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Effect of Rock on Aquathermolysis Reactions at Laboratory Scale (A Review)

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Abstract—Enhanced recovery methods are an important stage in the oilfield development and exploitation program. They allow generating an increase in production related to the decrease of the remaining oil in the reservoir, which was not possible to remove in previous production schemes. Among them, steam injection is one of the methods used, whose main objective is to reduce the viscosity of the crude oil. In the literature there are researches where is evident an interaction between the steam and the crude oil in the reservoir, giving way to the occurrence of chemical reactions called Aquathermolysis. This transformation is a chemical result that occurs at temperatures between 200 to 325°C typical for steam injection. However, the investigations have been focused more on the fluid-fluid interaction than the rock-fluid synergy. The present work aims to better understand the synergy generated in the fluid-rock interaction through a systematic review of the research found in the literature associated with the use of rock fragments, minerals, or porous media in steam injection conditions. For analyzing the data, a descriptive bibliometric study was made with the selected studies where a rock sample was used. As a result, the addition of the mineral and rocks over the reactions generates a catalytic effect observed in the physical and chemical crude oil properties changes. This additional effect is generated for the presence of some minerals in the rock sample and this behavior could change according to its composition. Also, the gas production and its variation under different operational parameters are evidence of rock presence benefits over the process.

Keywords: systematic review, aquathermolysis, enhanced oil recovery, laboratory test, fluid-rock interaction

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INTRODUCTION

The oil industry today has awakened an interest in strengthening the development of technologies that allow the production of heavy and extra-heavy crudes [1–3]. This behavior is a result of the decrease in light hydrocarbon reserves and an increase in the difficulty of crude oil extraction in the world's fields [4]. At the international level, the outlook of reserves distribution indicates that 70% of its resources are of unconventional type, where 25% corresponds to heavy crudes and the remaining 45% is related to extra heavy crudes and bitumen [5]. This is why thermal recovery as a method of injecting hot fluids into the formation has been a widely applied technique in heavy crude oil fields around the world [6–9].

One of the most representative processes is steam injection, in which water is injected into the formation at saturated steam conditions under different modalities: cyclic, continuous, SAGD, among others. During its implementation, an interaction is generated between the injected water and the hydrocarbon located in the reservoir. This phenomenon occurs in conditions of high temperatures between 200 and 325°C, giving rise to a chemical process which groups a series of reactions called aquathermolysis [10]. The reactions generate a transformation of the oil, from the energy supplied by steam and the effect of the mineralogy present in the reservoir, originating a series of products in which chemical compounds such as methane (CH_4), hydrogen sulfide (H_2S), carbon dioxide (CO_2), carbon monoxide

(CO), hydrogen (H_2) and hydrocarbons of lower molecular weight (HCS) stand out [11].

Several authors have studied the addition of chemical agents to the process with the aim of reducing the high energy requirements, making possible the occurrence, acceleration, and improvement of the reaction. This is why catalytic aquathermolysis arises as a technique in which a catalyst is added to a steam injection process generating a lower energy requirement to obtain its products in a shorter time [12]. In turn, additional effects associated with the reaction such as pyrolysis, isomerization, ring opening, oxygenation, alcoholization, esterification and depolymerization are manifested during the process [13]. As a consequence, the transformed hydrocarbons maintain the changes in their properties permanently, which is evidenced in the variation of physicochemical properties such as viscosity, API gravity and structural change in the crude oil with respect to its original conditions [14–16].

Hydrocarbon fields in the world present structural and mineralogical differences in their reservoirs, as well as compositional variations in the crude oil and water present in their formations. This is why the need arises to try to know the behavior of the processes at field scale using an approximation through laboratory tests. In the literature, there are studies in which analysis of parameters inherent to the catalytic aquathermolysis tests and their respective sensitization have been carried out. Reaction times, operating temperatures, equipment used in the tests, pressurization gases, added chemical agents and their concentration have been the main parameters studied in the investigations. Likewise, the representation of the porous medium has also been considered as a parameter by some authors.

The present work aims to better understand the synergy generated in the fluid-fluid interaction of steam with heavy hydrocarbon when in contact with a porous medium, which in this case would be the rock matrix of the formation of interest. For this reason, a systematic literature review was carried out, from which the researches associated with the use of rock fragments, minerals or porous media for the evaluation of rock-fluids tests under steam injection conditions were selected. As a result, a descriptive bibliometric analysis of the sources, the obtaining of the rock samples used, and the main operational parameters of the tests was carried out. Likewise, a review on the effect of mineralogy and rocks on viscosity changes, gas production and SARA fractions of the analyzed studies is presented.

METHODOLOGY

Systematic reviews have been mainly employed in the health area before the need to ensure that their decisions do not affect the integrity of life, having an understanding of the relevant published scientific evidence to date [17]. These processes aim to provide an updated view of the state of knowledge found in the literature analyzed on a particular topic of interest, found through primary searches, trying to avoid bias in the search and selection of information. In the present study, the primary research allowed delimiting the scope of the investigation to the study of the influence of rock fragments in the physical representation processes of steam injection at laboratory scale, generating the following research question: What is the effect of the rock and its mineralogy on the properties of the resulting crude oil subjected to upgrading processes? For this systematic review were used software that allowed the management of the information such as Mendeley for the referencing of the documents, Excel data sheets to group and filter the findings of the articles, and VOSviewer for the bibliometric analysis of the data.

Inclusion and exclusion criteria. In this review, full research articles found in academic journals and conference papers published as proceedings of events were considered. Articles with no experimental part developed were not considered as the first exclusion criterion. Likewise, papers that were not focused on crude oil upgrading under steam injection conditions, those whose emphasis was purely geological, reports of field pilots of the techniques or research that did not employ rock samples were not considered. It should be noted that some research theses were additionally consulted with the objective of deepening the understanding of the experimental processes exposed in the articles found in the review.

