a unique combination of cinnamaldehyde as a filming additive, and an organo-sulfur compound for petroleum wells and water wells subjected to stimulation with acid solutions is presented. cinnamaldehyde-contained poly(methyl methacrylate) nanocolloid was prepared in microemulsion system and characterized with dynamic light scattering (DLS) and zeta potential analysis. The new acid corrosion inhibitor with enhanced performance by a synergistic action between the cinnamaldehyde/ organo-sulfur compound and poly(methyl methacrylate) nanoparticles provide a fewer instances of pitting and also reduced rate of corrosion than previous inhibitors which include cinnamaldehyde alone. The anti-corrosion performance was performed on the C1018 alloy surface based on the Gravimetric analysis using corrosion rate measurement with the decrease in metal weight during the reference time period. The experimental results based on the AFM analysis shows that the cinnamaldehyde molecule's adsorption on an alloy surface in 15% HCl align. The anti-corrosion performance data confirmed that the developed inhibitor exhibited more than 95% (using 4 Wt.% of inhibitor) corrosion inhibition efficiency in 4% and 15% aqueous HCl at 303-343 K.

Keywords: Cinnamaldehyde-contained polymer, Nanocolloid, Microemulsion, Acid corrosion inhibitor

1. Introduction

Based on the (Schlumberger Market Analysis), it has been estimated that a remarkable portion of the gas and world's oil reserves lie in carbonate reservoirs that these heterogeneous carbonate formations primarily consist of calcite, dolomite or combinations thereof [1,2]. For an enhanced production the use of suitable acid stimulation technologies useful for dissolving calcium and magnesium-based carbonates [3]. Some acid platforms have been suggested that include to use of strong mineral acids such as hydrochloric acid, gelled or emulsified acids, organic-based acids such as formic acid and acetic acid [4]. While these mythologies are effective, strong mineral acids are corrosive to metal substrates and steel that occurs at all stages from downhole to surface equipment and processing facilities such as leaks in tanks, casings, tubing, pipelines, and other equipment. So, this serious drawback consists a large portion of the total costs for oil and gas production and the need of the eco-friendly acid corrosion inhibitor is necessary. Recently, Choudhary et al. [5] synthesized cinnamaldehyde thiosemicarbazone as a corrosion inhibitor on the deterioration of mild steel in 1M and 15% HCl. Utilization of nanosized particles such as polymer nanoparticles for increasing the stability and activity of inhibitor is an effective technology. The nanoparticles, due to the small size, would stabilize the inhibitor by connecting the different phases with one another more strongly. Our opinion is that cinnamaldehyde containing poly(methyl methacrylate) (PMMA) nanoparticles can effectively block charge transfer at the metal–electrolyte interface and prevent corrosion of the metal substrate under aggressive saline conditions [6,7]. PMMA is a good polymer that was used in several application field such as water treatment [8-13], corrosive materials [14,15], hydrophilic/hydrophobic surface [16]. Particular attention may be paid to microemulsion as soft templet system to prepare polymer nanoparticles [17-20]. Oil-in-Water (W/O) microemulsion as nanoreactor is a simple route to control of size and morphology of nanoparticles with excellent monodispersity [21-24]. Amino acid based-microemulsion due to usage of amino acid with synergistic effect can be used for synthesis of anti-corrosive material.

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In the present research work, this new green type acid corrosion inhibitor described herein composed of an active corrosion intermediate, an internal intensifier, a surfactant, a solvent, a filming additive, and/or other additives. This type of inhibitor has highlighted the influence a unique combination of cinnamaldehyde-contained poly(methyl methacrylate) based on the microemulsion system on the deterioration of alloy surface in 4% HCl and 15% HCl at different temperatures 303 K-443 K, as this has not been investigated by researchers to the best of our knowledge.

2. Experimental

2.1. Material

Lecithin as stabilizer agent to produce oil-in-water microemulsion, methyl methacrylate monomer ($CH_2C(CH_3)COOCH_3$) (AR grade), were purchased from Merck Company. Cinnamaldehyde ($C_6H_5CH=CHCHO$) as filming agent, thiourea (CH_4N_2S) active corrosion intermediate, were purchased from Sigma-Aldrich. Potassium persulfate as a hydrophilic initiator for the polymerization of MMA was purchased from Haihang Industry Co. LTD. Deionized water (0.055 µS/cm) which was produced in our lab with PKA (Smart two pure) instrument. C1018 alloy that the chemical compositions of the alloys are summarized in Table 1, was received.

