

Pore-scale experimental simulation of water flow in acidified carbonate rock based on tomographic imaging

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Abstract. It presents a pore-scale experimental simulation of water flow in acidified carbonate rock based on tomographic images with the aim of investigating the variations in the rock petrophysical properties because of the acidizing as well as the efficiency of the wormhole pattern. The experimental configuration allows an understanding of the behavior of the acidified reservoir by means of comparisons between the experimental petrophysical analysis and the digital analysis of an acidified sample. Digital analysis accounts for mathematical models of reconstructing images by using computerized X-rays. The validation of the simulation is done by comparing the experimental petrophysical results with the digital petrophysical analysis before and after the acidizing. This research is motivated by the need to evaluate the permeability and porosity characteristics of the rock and the digital rock derived from the simulation. Moreover, the simulation is used to evaluate the efficiency of the wormhole pattern (dissolution channels structure) and, consequently, validate the acidification process.

Keywords: Experimental simulation, matrix acidizing, wormhole, digital petrophysics, tomographic imaging.

1 Introduction

This contribution presents a pore-scale simulation based on experiments that investigate the flow of acid water in a carbonate subjected to the acidification process. Using tomographic images of the internal structure of the digital rock, the study aims to examine the effects of acidification on the petrophysical properties of the rock, as well as to evaluate the impact on the acid treatment efficiency, which can be affected by the formation of channels of high permeability and porosity in the porous structure of the rock, called wormholes. Acidification arises as a treatment for damage to the formation, an undesirable operational and economic problem that can occur during the various stages of hydrocarbon recovery. The matrix acid treatment carried out in carbonate reservoirs is a well-known stimulation technique used in the oil and gas industry to increase the permeability a few meters from the well and, thus, economically improving its productivity. This increased permeability is evidenced by the creation of branches or wormholes.

Yoo et al [1] reports that well productivity strongly depends on the length, diameter, and distribution of wormholes along the well near the field. The configuration of wormholes can be controlled by the injection rate, which is designed based on the fluid-rock interaction. In carbonates, acid treatment is carried out by injecting reactive fluids, most often hydrochloric acid (HCl) due to its availability, cost, and high efficiency in dissolving rocks.

composed of calcite and dolomite (Al-Arji et al [2]).

The volume of acidic fluid used in the acidification treatment is highly dependent on the injection rate. In general, at low flow rates, much of the acid reacts before penetrating the rock and thus the face of the formation is abundantly consumed (face dissolution). While at high flow rates, the acid can more deeply penetrate the formation without reacting with the face, as discussed in Ali and Ziauddin [3]. In this perspective, the search for the optimal injection rate will ideally produce a dominant wormhole pattern, that is, a deep and narrow dissolution channel that needs a minimum volume of acidic fluid to increase hydrocarbon productivity.

In this context, the present work seeks to understand the behavior of the reservoir in the treatment of acid water flow in porous media through the comparison of experimental and digital permoporous analyses. The analyses were carried in a carbonate rock sample to evaluate the injection efficiency, controlling the reaction between 5% hydrochloric acid and the rock matrix.

2 Theoretical background

2.1 The acid-rock reaction mechanism

The carbonate rocks are characterized by the presence of calcite $[(CaCO₃)]$ and dolomite $[(Ca, A)$ $Mg(CO₃)₂)$, that in the presence of acids are highly soluble minerals, as seen in Equations (1) e (2). The rock used was Indiana Limestone composed predominantly of calcite (99%), according to Churcher et al [4].

$$
CaCO_3 + 2HCl \rightarrow CaCl_2 + H_2O + CO_2 \uparrow
$$
 (1)

$$
CaMg(CO3)2 + 4HCl \rightarrow CaCl2 + MgCl2 + 2H2O + 2CO2 \uparrow
$$
\n(2)

From the acid attack on the core of the sample, dissolution channels are formed. The final dissolution pattern can be face dissolution, conical, wormhole dominant, branched, or uniform dissolution. Furthermore, the dissolution channels structure varies considerably, depending on parameters such as injection rate and mineralfluid properties.

