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**BIBLIOMETRIC ANALYSIS, SYSTEMATIC REVIEW AND  
EXPERIMENTAL STUDY OF SURFACE AND MATERIAL EFFECT IN  
CALCIUM CARBONATE INCRUSTATION IN OFFSHORE OIL  
EXPLORATION**

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**Abstract.** *Calcium carbonate scaling is a problem that leads to many losses in off-shore oil exploration, as it reduces the efficiency of the production process and increases operating costs. Several variables that influence the fouling process, such as temperature, pressure, chemical composition, surface finish, surface energy, among others. In this work, a bibliometric analysis, a systematic study and an experimental study of the main factors related to the influence of the surface on calcium carbide incrustation has carried out. The bibliometric analysis and systematic review study showed that surface energy and roughness exert a strong influence on the amount of encrusted material. In the experimental study, it was been observed that for ABNT 1020 steel, higher roughness values lead to higher incrustated mass values and greater corrosion.*

**Keywords:** *Off-Shore Petroleum, Fouling, Scale, Calcium Carbonate, Surface.*

## 1. INTRODUCTION

Among the problems faced in off-shore oil extraction, mineral fouling is one of the phenomena of great relevance, as it reduces the efficiency of the production process and increases operating costs (Lai et al., 2020).

Among the types of incrustation, calcium carbonate is one of the main problems in oil exploration, occurring mainly in places where there is pressure drop, such as valves and reductions, rendering the component unusable (Kohler et al., 2001; Kamal et al., 2018).

For a petroleum environment, salts dissolved in aqueous solution are always been expected, with concentrations ranging from 10,000 to 400,000 mg/l. Therefore, in offshore oil exploration, fouling becomes one of the main difficulties faced (Cosmo, 2013).

When analyzing the water present in Petrobras extraction wells, it was observed that due to the chemical composition, the main incrustation is calcium carbide ( $CaCO_3$ ), thus, understanding the factors that influence the formation, growth and Crystal precipitation is of great technical and economic importance (Cosmo, 2013).

According to Macadam and Parsons (2004), the fouling phenomenon causes high losses in several countries, for example, for the United States it cost 8,000-10,000 million dollars in 2004, it is estimated that the costs for the world in this same year were around 26,850 million dollars. The losses associated with fouling reach, on a global scale, the order of 1.4 billion dollars per year (Frenier and Ziauddin, 2008).

The fouling phenomenon can be divided into stages (Figure 1). For incrustation to occur, it is necessary that there are mineral ions present in solution, once present, these ions can react to form salt (such as calcium carbonate  $CaCO_3$ ). Under suitable thermodynamic conditions, the salts formed can nucleate, grow and later agglomerate. Finally, this particulate may precipitate and adhere to the pipe surface depending on the flow regime, surface finish, surface tension and material (Cosmo, 2013; Al-gailani; 2021b). When it comes to avoiding and/or mitigating the problem, action can be taken in any of the phases of the process.

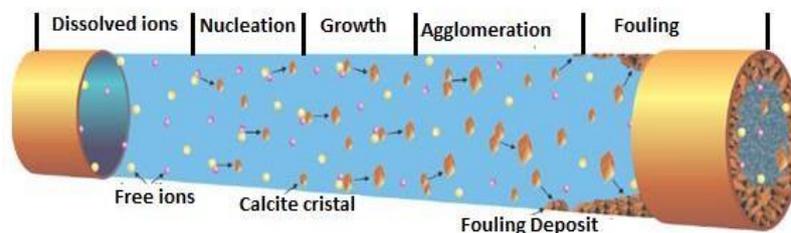


Figure 1: Steps of the saline fouling process. Source: Cosmo (2013).

The final step of encrustation has a strong influence on surface factors such as contact angle, surface free energy, surface finish, material and flow regime (Charpentier, 2014; Hamid et al., 2016 ;Cheong et al. 2008 ; Sun et al., 2019). Given the economic importance in the oil production process, increasing efficiency and mitigating losses becomes a key factor for the use of the resource in the future.