Table 1. The chemical compositions of the exemplary alloy

Chemical composition (%) of C1018 alloy									
C	Si	Mn	P	S	Cr	Ni	Cu	Mo	V
0.14-0.20	-	0.60-0.90	≤ 0.040	≤ 0.050	-	-	-	-	-

2.2. Preparation of PMMA nanocolloid in W/O amino acid based- microemulsion system

In this section, for the preparation of the PMMA nanoparticles in oil-in-water microemulsion system as a stable polymer nanocolloid, the MMA oil-in-water microemulsion systems consisting of lecithin as a surfactant, MMA as apolar phase, and water should be obtained. Briefly, a clear aqueous solutions of lecithin amino acid with concentrations of the 0.02 M were prepared. After that, based on the previous reports by Mirhoseini and Salabat [25], the certain amount of the MMA monomer oil that was adjusted at %2 (Wt.) was added to surfactant solutions and vigorously stirred to form a clear MMA-in-Water reverse microemulsion. Potassium persulfate as an oxidizing agent was added to the amino acid based microemulsion MMA nanodroplets system and the vial were put at 60°C for 4 h to start the polymerization process [14,26,27]. Finally, the cloudy and stable PMMA colloid was obtained and this color change confirmed the synthesis of the PMMA nanoparticles in amino acid-based micelles as soft template nanoreactors was successful.

2.3. Preparation of cinnamaldehyde-contained PMMA microemulsion

To prepare cinnamaldehyde contained PMMA nanoparticles, the water-in-cinnamaldehyde oil microemulsion system was used that an aqueous PMMA nanocolloid was replaced to the polar phase. In this procedure, for the first time, a clear solution of amino acid in cinnamaldehyde oil with concentrations of the 0.02 M were prepared. After that, the adjusted amount of the PMMA nanocolloid that was fixed at 5 % (Wt.) was added to surfactant solutions and vigorously stirred for the formation the cinnamaldehyde contained PMMA nanoparticles reverse microemulsion. A control sample was also prepared without PMMA nanoparticles to confirm the role of polymer nanoparticles in acid anti-corrosion activity.

2.4. Preparation of the acid anti-corrosive

To prepare a final acid anti-corrosive, the above cinnamaldehyde contained PMMA nanocolloid was mixed with thiourea as organic-sulfur compound that the molar ratio of cinnamaldehyde to thiourea was fixed at 3.

2.5. Characterization

Zeta-average diameters (Dz), particle Size distributions, polysispersity index, and also zeta potentials to confirm the stability of polymer nanocolloid and particle charge were studied by a dynamic light scattering (DLS) using a Zeta plus−Zeta potential Analyzer (Brookhaven Instruments Corporation).

2.6. Corrosion study with gravimetric analysis

In order to study the impact of acid corrosion inhibitor, inhibitor concentration, and also impact of temperature, weight loss-based C1018 alloy surface corrosion analysis for all experiments were carried out at different temperatures from 343 K to 373 K in 4 and 15% HCl in the absence of inhibitor as control sample and the presence of new green inhibitor at 2-5 Wt.% concentrations, respectively that the total exposure time of 4 h was selected. The experiment condition are as follows: 50 ml of uninhibited or control sample and working solution (inhibited corrosion solution) was employed, the size of the metal surface C1018 samples was 8.7 cm × 1.4 cm × 0.1 cm, Fig. 1.

Fig. 1. The image of the used C1018 alloy sample 8.7 cm × 1.4 cm × 0.1 cm.

The following equation (Eq. 1) was utilized for the calculation of the corrosion rate of the prepared green inhibitor and control sample.

$$\text{Corrosion rate (\%)} = w_o - w / w_o \times 100 \quad (1)$$

where w_0 and w are the weight loss of C1018 alloy in the absence and the presence of the acid anti-corrosive, respectively. In corrosion study, an acid corrosion inhibitor provides sufficient corrosion inhibition so that a pitting index has definitions of the integers. A pitting index estimates pitting (for example, localized corrosion) susceptibility with integer values ranging from 1 through 5, where a value of 1 designates a low susceptibility to pitting and 5 is a high susceptibility. Table 2 shows the definitions of the integers for the pitting index [4].

Table 2. Definitions of the pitting index

Rank	Description of pitting
0	No pits. Surface similar as before test.
1	Intergranular corrosion on cut edges of coupon. Trace mount of pits of major surfaces.
2	Small, shallow pits on surface. Or etching of surfaces.
3	Scattered, deep pits on surface; less than 10.
4	Deep pitting and etching on surfaces.
5	Layers of metal degraded
6	Coupon almost diminished.

