



Application of Gambier Concentrate and Coconut Shell Smoke Condensate as Eco-friendly Inhibitors Against Magnesium Carbonate Precipitation

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Abstract

This study focused on inhibiting the formation of magnesium carbonate ($MgCO_3$) scale using a green inhibitor obtained from a blend of gambier concentrate and liquid smoke. The experiment utilized the unseeded method, and the inhibitors were characterized through Fourier transform infrared spectrophotometry and gas chromatography-mass spectrometry. A 0.050 M $MgCO_3$ growth solution was treated with a combination of gambier concentrate and grade 2 smoke condensate derived from coconut shell at varying concentrations. The findings demonstrate that this inhibitor combination is of high quality and effectively suppresses $MgCO_3$ scale formation, achieving an inhibition rate exceeding 90%. This conclusion is supported by the observed alterations in the crystal morphology and particle size distribution of $MgCO_3$, which were examined using particle size analyzer and scanning electron microscopy. The inclusion of inhibitors led to the transformation of crystal structure, resulting in thin and small sheets instead of the original granular form. Further analysis of the $MgCO_3$ crystal structure with X-ray diffraction disclosed notable intensity variations upon the addition of inhibitors. Consequently, the gambier concentrate and grade 2 smoke condensate combination from coconut shell in all its variations enhance the quality of the inhibitor mixture. This combination shows promise as a green inhibitor for preventing $MgCO_3$ scale formation to save our environment.

Keywords: $MgCO_3$ scaling, green inhibitor, gambier concentrate, smoke condensate, coconut shell

1. INTRODUCTION

Pipes are commonly used in various industries, including oil and gas companies, drinking water systems, and chemical industries, to facilitate the transportation of fluids from one location to another [1]-[6]. One recurring issue in hospital boilers, drinking water systems, chemical industries, and oil and gas pipelines is the formation of deposits called scale, which leads to corrosion and disrupts the ionic composition, pH, pressure, and temperature of the water [7]-[13]. The costs associated with scale deposits are significant, with an estimated value of around 1.5 billion Euros annually in France [14], 0.8 billion USD in the UK, and 9 billion USD in the United States [15]. Additionally, the cost for replacing PT Pertamina Geothermal Energy's geothermal well pipes in Indonesia is estimated to

be 6–7 million dollars every 10 years [16]-[18]. Additives are commonly employed as anti-scalers to control scale deposition and minimize its detrimental effects [19]-[23].

The concentrate derived from *Uncaria Gambier* Roxb., known as gambier concentrate, contains natural compounds including tannic acid, catechin, and quercetin [24][25]. These compounds have been recognized as environmentally friendly inhibitors for inhibiting the growth of scale [18]. However, gambier concentrate has limitations, including vulnerability to mold and a short storage life [17]. To overcome these challenges, gambier concentrate can be combined with coconut shell liquid smoke, which is known to contain phenolic compounds, acids, aldehydes, and ketones [26]-[28]. These compounds are believed to inhibit fungal growth and enable long-term usage as a scale inhibitor. Additionally, the combination of gambier concentrate and smoke condensate from coconut shells offers a cost-effective and eco-friendly alternative. The novelty of this research lies in the application of a combination of gambier concentrate and smoke condensate from coconut shells. The unique properties of gambier concentrate, known as an environmentally friendly inhibitor, are acknowledged along with its limitations [17][18]. The combination with liquid smoke from coconut shells is proposed to overcome these challenges,

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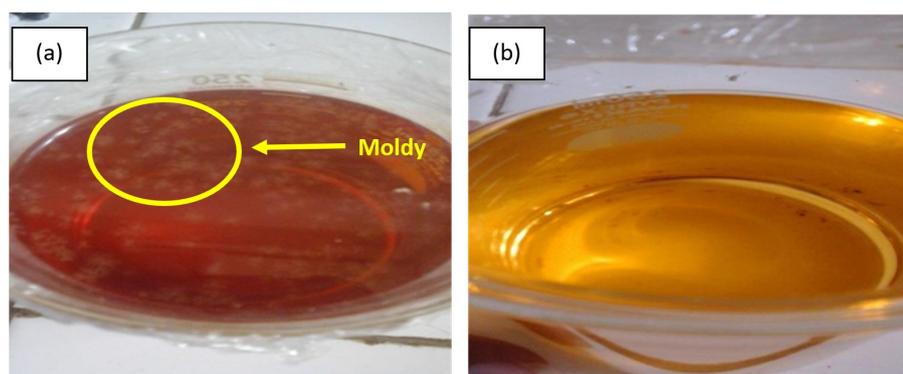
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Table 1. Concentration comparing of gambier concentrate (G) and grade 2 liquid smoke of coconut shell (L) mixtures.

Ratio of Concentration (G:L)	Concentration of G (ppm)	Concentration of L (ppm)
5 : 1	250	50
5 : 3	250	150
5 : 5	250	250
5 : 7	250	350
5 : 9	250	450

**Figure 1.** Changes in gambier concentrate (a) without the addition of grade 2 liquid smoke of coconut shell; (b) with the addition of grade 2 liquid smoke of coconut shell after being kept for 2 weeks.

offering an environmentally friendly and cost-effective alternative.

This research aims to assess the effectiveness of a combination of gambier concentrate and coconut shell smoke condensate as an environmentally friendly inhibitor in preventing the growth of magnesium carbonate (MgCO_3) scale. The study investigated various concentration ratios of gambier concentrate and smoke condensate in a 0.050 M MgCO_3 development solution, applying the seed-free approach. The rate of MgCO_3 crystal formation was assessed by observing the accumulation of sediment over time. The resulting precipitated magnesium carbonate was then analyzed through X-ray diffraction (XRD), scanning electron microscopy (SEM), and particle size analyzer (PSA) to gain insights into its morphology and size. The research to produce this article was conducted at the Department of Chemistry of the University of Lampung (Bandar Lampung, Indonesia), the Organic Chemistry Laboratory of University of Gadjah Mada (Yogyakarta, Indonesia), and the Integrated Laboratory of Sepuluh Nopember Institute of Technology (Surabaya, Indonesia).

