RESEARCH PAPER

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The effect of amino acid type on the anti-corrosion activity of poly(methyl methacrylate) nanocolloid in boiler

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Abstract

The main aim of the present work relates to the preparation of a chemical water-treating agent based on a new polymeric nanocolloid capable of preventing the general corrosion and local corrosion of boiler systems and providing excellent heat transfer properties. The water treating agent is constituted of poly(methyl methacrylate) as a main component which were prepared in a reverse microemulsion with two type of amino acids composed of glutamic acid, N-(ethoxycarbonyl) alanine and also mixed amino acid with a compound coating film forming agent, an oxygen scavenger. The prepared nanocolloid were characterized with DLS and zeta potential analysis and a typical sample was labeled as CAG characterized with AFM to approve the DLS result. Before the anti-corrosion study of all prepared samples, AFM analysis was used to confirm the formation of the nanolayer film on the coupon surface. The water-treating agent is injected into boiler feed water to perform the prevention of the corrosion of the heat transfer surface of the boiler. Thus, a coating film capable of suppressing corrosion is formed of the coating film forming agent on the heat transfer. The new area of the application represents an extension of the utility of a unique combination of polymeric nanocolloids and amino acids with synergistic effects with specific applications that need significant investigation in industrial and academic fields.

1. Introduction

Boiler systems are broadly employed in residential and industrial applications for supplying hot water and raising steam. In all boilers and their associated equipment, including piping, heat exchangers, valves, radiators, etc., the heated water or steam is circulated in a loop which may be open or closed [1,2]. Most industrial boiler and residential boiler systems are constructed of carbon steel and equipment corrosion is a continuing and serious problem that has to be addressed both with proper equipment design and operation along with periodic inspection and maintenance [3].

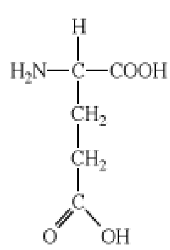
In the last two decades, a widely researched field of corrosion inhibitors based on amino acids has been considered due to they are environmentally friendly, relatively cheap, non-toxic, and biodegradable [4]. Amino acids are molecules that possess at least one amino (single bond- NH_2) group and one carboxyl (single bond -COOH) group that can coordinate with metals through an amino group and a carboxyl group, and consequently act as a corrosion inhibitor. Ibrahimi et al. [5] reported a review paper on corrosion inhibitors for metals and alloys with amino acids and their derivatives. Pour-Ali et al. [6] investigated the corrosion inhibition performance of l-asparagine (L-Asp), l-isoleucine (L-Iso), and l-proline (L-Pro) amino acids as eco-friendly compounds were investigated on mild steel coupons immersed in 0.5 M HCl solution. They reported at 1000 ppm, L-Asp, L-Iso, and L-Pro show inhibition efficiencies of around 95%, 93%, and 91%, respectively. Gong et al. [7] investigated the role of glutamic acid in enhancing the corrosion inhibition of polyaspartic acid on Q235 carbon steel as a green inhibitor.

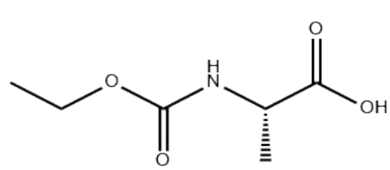
In continuation of previous work [8], the current work aims to study and compare the corrosion inhibition performance of a novel poly(methyl methacrylate) nanocolloid that was prepared in reverse microemulsion systems with two types of amino acids. Among the colloidal systems microemulsion as a soft template with a controllable size, nanoreactors were widely used to prepare nanocolloids [9-11]. Microemulsions were composed of polar phase, apolar phase, and surfactant [12-14]. Microemulsions were also used to fabricate nanoparticles or hybrid nanoparticles [15-19], engineering nanostructures with different morphology such as nanotube-shaped, core-shell or hollow nanospheres [20-22], polymer nanomaterials with a tunable size and morphology [23]. To the best of our knowledge, there are no studies investigating the corrosion inhibition performance and their mechanism of action in boiler systems. To fill this gap, in this work, three simple evaluations based on the Fe level of feed water measurements, the amount of Fe deposited on the thermal conduction surface, and test piece corrosion rate examinations were used to assess the anti-corrosion activity of each compound. So, at first, two types of PMMA nanocolloids were prepared and characterized with DLS and zeta potential technique. then their anti-corrosion performance was evaluated in a pilot-scale study in a fire tube boiler.