2.7. Effect of temperature on corrosion

In order to examine the impact of temperature on the deterioration of the used C1018 alloy surface in 4% and 15% HCl with/without the corrosion inhibitor, the gravimetric method was also employed at different temperatures. The experiment parameters are as follows: 50 ml of uninhibited and inhibited corrosion solution was used as a working solution, the size of the samples was 8.7 cm × 1.4 cm × 0.1 cm, the temperature was between 303 K to 343 K, and the exposure time was 4 hours.

2.8. Surface characterization

The surface evaluation was performed on the C1018 alloy surface samples after they were exposed to the various concentration of corrosion inhibitor to capture the surface changes. Primary, the alloy surface was immersed in the anti-corrosive (with/without) at room temperature for 4 hours, washed with water, dehydrated with a hot air gun, and used for surface characterization. The surface appearance of the specimens was assessed using the atomic force microscope (AFM) (BRISK).

3. Results and discussion

3.1. DLS and Zeta potential analysis of PMMA nanocolloid prepared in O/W microemulsion

Fig. 2 shows the DLS analysis of the cinnamaldehyde contained PMMA nanoparticles prepared in microemulsion system at fixed surfactant concentration. based on our previous report that the PMMA nanocolloid was prepared in microemulsion system composed of lecithin surfactant, the particles size was obtained 67 nm [14]. As can be seen from Fig. 2, it is clear that the PMMA particle size controlling is mainly dependent on surfactant concentration and physic-chemical properties of cinnamaldehyde as apolar phase and the average PMMA particle size is 86 nm in this system. Based on the DLS and also zeta potential analysis, the desirable characteristics, such as low size Z-average diameter (Dz) (<101 nm), low polydispersity index (0.05), high positive zeta-potential (-28.34 mV) were observed for the polymer nanocolloid prepared in O/W system as soft template method [29].

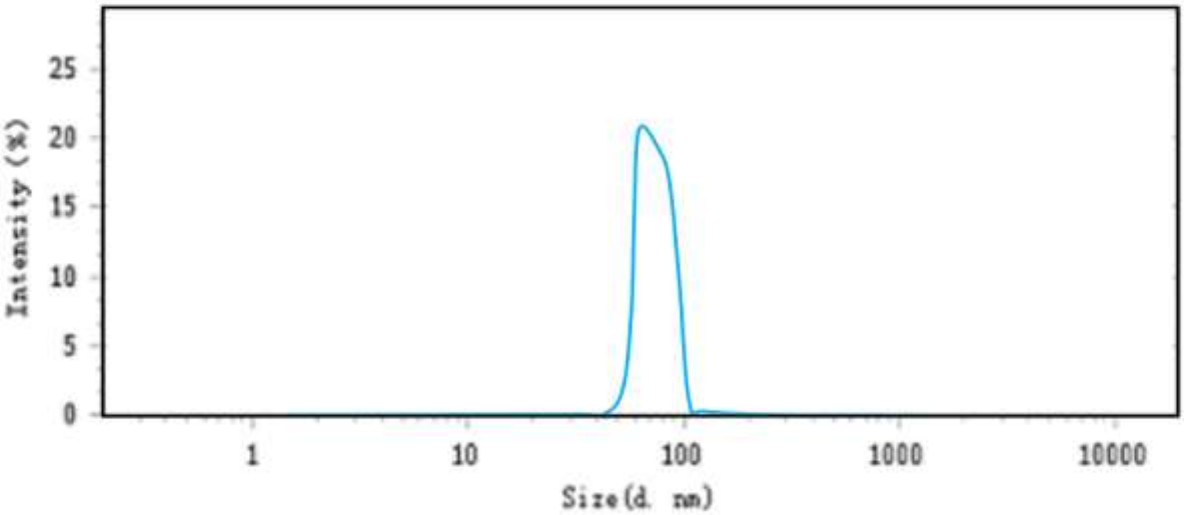

Fig. 2. DLS analysis of cinnamaldehyde contained PMMA nanoparticles prepared in microemulsion system.