Search and selection. For the selection of the appropriate databases for the review, a previous analysis was carried out in different portals of these pages with the aim of identifying which ones yielded articles of interest and contribution to the research. The databases selected were OnePetro, ScienceDirect, Taylor & Francis, EBSCOhost, ACS Publications and Scopus, with no language or publication date restrictions. The search of these documents was carried out by using a search equation considering keywords related to aquathermolysis

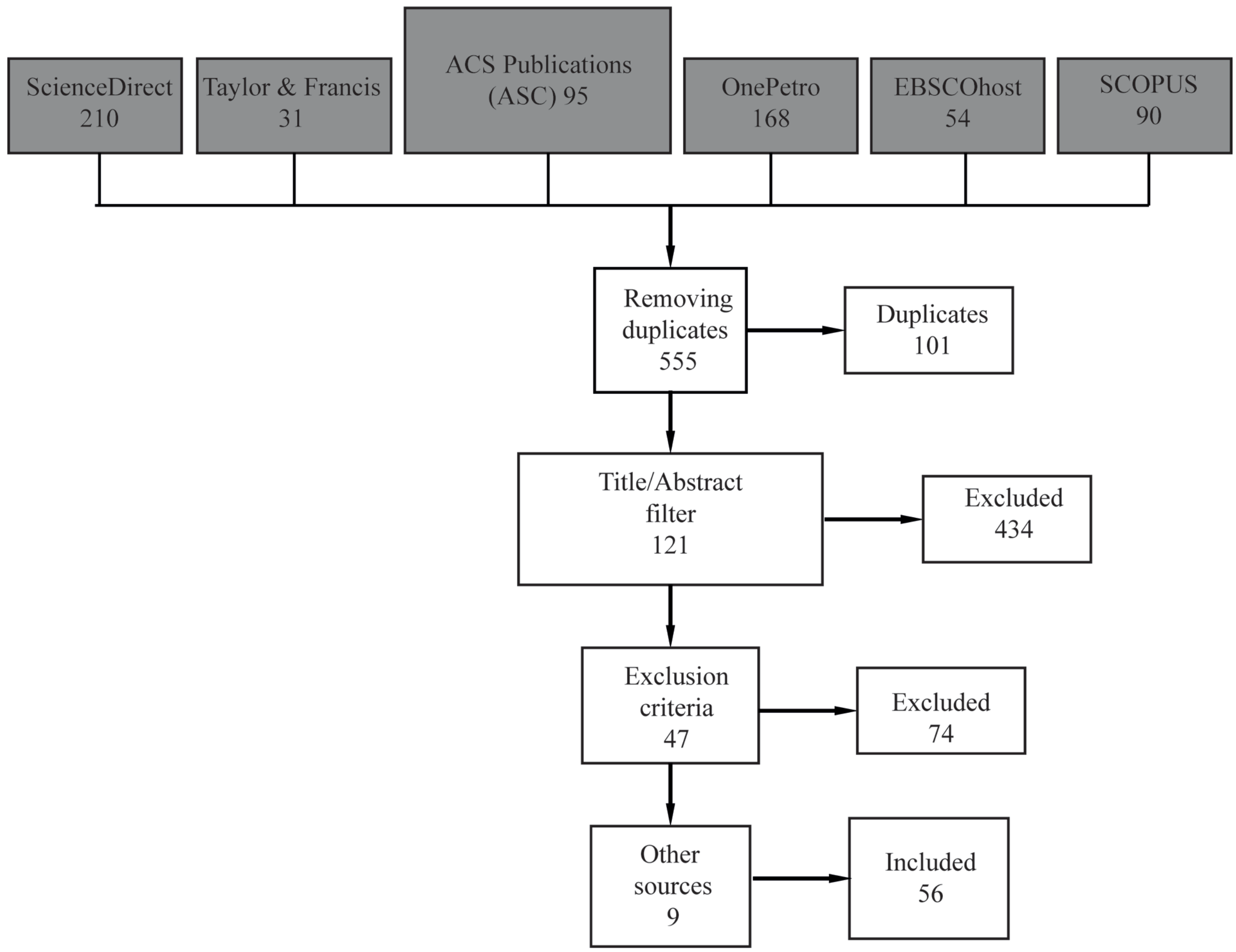

Fig. 1. Database review workflow based on PRISMA methodology.

reactions, crude handling and experimental tests as follows: (aquathermolysis OR aquathermolytic) AND (petroleum OR oil OR crude) AND (test OR experiment). It is important to note that the equation did not include keywords related to rock, since during the previous primary review it was observed that important studies were outside the scope of the search engine. The databases were initially consulted in December 2019, with renewal of the information in June 2020 and as a last search in May 2021. Likewise, the guidelines established by Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) for searching databases, registries and other sources were considered in the present review.

RESULTS

A total of 656 studies were found by reviewing the databases using the above-mentioned search equation. After excluding duplicate research, a scan of the title and abstract of 555 researches was performed where 121 were selected for a complete document review. After this process, 74 studies were excluded based on the previously defined criteria for a total of 47 studies obtained from databases. In addition, 9 studies obtained from other sources were added, consulted through the use of cross-references of the studies analyzed, finding research of weight for the review in question, for a total of 56 studies to be analyzed. Figure 1 shows the summary of the search process and selection of the studies to be included in this review, as shown in Table 1.

Bibliometric analysis of the research included in the study. The first publication analyzed dates from 1988, although after 2010 75% of the papers are concentrated, with peaks of greater frequency in 2011 and 2020 (Fig. 2). Of the selected papers, a large amount of the information has been published in academic journals (61%) or in conference proceedings (30%), completing more than

Table 1. Studies considered for the review were selected after applying the inclusion and exclusion criteria