2. MATERIALS AND METHODS

2.1. Materials

The experimental setup consists of various instruments, including an analytical balance (Kern & Sohn GMBH ABT 220-4M, Germany), a water bath (Haake S21, Thermo Fisher Scientific, USA), an oven (Thermo Fisher Scientific, United Kingdom), glassware, plastic bottles, and a magnetic stirrer. The inhibitor characterization was performed using gas chromatography-mass spectrometry (GC-MS) and Fourier transform infrared (FTIR) spectrophotometer (Shimadzu FTIR-8400, Japan). Meanwhile, the crystal characterization was obtained using a PSA (Beckman Coulter LS 13 320 MW, manufactured in the USA), XRD (Philip Analytical, manufactured in the Netherlands), and SEM (JSM 6360 LA, JEOL, Japan). The materials used in this research comprised gambier concentrate acquired from a traditional market in Bandar Lampung (sourced from certified organic farms in West Sumatra, Indonesia), NaHCO_3 and MgCl_2 (analytical grade ($\geq 99\%$ purity) obtained from Merck, Germany), grade 2 coconut shell smoke condensate sourced

from the coconut processing plant in Bandar Jaya, Lampung, Indonesia. The purification of liquid smoke to produce grade 2 liquid smoke was carried out through distillation at 130–145 °C with a distillation time of 4 h. Distillation was conducted to ensure the absence of impurities such as tar. Grade 2 liquid smoke is clear yellow in color and clearer than grade 3 liquid smoke.

2.2. Methods

2.2.1. Processing of Gambier Concentrate and Smoke Condensate

The process of making gambier concentrate in this experiment was obtained from solid gambier. Gambier leaves (*Uncaria Gambier* Roxb.), after being cleaned, were soaked to remove unwanted substances and prepare them for the concentrateion process. Subsequently, 1 kg of gambier leaves was boiled in 6 L of water at 90 °C for 3 h to concentrate the desired compounds. After heating, the still liquid gambier concentrate was filtered from the leaves. Then, the concentrate was concentrated and dried in an oven at 105 °C for 3 h to produce solid gambier (257 g). The obtained solid gambier was then finely ground into a powder form. To produce the gambier concentrate for these experiments, a powder of gambier (0.25 g) were gone into solution of purified water (1 L). The obtained solution was then agitated for 3 h at 90 °C

taking a magnetic stirrer and afterwards filtered through filter paper (Whatman No. 42 filter paper under vacuum filtration). This filtration process yielded the gambier concentrate with a concentration of 250 ppm [16].

The process of making liquid smoke begins by cleaning coconut shells from dirt or unwanted substances. A total of 5 kg of coconut shells was then burned in a pyrolysis process at 400 °C for 4 h. The smoke produced during combustion was collected and condensed into liquid form. The collected liquid smoke was then separated from solid particles and other possible impurities through filtration. The obtained liquid smoke, totaling 3.7 L, is classified as grade 3. To obtain grade 2 liquid smoke, grade 3 liquid smoke was distilled at 130 °C for 6 h. The resulting grade 2 liquid smoke is clear yellow with a lighter scent compared to grade 3. For the grade 2 smoke condensate derived from coconut shell, a 50 ppm concentration was obtained by dissolving 50 mg of grade 2 smoke condensate in purified water using a measuring flask, followed by thorough mixing. The same procedure was repeated to prepare inhibitor solutions with concentrations of 150, 250, 350, and 450 ppm [29].

2.2.2. Evaluation of the Effectiveness without the Inclusion of Inhibitors

A growth medium of MgCO_3 (0.050 M) was produced by combining 0.2 M solutions of NaHCO_3

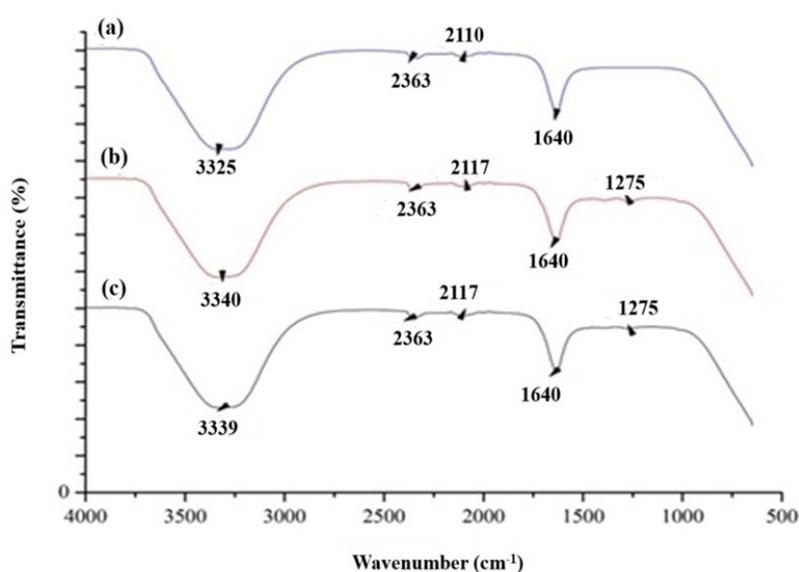


Figure 2. FTIR spectrum of (a) gambier concentrate; (b) grade 2 liquid smoke of coconut shell; (c) gambier -grade 2 liquid smoke of coconut shell (concentration ratio 5:9).

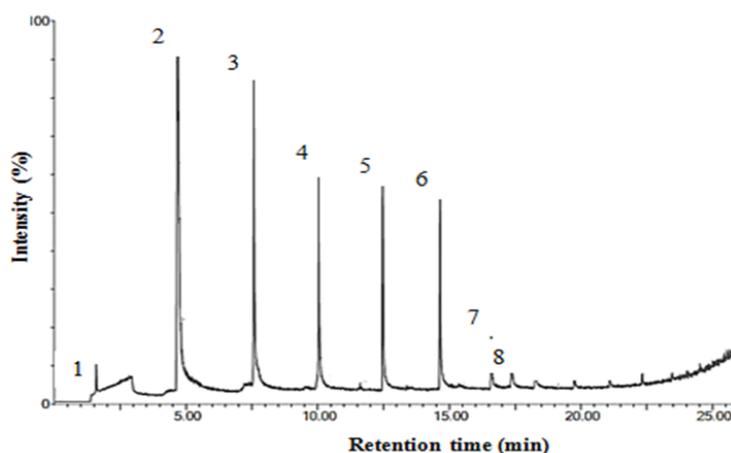


Figure 3. Characterization of gambier concentrate by GC-MS.