3.2. Acid anti-corrosive study

Table 3, displays the (%) based on the weight loss values for C1018 alloy surface corrosion that were exposed to 4 and 15% HCl in the absence and presence of 2-5 Wt.% of inhibitor and the pitting index are also listed at different temperature from 303 to 443 K for all condition as well as control sample that was prepared without PMMA nanoparticles in the formulation of corrosion inhibitor. As shown, the presented results imply that C1018 alloy surface in an uninhibited solution (without inhibitor) experiences an increase in metal loss as temperature in creases with bad pitting index.

The metal weight loss values for the C1018 alloy surface in the corrosion inhibited solution, namely 4% and 15% HCl + 4% corrosion inhibitor, experiences extremely low metal loss, and the metal loss gradually rises with temperature. The obtained results confirmed that the prepared green corrosion inhibitor based on the cinnamaldehyde-contained PMMA nanoparticles by forming an insoluble protection amine layer on the surface of C1018 alloy surface has extremely activity to control of the alloy surface deterioration in acidic media. In considering 15% HCl, 4 Wt.% of inhibitor exhibited outstanding anti-corrosion performance against C 1018 alloy surface with η of 96.40% at 303 K, and it dropped to 94.10% at 343 K. The excellent results of this research work evidently approved that the green cinnamaldehyde-contained PMMA nanoparticles with a synergistic effect of lecithin as surfactant in microemulsion formulation to form nanomicelles has the potential to mitigate the corrosion of alloy surface with a temperature range from 303K to 343K in a variety of acidic solution media. Control sample that at above was mentioned as the inhibitor without polymer nanoparticles shown low inhibition at all condition.

Table 3. The results of anti-corrosion study at different conditions

Corrosion inhibitor (Wt.%)	Temperature (K)	Solution	η(%)	Rank of pitting		
				With inhibitor	Control	Without Inhibitor
2	303	4% HCl	22.8	3	4	6
		15% HCl	17.3	6	6	6
	323	4% HCl	19.5	5	6	6
		15% HCl	7.2	6	6	6
	343	4% HCl	6.6	6	6	6
		15% HCl	2.2	6	6	6
3	303	4% HCl	89.8	2	4	6
		15% HCl	77.6	3	5	6
	323	4% HCl	82.5	3	5	6
		15% HCl	70.4	3	5	6
	343	4% HCl	80.1	4	6	6
		15% HCl	65.2	5	6	6
4	303	4% HCl	98.8	0	1	6
		15% HCl	96.4	0	3	6
	323	4% HCl	98.0	0	1	6
		15% HCl	95.7	0	3	6
	343	4% HCl	96.3	0	2	6
		15% HCl	94.1	0	4	6
5	303	4% HCl	96.0	0	1	6
		15% HCl	92.8	0	3	6
	323	4% HCl	91.8	0	1	6
		15% HCl	84.5	1	3	6
	343	4% HCl	90.2	0	2	6
		15% HCl	84.0	1	4	6

3.3. AFM study

The surface roughness of the thin films formed on the surface of alloy is strongly dependent on the concentration of the applied amine and AFM analysis is a simple technique to investigate of the surface [31,32]. As pictured in Fig. 3 b compared to the Fig. 3(a), the surface is relatively smooth with fine needle-like features for treated-C1018 alloy with inhibitor after 4h was observed. AFM analysis provides images with near-atomic resolution for measuring surface topography. AFM is also referred to as Scanning probe microscopy. Atomic Force Microscopy is capable of quantifying surface roughness of samples down to the angstrom-scale. The observed image confirm that a thin layer of amine compound formed on the C1018 alloy surface that can be protected of alloy under acidic condition and in presence of the green inhibitor no deterioration and pitting were detected on the alloy surface. Applications of AFM in the field of solid-state physics and corrosion test study include (a) the identification of amine atoms at a surface, (b) the evaluation of interactions between a specific atom and its neighboring atoms with anti-corrosion agent and also amine groups, and (c) the study of changes in physical properties arising from changes in an atomic arrangement through atomic manipulation as well as amine film formation. However, if a few monolayers of adsorbed fluid are lying on the surface of a rigid sample, the images may look quite different.