2. Experimental

2-1. Materials

Commercial glutamic acid ($C_5H_9NO_4$) (99.0%), N-(ethoxycarbonyl) alanine ($C_{15}H_{21}NO_4$) (99%) (scheme. 1) 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 and 2-(2-aminoethoxy)ethanol as amine source was purchased from Sigma-Aldrich. Potassium persulfate as a hydrophilic initiator for the polymerization of MMA was purchased from Haihang Industry Co. LTD, and sodium metabisulfite was received from Qingdao Tianya Chemical Co., Ltd. Deionized water (0.055 µS/cm) which was produced in our lab with PKA (Smart two pure) instrument.





Glutamic acid N-(ethoxycarbonyl) alanine

Scheme. 1. Molecular structures of amino acids used to prepare PMMA nanocolloids

2-2. PMMA nanocolloid preparation

To prepare PMMA nanocolloid, two oil-in-water microemulsion systems consisting of N-(ethoxycarbonyl) alanine and glutamic acid as a surfactant, MMA as apolar phase, and water should be obtained. In this procedure, for the first time, two clear aqueous solutions of amino acids with concentrations of the 0.02 M were prepared. After that, same as in our previous works, the adjusted amount of the MMA that was fixed at 2%Wt was added to surfactant solutions and vigorously stirred to form MMA-in-Water reverse microemulsions. Potassium persulfate as an oxidizing agent was added to the two above amino acid stabilized MMA colloidal systems and all vials were put at 60°C for 4 h to start the polymerization process [24-26]. 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 was successful. Then the prepared PMMA nanocolloids were labeled as CG and CA based on the name of the usage amino acids (CG for colloid prepared with glutamic acid and CA for colloid prepared with N-(ethoxycarbonyl) alanine) and then characterized with DLS analysis. Furthermore, a microemulsion was prepared by a solution of both amino acids (50% V/V), and then MMA oil was added and the polymerization process was completed after 4 h. This PMMA nanocolloid sample was labeled as CAG. Fig. 1 shows the prepared PMMA nanocolloids at different stabilizer agent types. As can be seen in Fig. 1-(b) (CG sample) the poorly agglomerated PMMA nanoparticles were observed in contrast to other samples (CA and CAG).

2-3. Preparation of the anti-corrosion inhibitor

To prepare anti-corrosion inhibitors, two mixture of 2-(2-aminoethoxy)ethanol (3 g) and sodium metabisulfite (6.5 g) as oxygen scavengers was prepared, separately. Then 0.5 ml of both PMMA nanocolloids composed of CG and CA samples were added to both mixtures. Finally, a fine powder was obtained and aged at room temperature. As a control sample, a sample was prepared without PMMA nanocolloid to prove the role of PMMA nanoparticles capped with glutamic and N-(ethoxycarbonyl) alanine stabilizing agents.

2-4. Preparation of anti-corrosion-PMMA colloid

For this purpose, 100 g of both above-mentioned powders were dissolved in 1 L pure water (ion-exchange water) feed and used for the corrosion tests in the boiler.

2-5. Characterization of PMMA nanocolloid

To confirm and determine of average particle size and polydispersity index (PDI) of both samples (CG, CA, and CAG) DLS analysis as a common technique with Bettersize (Optical nanoparticle size analyzer Nanoptic 90) was employed. Before DLS analysis, all samples (CG, CA, and CAG) were sonicated for 6 min at room temperature. The prepared PMMA nanocolloid was prepared in a mixed amino acid system as a typical sample (CAG), and to determine the average roughness in cleaned coupons in corrosion test an atomic force microscope (AFM, Ara AFM) was used.