Study type	Source			Disposition		Test type		References
	core	artificial	aggregate	synthetic core	core mixture	static	dynamic	
C	X	–	X	X	–	X	X	[18]
J	X	–	–	–	X	X	–	[19]
J	X	–	–	–	X	X	–	[20]
J		X	–	X	–	–	X	[21]
C	X	–	X	–	–	X	–	[22]
J	X	–	X	–	–	X	–	[23]
C	X	–	X	–	–	–	X	[24]
J	–	X	X	–	–	X	–	[25]
J	–	X	X	–	–	X	–	[26]
J	–	X	X	–	–	X	–	[27]
C	X	–	X	–	–	X	—	[28]
J	X	–	X	–	–	–	X	[29]
C	–	X		X	–	–	X	[30]
D	–	X		X	–	–	X	[31]
J	X	–	X	–	—	X		[32]
C	–	X–	–	X	–	–	X	[33]
J	X	–	X	–	–	X	–	[34]
J	–	X	–	X	–	–	X	[35]
D	–	X	–	X	–	–	X	[36]
J	X	–	X	–	–	X	–	[37]
J	X	–	X	–	–	X	–	[38]
C	X	–	X	–	–	X	–	[39]
J	–	X	—	X	–	X	X	[40]
J	–	X	–	X	–	X	X	[41]
M	X	–	X	–	–	X	–	[42]
C	X	–	X	–	–	X	–	[43]
J	–	X	–	X	–	–	X	[44]
J	–	X	X	–	–	X	–	[45]
J	X	–	–	–	–	–	X	[46]
C	–	X	–	X	–	–	X	[47]
C	X	–	–	–	–	–	X	[48]
C	–	X	–	X	–	–	X	[49]
J	X	–	X	–	–	X	–	[50]
C	X	–	–	–	–	–	X	[51]
D	X	X	X	X	–	–	X	[15]
C	–	X	–	X	–	–	X	[52]
M	–	X	–	X	–	–	X	[53]
J	–	X	X	–	–	X	–	[54]
J	X	–	X	–	–	X	–	[55]
J	–	X	X	–	–	X	–	[56]
J	–	X	X	–	–	X	–	[57]
J	X	–	X	–	–	X	–	[58]
J	–	X	–	X	–	–	X	[59]
J	X	–	X	–	–	X	–	[60]
C	X	–	–	–	–	–	X	[61]
J	X	–	X	–	–	X	–	[62]
J	X	–	X	–	–	X	–	[63]
J	X	–	X	–	–	X	–	[64]
J	X	–	–	–	X	X	–	[65]
C	–	X	X	–	–	X	–	[66]
C	–	X	X	–	–	X	–	[67]
C	–	X	X	–	–	X	–	[68]
J	–	X	X	–	–	X	–	[69]
J	X	–	X	–	–	X	–	[70]
J	X	–	–	–	X	X	–	[71]
J	X	–	–	X	–	–	X	[72]

[a] Conference proceeding (C), Journal (J), Master thesis (M), Ph.D. Thesis (D).

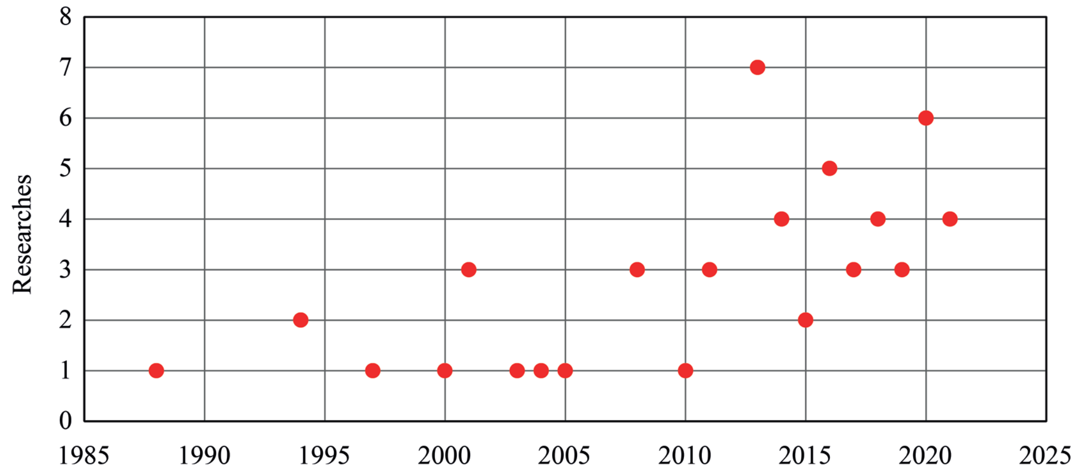

Fig. 2. Timeline of the research included in the systematic review.

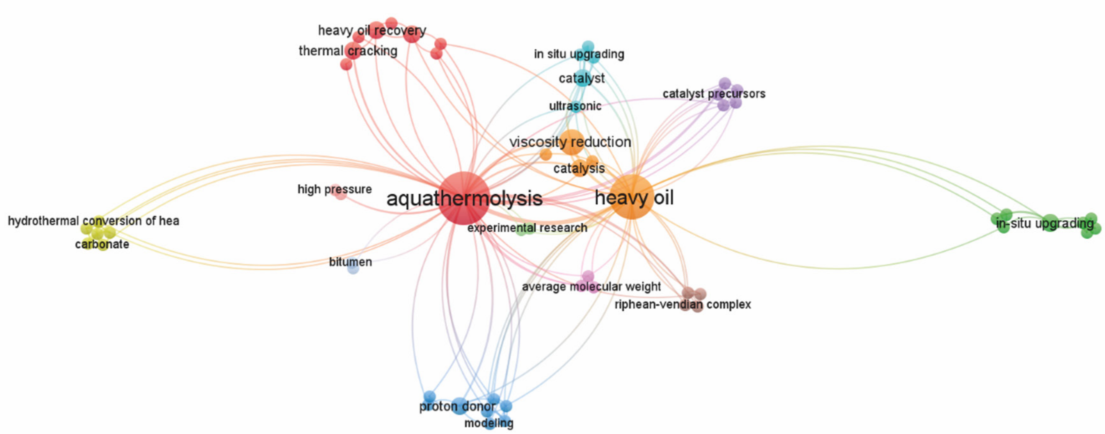

Fig. 3. Bibliometric map of cooccurrence using keywords.

90% of the sources consulted, while the remaining 9% corresponds to research theses. Thirty-six percent of the studies included were obtained through the OnePetro database, followed by ScienceDirect with 13% of the total sample.

Using a co-occurrence analysis through the use of the keywords of the consulted documents, Fig. 3 was obtained. Aquathermolysis is the main node of this bibliometric study, which is connected to seven alternate nodes formed by heavy crudes, in-situ upgrading, hydrothermal conversion, hydrogen donor, average molecular weight, catalyst precursors and heavy crude recovery. Although the present study wished to observe the effect of mineralogy or rock fragments on the reactions, these keywords were not widely used in the papers, which is why a main node that brings these words together is not observed.