In this work, it is proposed to carry out a bibliometric study and a systematic review to investigate the influence of surface parameters on the calcium carbonate fouling process, to identify and discuss how surface variables influence the fouling process. This work aim to provide to the reader a consistent database, discuss which are the main countries working with the theme, main institutions and provide a synthesis of the main works produced in the last 10 years.

The experimental part of the work investigates the effect of the surface finish of ABNT 1020 carbon steel on calcium carbonate inlay in a static solution.

## 2. MATERIALS AND METHODS

### 2.1 Methodology of bibliometric analysis

The research methodology has based on the works of Da Conceição Moreira (2020), where the VOSviewer software has selected for bibliometric analysis. The processes consist of selecting a research base, refining it according to the desired scope, selecting the most relevant articles, performing a statistical analysis, and finally carrying out an analysis of the content of the works, discussing and comparing the results (Figure 2).



Figure 2: Flowchart of the methodology used in bibliometric. Source: Moreira (2020).

For the elaboration of the systematic bibliometric study on the surface effect on the calcium carbonate fouling rate in offshore oil exploration applications, the Web Of Science database was used. The search keywords were Off-Shore Petroleum; Fouling; Scale; Cabonate Calcium and Surface.

The search was restricted to the last 10 years and only articles were been analyzed. Where 730 articles were been found. When the titles and abstracts were been analyzed, it was observed that only 31 articles were close to the scope of the study. The selected works were been statistically analyzed through VOSviewer, where the origin of the works, relationships between studies and countries were observed.

Finally, the analysis of the 31 articles selected by the bibliometric analysis was carried out and their results were compared and discussed, 3 articles were judged with content unrelated to the problem investigated, and for reasons synthesis are not presented in the discussion.

### 2.2 Experimental methodology

After the bibliometric analyses and systematic study, an experimental study has carried out to evaluate the effect of surface finish on calcium carbide inlay. The experiment a static solution condition was choose in order to eliminate the effects of flow. The material used was ABTN 1020 carbon steel, with dimensions of 50.5mmx12.5mmx7.3mm. To vary the surface finish, a PLF model Fort Polisher was been used, with 80, 320, 600 and 1200 sandpaper. Four specimens with different finishes were tested. The surface finish was been measured using a Mitutoyo SJ-210 rugosimeter,

The tests were been performed using a solution of 4.16g of calcium chloride in 500ml of demineralized water, and 5.3g of sodium bicarbonate in 500ml of demineralized water. The specimens were been weighed before the encrustation process, after one week in solution and after two weeks in solution. For mass measurement, the Shimadzu AUY220 analytical balance was been used.

### 3. RESULTS AND DISCUSSION

#### 3.1 Bibliometric results and discussion

Analyzing the data from the Web of Science website, an increasing number of publications related to calcium carbide inlay was observed, comparing the number of publications from 2012 to 2022, it is observed that the number of works increased by more than 100% (Figure 3), revealing a growing interest in the topic. The growing interest is due to its various technical applications, such as offshore oil exploration, application in desalination, water heaters, seawater desalination and heat exchangers, among others. It should be noted that the year 2022 had not yet ended at the time of the bibliometric research (March 2022), so there are fewer published works.