Table 2. Chemical compounds of gambier concentrate.

No.	Retention time (min)	% Area	Chemical compounds
1	2.86	15.83	1,3-Benzendicarboxylic acid
2	4.68	33.04	5-[(4-Methylphenoxy) methyl]phenol
3	7.82	1.20	(4-Fluorophenyl)-(2-thienyl) methanol
4	10.03	8.83	Benzothiophene
5	12.46	7.60	Pentanoic acid
6	14.65	6.83	2,6-Dihydroxyacetophenone
7	16.59	2.98	1 <i>H</i> -Purine-6-amine
8	17.36	1.84	Patchouli alcohol

and MgCl_2 , each in 200 mL of purified water. The solution was mixed for 15 min at 60 °C with a magnetic stirrer. After thorough mixing, the solution was poured into six plastic container, by every container conceiving 50 mL of the growth medium. These bottles were then located in a water bath at 60 °C. The experiment was observed for 65 min, with the initial observation taken at 15 min and subsequent observations made at 10-min intervals.

To measure the weight of the formed precipitate, the mixture from each container was filtered through filter paper, rinsed by deionized water, and then placed for 3 h in an oven at 105 °C. The resulting crystals were identified, and the most optimal sample was selected for SEM analysis (Accelerating voltage of 15 kV, magnification range 1000–5000 \times) and XRD analysis (scan range of 10°–80° (2 θ), scan rate of 2°/min, Cu $K\alpha$ radiation source ($\lambda = 1.5406 \text{ \AA}$)) to examine the crystal morphology and structure, as well as PSA analysis to determine the particle size distribution (PSD) within the precipitate.

2.2.3. Evaluation of the Effectiveness in the Presence of Inhibitors

The inhibitor solutions were prepared by combining 200 mL of the gambier concentrate (G) with 200 mL of the smoke condensate (L) at different concentration ratios. The effectiveness of the gambier concentrate and smoke condensate mixture (GL) was evaluated by varying the concentration of the smoke condensate while keeping the gambier concentrate concentration constant. The concentration ratios of the smoke condensate mixture are detailed in Table 1. The GL solution was stirred for 15 minutes with a magnetic stirrer, then refrigerated and stored in an opaque container. Each mixture was examined to assess its potency in preventing the forming of MgCO_3 crystals in a 0.050 M MgCO_3 growth solution.

To prepare the growth solution, a combination of inhibitors consisting of G and L by varying the proportion of 5:1 was combined with 0.2 M MgCl_2 and 0.2 M NaHCO_3 solutions. The resulting blend was stirred for 15 min at 60 °C using a magnetic

stirrer. Once the solutions were thoroughly stirred, they were poured into six plastic containers, with each container receiving 50 mL of the growth medium. The bottles were afterwards placed in a water bath set at 60 °C. Monitoring was carried out over a period of 65 min, starting with an initial check at 15 min and followed by additional checks every 10 min. To measure the mass of the produced crystals, the mixture from each container was filtered through filtration paper, rinsed with deionized water, and then placed in an oven at 105 °C for 3 h. This procedure was replicated with different concentration ratios of G and L, specifically at ratios of 5:3, 5:5, 5:7, and 5:9. The resulting crystals were identified, and the most effective sample was selected for SEM analysis to study the crystal morphology and PSA analysis to assess the PSD within the precipitate. All data of these experiments were obtained from triplicate experiments (n=3).

2.2.4. Data Analysis

In order to determine the efficiency of inhibitors in preventing the formation of MgCO₃ scale [30] can be employed. The effectiveness of the inhibitor (%) can be identified by the Eq. (1);

$$\text{Effectiveness of inhibitor (\%)} = \frac{(C_a - C_b)}{(C_c - C_b)} \times 100\% \quad (1)$$

where, C_a is precipitation amount of MgCO₃ at equilibrium after the addition of the inhibitor (g L⁻¹), C_b is precipitation amount of MgCO₃ at equilibrium without the inhibitor (g L⁻¹), and C_c is initial precipitation amount of MgCO₃ (g L⁻¹).

3. RESULTS AND DISCUSSIONS

3.1. Quality Assessment of the Inhibitor Mixtures

While gambier is able to be employed as a inhibitor for scale formation, it is prone to rapid degradation caused by fungal growth. To address this limitation, a combination of G and L inhibitors was utilized. This combination aimed to mitigate

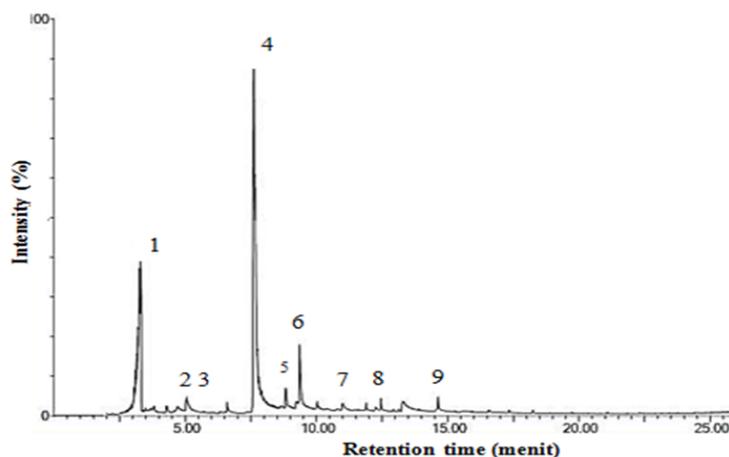


Figure 4. Characterization of grade 2 liquid smoke of coconut shell by GC-MS.