The crude oils used in the research were mainly of the heavy type with high viscosities, around 100 000 cP measured between 20 to 50°C. These samples come mainly from three countries: China, Canada, and Venezuela, with more than 5 samples used in the studies. In addition, the literature reports a smaller number of studies using samples from other countries, such as Russia, Turkey, United Kingdom, Iran and Oman. With respect to the operational parameters, 84% of the reaction times used were less than 100 h, with maximum values of 1680 h and minimum values of 2 h. Operational

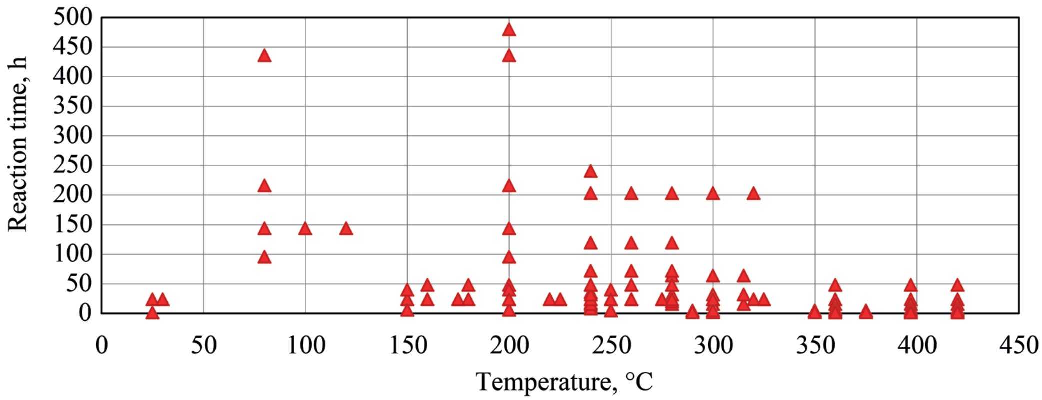

Fig. 4. Operating temperatures and reaction times of the tests analyzed.

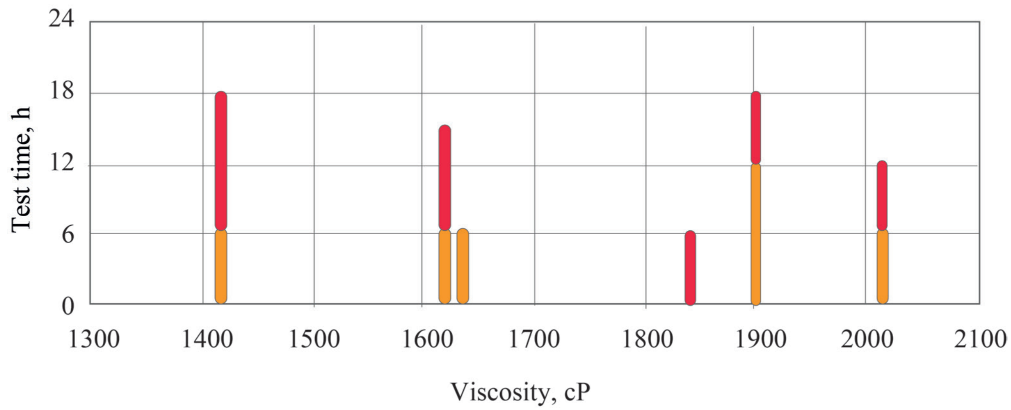

Fig. 5. Changes in viscosity under different temperatures and reaction times [22].

temperatures ranged from 25 to 420°C, with 78% of the temperatures used being less than 325°C. Figure 4 shows the distribution of time-temperature values used in the studies analyzed, where it is highlighted that there is a high density of tests between 150 to 400°C, typical for the aquathermolysis processes with times of less than 50 h.

DISCUSSION

Effect over viscosity. Viscosity is one of the main physical measurements applied to crude oil characterization processes in laboratory tests. This trend is associated with the recovery mechanism inherent to the modification of this property, which causes the improvement of the mobility ratio in the reservoir represented in an increase in production. Studies found in literature show that the variation of viscosity has a greater dependence on temperature changes with respect to changes in reaction times [23, 27, 73].

The magnitude of the change in viscosity at different operating temperatures and reaction times in the tests are related to the reactivity of each of the crudes. For example, Fig. 5 shows a crude oil from a Canadian field near Fort McMurray. The height of the bars indicates the duration of the test at the specific temperatures to which the samples were subjected, 80°C represented by the orange color and 200°C by the red color. It is evident that the lowest viscosity reduction occurs when there is greater exposure to high temperatures, allowing the bonds to break and stabilize. However, exposure to high temperatures for short periods of time leads to the appearance of the acid polymerization effect due to the breaking of free radicals and their inability to stabilize.

The effect generated by the presence of the porous medium on the aquathermolysis reactions can be seen in Fig. 6. In this case, the original crude oil (OS) has a viscosity value close to 70 000 cP, which decreases

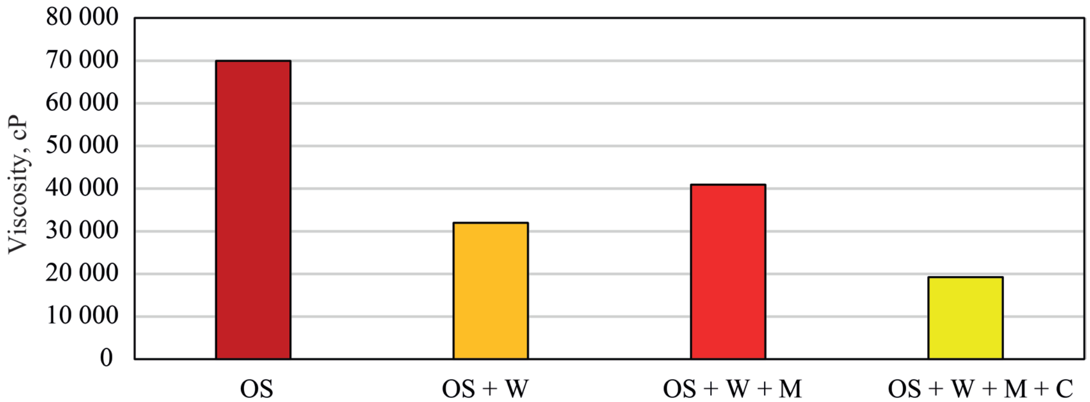

Fig. 6. Variations in viscosity of different tests in the presence and absence of rock [35].

by 54.3% under steam injection conditions (OS + W) at 200°C for 24 h. The addition of rock fragments (OS + W + M) in the process generated an improvement, although of a smaller proportion with 41.5%. This difference may be related to the catalytic effect of the minerals present in the porous medium, which could generate greater free radical breakage that, due to the test conditions, could not be stabilized in the best way. The opposite case occurs when a catalyst is incorporated to the process (OS + W + M + C), since free radicals are stabilized in the presence of hydrogen in the system forming a stable molecule [16].