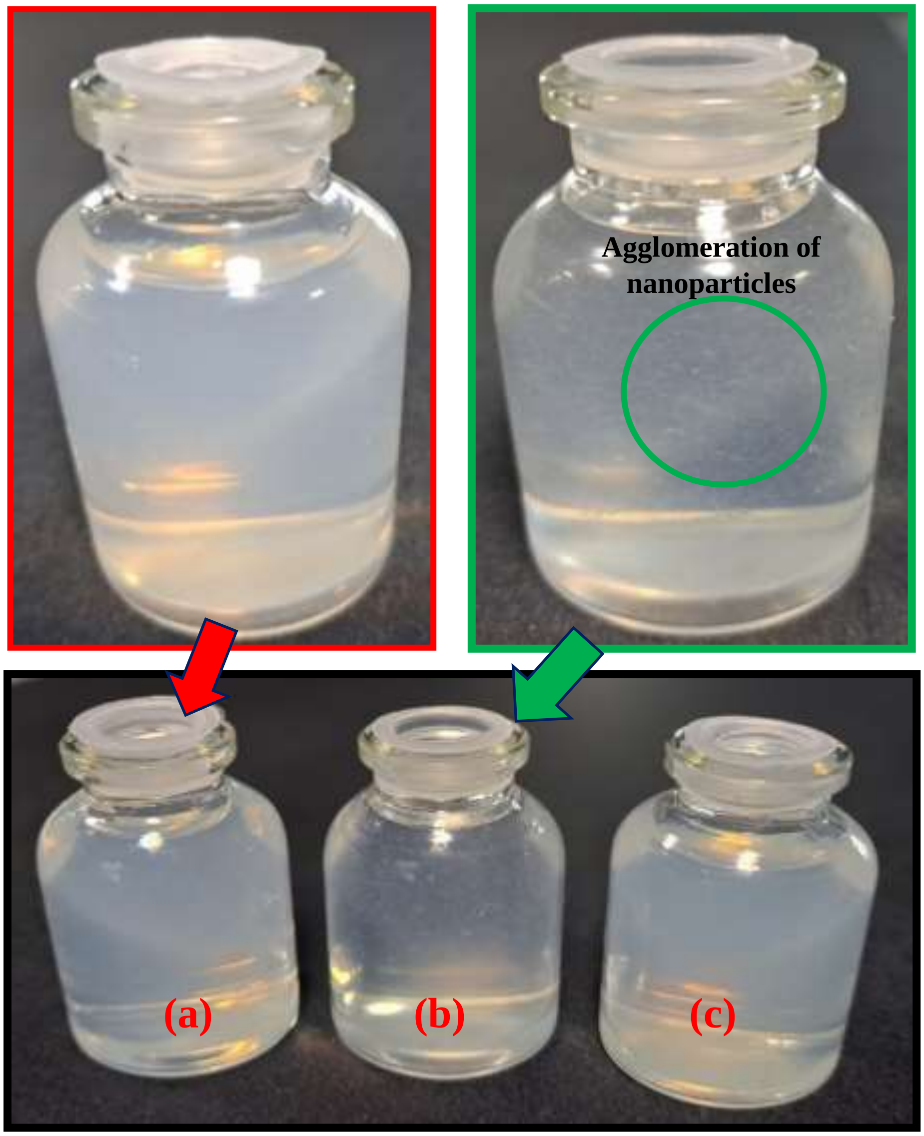


Fig. 1. The PMMA nanocolloids prepared based on the microemulsion system at different stabilizing agents (a) glutamic acid (CG) (b) N-(ethoxycarbonyl) alanine (CA), and (c) mixed CA-CG (50% V/V).

2-6. Corrosion study

In this section, the corrosion inhibition examination was first investigated to confirm the role of the prepared nanocolloid inhibitors and the formation of the thin film on the coupon's surface (the chemical composition of coupon is 0.22 C, 0.3 Si, 1.4 Mn, 0.01 Cr, 0.01 Ni, 0.045 P, 0.05 S, and Fe balance (wt %)) with the rotating hanging piece method and compared with a blank sample (only water) [27, 28]. Briefly, three coupons in 3 groups of nanocolloid inhibitors were immersed in simulated boiler water at 100 rpm and 35 ±1 °C for 72 h. Then, all coupons were collected and cleaned with 2.4 M HCl, 1.5 M NaOH, and ethanol to remove the corrosion products on the surface Fig. 2 (a-c).