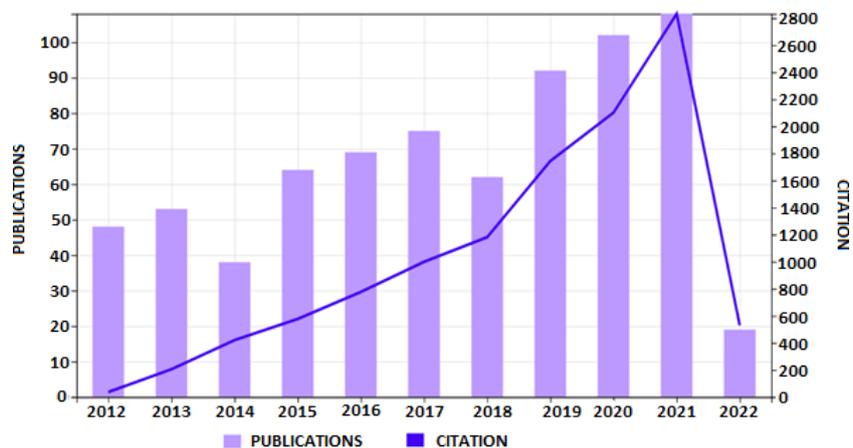


Figure 3: Evolution of publications and citations per year. Source: Web of science (2022).

After selecting the 31 articles most related, the data were been processed with VOSviewer, revealing the links between authors, the authors with the largest publications being highlighted (Figure 4). It is possible to observe with the color scale the authors who published the most recently, being the yellow coloring for more recent publications and blue for the publications closer to 2012.

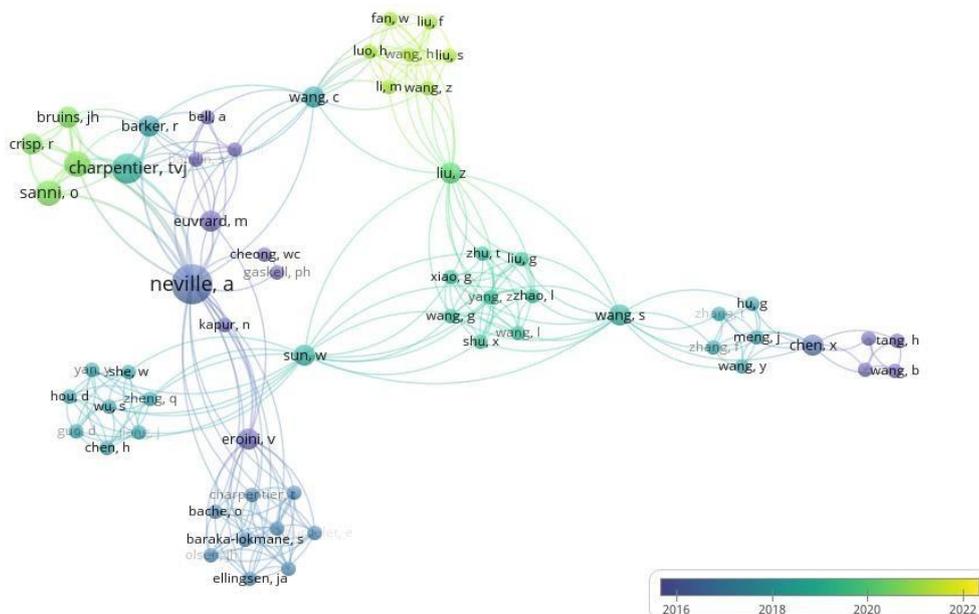


Figure 4: Authors with the highest number of publications, links and period of activity.

Table 1 presents data from the main countries where there is technical-scientific interest in the subject, it is observed that England appears in first place with 7 publications out of 31, with 112 citations and a strength of links of 6, followed by France with 5 publications and 115 citations.

Table 1: Countries with the highest number of publications and citations.