Table 3. Chemical compounds of liquid smoke (grade 2).

No.	Retention Time (min)	% Area	Chemical Compounds
1	3.30	8.19	Acetone
2	4.86	0.31	Butanoic acid
3	5.05	2.66	3-Furaldehyde
4	7.61	45.04	Phenol
5	9.36	4.57	2-Methoxyphenol
6	12.25	0.30	2,5-Dimethylfuran
7	13.28	2.93	2,4-Dimethoxyphenol
8	14.63	1.22	Methyl salicylate

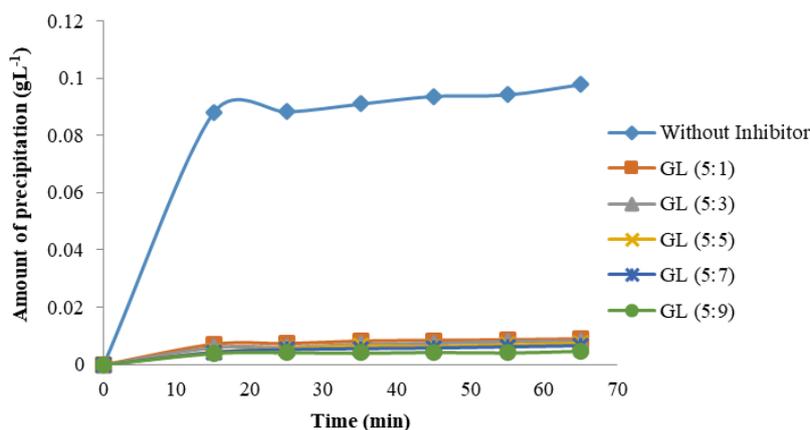


Figure 5. The growth of MgCO_3 scale at a growth solution concentration of 0.050 M versus time at varied ratios of GL inhibitor concentrations.

Table 4. Inhibitor efficiency (%) at different concentrations of inhibitor added to a growth solution of 0.05 M.

Ratio of Concentrations (G:A)	Effectiveness of Inhibitor (%)
5:1	91.34
5:3	92.19
5:5	93.29
5:7	94.00
5:9	95.44

the susceptibility of gambier concentrate to mold formation and enhance its effectiveness in preventing MgCO_3 scale formation. Figure 1 presents the resilience of the inhibitor solution and illustrates the gambier concentrate without the addition smoke condensate, which exhibited fungal growth and the presence of impurities within a two-week period. Conversely, the combination of G and L remained clear without any impurities or fungal growth. The inclusion of benzoic acid and cinnamic acid in grade 2 liquid smoke of coconut shell contributes to slowing down the deterioration of gambier concentrate by inhibiting fungal growth. These components possess antimicrobial properties [29].

3.2. Characterization of Inhibitor using FTIR Spectrophotometer

The characterization process employing FTIR spectrophotometry is conducted to recognize the functional groups present in the G and L. Figure 2 (a) displays multiple absorption bands that correspond to the chemical groups of the organic

substances in G. A broad absorption band in the area of 3417 to 3363 cm^{-1} exhibits the appearance of a hydroxyl group (O-H). Furthermore, a stretching vibration associated with O-H group found in tannic acid is observed at a wavenumber of 3324 cm^{-1} . The stretching vibration at 1640 cm^{-1} signifies the existence of a carbonyl (C=O) group in tannic acid. Peaks observed at wavenumbers 2110 and 2363 cm^{-1} confirm the presence of a C=C bond in aromatic molecules. These findings suggest that G contains various chemical compounds, including tannic acid, which can serve as inhibitors for MgCO_3 scale formation. These results indicate that the G contains a variety of chemical substances, including tannic acid, that have the potential to function as inhibitors of MgCO_3 scale formation.

Figure 2(b) demonstrates absorption bands associated with the functional groups present in smoke condensate. An absorption peak at a wavenumber of 3340 cm^{-1} indicates the stretching vibration of O-H group, which corresponds to the phenol component commonly found in grade 2 liquid smoke of coconut shell. The stretching

vibrations observed at 1640 cm^{-1} show the existence of a C=O group derived from carboxylic compounds (acetic acid) and ketones. Additionally, a peak at 1275 cm^{-1} with a bending vibration suggests the existence of an alcohol (C–O) group. These observations indicate that smoke condensate contains chemical substances that may act as inhibitors for MgCO_3 scale formation. Additionally, absorption peaks with their stretching vibrations at 2117 and 2363 cm^{-1} suggest the existence of a C=C bond in alkene.

In this experiment, the most effective inhibitor was a blend of G and L with a proportional concentration of 5:9. The GL inhibitor was too analyzed in more detail utilizing an infrared spectrometer, as illustrated in Figure 2(c). The investigation identified an absorption peak for the O–H group at 3340 cm^{-1} , the C=O group at 1640 cm^{-1} , and the C=C group at 2117 and 2363 cm^{-1} . Additionally, a peak at 1275 cm^{-1} with bending vibrations presents the existence of alcohol groups. These characterization results demonstrate the

presence of active groups in the mixed GL inhibitor (5:9) that can effectively function as inhibitors for MgCO_3 scale formation.