3 Experimental setup and procedure

3.1 Materials

The three relevant experimental steps of this work were: (1) petrophysical analysis of carbonated rock, (2) matrix acidizing experiments in the core flooding system and (3) obtaining digital petrophysics by X-ray computed tomography (Fig. 1). The characterization of the sample aims to standardize its dimensions, mass, and evaluate its petrophysical properties, such as porous volume and permeability. The sample was randomly selected from a set of 20 carbonate plugs, vacuum dried, and then the porosity volume and permeability were measured.

Figure 1. The three relevant experimental steps of this work

A 5% w/w solution was prepared from 37% HCl (concentrated hydrochloric acid), according to [2] guidelines, considering that the acidic properties of HCl change significantly according to the concentration (Harris et al [5]). This solution was standardized by an acid-base reaction with a sodium hydroxide solution which, in turn, was standardized with a potassium hydrogen phthalate solution.

3.2 Core petrophysics preparation and procedure

The Indian Limestone rock was molded into a plug 3 inches long by 1.5 inches in diameter and dried for 24 hours in a vacuum oven (Vinci Technologies, model DR3003). Then, the plug was weighed on a digital scale (Shimadzu, model ATY224), and its dimensions were checked with a digital caliper (Mitutoyo). Pore volume was determined using a helium porosimeter (Vinci Technologies, model HEP-E) based on Boyle's Law. Permeability was determined on a steady-state gas permeameter (DCI Test System) which enables the user to perform the Klinkenberg correction.

3.3 Experimental preparation of matrix acidizing test

The matrix acidification test was performed in a core flooding system (Fig. 2). Fluid injection was performed with a single piston pump capable of maintaining a constant flow rate between 0.01 and 10 ml per minute (ml/min or cm³/min).

The study of the performance of the formulation in the carbonate porous medium goes through three steps: saturation of the plug with brine, acid flow until the moment of rupture (breakthrough) and water flow to remove the acid from the porous medium, ceasing its reaction with the carbonate. In the brine saturation step, the plug spent 24 hours in the saturator, in a 2% KCl solution, under pressure, so that the maximum number of pores were filled by the solution.

In the test, the solution was injected at a flow rate (Q_{inj}) of 5 ml/min until the moment of breakthrough, as recommended by Rodrigues et al [6]. The process ends when there is a change in the pH of the produced fluid, after passing through the rock. In this way, the volume of solution injected by the pump was verified, and from this total value, the dead volumes of the flow lines were deducted to find the volume that passed through the plug. After the acidification step, 10 pore volumes of distilled water were injected at a flow rate of 5 ml/min to remove the remaining acid present in the plug. The reaction of the acid with the carbonate must be stopped to not influence the wormhole formed and prevent disparity with the image analysis performed by computed tomography.

All the steps of the experiment were at room temperature (T) , with a confining pressure (P_{conf}) of 2000 psi and back pressure (P_{BP}) of 1000 psi to prevent the release of CO_2 from rock dissolution, which can affect the hydrodynamic behavior of the system, as seen in Yoo et al [7]. Table 1 details the operational parameters, in the three stages, used in the acidification test.

Figure 2. A schematic of the experimental apparatus (adapted of [5])

3.4 X-ray CT images analysis

To evaluate the wormhole pattern created by the action of acid inside the carbonate rock, the rock was mapped using images obtained by X-ray computed tomography with a tungsten tube (Nikon, model XT H 225 ST), equipped with a 0.8 mm beryllium inherent filter, capable of reaching maximum voltage and current of 225 kV and 1000 μ A, respectively. For the analysis, a voltage of 150 kV, current of 80 μ A, resolution of 50 μ m, and an additional 0.5 mm thick aluminum filter were used. In addition, the software CT PRO 3D XT (Nikon Metrology NV) was used in the image reconstruction procedure.