This effect may have variations based on the mineralogies present in the samples added to the process and the reactivity of the evaluated crude oil. Tavakkoli Osgouei and Parlaktuna [58] performed steam injection processes in a static system at 250°C realizing a variation in the mineralogies of the rock samples, sandstone composed mainly of quartz, and the other limestone with calcite as its main component. Their research proposal posed two modalities, the first with the addition of 10% by weight of rock and the second using 8% by weight of rock plus 2% by weight of clay minerals, bentonite, kaolinite and sepiolite. In general, the authors evidenced that the use of rock and mineral samples generate a decrease in the quality of the crude obtained during the fluid-fluid tests under steam injection conditions. This was observed with the increase in the viscosity value of the produced oil regarding to the initial value of the crude oil feedstock. In this case, a different behavior to that obtained in the study of Xu and Pu [35] is presented, this is caused by the differences in the properties of the crude oil, taking into account their variations in viscosities as a physical aspect and the changes in the reactivities of each one due to their chemical composition.

X. Zhang et al. [32] conducted a study to evaluate the effect of different minerals (illite, montmorillonite, kaolinite, quartz, plagioclase, potassium feldspar) on the viscosity reduction of base oil under steam injection conditions. Within their experimental design they performed three main sensitizations of operational parameters such as operating temperatures (160–260°C), reaction times (8–48 h) and mineral dosage (1–30 wt %). The results showed that the increase in operating temperature generates an increase in viscosity reduction in the presence of all minerals, where the greatest changes are generated by illite, montmorillonite and kaolinite. Regarding reaction times, it was observed that the tests did not show significant changes in the property values after 24 h and the greatest reductions in viscosity were obtained by the addition of the minerals illite, kaolinite and montmorillonite, reaching a reduction of more than 30% [32]. As a last analysis, they found that although at low dosages the greatest incidence was generated by plagioclase and potassium feldspar, once their concentration is increased, their participation in viscosity reduction is overshadowed by illite, montmorillonite and kaolinite.

Gas production. Several studies have shown the effect of rock samples or mineralogy on gas production in tests. One of them has been that of Fan et al. [25] which used a sample of 100 g of heavy crude oil from the Huanxiling field (OS), 10 g of rock samples (M) composed of a mixture of mainly quartz, potassium feldspar and plagioclase and 10 wt % of clay components, 10 g of

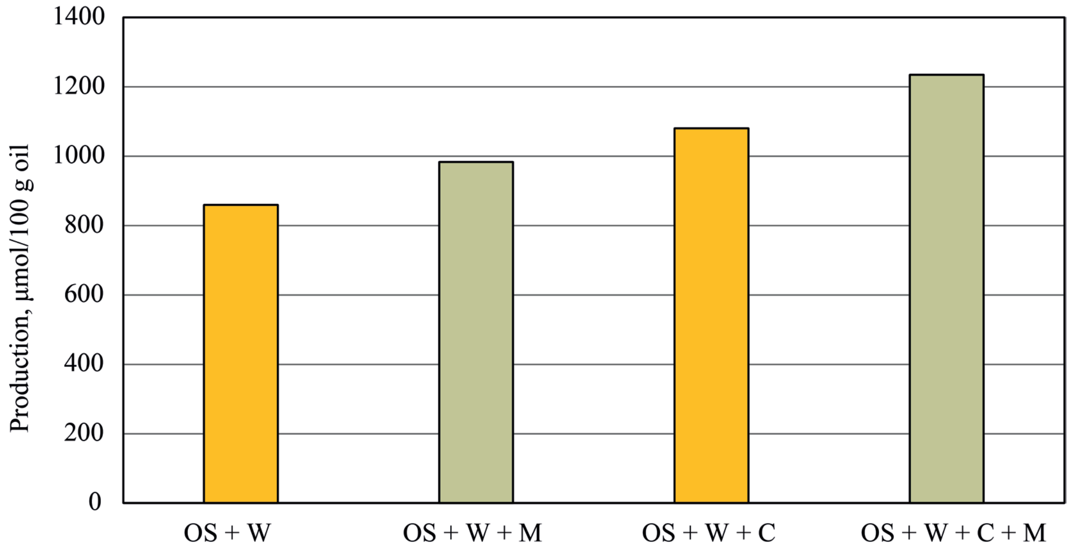

Fig. 7. Gas production under different proposed scenarios [25].

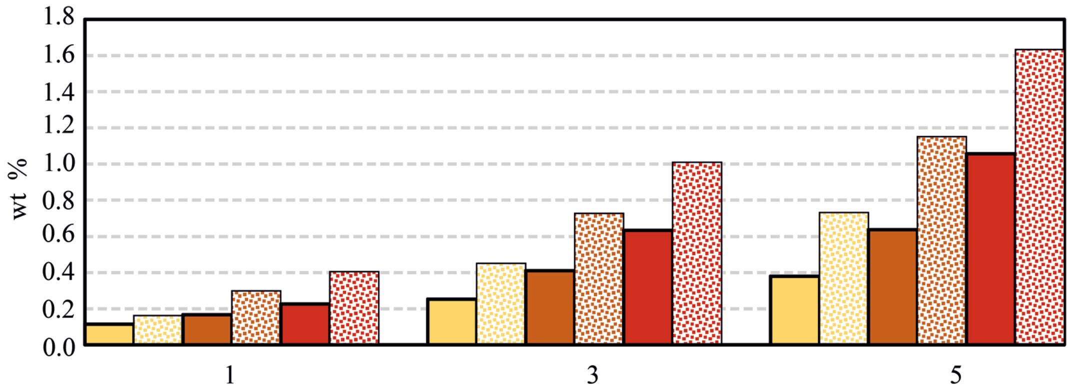

Fig. 8. Variation in hydrogen sulfide production under different test schemes [64].

water (W) and 10 g of catalyst (C) with composition: vanadium, nickel and iron metallic agents in a molar ratio of 1 : 1 : 5 respectively. Figure 7 shows the variation in gas production under different arrangements of the proposed tests. The presence of sand as observed in the results generates a considerable increase in the amount of gases, 14% under the oil-water scenario, and 44% with oil-catalytic solution.

