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Evaluation of Saline Solutions and Organic Compounds as Displacement Fluids of Bentonite Pellets for Application on Abandonment of Offshore Wells

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Abstract: One of the operational challenges for the use of bentonite pellets as a sealing material in the abandonment of offshore fields consists in their disposition inside the well. This study aims to analyze the interaction of fluid media, consisting of saline solutions (NaCl, $CaCl_2$ and KCl) and organic compounds (diesel, glycerin and olefin), with bentonite pellets, aiming its application as displacement fluid in offshore oil well abandonment operations. The physical integrity of the bentonite pellets in contact with the fluids was verified through visual inspections and dispersibility tests. Linear swelling tests were also performed to evaluate the potential for swelling of pellets in deionized water after contact with the fluid media. The results indicated that NaCl, $CaCl_2$ and KCl solutions completely compromise the physical integrity of the pellets, while diesel and olefin showed the best responses regarding the structural preservation. Futhermore, the linear swelling tests showed that, even after the contact with diesel and olefin for 1 hour, bentonite pellets have reached a total swelling in water of 78%, after 24 hours. In this way, diesel and olefin proved to be highly promising alternatives to be used as displacement fluids for bentonite pellets in wells to be abandoned in a submarine environment.

Keywords: compacted clay; clay swelling; physical integrity; displacement fluid; solids transportation

1. Introduction

Well abandonment operations are generally associated with the end of the productive life cycle of a field, when it becomes necessary to restore the perfect isolation between the different permeable intervals of the oil and/or gas zones and existing aquifers, preventing the migration of fluids between formations or to the surface or seabed [1]. The isolation of these zones, which may be located close to the perforations, as well as in intermediate zones of the well or close to the surface, has been commonly performed by cement plugs [1].

The use of cement as a sealing material, although considered effective and required by most international regulatory bodies, has limitations related to the tendency of shrink and crack and mechanical or chemical degradation, compromising its integrity and causing high costs for repair the plug [2,3]. In addition, the use of this type of material requires additional costs due to the need of a work-over rig to carry out the pumping and setting of the plug, which represents a substantial plugging cost [3].

One of the alternatives suggested to the use of cement plugs in oil well abandonment is the use of hydrated bentonite clay, which, due to its high swelling capacity and low permeability, has also been widely used in the plugging of water wells, seismic shot holes and geological repositories for radioactive waste ([2,4–6]. The advantages of using bentonite plugs, when compared to cement, include economic, environmental and aspects related to safety and health. In addition, the plasticity of this material make it more reliable in the case of formation of cracks, since it tends to heal, ensuring the integrity of the formation [1,7].

Several studies agree that hydrated bentonite is effective in isolating formations with different pressure gradients [4,8–10]. However, there are still gaps that need to be filled to ensure its effectiveness and safety, especially in offshore oil wells.

An operational challenge related to bentonite application in deep oil wells is its proper disposal up to the intervals defined in the abandonment project. One of the ways to ensures the formation of the bentonite plug in the proper location of the well, preventing premature swelling and ensuring its proper setting, is to use its compacted form, also known as pellets, since kinetic hydration is closely related to its physical conformation [1,11]. Additionally, the transportation of pellets should take place by means of a displacement fluid and exclusively hydromechanically, avoiding chemical interactions that could result in premature swelling or disintegration [2,12].

Many studies in the literature have been developed in order to characterize the swelling mechanisms of compacted bentonite based on parameters such as types of fluid for hydration, environmental conditions and physical aspects [13–16]. However, few studies analyze the operational performance prior to the swelling stage, which is crucial, since it is related to the interaction between the pellets and the fluids that must provide their hydraulic displacement efficiently. In this sense, this study aims to analyze the interaction of fluids, consisting of saline solutions (solutions of NaCl, $CaCl_2$ and KCl at different saturations) and organic compounds (diesel, glycerine and olefin), with bentonite pellets, targeting its application as displacement fluid in abandonment operations of offshore oil wells.

2. Materials and Methods

2.1. Materials

Sodium bentonite compacted pellets was used in this work. As displacement fluid, saline solutions and organic compounds were evaluated. The saline solutions were prepared from sodium chloride (NaCl), calcium chloride (CaCl2) and potassium chloride (KCl) brines, at different saturations (10, 20, 30, 40, 50, 60, 70, 80, 90 and 100%). The organic compounds were diesel, olefin and glycerin. The materials were provided by CENPES/PETROBRAS (Rio de Janeiro – Brazil).