Fig. 2. The coupons surface after treated with anti-corrosion inhibitor nanocolloids

Based on our previous report, a test boiler was operated under the following conditions: a pressure of 4 MPa, a blow rate of 1%, a condensate recovery of 20%, pure water (ion-exchange water) feed, and the use of a heater-degasser [outlet degassing capacity (dissolved oxygen level: DO) of 0.03 mg/L] was used to study the corrosion tests. In the corrosion test, to the feed water, the anti-corrosion-PMMA colloid with different PMMA nanocolloids (CG and CA) in the formulation of inhibitor were added in an amount of 10 mg/L, separately. As a result, the pH of boiler water was 10.20-10.57.

During the test period of one week, the boiler was operated for 5 days and stopped for 2 days. The procedure was repeated. That is, the test was performed for 14 days. By the above test, the iron level of the feed water (average) (mg/L), the amount of iron deposited on the thermal conduction surface (mg Fe/cm2), and the corrosion rate (mdd) of a steel test piece placed in the test boiler were evaluated for both anti-corrosion inhibitors.

As a comparative study, the corrosion study was tested in the same condition but with a solution composed of 2-(2-aminoethoxy)ethanol, sodium metabisulfite, and water without PMMA nanoparticles. The unit (mdd) of the corrosion rate of a test piece is based on the daily amount of corrosion (mg) for an area of 100 cm^2.

Test piece corrosion rate (mdd) was calculated based on the thickness measurement data with Thickness Meter PCE-CT 80-FN0D5 with accuracy after foil calibration: ±(1% of reading + 1 µm); for many materials such as iron, steel, aluminum, copper, brass, stainless steel; Diameter: 17mm / 0.67). Iron Determination by colorimetric method Using O-Phenanthroline was performed [29].

3. Result and discussion

3-1. DLS and zeta potential analysis

Fig. 3. presents the zeta potential of all PMMA nanocolloid samples that were sonicated for 6 min. As can be seen, the zeta potential of prepared PMMA nnaocolloids (CA, CG, and CAG) are -10.23, -6.75, and -15.44, respectively, and depends upon the electrical potential and the surface charge of the used amino acids. The surface potential of

charged particles and stability for CAG samples increases with an increase in zeta potential [30]. The negative charge is due to the presence of the carboxyl radical (–COOH) of amino acids and the carbonyl group of PMMA. For the CG sample, the stability is low and the PMMA nanoparticles tend to agglomerate. The process of agglomeration is ruled by thermodynamics. Depending on the sign of the enthalpy of interaction, ensembles consist of (repelling) poorly agglomerated particles [30].