3.3. Characterization of Inhibitor Concentrates by GC-MS

The objective of characterizing the gambier concentrate and grade 2 liquid smoke of coconut shell using GC-MS is to identify the chemical components present in these compounds according to the gains of the functional group investigation conducted with FT-IR. The GC-MS characterization results for the gambier concentrate and grade 2 liquid smoke of coconut shell are showed in Figures 3 and 4, serially. Figure 3 illustrates the separation of chemical components obtained from the characterization of the gambier concentrate, as evidenced by the chromatogram peaks observed in the GC. These peaks are detected within the retention time range of 2.86 to 14.65 min. The chemical composition contained in the gambier concentrate as a result of GCMS analysis

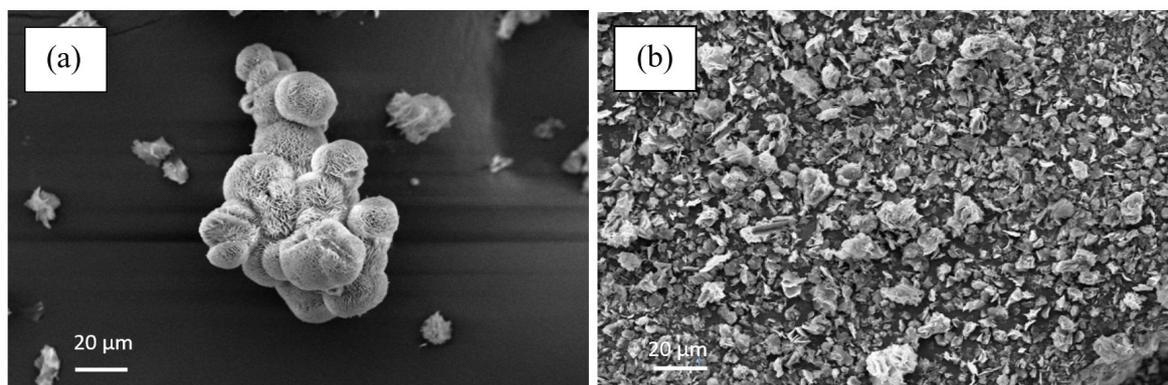


Figure 6. Morphology of MgCO_3 crystals (a) in the absence and (b) in the presence of GL inhibitors with a concentration ratio of 5:9 at magnification of $1000\times$.

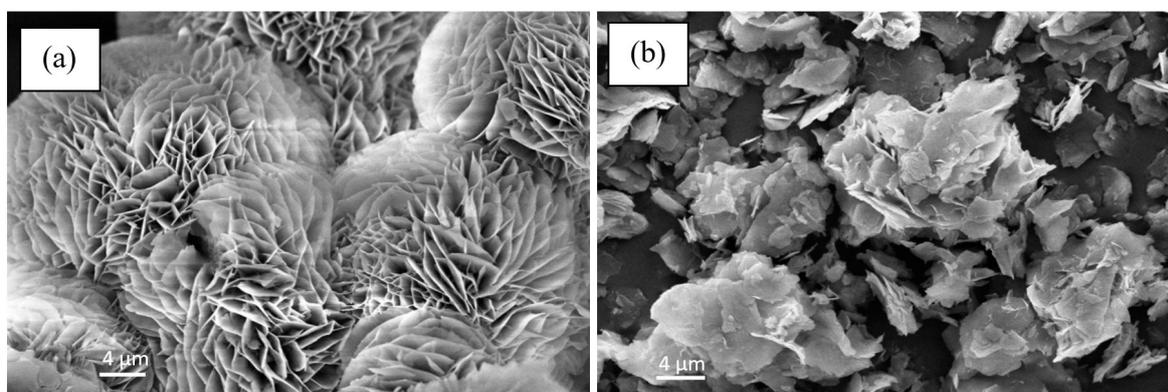


Figure 7. Morphology of MgCO_3 crystals (a) in the absence and (b) in the presence of GL inhibitors with a concentration ratio of 5:9 at magnification of $5000\times$.

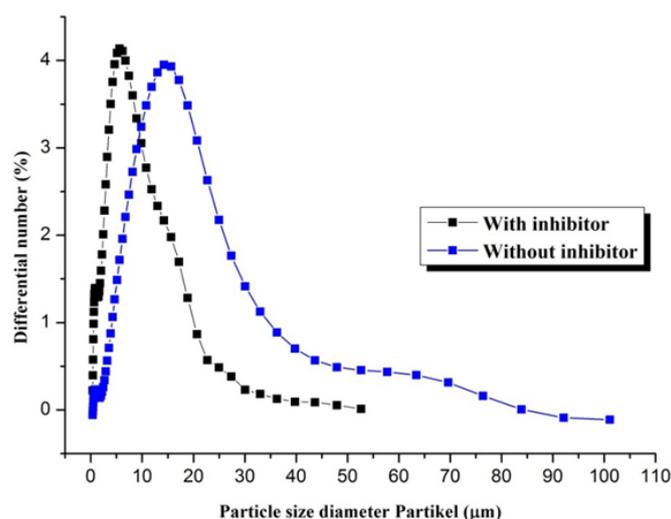


Figure 8. Distribution of particle sizes for MgCO_3 crystals in the absence and in the presence G:L Inhibitor with a concentration ratio (5:9) at a concentration of 0.05 M in the MgCO_3 growth solution.

can be seen in Table 2. From the GC-MS analysis results, 8 chemical compounds were obtained in the retention time range of 2.86 to 17.36 min. The compound with the largest percentage area (33.04%) was identified as 5-[(4-methylphenoxy)methyl]phenol. On the other hand, the compound with the smallest percentage area (1.20%) was (4-fluorophenyl)-(2-thienyl)methanol. The second-largest (15.83%) and third-largest (8.83%) percentage areas belonged to 1,3-benzendicarboxylic acid and benzothiophene, respectively. Percentage area provides a quantitative representation of how much a specific compound or component is present in the analyzed sample. The larger the percentage area, the more of that compound or component is present in the sample. Major components in the sample can be identified based on the percentage area of their peaks in the chromatogram. Higher or larger percentage areas indicate the presence of a compound in greater quantities. In the context of this research, the percentage area can provide an insight into how effective a compound or mixture is in inhibiting or influencing the amount of target compounds in the sample.