2.2. Methods

2.2.1. Interaction under static conditions

The interaction of bentonite clay with each fluid was analyzed by immersing a single pellet of approximately 1.5g in 15ml of each fluid. The physical cohesion of the bentonite pellet was visually verified throughout the test for a maximum time of 120 minutes. Furthermore, in order to compare the results, its swelling in deionized water under the same conditions was evaluated.

2.2.2. Physical integrity under dynamic conditions

From the previous analysis of the physical cohesion of the pellets in static conditions, dynamic tests were conducted, using compounds in which the pellets showed better maintenance of their integrity. These tests were performed based on the ISO 10416:2008 Standard [17].

To carry out the tests, a stainless steel cell was filled with 350mL of fluid and 20g of bentonite pellets. The cells were exposed to shear in a Roller Oven 704 ES Fann, with rotation of 50rpm for 2 hours. At the end of this time, the cell content (fluid and pellets) was carefully filtered through an ABNT number 6 sieve.

The material not retained in the sieve was considered as part of the physical disintegration of the pellet, while the material retained followed to a hot air drying oven, where it was subjected to a temperature of 60°C for 24 hours. After 24 hours, the material was weighed on a high precision analytical balance. To standardize the test conditions, 20g of bentonite pellets without previous immersion was also placed in an oven at a temperature of 60°C and weighed after 24 hours. Equation 1 quantifies the disintegration of the sample.

$$D = \frac{(m_i - m_r)x100}{m_i} \quad (1)$$

where D is the dispersibility or disintegration rate (%), mi is the initial sample mass (g) and mr is the mass of pellets retained on the sieve after drying (g).

2.2.3. Linear swelling

The swelling of pellets in deionized water was evaluated after the contact with the fluids for which it was observed pellets physical preservation under dynamic conditions. Furthermore, in order to compare the results, its swelling in deionized water, without previous contact with such fluids, was also evaluated.

For this, linear swelling tests were carried out using the Linear Swell Meter (LSM) - Fann Instrument Company, model 2100. The tablets needed for the test were made pressing 10g of bentonite pellet, with a granulometry of less than 0.075 mm, corresponding to ABNT number 200 sieve, at 10,000psi for 1 hour. Linear swelling measurements were performed using an automatic-digital transducer that allows direct reading.

The test was carried out in two conditions: in the first condition, the tablets were placed in the equipment and the containers were filled with the fluid media analyzed as displacement fluid. After one hour, the fluid medium was removed and replaced for deionized water, which remained in contact with the tablets for 24 hours. The total test time was 25 hours. In the second condition, the degree of swelling of the pellets in contact with deionized water for 25 hours was evaluated, in order to standardize the total test time. The data obtained in the tests were processed by the software LSM 2100.

3. Results and Discussion

3.1. Interaction under static conditions

The graph in Figure 1 shows the time for which was observed total loss of physical integrity of the pellet, in seconds, for each of the saline solutions analyzed at different saturations.

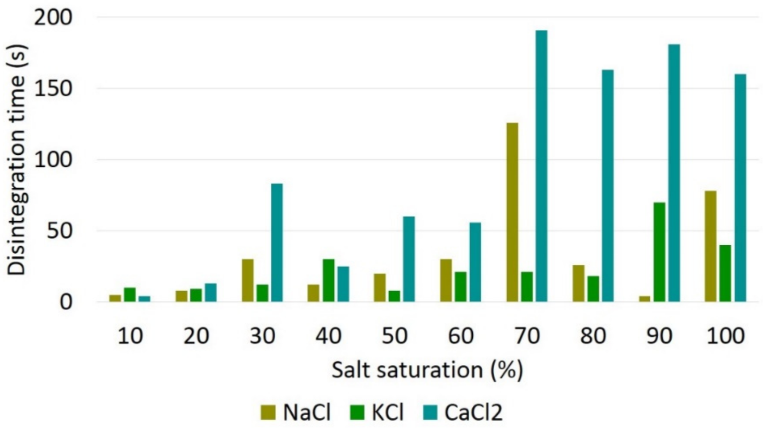

Figure 1. Disintegration time of pellets for saline solutions of different saturations.

In an oil field, the displacement of the pellets inside the well takes approximately 120 minutes. Thus, the same time was estimated for the duration of this test. However, for all saline solutions analyzed, regardless of saturation, the maximum disintegration time was 180 seconds (3 minutes).

The physical appearance of the bentonite pellets, before contact with any type of fluid, is shown in Figures 2–4 shows the appearance observed at the end of the test for the samples that presented the shortest and longest disintegration time for each brine, respectively.

Figure 2. Physical aspect of bentonite pellets.