Selected	Country	Documents	Citations	Total link strength
<input checked="" type="checkbox"/>	england	7	112	6
<input checked="" type="checkbox"/>	france	5	115	6
<input checked="" type="checkbox"/>	netherlands	2	6	2
<input checked="" type="checkbox"/>	norway	1	13	2
<input checked="" type="checkbox"/>	scotland	1	48	2
<input checked="" type="checkbox"/>	usa	2	47	2
<input checked="" type="checkbox"/>	algeria	1	14	1
<input checked="" type="checkbox"/>	brazil	1	0	1
<input checked="" type="checkbox"/>	canada	1	30	1
<input checked="" type="checkbox"/>	chile	1	0	1
<input checked="" type="checkbox"/>	finland	1	30	1
<input checked="" type="checkbox"/>	india	2	16	1
<input checked="" type="checkbox"/>	mexico	1	0	1
<input checked="" type="checkbox"/>	peoples r china	11	201	1
<input checked="" type="checkbox"/>	germany	3	18	0
<input checked="" type="checkbox"/>	iran	2	24	0
<input checked="" type="checkbox"/>	japan	1	3	0
<input checked="" type="checkbox"/>	spain	1	0	0

After the selection, statistical analysis of the articles, the content analysis has carried out, where the main results were summarized in Table 2 and 28 of the 31 were selected for presentation of the synthesis.

Table 2: Summary of the analyzed works

Author	Work Brief
Contreras et al. (2022)	Use a chamber term to evaluate the effect of deposition and fouling in a pumping system with the carbon test for pumping. The steel used was 304L stainless steel with a relative roughness of 0.0008327949 Å. Observe two distinct mechanisms; agglomeration, a process dominated by gravity and encrustation, a phenomenon influenced by diffusion. Through the heterogeneous nucleation of the crystals, they nucleate on the pipe surface in the allotropic forms of calcite and aragonite.
Palanisamy et al. (2020)	They evaluated the variation in fouling conditions on copper and aluminum surfaces both under the effect of the fouling inhibitor DTPA. No morphological differences were observed in calcite crystals due to surface change under the same conditions.
Ahmadi et al. (2018)	They used carbonate nanoparticles as a way to reduce surface energy at the oil and solid interface.
Pääkkönen et al. (2015)	They carried out a study on the rate of fouling on a heated stainless steel surface (316L). The encrusted mass was measured by varying the thermal resistance of the surface. In the study, comparisons were made between models predicted in the literature and experimental measurements, it was observed that the incrustation process is strongly dependent on hydrodynamic conditions and surface finish. Greater roughness provides a greater surface for heterogeneous nucleation in addition to a greater thermal exchange, heating the fluid and thus favoring the encrustation process.
Al-gailani et al. (2020).	They evaluated the effect of ionic constituents on calcium carbonate fouling on a heated aluminum surface. It was observed that ions $SO_4^{2-}$ and carbon in solution act by reducing the fouling rate, while $Na^+$ e $Cl^-$ increasing the fouling rate.
Zhao et al. (2019)	They used ICVD polymeric surface coatings as a way to inhibit the fouling process. By reducing the surface energy through the polymeric coating, a smaller amount of calcium carbonate mass adhered was observed. Experimental results suggest that surface energy is a fundamental variable when estimating the effects of fouling.
Schilling et al. (2021)	They carried out a comparative study between the calcium carbonate fouling rate on a graphite-doped polypropylene polymeric surface with duplex and 316L stainless steel substrate, the tests were conducted hot. The study suggests that polymeric surfaces have a lower rate of fouling due to the characteristic of lower roughness and lower surface energy, in addition, the lower thermal conductivity of the polymeric material led to lower surface heating, which disfavored the precipitation and crystallization of carbonates. of calcium that exhibit solubility inversely proportional to temperature.