The major constituents found in the gambier concentrate include 5-[(4-methylphenoxy)methyl]phenol (33.04%), 1,3-benzendicarboxylic acid (15.83%), 7,7-bis(methylthio)-6-methyl-1-(2-thienyl)-2,4,6-heptatriene-1-one (15.04%), and benzothiophene (8.83%). The significant presence

of phenol content in the gambier concentrate, as indicated by the GC-MS characterization, suggests its potential as an inhibitor for MgCO_3 scale formation. These results align with the findings from the functional group identification by FT-IR, which identified the existence of the O-H group in compounds such as 5-[(4-methylphenoxy)methyl]phenol and (4-fluorophenyl)-(2-thienyl)methanol, the C=O group in compounds such as 1,3-benzendicarboxylic acid, pentanoic acid, and 2,6-dihydroxyacetophenone, and the C=C group in benzothiophene. Figure 4 showcases the separation of chemical components obtained from the analysis of grade 2 liquid smoke of coconut shell, as observed through the chromatogram peaks in the GC. These peaks are detected within the retention time range of 1.579 to 16.571 min. The results of the GC-MS analysis indicate that grade 2 coconut shell liquid smoke contains 8 chemicals. Table 3 presents that phenol (45.05%) has the largest percentage area, followed by 2,5-dimethylfuran (0.30%) with the smallest percentage area. Additionally, Methyl salicylic acid and butanoic acid are also present in grade 2 coconut shell liquid smoke with percentage areas of 1.22% and 0.31%, respectively.

The major constituents found in smoke condensate include phenol (45.05%), acetone (8.19%), 2-methoxyphenol (4.57%), and 3-furaldehyde (2.66%). The high content of certain functional groups, identified through GC-MS

analysis, suggests that smoke condensate may serve as an inhibitor for MgCO_3 scale forming. These results align with the findings from the functional group investigation by FT-IR, which identified the existence of the O–H group in phenolic compounds, 2,4-dimethoxyphenol, and 2-methoxyphenol, the C=O group in compounds such as acetone, 3-furaldehyde, propanoic acid, butanoic acid, and methyl salicylic acid, the C=C group in 2,5-dimethylfuran, and the aromatic C–H group in phenolic compound.

3.4. The Examination of Inhibitor Mixtures in Preventing the Formation of MgCO_3 Scale

The efficacy of the inhibitor blend, comprising G and L, in slowing down the growth rate of MgCO_3 scale was assessed through experiments involving various inhibitor concentrations in a MgCO_3 growth medium of 0.050 M employing the seed-free procedure and in the plastic bottles. The choice of using plastic bottles instead of glass to study the growth of MgCO_3 crystals because plastic bottles are generally chemically inert, meaning they are less likely to react with the substances inside them. This property is crucial in experiments where the goal is to study the crystal growth of MgCO_3 without interference from the container material. Figure 5 visually represents the outcomes of these experiments.

The conditions of temperature (105 °C) and time (3 h) for drying the precipitate in an oven (Thermo

Fisher Scientific, United Kingdom) after the growth solution experiment are selected to ensure the complete removal of water from the formed crystals. This step is crucial for obtaining accurate and consistent measurements of the weight of the crystals. The drying process at 105 °C is chosen because it is sufficiently high to facilitate the evaporation of water but is not so extreme as to cause decomposition or undesirable changes in the crystal structure. The 3-h duration allows for thorough drying, ensuring that the crystals reach a constant weight, and any remaining moisture is effectively removed. By employing these specific conditions, the experiment aims to achieve reliable and reproducible results in terms of crystal weight, which is essential for subsequent analyses such as SEM and PSA. Additionally, maintaining consistent and controlled conditions helps ensure the accuracy and comparability of the findings throughout the experiment.

Figure 5 illustrates that the growth of 0.050 M MgCO_3 scale was reduced when the mixed GL inhibitor was used at different concentration ratios. To determine the effectiveness of the mixed GL inhibitor, Eq. (1) and observational data were utilized, resulting in the calculations presented in Table 4. These findings align with prior studies indicating a direct correlation between inhibitor concentration and its inhibitory effect on MgCO_3 scale formation [31]. The most significant decrease was observed when with the GL inhibitor mixture at

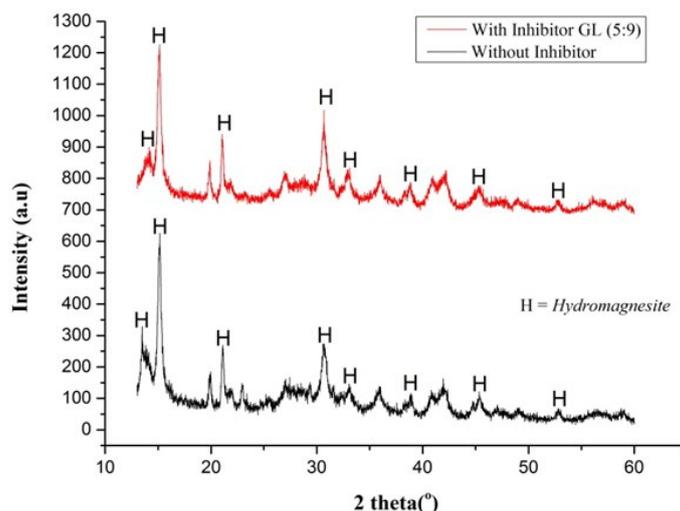


Figure 9. XRD graph of MgCO_3 crystals in the absence and in the presence G:L Inhibitor with a concentration ratio (5:9) at a concentration of 0.05 M in the MgCO_3 growth solution.

Table 5. Intensity of MgCO₃ diffraction pattern without inhibitor and with the addition of GL inhibitor (5:9).

Intensity of MgCO ₃ Diffraction Pattern			
Without Inhibitor		With Inhibitor GL (5 : 9)	
2θ (°)	Intensity (a.u)	2θ (°)	Intensity (a.u)
13.51	176	13.25	91
15.17	487	15.10	430
21.98	45	21.90	41
30.64	164	30.60	147
33.06	56	33.03	42
38.88	48	38.82	47
45.36	70	45.28	56
46.99	22	45.43	17
52.90	21	52.05	20

a proportion of 5:9, resulting in a 95.44% reduction in the MgCO₃ growth medium of 0.050 M. The surface morphology MgCO₃ crystals at 0.050 M was examined using SEM, and the results are portrayed in Figures 6 and 7, revealing the alterations in the surface structure of 0.050 M MgCO₃ crystals when treated with the mixed GL inhibitor (5:9). Prior to the addition of the GL inhibitors (5:9), MgCO₃ crystals exhibited plate-like structures that fused together, forming granules. However, after the addition of the GL inhibitors (5:9), MgCO₃ crystals transformed into thin and small sheets. These results corroborate previous studies indicating that magnesium carbonate crystals adopt a spherical shape composed of small plates fused together, resembling the arrangement of flower petals [1]. Hence, it can be inferred that the presence of GL induces notable changes in morphology of MgCO₃ crystals.