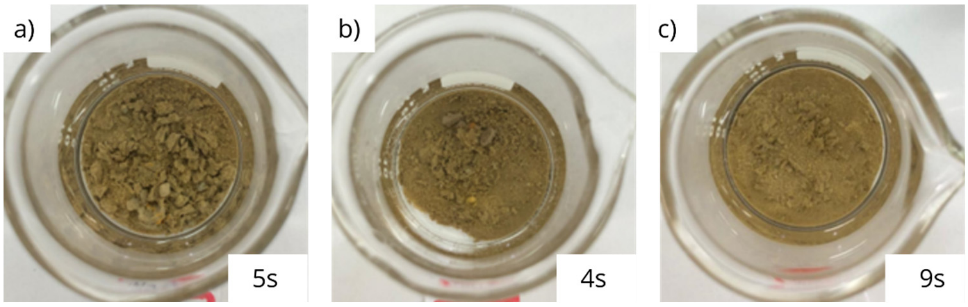

Figure 3. Physical appearance and disintegration time of bentonite pellets after immersion in 10% sodium chloride brine (a), 10% calcium chloride brine (b) and 20% potassium chloride brine (c).

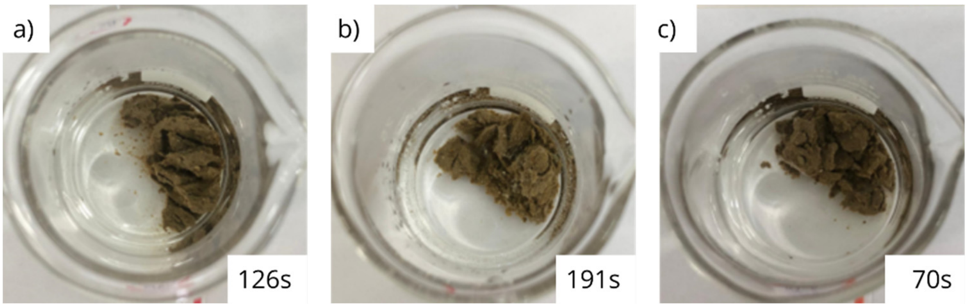

Figure 4. Physical appearance and disintegration time of bentonite pellets after immersion in 70% sodium chloride brine (a), 70% calcium chloride brine (b) and 90% potassium chloride brine (c).

As seen in Figs. 3 and 4, there was a significant physical change on the pellets in contact with saline solutions, compromising their integrity. This may occur due to changes in the microstructure of compacted bentonite promoted by exposure to saline solutions, once the concentrated saline media promotes an osmotic suction, which can act as an internal compaction tension, causing the particles to approach each other [18]. Associated with this mechanism, there is a minimization of the repulsion of the double layer, caused by the presence of the high content of cations in the solution,

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resulting in an increase in the attraction between the lamellae and consequent aggregation of the clay particles. Furthermore, the formation of aggregated structures by the clay particles becomes even more significant when the cations in the saline solution have a greater number of electrons in the valence layer, exerting greater compaction of the double layer. This effect is observed in Fig. 4.

In the images presented in Fig. 4, it was also observed that pellets exposed to higher saline saturations, presented a less expressive disintegration when compared to that presented by the pellets exposed to saline solutions with saturations varying between 10% and 20% (Fig. 3), indicating a correlation between the mechanical properties of clay and brine saturation. Zhang et al. (2016) observed a decrease in plastic compressibility of compacted clays with increasing saline concentration [14]. This behavior would also be related to the osmotic suction, which is proportional to the salt concentration, resulting in the induction of the formation of aggregates and inter-aggregates in the pores, also contributing to the decrease in deformability [14,19]. Destabilization of bentonite particles may occur when in contact with saline solutions of NaCl and $CaCl_2$ at concentrations even lower than those used in this study [20]. It was observed that in concentrations in the order of 10-5 to 10-3 mol/L there is flocculation of the bentonite and, consequently, the sedimentation of its particles.

Although saline solutions (especially KCl solution) are commonly used in drilling fluids, during the well drilling, when reactive formations are encountered [21], this practice should not be extended to the abandonment of wells, since the physical integrity of the pellets is compromised when in contact with NaCl, $CaCl_2$ and KCl solutions, regardless of their concentration.

For the evaluated organic compounds, i.e. diesel, olefin and glycerin, the immersion of the pellets resulted in the physical aspects recorded in the images presented in Figures 5–7, respectively.