Most studies show gas production as an accumulated value; however, it is important to know how the presence of rock affects the generation of each gas and its performance. J. Zhang et al. [64] set up their experimental tests with the objective of quantifying the effect of rock samples on aquathermolysis in the production of gases. Within their experimental design, they also carried out sensitization of operating parameters such as temperature at 240, 260, and 280°C; and reaction times of 24, 72, and 120 h in the presence and absence of rock samples. Within the measurement of the gaseous effluents, the authors divided the gaseous products resulting from the aquathermolysis as shown in the following figures.

To read the results in Figs. 8–13 the following designations are used:

—The operating temperatures are identified by color: 240°C with yellow, 260°C with brown and 280°C with red.

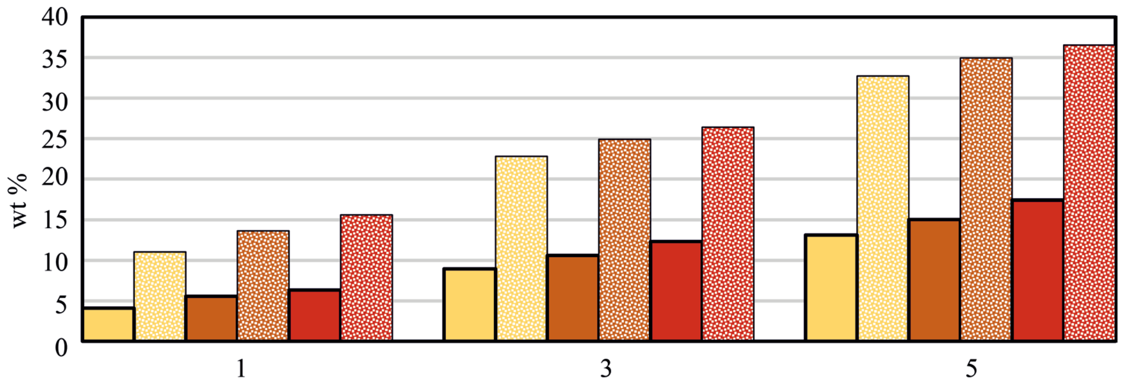

Fig. 9. Variation in carbon dioxide production under different test schemes [64].

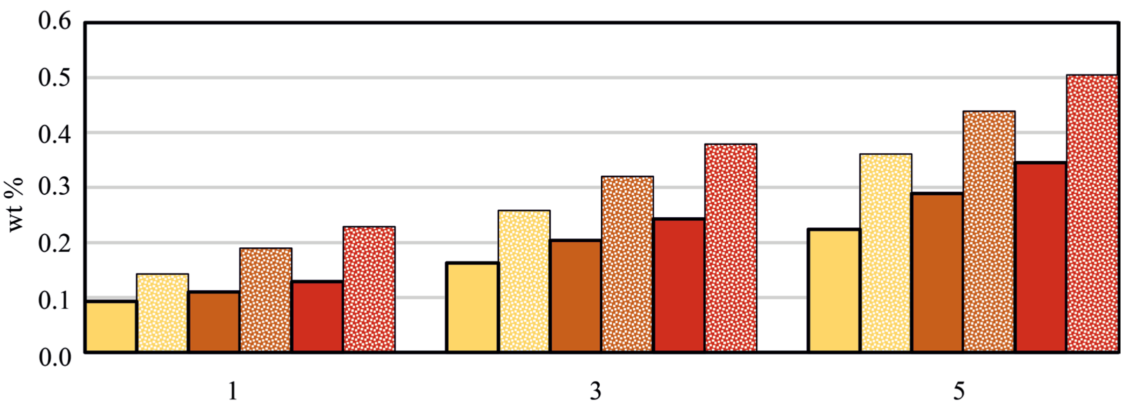

Fig. 10. Variation in carbon monoxide production under different test schemes [64].

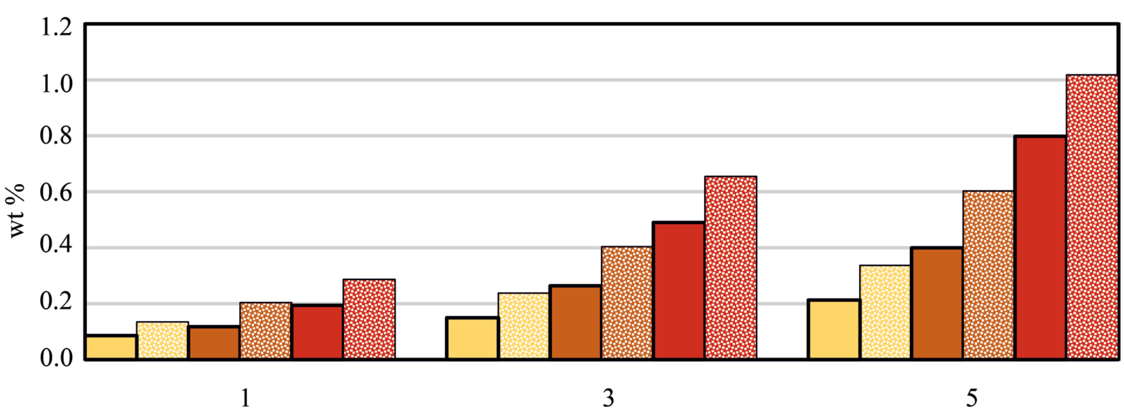

Fig. 11. Variation in hydrogen production under different test schemes [64].

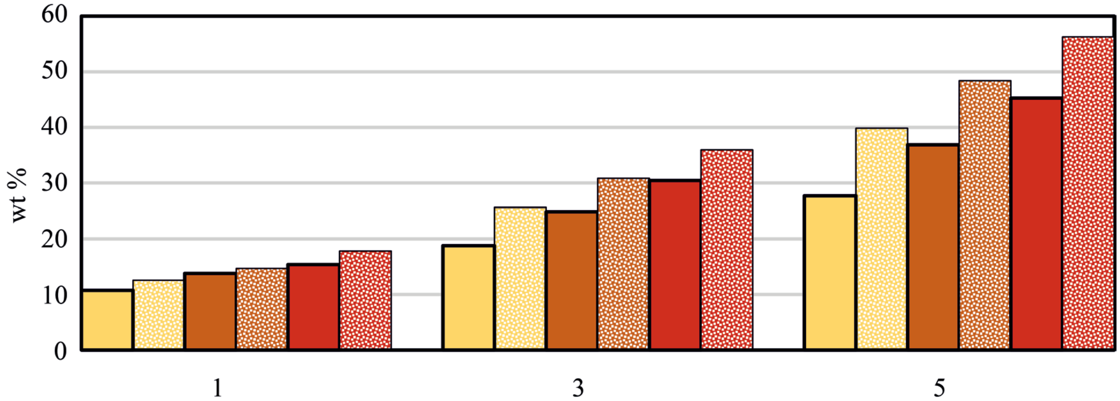

Fig. 12. Variation in methane sulfide production under different test schemes [64].

