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Modification of Gambier extracs as green inhibitor of calcium carbonate ($CaCO_3$) scale formation

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ABSTRACT

The addition of the Gambier extract (*Uncaria gambier* Roxb leaves) modification (Gambier:benzoic acid:citric acid/2:1:2) as a green inhibitor on the formation of $CaCO_3$ scale at various concentrations was carried out using a seeded experiment method. The experiments were performed with observing the precipitation change of the $CaCO_3$ crystals growth obtained. In order to prove the efficiency of the inhibitor in inhibiting the formation of the $CaCO_3$ crystals, the changes of the crystal morphology were investigated by scanning electron microscopy (SEM) and the changes of the crystal size distribution were analyzed by particle size analyzer (PSA). The research results showed that the Gambier extract modification was able to inhibit the formation of the $CaCO_3$ scale indicated with the morphology change of the $CaCO_3$ crytals and smaller crystal size distribution after the addition of this inhibitor.

1. Introduction

Scale formation causes a serious issue encountered by many industries in Indonesia and other countries such as; the industry of oil or gas production, water transport, power generation, and batch precipitation [1–6]. Faced with this issue; PT Pertamina Geothermal Energy (PGE) which is one of subsidiary of Indonesian Oil Company (PT. Pertamina) has spent US$ 6–7 million to rejuvenate the pipe installation of the Geothermal Energy Industry for 10 years. In order to solve this problem, several researchers studied many additives to prevent the scale formation or to modify the crystal morphology [7–12]. The addition of the additive in a growth solution of the scale formation is looked a cheaper method and an effective enough to inhibit the scale formation of an inorganic material.

Gambier extract from *Uncaria gambier* Roxb leaves was reported as a green inhibitor of $CaCO_3$ scale formation [13]. Nevertheless, the Gambier extract has several problems, such as; it is easily moldy and cannot be stored for a long time. In order to control these problems, the Gambier extract was mixed with citric and benzoic acid. Previous research reported that the citric and benzoic acid can play a role as an inhibitor also to inhibit the scale formation [14–16]. In addition, these mixtures are potential as a green inhibitor of the scale formation because they have some chemical compounds such as tannic acid, catechin, and quercetin [13] which are effective inhibitors of scale formation of $CaCO_3$ beside the citric and benzoic acid. Other advantages of these mixtures are cheaper and they can be applied as a green inhibitor like other materials which have been used previously [17–27] to keep our environment.

The effects of the Gambier extract modification as the inhibitor of $CaCO_3$ scale formation at various concentrations from 0 to 300 ppm on the precipitation rate of calcium carbonate was observed at temperature of 80 °C and at a growth solution of 0.1, 0.3 and 0.6 M under seeded experiment. The seeded experiments were applied by previous researchers to investigate the inhibition of the calcite crystal growth with using phosphonates additive and they monitored the rate of crystallization by measuring the concentration of calcium ion as a function of time [28]. In this experiment, the rate precipitation of the calcium carbonate seed crystals was monitored by weighing the amount of the calcium carbonate precipitated as function of time [13]. The precipitation obtained was characterized using scanning electron microscopy and particle size distribution to give an overview of its composition, size and morphology of the $CaCO_3$ crystals was analyzed by particle size analyzer.

2. Experimental procedure

2.1. Preparation of Gambier extract modification

Preparation of Gambier extract modification was made with the ratio of 2:1:2 (Gambier:benzoic acid:citric acid). The benzoic and citric

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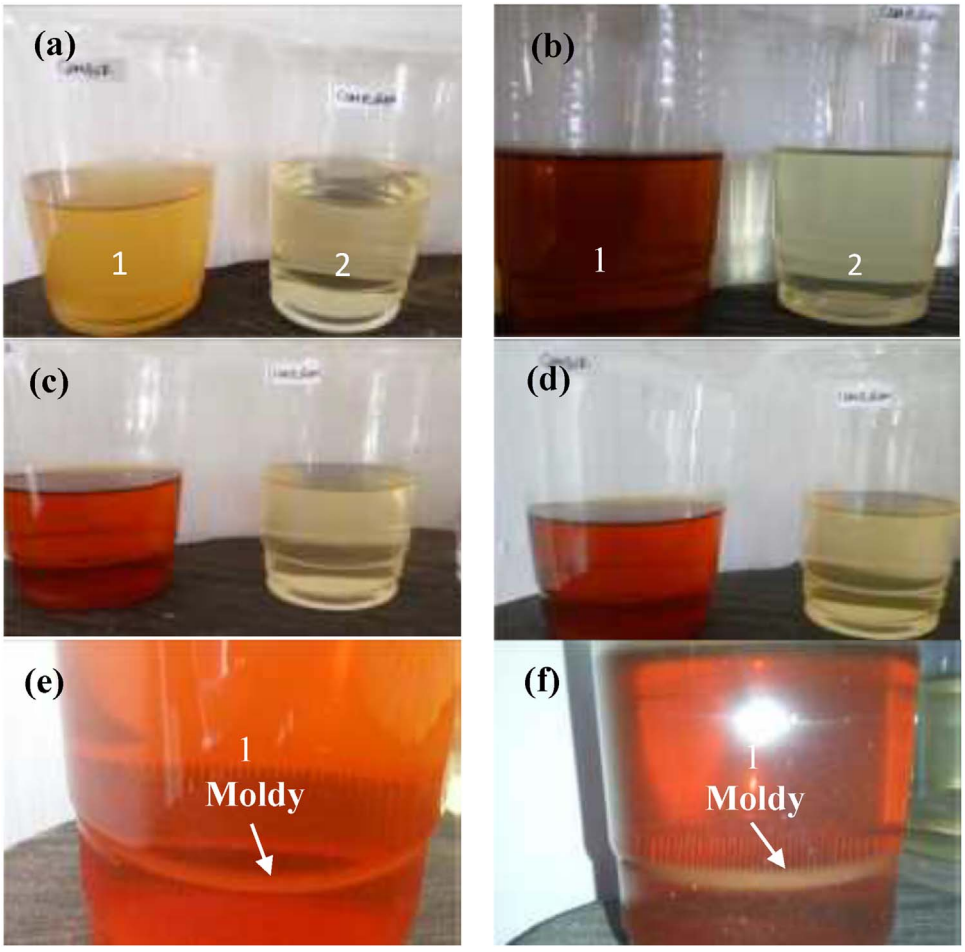