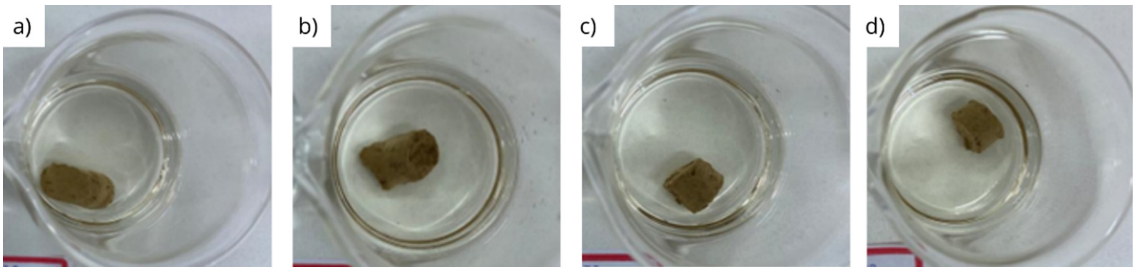

Figure 5. Physical appearance of bentonite pellets after immersion in diesel for 5 (a), 30 (b), 60 (c) and 120 (d) minutes.

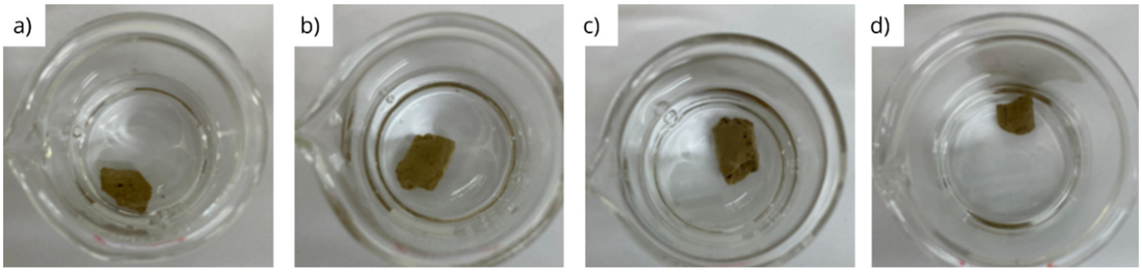

Figure 6. Physical appearance of bentonite pellets after immersion in olefin for 5 (a), 30 (b), 60 (c) and 120 (d) minutes.

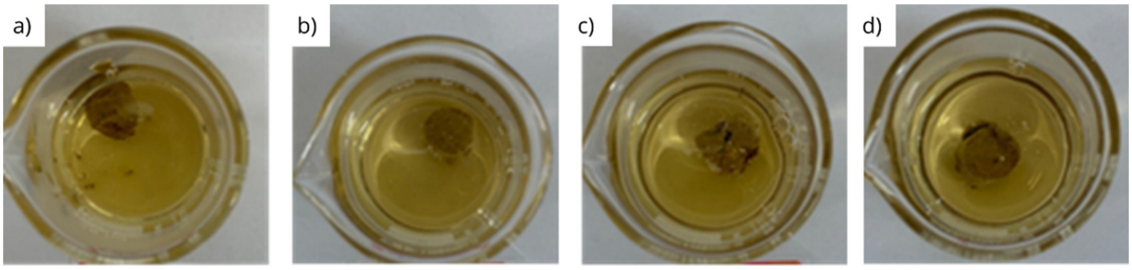

Figure 7. Physical appearance of bentonite pellets after immersion in glycerin for 5 (a), 30 (b), 60 (c) and 120 (d) minutes.

Through the images presented in Figures 5 and 6, it is observed that physical integrity of pellets immersed in diesel and olefin is preserved. However, the presence of small clay fragments was observed, deposited at the bottom of the container, when the pellets were immersed in glycerin (Figure 7), indicating disintegration of the pellets when immersed in this organic medium.

Natural bentonite clays have a hydrophilic surface and, therefore, do not adsorb hydrophobic liquids, such as diesel and olefin, since these clays contain exchangeable inorganic cations that are highly hydratable only in aqueous media [22]. Therefore, the occurrence of disintegration into glycerin, although in low proportion, may be associated with the intermolecular interaction between glycerol, the main component of this compound, due to its hydrophilicity, presenting chemical affinity with the clay surface [23]. However, this chemical affinity does not constitute a possibility for the occurrence of premature swelling of the pellets in this medium, once glycerin seems to have a limited invasion radius in bentonite specimens [24].

The physical appearance of a bentonite pellet after immersion in deionized water for times of 60 and 120 minutes can be visualized through the images shown in Figure 8. It was observed that there are no significant differences in pellet swelling after times of 30 and of 60 minutes, proving that almost total hydration and swelling occur in the first 60 minutes of contact with deionized water. This time is greater than expected for pulverized particles of bentonite, which is in order of minutes [25].