In order to confirm deeply the alteration in the size of MgCO₃ crystals with and without inhibitors of GL, PSA was undertaken on the gained MgCO₃ crystals, as shown in Figure 8. Figure 8 presents a comparing of the PSD of MgCO₃ particles at 0.05 M with and without the presence of the GL inhibitor, revealing a noticeable shift in the graph between the two cases. In the absence of inhibitors, the MgCO₃ crystals have a median particle size and an average particle size of 12.87 and 16.09 μm, respectively. When the GL mixtures (5:9) were introduced, the particle size of the MgCO₃ crystals

decreased, with a median and an average to be 5.38 and 8.04 μm, respectively. The PSD of MgCO₃ scale, both with and without inhibitors, varied based on the median value (7.49 μm) and the average value (8.05 μm). The PSD provides insights into the inhibitory effectiveness of inhibitors on crystal growth by comparing the growth of crystals in the presence and absence of inhibitors in the growth medium [1]. The change in the average and median measurements of the MgCO₃ scale by the presence of inhibitors is likely in consequence of the interaction between magnesium ions and the active compounds in the GL inhibitor substances, like C=O and O-H groups. This interaction influences the particle size, which correlates to the crystal size of the MgCO₃. Tannic acid acts as the compound in gambir concentrate responsible for suppressing the formation rate of MgCO₃ scales, while phenol and acetic acid play a role as compounds in smoke condensate suppressing the formation rate of MgCO₃ scales. The development solution was made by separately dissolving MgCl₂ and NaHCO₃ in GL inhibitor solutions. This suggests that Mg²⁺ ions engage with the inhibitors prior to interacting with CO₃²⁻ ions to shape MgCO₃ crystals. The Mg²⁺ ions bind to the active C=O and O-H groups derived from GL inhibitors.

The modifications in the intensity of diffraction peaks of MgCO₃ crystals indicate noticeable changes in the crystal shape when comparing samples treated with the mixed GL inhibitor (5:9) to

those without any inhibitors. This observation was made during XRD testing, as depicted in Figure 9. By analyzing the XRD data using the Match program, it was determined that the resulting magnesium carbonate compound took the form of hydromagnesite. A closer examination of Table 5 reveals a general reduction in intensity across multiple 2θ diffraction peaks when GL inhibitors (5:9) were introduced. This finding suggests that MgCO₃ crystals treated with inhibitors exhibited imperfect growth compared to crystals that did not have inhibitors added. Consequently, the GL inhibitors (5:9) contributed to the distortion and damage of MgCO₃ crystals.

3.5. Inhibition Mechanism

Crystal distortion is responsible for the observed morphological changes in MgCO₃ crystals, regardless of the presence or absence of inhibitors. Scale inhibitors play a crucial role in this distortion mechanism as they disrupt the arrangement reaction and hinder crystal growth, resulting in the formation of irregular and non-crystalline structures. The presence of inhibitor molecules at active crystal growth sites obstructs further growth, while the crystal lattice defects caused by distortions generate internal stresses, making the crystals brittle and fine. Consequently, these delicate crystals are easily dislodged from the surface due to the mechanical forces exerted by water [32]-[37].

Figure 10 illustrates the mechanism by which GL inhibitors impede the rate of MgCO₃ crystal growth. The diagram demonstrates how GL inhibitors adsorb onto the crystal growth sites of crystal seedlings, inhibiting crystal growth and leading to the formation of irregular crystal shapes. Gambier concentrate and coconut shell liquid smoke are rich in bioactive compounds containing various functional groups, such as O–H, C=O, and carboxylate (–COO[–]). These functional groups can directly interact at the molecular level with magnesium ions (Mg²⁺) and carbonate ions (CO₃^{2–}) in solution, making them effective in inhibiting the formation of MgCO₃ scale. The hydroxyl group contains a lone pair of electrons on the oxygen atom. This oxygen can form coordination bonds with Mg²⁺, resulting in a stable complex that reduces the amount of free Mg²⁺ ions in solution. This interaction hinders the nucleation and crystal growth of MgCO₃ through the reaction: Ar–OH + Mg²⁺ → Ar–O–Mg⁺.

The carbonyl group also has an electronegative oxygen that can bind to Mg²⁺. When present alongside hydroxyl groups, such molecules can form bidentate chelate complexes, increasing the stability of the interaction with Mg²⁺. Additionally, the carbonyl group may engage in polar interactions with carbonate ions. Carboxylic acids found in both liquid smoke and gambier concentrate ionize in solution into their –COO[–] form. These groups are

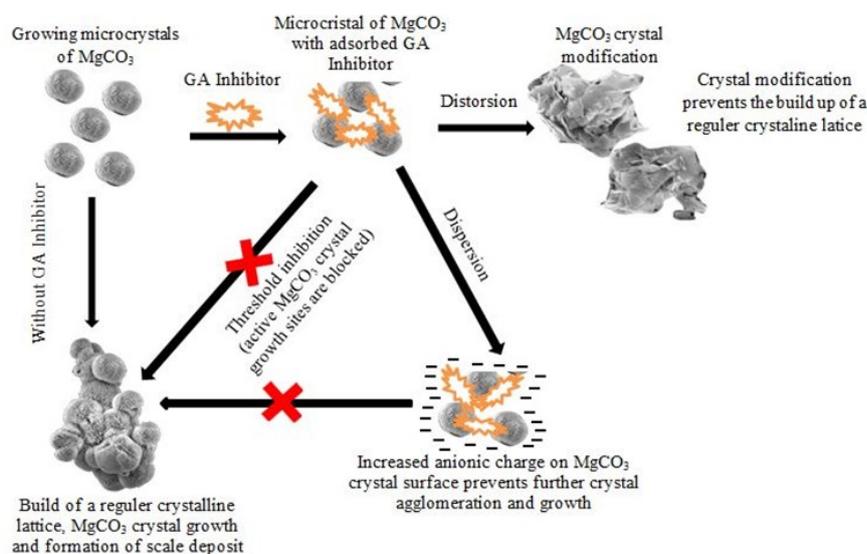


Figure 10. The mechanism of GL inhibitors in inhibiting the growth rate of MgCO₃ crystals in the growth solution.