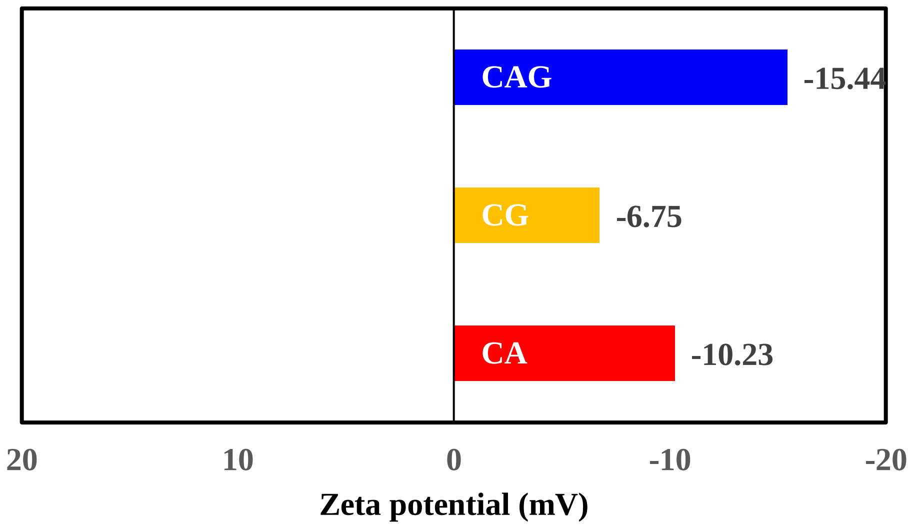


Fig. 3. The zeta potential analysis of the PMMA nanocolloids

Fig. 4-(a-c) shows the DLS analysis of the PMMA nanocolloids prepared in microemulsion with different stabilizing agents composed of glutamic acid, N-(ethoxycarbonyl) alanine, and a mixture of both amino acids. DLS particle size analyzers are used to measure the size and hydrodynamic radius (R_h) of very small particles (0.6 nm to 6 um) in solution. The particle size range of DLS depends on the properties of the analyzed species, such as