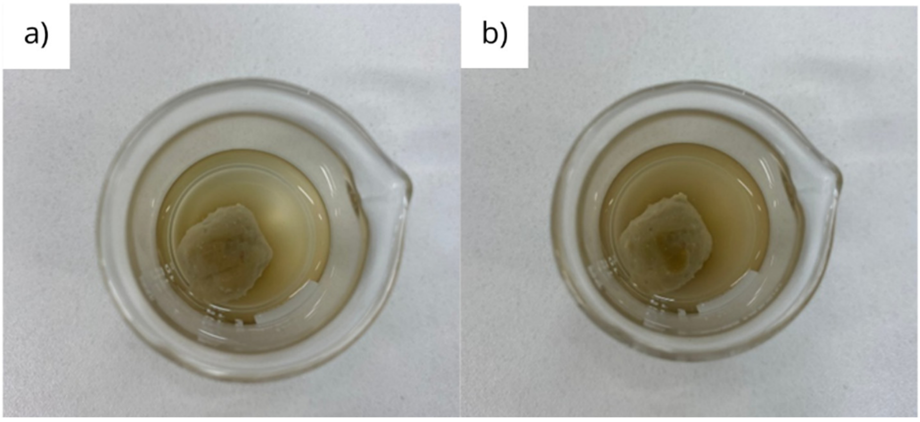

Figure 8. Physical aspect of bentonite pellet after immersion in deionized water for 60 (a) and 120 (b) minutes.

3.2. Physical integrity under dynamic conditions

In view of the results obtained under static conditions, the organic compounds, diesel, glycerin and olefin, showed a better tendency to maintain the cohesion of the pellets, with total physical preservation when the organic medium was diesel and olefin, and partial preservation when glycerin was used. In this sense, the integrity of the pellets in contact with these fluids was also investigated through tests conducted under dynamic conditions, which are close to the turbulent conditions in which the pellets are transported into the well.

Figure 9 presents images of the organic fluid media (diesel (a), olefin (b) and glycerin (c)) filtered after the tests to evaluate the physical integrity of the pellets under dynamic conditions.

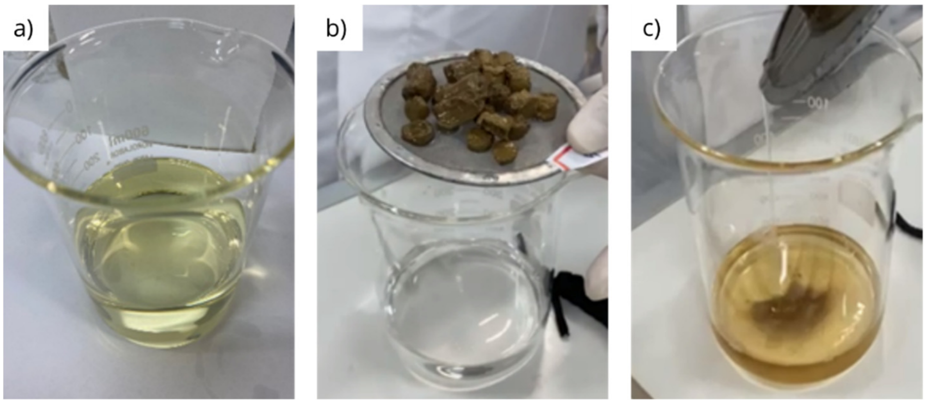

Figure 9. Visual appearance of diesel (a), olefin (b) and glycerin (c) filtered after testing under dynamic conditions.

The dynamic test significantly enhanced the disintegration of the pellets in glycerin (Figure 9(c)), which was also observed, in a lesser extent, under static conditions. This behavior can be clearly noted by the significant volume of fragments visualized at the bottom of the beaker containing the filtered glycerin after the test, and is probably due to the combined action of shear with the longer exposure time of the test, when compared to the test carried out under static conditions. In contrast, no fragments were observed in the filtered material when diesel and olefin were used (Figures 9(a) and (b)).

Table 1 presents the initial mass of the pellets, the residual mass obtained at the end of the test (final mass), and the disintegration rate for each of the analyzed organic compounds.

Table 1. Disintegration rate of pellets.

Fluid	Initial mass (g)	Final mass (g)	Disintegration rate (%)
Diesel	20.00	17.98	10.10
Olefin	20.00	17.49	12.55
Glycerin	20.00	17.13	14.35

The results obtained demonstrate a greater disintegration for the pellets immersed in glycerin, calculated at 14.35%. The disintegration rates calculated for diesel and olefin were 10.10% and 12.55%, respectively. These values are higher than expected, since the filtrate obtained after the tests are clear, with no visible evidence of pellet fragments (Figure 9).