Stark et al. (2015)	They evaluated calcium carbonate scale in pipes used for seawater desalination. The materials used were: aluminum alloys, brass and stainless steel. It was observed that higher surface energies favor the carbonaceous encrustation process.
Keogh et al. (2017)	Using surface coatings to reduce mineral fouling, it was observed that hydrophobic surfaces of nano fluorine polymers have the ability to drastically reduce the fouling rate due to the reduction of surface roughness and energy.
Dreiser et al. (2015)	They carried out a comparative study between the calcium carbonate fouling rate on a polymeric surface of PEEK 1000 and stainless steel. It was observed that the incrustation in the polymeric material was considerably smaller than in the stainless steel, one of the reasons attributed by the authors was the difference between the surface finishes, which had a roughness of 0.028 $\mu\text{m}$ in the polymer and 0.121 $\mu\text{m}$ in the stainless steel.
AL-Gailani et al.(2021a)	They promoted a study applied to the precipitation of calcium carbonate on metallic surfaces. It was observed that the chemical nature of the substrate and the surface finish considerably affect the amount of precipitated mass.
AL-Gailani et al.(2021b)	They studied the effect of temperature, roughness and pressure on the calcium carbide embedded mass on an aluminum surface. It was observed that due to the reverse solubility of calcium carbide, higher temperatures favor the incrustation process. The variable pressure acts on the solubility of $CO_2$ , when the pressure is reduced, carbon dioxide is released from the liquid solution, favoring precipitation. Finally, higher roughness values promote a greater number of heterogeneous nucleation sites, favoring the fouling process.
Belarbi et al. (2016)	They evaluated the influence of Paronychia Argentea type fouling inhibitors using the rotating electrode technique. It was observed that the inhibitor acts by modifying the morphology of calcium carbide crystals, reducing the surface energy and thus hindering the encrustation process.
Morita and Umezawa (2016)	Assessed the morphology of calcium carbonate crystals formed on the inner surface of a carbon steel pipe used to transport hot water. Corrosion has been observed to help accentuate the fouling process.
Charpentier et al. (2015)	They used polymeric surface coatings to reduce calcium carbide scale. The coatings used were PTFE, PPY, Fluorinert FC-70 and BMIm, which were compared with a 316L stainless steel surface. The results showed that the polymeric coatings were effective in reducing the calcium carbide encrusted mass.
Cheong et al. (2013)	They selected 10 surfaces to evaluate the surface effect on fouling rate using the rotating electrode technique. It was observed that the surface that exhibited a lower rate of fouling was the one that had the lowest surface energy. The authors suggest that the contact angle and surface energy have a notable effect on the fouling rate, being able to be dominant in relation to the roughness in certain applications.
Li et al. (2014)	They evaluated the effect of the surface on the precipitation of calcium carbide. In the study, two minerals were used as substrate, Quartz and Mica. The results showed that the chemical nature of the substrate changes the surface energy, causing differences in precipitation on different surfaces analyzed. It was observed that the Veretita polymorphic phase was favored in both substrates while calcite was less favorable. Mica substrate favored calcium carbide precipitation in both polymorphic phases compared to Quartz.
Zhu et al. (2019)	They analyzed the effect of surface corrosion on calcium carbide fouling rate. Ferrous ions play important roles in $CaCO_3$ polymorphism, regions of localized corrosion favored the Aragonite phase, which was deposited in cathodic microregions. It can be inferred that greater corrosion favors the calcium carbide fouling rate, providing a greater number of sites for heterogeneous nucleation.
Sun et al. (2019)	They investigated the influence of roughness, surface energy, material and surface angle on calcium carbonate fouling rate. Six metallic materials were used, namely: 316L stainless steel, 13Cr stainless steel, aluminum, copper and galvanized steel. In the study, it was observed that the roughness is an insufficient parameter for surface characterization, as there may be peaks and small dimensions not detected in the roughness measurement, which contribute significantly to the fouling. It was observed that higher surface energies showed higher values of encrusted mass. The difference between the materials altered the morphology of the nucleated crystals on the surface.
Zhang et al. (2018)	They used nano-structured hydrophobic surface coating in capillary format as a way to avoid calcium carbide incrustation adhesion. Tests showed a significant reduction (about 90%) in surface fouling.
Hoseinieh et al. (2017)	They investigated the influence of sea water on the morphology of precipitated calcium carbide crystals on the surface of 316L stainless steel. The variation of the ion alters the precipitated phase leading to different adhesions of the encrusted crystals. The reduction of the calcium ion disfavors the Aragonite phase.
Wang et al. (2021)	They proposed a nanostructured surface coating as a way to reduce the calcium carbide fouling rate and prevent corrosion of the metallic substrate. The tests showed that the coating was efficient in reducing the encrusted mass.
Jiang et al. (2018)	They evaluated the influence of ions in solution on the morphology of calcium carbonate crystals. It was observed that the different substrates provide different activation energies, which influence