Fig. 1. The qualitity of Gambier extract (1) and modified Gambier (2) in the room temperature at (a) First day, (b) 7 days, (c) 14 days, (d) 25 days, (e) 14 days (drawn closer), and (f) 25 days (drawn closer).

acid used was produced by commercial products of Merck, Germany. Gambier was prepared from dry leaves of *Uncaria Gambir* Roxb from West Sumatra, Indonesia. The amount of 2 g powdered Gambier, 1 g benzoic acid, and 2 g citric acid was placed into a 1 L volume glass added water up to 1 L. The mixtures were heated at temperature of 80 °C for 1 h and left for 1 night. Then, the mixtures were filtered and the filtrate was used as an inhibitor of $CaCO_3$ scale formation. Similar with previous research, the Gambier extracts was identified containing 40% of tannic acid, 25% of catechin, and 12% of quercetin [13].

2.2. Seed crystal production

The crystal seeds of $CaCO_3$ were made with mixing each $CaCl_2$ anhydrate solution (1 M) and Na_2CO_3 solution (1 M) in 50 mL of water. The $CaCl_2$ anhydrate and Na_2CO_3 used was purchased by commercial products of Merck, Germany. The mixture was stirred by magnetic stirrer and it was left for 2 h to produce seed crystals continued with separating the seed crystals from the liquor by filtration through a 0.45 μm Millipore filter. The seed crystals obtained were cleaned thoroughly with water and dried at room temperature for at least two days. This work was replicated a bunch of times to produce the required amount of the $CaCO_3$ seed crystals for the work.

2.3. Experiments of crystallization

An amount of 200 mL of $CaCl_2$ anhydrate solution (0.2 M) was mixed into 200 mL of Na_2CO_3 solution (0.2 M) in 500 mL Nalgene polypropylene bottles followed by shaking to produce a homogeneous solution. This procedure was to prepare the growth solution of 0.1 M $CaCO_3$. The homogeneous solution produced was separated through a 0.45 μm Millipore filter. The solution was fed into 250 mL Nalgene polypropylene bottles, each containing 50 mL consisted of 7 bottles. The bottles were fed back to the bottle-roller bath adjusted the temperature at 80 °C and the rotating at 40 rev min^{-1} continued with adding together 100 mg of the $CaCO_3$ seed crystals into each bottle. Over the 90 min the experiment run, each bottle was picked up for every 15 min. The crystals obtained were cleaned thoroughly with water and placed in oven at temperature of 105 °C for one day. The weight of the $CaCO_3$ crystals obtained from each bottle was measured and the amount precipitated of $CaCO_3$ crystals was calculated.

For each crystallization experiment, a blank containing no inhibitor was performed in conjunction with the presence of inhibitor. The similar experiments were carried out for concentration variety of $CaCO_3$ growth solutions for each 0.3 and 0.6 M. Following the similar experiment applied for the 0.1 M $CaCO_3$ growth solution concentration, a growth solution concentration of 0.3 M $CaCO_3$ was made with mixing 200 mL of $CaCl_2$ anhydrate solution (0.6 M) and 200 mL of Na_2CO_3 solution (0.6 M), and a growth solution of 0.6 M $CaCO_3$ was made with mixing 200 mL of 1.2 M $CaCl_2$ anhydrate solution and 200 mL of 1.2 M Na_2CO_3 solution.

2.4. Presence of additives

The influence of various additive concentrations was observed by adding different amount of the additive (0, 50, 100, 200 and 300 ppm) in the similar treatment. The weight of the crystals was measured and