Thus, in order to understand and elucidate the results obtained, additional tests were carried out with the aim of obtaining the average mass reduction by loss of moisture in the pellets under the same test conditions, without prior contact with any organic compound. In addition, specific measurements of the interaction between the pellets and the liquids were carried out, with the pellets being immersed in the media for 2 hours before submitting them to the dispersibility test, excluding the loss of mass resulting from the disintegration of the pellets. Tables 2 and 3 present the results obtained in these additional tests.

Table 2. Moisture loss of pellets after drying at 60°C for 24h.

Initial mass (g)	Final mass (g)	Moisture loss (%)
20.00	17.22	13.90%

Table 3. Mass variation of pellets after immersion on fluid and drying at 60°C for 24h.

Fluid	Initial mass (g)	Mass after immersion in fluid (g)	Final mass (g)	Mass variation (%)
Diesel	20.00	20.71	17.86	- 10.70
Olefin	20.00	20.45	17.55	-12.25
Glycerin	20.00	26.06	23.57	+17.85

The results obtained show an average mass reduction due to moisture loss of the pellets without prior immersion of 13.90% (Table 2). This value is significantly close to the disintegration rate obtained for the test carried out under dynamic conditions for the previously immersed samples in diesel and in olefin, which are 10.10% and 12.55% (Table 3). Similarly, the mass variations obtained when performing the static immersion of the pellet in diesel and olefin, once again represent losses similar to those obtained in the other tests, calculated at 10.70% and 12.25%. Thus, the water physically attached to the edges and external surfaces of the pellets is removed during drying and considered in the calculations to obtain disintegration. As this amount of water removed does not compromise the physical cohesion of the pellets, a correction considering the moisture loss is required in order to obtain the actual rate of disintegration.

Although the temperature at which the pellets were exposed during drying (60°C) is lower than that at which water boils, the moisture loss verified is consistent with thermogravimetric tests conducted with the clay mineral montmorillonite, the main component of bentonites, which demonstrate that the thermal transitions associated with the elimination of adsorbed water can occur at temperatures below 100°C [26,27].

It was also possible to observe that, both for the disintegration test and for the test in which there was static immersion in diesel and olefin, the values of lost mass (Tables 1 and 3) are lower than the values of mass reduction due to loss of moisture presented in Table 2. This behavior is probably related to the disposition of organic compounds on the surface of the pellets, even in small amounts, given the increase in mass registered after immersion, as shown in Table 3. This result demonstrated that, although the affinity between the organic compounds and the pellets is not expressive, this contact might form a physical barrier capable of hindering, even in a minor way, the elimination of water molecules during drying.

For the pellets immersed in glycerin, it was observed absorption of this fluid as a result of the interaction in the first two hours of the test in static condition, resulting in a mass of 26.06g. This mass increase is expressively significant, which represents a percentage of about 30.30% in relation to the mass of the dry sample (Table 3) and is attributed to the strong intermolecular interactions between glycerin, which has a hydrophilic nature, and the surfaces of the clay mineral [23].

This behavior, combined with the high boiling temperature of glycerin, which is 290°C, inhibited the elimination of organic molecules adsorbed under the test conditions. Thus, the increase in mass verified after drying, of approximately 17.60%, corresponds to the compensation between the absorption of organic molecules, which remain in the system and are responsible for the moistened appearance of the sample, and the elimination of water molecules, which corresponds to 13.90% (Table 2). Thus, the physical disintegration value of 14.36%, presented in Table 1, must be corrected, adding the mass gain resulting from the interaction between the pellets and glycerin (17.60%), so that the actual physical disintegration rate obtained is approximately 32%.

Table 4 presents the corrected values for the physical disintegration of the bentonite pellets in organic compounds, exclusively considering the loss of structural integrity provided by contact with these fluids. These results are based on observations during the performance of the test and on the interpretation of the results obtained for the additional tests performed targeting the calculation of moisture content of the pellets.

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Table 4. Corrected physical disintegration rate of pellets.