	the morphology of the solid formed. It was also found that the morphology of the calcite phase is controlled by the amount of $Ca^{2+}$ and $CO_3^{2-}$ ions.
Eroni et al. (2013)	They evaluated the effect of temperature on calcium carbonate precipitation. To measure the precipitated mass, the variation of water turbidity was associated with precipitation of calcium carbonate. It was observed that higher temperatures favor the chemical kinetics of the formation of calcium carbonate crystals as well as causing a greater mass encrusted on the surface.
Jiang et al. (2015)	They designed hygroscopic and hydrophobic nanostructured surfaces in order to evaluate the resistance to calcium carbide incrustation. The tests were carried out with a supersaturated solution under static conditions at a temperature of 90°C. The hygroscopic surface had a low contact angle and high surface energy (4.5° and 74.8mJ/m <sup>2</sup> ), while the hydrophobic surface had a high contact angle and low surface energy (154° and 0.2mJ/m <sup>2</sup> ). The results revealed a greater tendency for nucleation and growth of calcium carbonate crystals on the higher energy surface. The morphology of carbonate crystals and the growth mechanism was also drastically changed with the change of substrate.
Zhang et al. (2015)	They investigated the polymorphic variation of calcium carbide in aqueous solution when alcohol and PSS polymer were added. The results show that the dominant phase became Varette, morphological structure in the form of spheres, as a consequence, the new phase has mechanical resistance and surface adhesion inferior to calcite crystals. Polymorphic changes alter the bond strength between the inlay and the surface.
Palanisamy e Subramanian (2016)	They analyzed the effect on calcium carbide scale when using EDTA acid in aqueous solution. The results showed that the morphology changes, making the adhesion of the crystals to the copper metallic surface weaker.

According to the analyzed studies, the variables surface energy, roughness, temperature and partial pressure have a considerable impact on the calcium carbonate fouling rate (Table 3). The table lists the variables, with the change in intensity, with two levels, low and high levels of fouling, in the column on the left provides the related authors.

Regarding the deposition process, later nucleation and crystal growth, the surface energy plays a fundamental role, low surface energies provide weakly bound crystals, which can later be removed by the shear stress imposed by the flow, high Surface energies provide strongly adhered crystals making it difficult to remove the crystals by the fluid flow.

The roughness variable must be analyzed with caution, some authors consider it insufficient to describe the encrustation process (Sun et al., 2019), because at levels smaller than microscopic, a surface with less roughness can provide a more irregular surface than a surface. surface considered rough. In general, the rougher the surface, the greater the number of sites for heterogeneous nucleation, the greater the adhesion of crystals, thus, higher levels of fouling.

The temperature variable influences the rate of precipitation of calcium carbide in solution, the higher the temperature, the lower the solubility of the carbide, providing a greater amount of precipitated material, where it will nuclear and grow on the surface of analysis. The partial pressure of  $CO_2$ . influences the solubility of calcium carbide, lower pressure, leads to lower solubility, providing greater amount of particulate available for encrustation. The study by Cosmo (2013) provides more details about the variables that affect carbide precipitation.

It should be noted that partial pressure and temperature are not surface parameters, but variables that influence the amount of carbide that will precipitate, which will be necessary for fouling to occur, the main variables related to the surface are surface energy, roughness and material (Jiang et al., 2015 ; Li et al., 2014 ; Cheong et al. 2013 ; Charpentier et al., 2015).

Table 3: Variables that influence the calcium carbonate scaling process.