After this procedure, with the acquisition of the reconstructed images, it was possible to simulate the digital petrophysics analysis of the acidified sample using the VGSTUDIO MAX software, using the median filter to eliminate possible noise from the tomographic reconstruction and, thus, minimizing the presence of artifacts in the images.

4 Applications

Experimental and digital permoporous analyses of the carbonate plug were performed before and after the acidification process. The pore volume analysis performed on the physical equipment found that the plug used showed porosity values ranging from 20.64% (before acidification) to 20.99% (after acidification). While in digital petrophysics, porosity ranged from 19.34% to 23.42%. The digital porosity presented a higher value in relation to the porosity measured in the physical equipment. This is explained by the fact that porosity via tomography reaches smaller and isolated pores (configuration limited by voxel size), while in gas porosimetry these pores are not accessed (equipment limitation). Figure 3 shows the pore distributions (blue) of the acidified sample in three sections:

(A) top, (B) center, and (C) base.

The permeability found in the physical equipment was around 773 mD (before acidification) and infinity (after acidification). The discrepant value of permeability before acid treatment possibly occurred due to the

heterogeneity of the plug, which is characteristic of carbonate rocks, according to Mahdaviara at al [8]. While the value that tended to infinity is explained by the formation of the dissolution channel inside the plug after the acid attack. The digital permeability values are in progress. Here, the streamlines simulations were performed to evaluate the fluid flow through porous media and wormholes. The objective of the next step of this study is to evaluate the difference between the experimental and digital results in terms of permeability.

Figure 3. Volume distribution of pores of Indiana Limestone plug after acidification in three sections: (A) top, (B) center and (C) base

Figure 4 shows the inlet and outlet faces of the plug after the acid treatment. It is possible to observe in Fig. 2A two openings on the entrance face, indicating the origin of the wormhole. It is also noted that for the injection rate used, there was no excess dissolution of the plug inlet. This can be explained by two reasons: (1) the injected flow rate was high, that is, a short contact time between the fluid and the rock and (2) the used concentration of hydrochloric acid was low.

The result of these two actions favors the penetration of the acid into the rock and, consequently, allows the formation of channels. Fig. 2B confirms that a dissolution channel was formed, as the outlet face has an opening emphasizing that the breakthrough occurred.

Figure 4. Plug inlet and outlet faces after acidizing

Figure 5 shows the images generated by computed tomography to assess the wormhole pattern formed inside the plug after the acidification process. When interpreting it, it is noted that the treated sample presented a robust

and prominent wormhole pattern, which demonstrates greater consumption of solution for the breakthrough. Thus, it becomes a disadvantage to use a treatment that needs larger volumes of solution to reach great depths of the plug, which increases the cost of acid treatment, in addition to increasing the corrosion of pumping equipment and piping.

After the acidification process, tomographic images were essential to clarify the wormhole pattern found. A conical dissolution was observed in the treated plug. This type of dissolution is considered inefficient since large volumes of acid are consumed to have the breakthrough. Although the permeability of the plug was out of the measurable range, the analysis of the flow velocity through streamlines simulation at pore scale found an infinity permeability, consistent with what was observed in the experiment and with the results available in the literature. The streamlines simulation was performed using the VGSTUDIO MAX software (transport phenomena module), where in Figure 5 (right) it is possible to observe the flow velocity field emphasizing the presence of a pronounced wormhole in the center of the specimen, along its entire length.

Figure 5. X-ray CT image of the Indiana Limestone plug (on the left) and flow velocity field from streamline simulation with VGStudio MAX software (on the right).

5 Conclusions

In this study, using a core flooding system, a low-concentrated hydrochloric acid solution was injected into a carbonate rock plug at a high injection rate to obtain the wormhole pattern. The characterization tests showed that the acid attack occurred satisfactorily since there was a reduction in the dry mass of the tested plug. It was found that the high injection rate delays the contact time between the acidic fluid and the rock, thus preventing the plug-inlet face from being excessively consumed, allowing the penetration of the acid, and forming the expected dissolution channels.