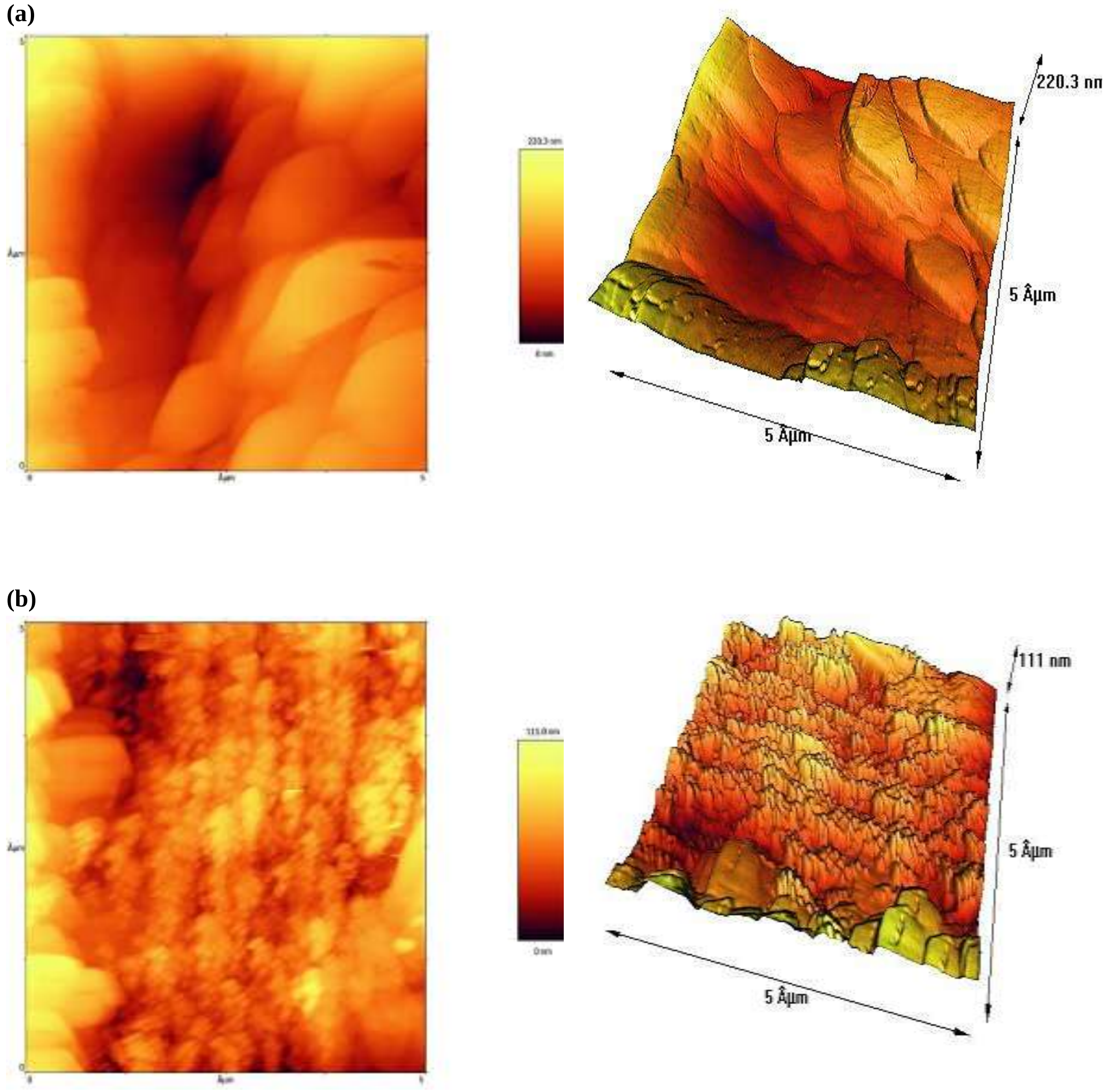

Fig. 3. AFM images of the (a) untreated and (b) treated-C1018 alloy with inhibitor after 4h in HCl 15%.

4. Conclusion

In this work we introduce a novel type of eco-friendly acid corrosion inhibitor composition based on the cinnamaldehyde-contained poly(methyl methacrylate) nanocolloid with a unique combination of cinnamaldehyde as a filming additive, and an organo-sulfur compound for petroleum wells and water wells subjected to stimulation with acid solutions. The DLS analysis presented the average particle size is 67 nm. The new acid corrosion inhibitor with enhanced performance by a synergistic action between the cinnamaldehyde/ organo-sulfur compound and acid droplet nanoparticles provide a fewer instances of pitting and also reduced rate of corrosion due to the formation the cinnamaldehyde molecule's thin film on the alloy surface in 15% HCl align. The anti-corrosion performance data confirmed that the developed inhibitor exhibited more than 95% (using 4 Wt.% of inhibitor) corrosion inhibition efficiency in 4% and 15% aqueous HCl at 303-343 K.

Conflicts of Interest

The author declares no conflict of interest.

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