refractive index or density, as well as the surrounding formulation, mainly the viscosity. As can be seen from Fig. 2, it is clear that the PMMA particle size controlling is mainly dependent on the stabilizing agent type, and when the capping agent is a mixture of glutamic acid and N-(ethoxycarbonyl) alanine the PMMA particle size is 41 nm it is smaller than only N-(ethoxycarbonyl) alanine (53 nm) and only glutamic acid (119 nm).

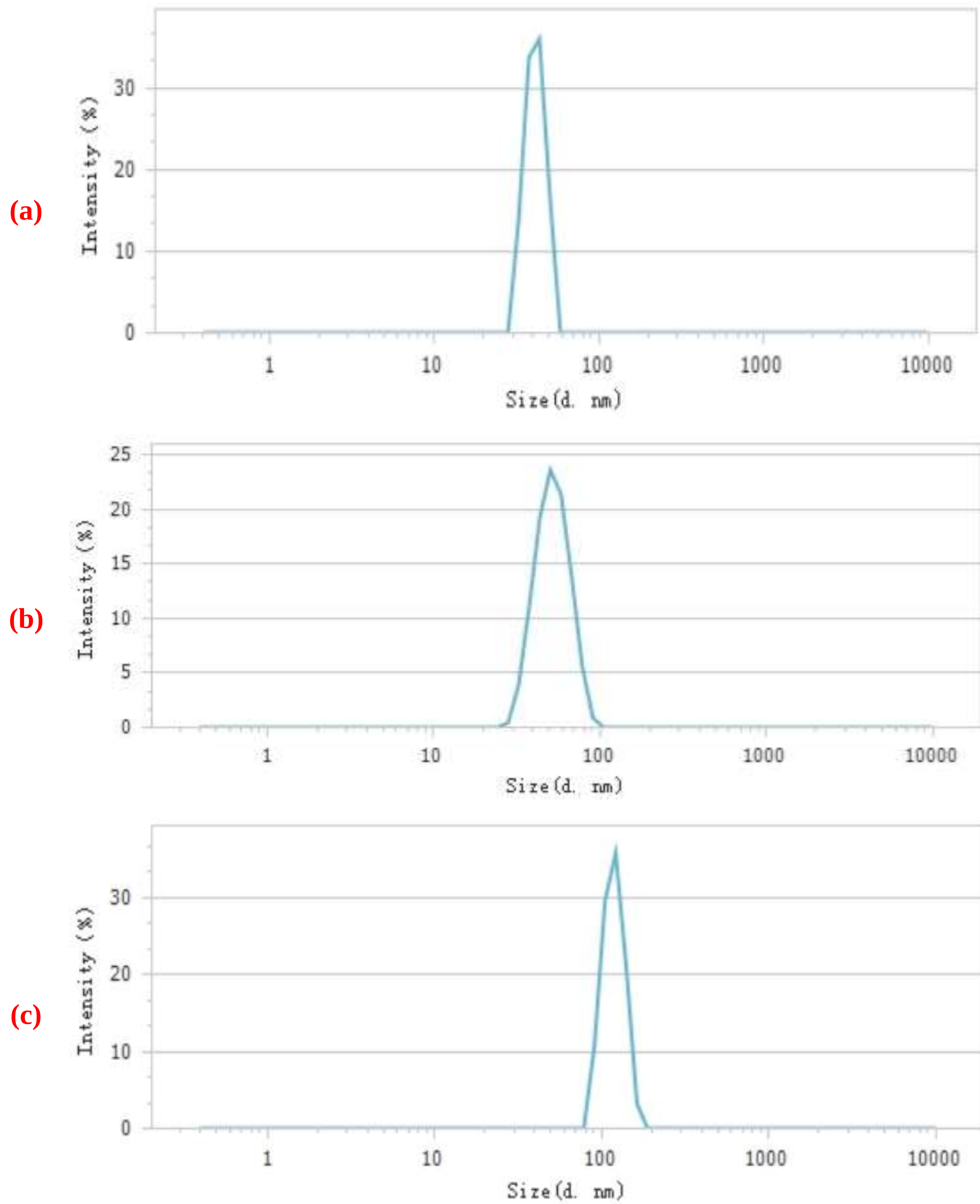


Fig. 4. DLS analysis of the PMMA nanocolloids (a) CA, (b) CG, and (c) CAG.