Variable	Intensity	Scale	Recommended authors
Surface energy	low	low	Jiang <i>et al.</i> (2015) ; Jiang <i>et al.</i> (2018); Li et al. (2014) ; Cheong et al. (2013) ; Charpentier et al. (2015) ;
Surface energy	high	high	Belarbi et al. (2016); AL-Gailani et al.(2021) ; Stark et al. (2015); Schilling et al. (2021)
Roughness	high	high	Contreras et al. (2022); Pääkkönen et al. (2015) ; Keogh et al. (2017); Dreiser et al. (2015);
Roughness	low	low	Charpentier et al. (2015) ; Zhu et al. (2019); Sun et al. (2019); AL-Gailani et al.(2021)
Temperature	high	high	AL-Gailani et al.(2021) ; Jiang et al. (2018); Eroni et al. (2013); Jiang et al. (2015) ; AL-Gailani et al.(2021)
Temperature	low	low	
Pressure CO2	high	low	(COSMOS, 2013); AL-Gailani et al.(2021)
Pressure CO2	low	high	

Synthesizing the analyzed results, the higher the surface energy, the higher fouling rates will be expected. Rough surfaces provide a greater area for heterogeneous nucleation to occur, thus, a higher fouling rate. Calcium carbonate has reverse solubility with temperature, that is, higher temperatures have lower solubility values, which provides higher rates of calcium carbonate deposition, which may eventually become encrusted depending on surface conditions. Reducing the partial pressure of  $CO_2$  in solution provides greater precipitation of calcium carbonate particles.

If the fluid is not static, the effect of flow can provide sufficient shear stress to remove the nucleated crystals from the surface, so higher values of velocity provided lower rates of surface fouling (Hamid et al., 2016).

Graham (2014) and Macadam et al. (2004) present studies regarding the effect of surface finish on fouling rate. Both works were not used in the bibliometric analysis, Macadam et al. (2004) for not being in the analyzed period and Graham (2014) for not being part of the Web of Science database. The authors concluded in their investigations that higher roughness favors the calcium carbonate fouling process and that the substrate material will influence the fouling rate.

### 3.1 Experimental Results and Discussion

The specimens produced presented the average roughness according to Table 4. The specimen 1 was finished with 80 sandpaper, the second was finished with 320 sandpaper, the third with 800 sandpaper and finally the fourth with less roughness was finished with sandpaper 1200.

Table 4: Roughness of the selected specimens.

	Roughness Ra in upright position	Roughness Ra in horizontal position
Specimen 1	$(0,921 \pm 0,001) \mu\text{m}$	$(0,352 \pm 0,001) \mu\text{m}$
Specimen 2	$(0,071 \pm 0,001) \mu\text{m}$	$(0,063 \pm 0,001) \mu\text{m}$
Specimen 3	$(0,039 \pm 0,001) \mu\text{m}$	$(0,030 \pm 0,001) \mu\text{m}$
Specimen 4	$(0,023 \pm 0,001) \mu\text{m}$	$(0,023 \pm 0,001) \mu\text{m}$

In the first week, it was observed that the specimens lost mass, this fact is due to the corrosion that the carbon steel suffered in contact with the solution. It was observed that the bodies with higher roughness values lost a greater amount of mass than the specimens of lower roughness. This fact is due to the greater surface area, providing a greater region for uniform surface corrosion to occur thus leading to greater loss of mass (Figure 5). This result is in agreement with Morita and Umezawa (2016) and Zhu et al. (2019).

After two weeks in the encrusted mass gain solution, a mass gain was observed at the initial mass of specimens. It was observed that the mass gain was greater in specimens with higher surface roughness values (Figure 5), a result in accordance with the studies by heterogeneous Pääkkönen et al. (2015), Schilling et al. (2021) and AL-Gailani et al. (2021b). Higher values of surface roughness provide a greater surface area favoring the heterogeneous nucleation of calcium carbonate crystals.