Fluids	Corrected disintegration rate (%)
Diesel	- 0.60
Olefin	0.24
Glycerin	31.96

For diesel and olefin, the corrected results showed that there was no relevant quantitative disintegration of the pellets, which is demonstrated by the visual appearance of these fluids, in which no disintegrated particles were observed. Furthermore, the negative value obtained for the disintegration rate of the pellets in diesel suggests that the mass of the sample immersed in this fluid, after drying, was greater than the initial mass of the sample without immersion, also after drying. Once there is no chemical interaction between the pellets and the diesel, it is inferred, therefore, that only the physical adsorption of the medium to the surface of the pellets occurs and this mass increase is attributed to the thin oily layer adhered to the surface of the pellets. For glycerin, in turn, an even higher disintegration rate was observed after correction, attributed to the strong intermolecular interactions between glycerin and the pellet surface [23].

3.3. Linear swelling

Percentage linear swelling measured for the pellets, initially immersed in the organic compounds for 1 hour and then immersed in deionized water for 24 hours, is shown in the graph in Figure 10.

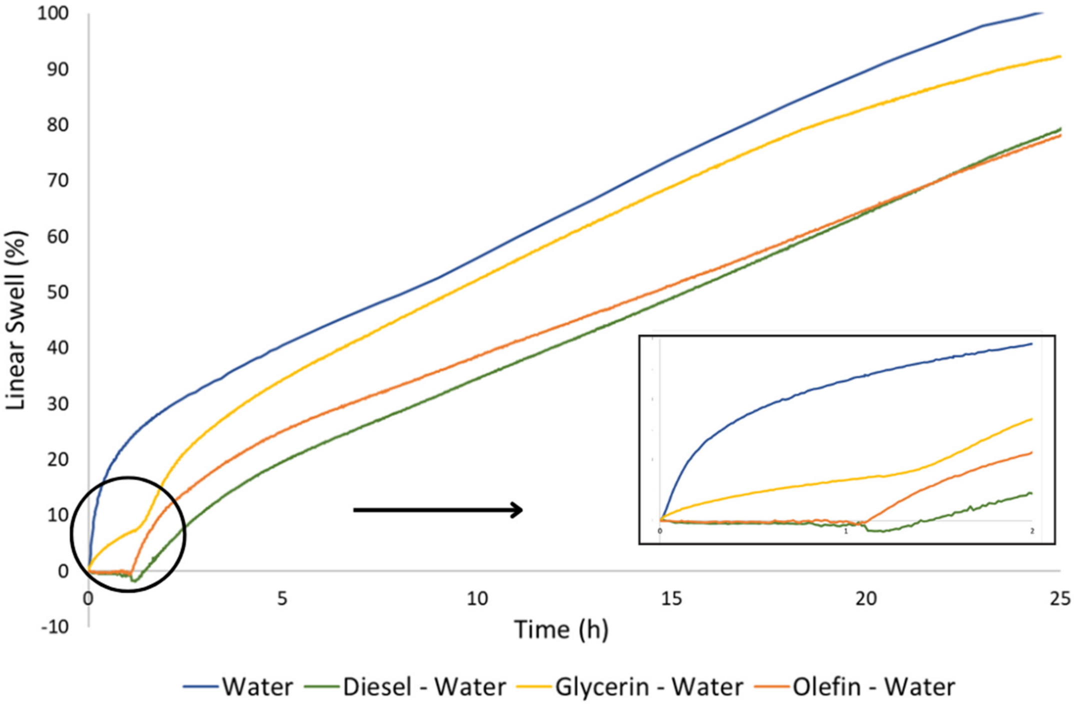

Figure 10. Percentage of linear swelling of bentonite pellets as a function of contact time in each organic compound (diesel, olefin and glycerin) followed by contact with deionized water.

During the first hour of test the bentonite pellets were in contact with the organic media and different behavior was observed. The samples immersed in diesel and olefin did not show swelling, while the sample immersed in glycerin showed swelling from the beginning of the test, accounting for a total swelling close to 7% in 1 hour.

As discussed previously, the interaction between organic compounds and bentonite might be attributed to the chemical properties of these substances. Glycerol, the main component of glycerin, has a hydrophilic nature [28], and when in contact with a mineral that is also hydrophilic, such as bentonite clay, develops interactions with the surfaces of particles that are electrically active [29], promoting an increase in the basal interplanar distance and, consequently, its swelling. In contrast, diesel and olefin are hydrophobic substances [30] and, therefore, do not develop interactions when in contact with a hydrophilic material [31].

After replacing diesel, olefin and glycerin with deionized water, a similar behavior was observed, regardless of the initial organic fluid: continuous swelling was recorded until the end of the 24 hours of testing. For tests carried out with samples previously immersed in diesel and olefin, linear swelling values of approximately 78% were obtained. For the test carried out with the sample previously immersed in glycerin, the linear swelling obtained was 92%. Comparing these values, there is a 14% greater linear swelling when glycerin was previously used. As already discussed, this greater swelling is most likely due to interactions between glycerol and clay particles that promoted an initial expansion between the clay layers. This first interaction made the layers more loosely bound, favoring the hydration mechanisms as well as the hydration rate of the clay particles.