3-2. AFM study

Fig. 5-(a) shows the AFM analysis for PMMA nanocolloid prepared in mixed amino acid stabilized microemulsion system. As can be seen from this Fig. it is clear that the PMMA nanoparticles have a fine and spherical morphology with very excellent monodispersity and confirmed the DLS result. Furthermore, the surface roughness of coupons in the primary test before the investigation in the boiler was analyzed with AFM. As shown in Fig. 3-(d), the addition of the PMMA nanocolloid (CAG) that was prepared in a mixed amino acid system reduced the surface roughness of coupons compared to CA and CG samples, confirming the formation of a protective layer on the coupon surface. Besides, higher roughness was observed in the CG (Fig. 3-b) and CA (Fig. 3-c), which could explain why the CAG nnaocolloid exhibited higher anti-corrosion performance than other samples.

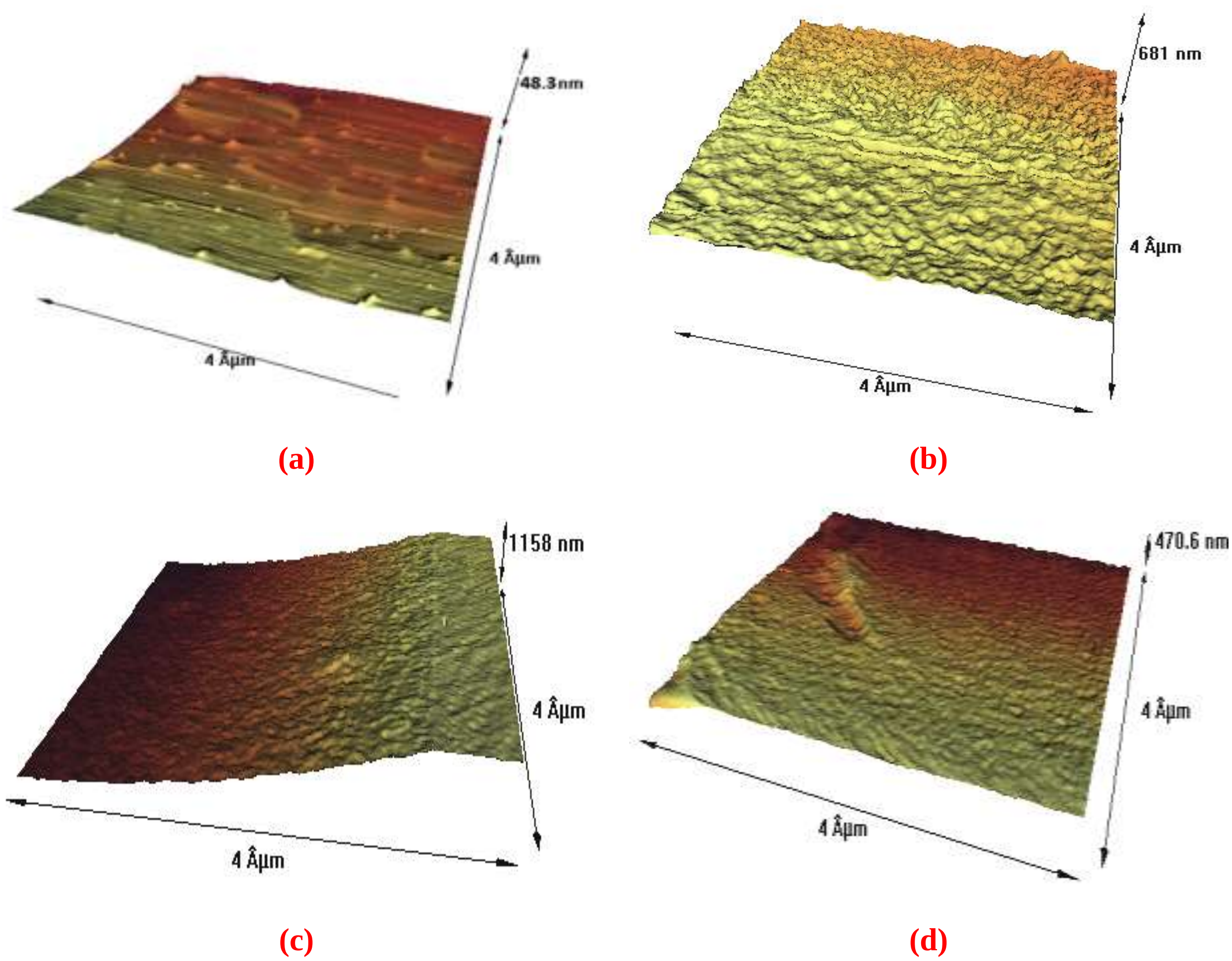


Fig. 5. The AFM analysis of (a) PMMA nanocolloid (CAG as a typical sample), the coupons surface after treated with anti-corrosion nanocolloid inhibitors (b) CA, (c) CG, and (d) CAG sample.

3-3. Corrosion study

Fig. 5-(a-c) presents the corrosion investigation based on the Fe level of feed water (mg/L), the amount of Fe deposited on the thermal conduction surface (mg Fe/cm^2), and the test piece corrosion rate examination. As can be seen from Fig. 4, the best anti-corrosion performances were observed for the anti-corrosion sample produced by PMMA nanocolloid (CAG) in all test examinations (Fe level of feed water, amount of Fe deposited, and corrosion rate) for the used boiler. The reason for this event relates to the increase of the PMMA surface area that is stabilized with N-(ethoxycarbonyl) alanine molecules. The carbonyl group of PMMA coordination with amine provided stable complexes against diffused oxygen. The interaction of glutamic acid or N-(ethoxycarbonyl) alanine molecules and amine molecules led to create a nanometric layer on the surface of metal.