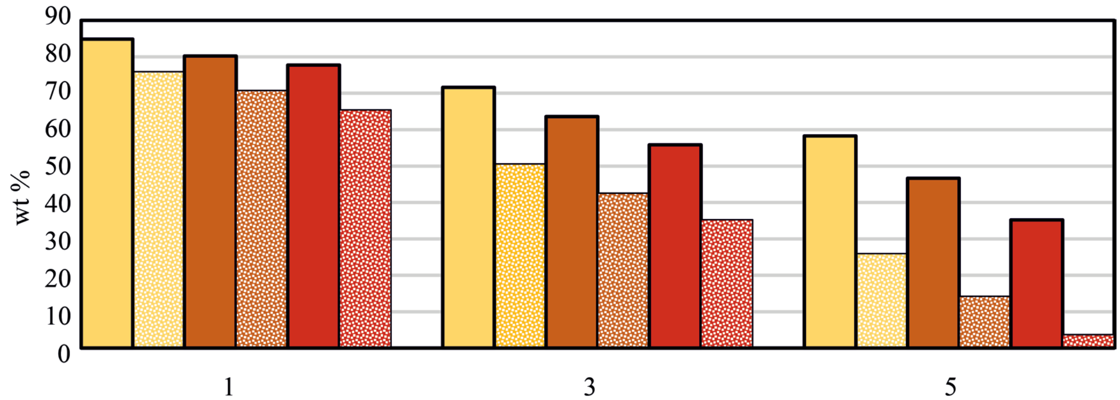

Fig. 13. Variation in higher molecular weight hydrocarbon gases production under different test schemes [64].

—The reaction times are grouped into three groups 24 h (1), 72 h (3) and 120 h (5).

—The addition of rock sample is identified by the columns with grain texture, and the absence of this material is represented in the columns with smooth texture.

Hydrogen sulfide. The production of hydrogen sulfide is increased by the presence of rock sample, as well as by increased reaction times and higher operating temperature. It is evident from Fig. 8 that the production of these gases increases in greater proportion with respect to temperature, rather than with the duration of the tests. Likewise, the magnitudes obtained from this gas did not exceed 2 wt % in the tests, thus considering that it is a small volume to treat on surface. The results exhibit a behavior found in other studies, where the presence of mineral matrix promotes the formation of H_2S from the sulfur molecules generated for the breakdown of asphaltene fraction during the process [50, 55]. Also, Zhao et al. [74] observed that the catalytic effect made for the mineral presence decrease the temperature of the reaction between the heavy oil and the formation water generating an increase in the compound content trough the steam injection process.

Carbon dioxide. The production of carbon dioxide is considerably increased in the presence of rock sample, exceeding in almost all scenarios the double of its content compared to those obtained in the absence of this material, as shown in Fig. 9. Likewise, the results show that the production of this gas is more dependent on the reaction times compared to the operating temperatures used. This behavior was also appreciated by Rivas et al. [18] in their experiments, where a Cerro Negro heavy oil sample was evaluated. The addition of sand to the experiments generated an increase over the 100% in

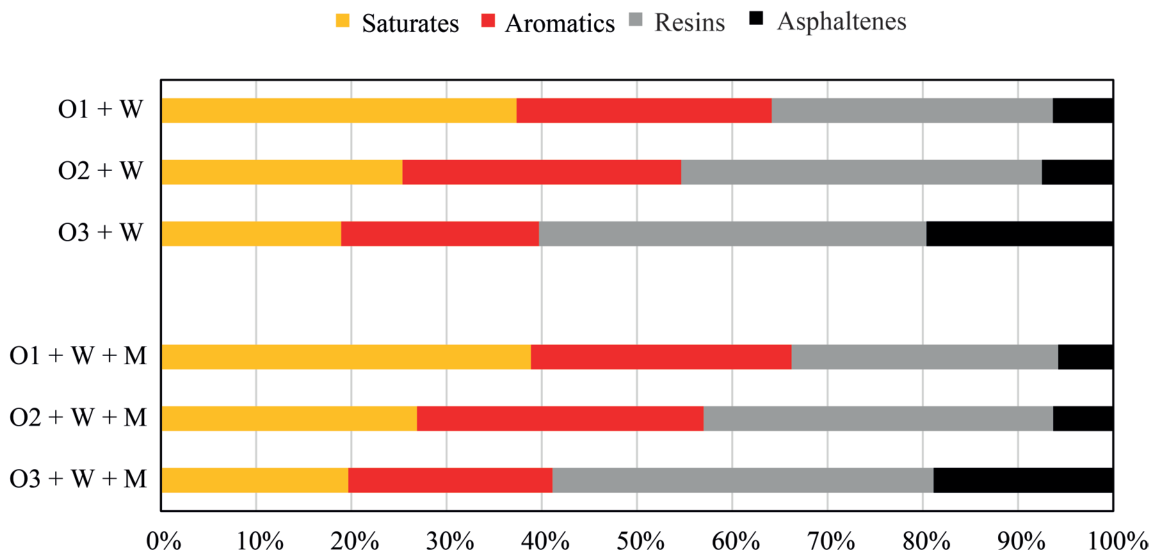

Fig. 14. SARA fractions of crude oil samples subjected to steam injection processes in the presence and absence of rock fragments.

the CO_2 presence in their gas effluents. Regarding the volumes of carbon dioxide obtained, these represent a variable to be considered when treating gaseous effluents on the surface, since they make up between 15 and 35% of the total production.