The linear swelling obtained for the samples that had previous contact with the diesel and the olefin evidenced that the immersion of the samples for a period of 1 hour promoted the formation of a membrane on the surface of the tablets and this, in turn, prevented the hydration and total swelling capacity of the samples. It is evidenced by the lower total linear swelling of the samples previously immersed in diesel and olefin, which was approximately 78%, while the swelling of pellets only immersed in water (blue curve in the graph in Figure 10) was 101% at the end of 25 hours.

Although the linear swelling test, carried out in the LSM equipment, registers the percentage of swelling of the sample vertically, it is possible to observe the swelling of the material radially through the metallic screen in which the sample is contained. This swelling was recorded and is shown in the images in Figure 11.

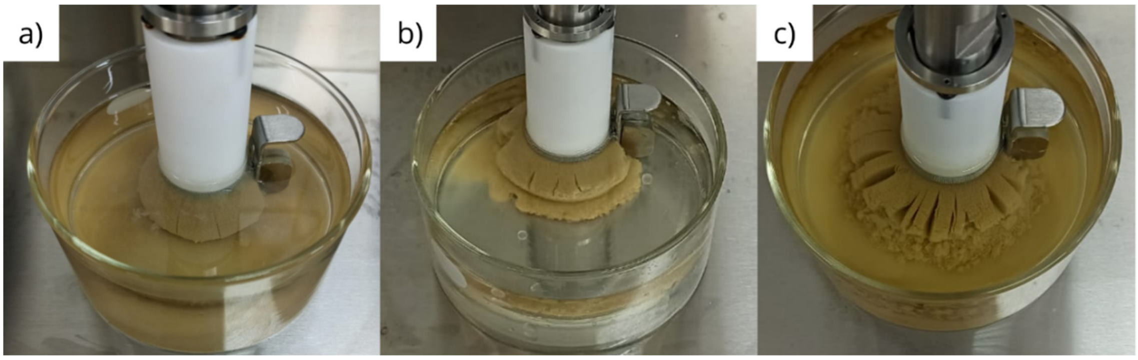

Figure 11. Visual appearance of bentonite pellets tablets after 24 hours of hydration, for samples previously immersed for 1 hour in a) diesel, b) olefin and c) glycerin.

Figures 11 (a) and (b) exhibit a similarity in the radial swelling of the tablets that were previously immersed in diesel and olefin. The material has a uniform and cohesive appearance, although small openings are observed. For the tablets previously immersed in glycerin (Figure 11(c)), many openings are observed, larger than that noted in tablets immersed in diesel and olefin, suggesting absence of cohesion between the particles.

The visual inspection of the pellets after 25 hours of testing in the LSM, together with the other tests carried out and the analysis of the results, showed that the contact of bentonite pellets with the glycerin favors the dispersion of the clay particles, compromising its physical integrity. On the other hand, diesel and olefin do not interact with the clay particles, do not interfere in the physical integrity of the pellets and, finally, do not affect their swelling, suggesting that these are promising fluid to be used on the displacement of bentonite pellets in offshore wells.

4. Conclusions

In this study, the compatibility of fluid media, consisting of saline solutions and organic compounds, with bentonite clay pellets was analyzed, aiming its application as displacement fluid in offshore oil well abandonment operations. Based on the results obtained, it was concluded that:

1. Saline solutions consisting of sodium chloride (NaCl), calcium chloride ($CaCl_2$) and potassium chloride (KCl) brines, in different saturations, compromise the physical integrity of the bentonite pellets and are unsuitable for use as displacement fluids;
2. Saline solutions with lower saturation, around 10 to 20%, promote faster and more significant disintegration of bentonite pellets;
3. The interaction between glycerin and bentonite pellets results in partial physical disintegration of the pellets, which is significantly enhanced by shear under dynamic conditions;
4. Bentonite pellets maintain their physical integrity when exposed to diesel and olefin, even under dynamic conditions;
5. Diesel and olefin seems to present minor effects on pellets hydration and swelling, since it was observed a linear swelling in deionized water of 78% after the previous contact with these fluids, which represents a decrease of only 20% on its total swelling capacity in water. It does not impair the formation of a hydraulically solid plug of bentonite in the well, neither the use of diesel and olefin as displacement fluids;
6. Among the fluids considered for displacement of pellets in offshore wells, diesel and olefin proved to be a highly promising alternative.

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