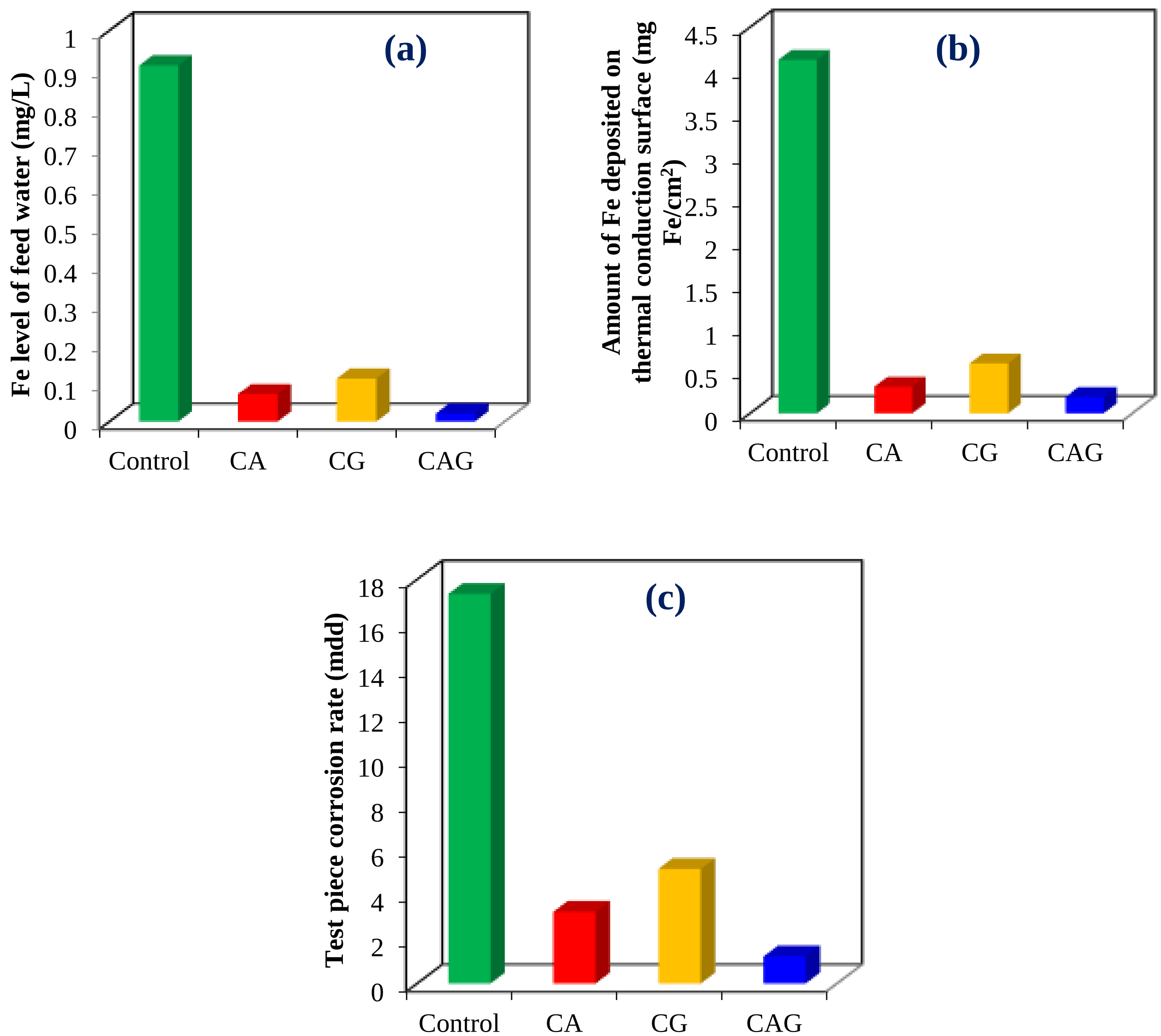


Fig. 6. (a) Fe level of feed water (mg/L), (b) Amount of Fe deposited on thermal conduction surface (mg Fe/cm^2) and test piece corrosion rate (mdd)

Amino acids, small-molecule compounds with amino and carboxyl groups, may react with each and PMMA nanoparticles with carboxyl groups and act as chains to link PMMA nanoparticles, which enhance the inhibition of nanocolloid on metal corrosion. In this study, it is clear that a combination of selected glutamic acid and N-(ethoxycarbonyl) alanine due to the potential synergistic effect of the amino acids and PMMA on corrosion inhibition has the best anti-corrosion performance. The corrosion inhibition of nanocolloid was promoted by PMMA nanoparticles stabilized with both glutamic and N-(ethoxycarbonyl) alanine with less weight loss and less pitting. This phenomenon showed that glutamic acid could bind with N-(ethoxycarbonyl) alanine at carboxyl groups and amino groups via donor–acceptor interactions and accelerate the diffusion of PMMA to the boiler surface. Furthermore, the binding between PMMA and the carbon steel surface of the boiler can be enhanced by a mixing of glutamic and N-(ethoxycarbonyl) alanine, resulting in a stable and dense protective layer or film on the surface [31,32]. To the best of our knowledge, this is the first investigation into the mechanism of two amino acids that were used as stabilizing agents for the production of stable PMMA nanocolloid as an enhancer to improve corrosion inhibition in the pilot-scale.

This work provides a new strategy to enhance existing green inhibitors, which would significantly reduce the cost of boiler water treatment and its adverse environmental impacts. The nanometric layer coated on the surface of the boiler is highly strong under dissolved oxygen and more effectively protects the surface of the boiler. Other samples CG and CA have weak anti-corrosive activity in the same condition for three tests.

4. Conclusion

This work relates to corrosion inhibiting compositions for aqueous systems comprising a mixture of amino acids, polymeric nanocolloid, and an amine compound having a specific structure including an amino group and a hydroxyl group in the molecule thereof, and oxygen scavenger. The novel amino acid stabilized polymer nanocolloid based-anti corrosion inhibitor is an effective corrosion inhibitor and also this is useful from an environmental standpoint because they are non-toxic, contain no heavy metals, and have no phosphorus.

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