highly effective at forming bidentate coordination bonds with Mg^{2+} , yielding very stable complexes. This bond formation blocks Mg^{2+} reactivity with CO_3^{2-} , thereby inhibiting precipitation via the reaction: $R-COO^- + Mg^{2+} \rightarrow [R-COO-Mg]^+$. In addition to binding with Mg^{2+} , hydroxyl and carboxylate groups can also form hydrogen bonds or ionic interactions with CO_3^{2-} . This may reduce the mobility or availability of carbonate ions in solution, lowering the rate of $MgCO_3$ formation. Beyond chemical binding, active molecules from gambier extract and liquid smoke can adsorb onto the surface of $MgCO_3$ crystals, preventing further growth through a blocking mechanism. This adsorption alters the morphology and size of the crystals, resulting in smaller, less sediment-prone particles. With the combined effects of these three functional groups, the mixture of gambier extract and coconut shell liquid smoke acts as a multifunctional inhibitor system that significantly reduces $MgCO_3$ precipitation through ionic complexation, crystal surface modification, and precipitation reaction inhibition.

3.6. Ecological Implications

The use of gambier (*Uncaria gambir* Roxb.) concentrate and coconut shell (*Cocos nucifera* L.) smoke condensate as eco-friendly inhibitors to prevent the formation of $MgCO_3$ scale has significant ecological implications, particularly in the context of sustainability, waste management, and resource availability. These two materials are abundant agricultural by-products in tropical regions, especially in Indonesia, and are often underutilized. Their use as raw materials for inhibitors supports the principle of waste valorization—transforming organic waste into value-added products—and contributes to reducing environmental burdens caused by biomass disposal [38][39]. Furthermore, the active compounds in gambier and coconut shell smoke condensate, such as tannins, flavonoids, acetic acid, and phenolic compounds, exhibit complexation abilities with Mg^{2+} ions, making them suitable replacements for synthetic chemical inhibitors that are toxic and non-biodegradable [40]. Utilizing renewable local resources in this way is not only cost-effective but also aligns with the principles of green chemistry and promotes the development of a more

sustainable chemical industry.

3.7. Industrial Application Challenges and Comparison with Commercial Inhibitors

The application of a mixture of gambier extract and coconut shell liquid smoke as an inhibitor of $MgCO_3$ scale on an industrial scale faces several challenges that must be seriously considered. One of the main issues is the consistency of the active compound composition. As natural products, the bioactive content in gambier and liquid smoke is highly influenced by geographical factors, harvest season, and extraction methods, which may lead to fluctuations in inhibition effectiveness if not properly standardized [39]. In addition, the availability and logistics of raw materials in large quantities pose another challenge, especially in ensuring a sustainable supply for industries operating continuously [38]. The scalability of the production process is also a crucial issue since traditional extraction methods may not be efficient or economical when applied to large-scale operations and often require specialized processing infrastructure [41]. On the other hand, the thermal and chemical stability of active compounds needs to be further tested, as industrial conditions such as high temperatures and variable pH can accelerate the degradation of natural organic compounds, thus reducing their effectiveness [42]. From a regulatory perspective, the use of natural materials as industrial additives requires toxicological testing and certification to meet current safety and environmental standards [40].

Compared to commercial inhibitors such as phosphonates and synthetic polymers, the mixture of gambier extract and coconut shell liquid smoke demonstrates competitive inhibition effectiveness (90–95% in laboratory tests), although it remains slightly below the maximum efficiency levels of commercial products, which often exceed 95% [43]. The main advantages of these natural inhibitors lie in their sustainable raw material sources, lower local production costs, and environmentally friendly biodegradable properties. In contrast, synthetic inhibitors, while more stable and easier to standardize, are often derived from non-renewable chemical sources and may pose long-term ecological risks [40]. Therefore, although the gambier and liquid smoke mixture has significant

potential as a green alternative, further development is needed to overcome challenges related to stability, formulation, and standardization to compete with established chemical inhibitors in industrial applications.

4. CONCLUSIONS

The major compounds found in the gambier concentrate in this study, based on GC-MS analysis, are 5-[(4-methylphenoxy)methyl]phenol (33.84%), 1,3-benzenedicarboxylic acid (15.83%), 7,7-bis(methylthio)-6-methyl-1-(2-thienyl)-2,4,6-heptatrien-1-one (15.04%), and benzothiophene (8.83%). Meanwhile, the major compounds found in grade 2 coconut shell smoke condensate are phenol (45.05%), acetone (8.19%), 2-methoxyphenol (4.57%), and 3-furfural (2.66%). The combination of these two natural materials plays a significant role as a green inhibitor in preventing scale growth of MgCO_3 scale formation, presenting a viable alternative as green inhibitor for inhibiting. The mixture of gambier concentrate and grade 2 coconut shell smoke condensate at a mixing ratio (5:9) as an inhibitor of MgCO_3 deposit build-up demonstrated the optimal efficiency, reaching 95.44% in a 0.050 M MgCO_3 growth solution applying the seed-free approach. Analysis using PSA indicated that the PSD of MgCO_3 crystals decreased after the addition of the GL (5:9) inhibitor, as observed from the average and median particle size of the MgCO_3 scale. Observations using SEM revealed significant changes between MgCO_3 crystals in the absence and the presence of GL mixtures. The presence of the GL mixtures can change the morphology of MgCO_3 crystals. The XRD analysis showed a decrease in intensity at several 2θ diffraction peaks, indicating damage to the MgCO_3 crystal structure.

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Conflicts of Interest

The authors declare no conflict of interest.

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