Carbon monoxide. Carbon monoxide production shows an increase in the presence of rock samples, although it does not exceed half the content in its absence, as shown in Fig. 10. The results show that there is no dependence on the operating temperature or the reaction time of the tests, since its increase is perceived in a staggered manner. The volume represented by this gas is quite small since at its highest production it does not exceed 1 wt %. These concentration values could be the result of the water gas shift reactions where the carbon monoxide interacts with water and generates more carbon dioxide [10].

Hydrogen. Hydrogen production in the presence of rock fragments is not as altered since the largest increase corresponds to 25 wt % in the best scenario as shown in Fig. 11. In this case the content of this gas is mainly affected by the temperature of the process rather than the duration of the process. In terms of volume, it is of low proportions, which is why in some processes it is necessary to add hydrogen donors to stabilize the free radicals fragmented by the aquathermolysis reactions [75].

Methane. Methane production in the presence of rock samples is affected to a greater extent when a prolonged reaction is generated, as shown in Fig. 12. It is evident that both the reaction time and the operational temperature have an effect on the concentration of this product. Regarding the volume produced, it is of great importance since this gas is of high utility in the industry and it would not be a waste but a product with added value. These values reflects the trends found in other studies, where the light gases increasing in the presence of minerals and more reaction time and temperature [18].

Higher molecular weight hydrocarbon gases. The production of hydrocarbon gases of higher molecular weight is quite affected by the presence of rock samples, mainly at high temperatures with prolonged reaction times as shown in Fig. 13. Without the presence of this material, production is believed to be equally affected by temperature and reaction time due to the linear trend in the reduction of its concentration. It is important to highlight that its decrease is due to the breakage of the chains of higher molecular weight by the catalytic effect of the minerals present in the rock, as well as the elevation of temperatures for prolonged periods of time that allow the upgrading of the chains into ones of smaller extension.

SARA fractions. These fractions refer to the division of crude oil into four components: saturates, aromatics, resins and asphaltenes [76]. Among the studies analyzed,

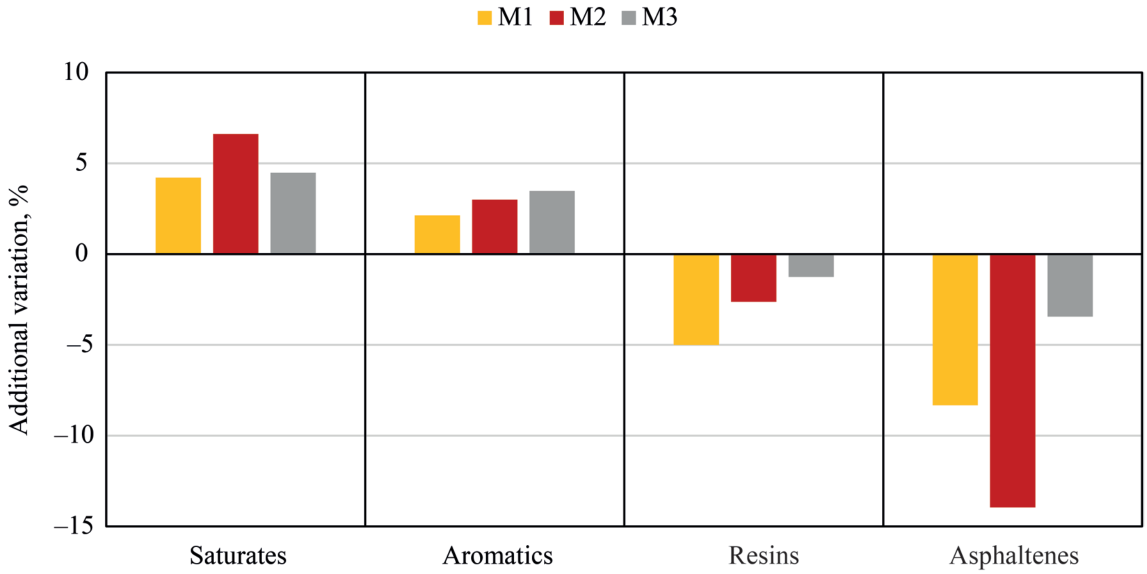

Fig. 15. Additional variation generated by the presence of rock fragments.

some of them performed SARA fraction measurements on the crude feedstock used, as well as on the crudes resulting from the steam injection tests and the crudes resulting from the addition of rock samples to the systems. The investigations considered for the effect of rock in the tests were those carried out by Chen et al., Kayukova et al. and Zhang et al. [32, 34, 70].

Figure 14 shows the effect of steam injection in the presence and absence of the rock samples on the SARA fractions of the samples analyzed (W/W+M). It is evident that in different magnitudes, but with the same tendency, the saturated and aromatic fractions presented an increase in their content, while the resins and asphaltenes decreased. Likewise, an additional effect generated by the rock on the variations in these fractions is observed as shown in Fig. 15.

From this analysis it can be evidenced that the main additional changes generated by the rock are in the asphaltene fractions. This is supported by the findings of Montgomery et al. [55], who expressed that the mineral matrix influences the oil chemistry during aquathermolysis by promoting the formation of H_2S from the smaller sulfur-bearing molecules generated by the catalytic decomposition of the high molecular weight asphaltenes fraction in the original crude oil.

LIMITATIONS

Among the limitations found in the present review is that a large number of studies, although they use rock samples in their experiments, these cannot be taken into consideration to evaluate the effect on the resulting crudes since there are no tests in the absence of these fragments with which to perform a comparative analysis.

CONCLUSIONS

The understanding of aquathermolysis reactions has allowed highlighting them as a chemical mechanism associated with steam injection, where the generation of acid gases and hydrocarbons of lower molecular weight are the main indicator. Therefore, for an adequate representation of crude oil upgrading, it is necessary to understand the effect of mineralogy and rocks on the physicochemical properties of the resulting crudes subjected to steam injection processes.

Due to the variety in crude types, as well as their differentiating reactivity, the magnitude of the effects generated by mineralogy and the presence of rock fragments may be altered. However, it is evident that there is a tendency to generate an additional improvement on the properties of the crude oil due to the catalytic effect related to its addition.

AUTHOR CONTRIBUTION

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Luis Miguel Salas-Chia, who also make the first draft of the manuscript with Paola Andrea León Naranjo. Adan Yovani León Bermúdez critically revised the work. All authors commented on previous versions of the manuscript, read and approved the final manuscript.

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