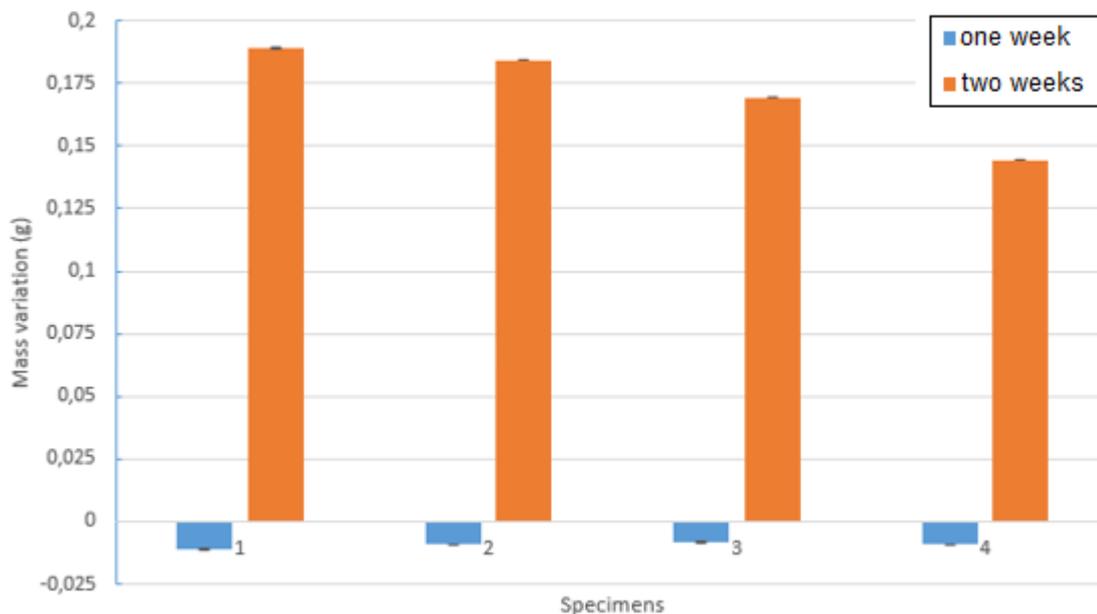


Figure 5: Mass variation in one week and two weeks in static solution.

The morphology of the formed carbide was not distinguished from the oxides present and on the surfaces, Zhu et al. (2019) discusses that the corrosion process and the ferrous ions present are fundamental for calcium carbide polymorphism. In this study, the evaluation of the phases present on the surface and the percentage of each one was not carried out, only the adhered mass was evaluated.

## 4. CONCLUSION

### 4.1 Bibliometric research conclusions

Bibliometric research is a useful tool for engineering studies, providing a fast, efficient and organized learning methodology, a consistent database, knowledge of which countries are interested in the topic, identifies which are the main researchers in the area.

In the study, it was possible to identify and investigate surface variables that affect the fouling process, they are: surface energy, roughness and material. As discussed, higher surface energy provides higher fouling rate, higher roughness values are generally associated with greater surface area which would provide greater interface for heterogeneous nucleation, consequently greater fouling. Different materials provide different types of chemical bonds between calcium carbide and the surface, which will influence the fouling rate.

However, for fouling to occur, it is necessary to supply particulate material, this supply is favored by the reduction of the partial pressure of  $CO_2$ . and temperature rise. It should be noted that the flow, as well as the presence of other solution ions, may hinder or facilitate the process of nucleation and crystal growth.

### 4.2 Experimental study conclusions

It was observed that the surface finish influenced the incrustation of calcium carbide in static solution, it was observed that higher values of roughness led to higher values of incrustation mass and greater corrosion, on the other hand, lower values of roughness led to lower values of encrusted mass. and less corrosion.

As a recommendation for future work, it is recommended to analyze the phases formed on the surface, carry out the tests on a fluid dynamic bench to evaluate the effect of flow and evaluate other metallic surfaces.

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