It was observed also that the porosities obtained from gas porosimetry and from the tomographic analysis are similar. The small discrepancy in these results is a consequence of both the limitation of the physical equipment and the configuration defined by the user, respectively. In addition, the simulation of the fluid flow velocity through streamline simulation was also satisfactory, since the wormhole pattern was of the conical type as expected given the parameters chosen in the flow test.

As the study is research in development, therefore, more plugs are being tested to evaluate the behavior and efficiency of the control of the reaction between hydrochloric acid (5% concentration) and the rock matrix under different injection rates (0.5, 3, and 5 ml/min). The variation in the injection rate reflects the change in the

flow velocity of the acidic fluid inside the plug and, consequently, in the fluid-rock contact time. The aim of the next steps of this work is to interpret the effects of these changes in injection rates on the petrophysical and mechanical properties of carbonate rocks, as well as on the wormhole pattern.

Acknowledgements. The authors would like to thank the Petroleum Reservoir Engineering Laboratory (LABRES/UFRN), at the Federal University of Rio Grande do Norte (UFRN), for providing the space, equipment, and techniques used in this manuscript, and for providing the Indiana Limestone plug. We also thank the Laboratory of Computed Tomography by X-Rays (Department of Nuclear Energy/UFPE) for the support in the images of the exams and in the technical discussions. The authors also thank CAPES (Coordination for Education of Higher Level Personnel, Ministry of Education, Brazil) and Energi Simulation (Calgary, Canada) for financial support to this research.

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References

[1] H. Yoo, Y. Kim, W. Lee, J. Lee, "An Experimental Study on Acid-Rock Reaction Kinetics Using Dolomite in Carbonate Acidizing". Journal of Petroleum Science and Engineering. vol 168, pp 478-494, 2018.

[2] H. Al-Arji, A. Al-Azman, F. Le-Hussain, K. Regenauer-Lieb, "Acid Stimulation in Carbonates: A Laboratory Test of a Wormhole Model Based on Damkohler and Peclet Numbers". Journal of Petroleum Science and Engineering. vol 203, paper n. 108593, 2021.

[3] M. Ali, M. Ziauddin, "Carbonate Acidizing: A Mechanistic Model of Wormhole Growth in Linear and Radial Flow". Journal of Petroleum Science and Engineering. vol 186, paper n. 106776, 2020.

[4] O. E. Harris, A. R. Hendrickson, A. W. Coulter, "High-Concentration Hydrochloric Acid Aids Stimulation Results in Carbonate Formations". Journal of Petroleum Technology. vol. 18, n. 10, pp. 1291-1296, 1966.

[5] M. A. F. Rodrigues, G. M. Mentges, D. C. da Silva, F. M. F. da Costa, M. F. P. de Brito, A. C. D. Antonino, A. de O. W. Neto, "Application of Nanionic Surfactant Nonylphenol to Control Acid Stimulation in Carbonate Matrix". Journal of Petroleum Science and Engineering. vol 203, paper n. 108654.

[6] H. Yoo, Y. Kim, H. Jang, J. Lee, "Propagation Characteristics of Optimum Wormhole in Carbonate Matrix Acidizing Using Micro X-ray CT Imaging". Journal of Petroleum Science and Engineering. vol 196, paper n. 108010, 2021.

[7] P. L. Churcher, J. C. Shaw, L. L. Schramm, "Rock Properties of Berea Sandstone, Baker Dolomite, and Indiana Limestone". Society of Petroleum Engineers. SPE paper n. 55563, pp. 431-446, 1991.

[8] M. Mahdaviara, A. Rostami, F. Keivanimehr, K. Shahbazi, "Accurate Determination of Permeability in Carbonate Reservoirs Using Gaussian Process Regression". Journal of Petroleum Science and Engineering. vol 196, paper n. 107807, 2021.