

Article



Possibilities of Limiting Migration of Natural Gas in Boreholes in the Context of Laboratory Studies

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Abstract: Gas migration through fresh and hardened cement slurry is an ongoing problem in the oil industry. In order to eliminate this unfavourable phenomenon, research is being conducted on new compositions of slurries for gas wells. The article presents the results of research for slurries with low and high resistance to gas migration. The proper selection of the quantity and quality of components makes it possible to design slurry with the required static structural strength values. In addition, the cement sheath of such anti-migration slurry has low porosity and a very low proportion of large pore spaces. Additionally, the mechanical parameters do not decrease during long-term deposition in borehole-like conditions. By obtaining these results, it was possible to design slurry whose cement sheath has high corrosion resistance. The new slurry has a lower water-cement ratio. Additionally, GS anti-migration copolymer, anti-filter additive and latex are used. The presence of n-SiO2 aqueous solution and microcement allows for sealing the microstructure of the hardened cement slurry. Such modifications significantly improve the technological parameters of the cement slurry and the cement coat formed from it.

Keywords: gas migration; well cementing; cement slurry; cement sheath; corrosion resistance; gas outflows

1. Introduction

Limitation of uncontrolled natural gas flows from the ring and/or annular space of a borehole is one of the crucial aspects during sealing of casing in boreholes drilled in formation with a higher risk of gas migration (the so-called exhalation). It relates to the main objective of casing sealing, i.e., ensuring durability and tightness between the lowered casing, on the one hand, and the drillhole wall and the previously cemented casing string, on the other [1–5]. As there are cases of gas invasion into the structure of the bonding slurry, resulting in leaks in the structure of the cement coating, it is necessary to carry out research and development work aimed at continually improving the quality of cement slurries formulations and the slurry injection technology [2,5–7]. During cement slurry bonding, in the process of its hydration, gel structure is created and at that time a clear decline of hydrostatic pressure in the ring space of a drillhole is observed. When slurry changes its structure, it is very likely that gas moves in the gelled slurry structure, which may consequently result in gas migration both through fresh and hardened cement slurry [2,3,5,8–13].

2. Uncontrolled Flows of Fluids in a Borehole

Interhorizontal flows of drilling fluids (natural gas, crude oil, brine, drilling fluid) are caused by the lack of balance between formation and hydrostatic pressures in the ring space of a wellbore.



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Uncontrolled natural gas flows within particular horizons being drilled are called migrations (drillhole no. 1, Figure 1), whereas gas flow from rock formation on the ground surface is called exhalation (drillhole no. 2, Figure 1).

Figure 1. Example of gas migration and exhalation.

The first issues of this kind were observed in the 1960s when making a borehole for gas storage. The topic of uncontrolled natural gas outflows from ring and/or annular spaces of a wellbore is very important in the context of the influence on safety, environmental protection, in terms of legal regulations, with regard to suitable management and exploitation of the borehole area. The results of studies conducted by oil companies (Schlumberger, Halliburton) prove that ca. 15% of failed cementing of a casing string in wellbores are caused by gas migration [4,14].

Presently, this problem occurs to a much higher extent, mainly on some gas formations and in boreholes prepared for gas storage. The above is confirmed by the analysis of the state of cementation of boreholes carried out (in the years 2008 to 2011) in the area of the Carpathian Foredeep and in the Carpathians, which is presented in Figure 2 [15].



Figure 2. Gas outflows from the ring space in boreholes drilled in the Carpathian Foredeep and Carpathians in the years 2008—2011. P-4—Pogwizdów 4, P-5—Pogwizdów 5, P-25—Pruchnik 25, T-6—Terliczka 6K, L-3—Łapanów 3, M-63—Mirocin 63.

In extreme situations, where an abnormally high pressure occurs after cementing of the casing on a wellhead, it may be necessary to decommission the borehole. In most cases, however, this problem is solved by using different techniques to cement the leaking zone in order to eliminate the gas flow or to reduce the pressure to a value that allows further work to be carried out in compliance with the applicable standards. Such procedures, however, are very costly, so the optimum solution is to minimise uncontrolled gas flows from the design stage of the formulation and the technological parameters of the slurry, while conducting laboratory tests [3,16–20].

3. Minimizing of Uncontrolled Gas Flows

The sealing slurry changes its physical state during hydration from the liquid phase during injection, through the transition phase—gel (slurry bonding), to the solid phase (bonded) slurry. Taking the technological properties of the sealing slurry as a criterion, it can be noted that the potential factors minimising the exhalation of natural gas after cementing include:

- Achieving the end of the slurry bonding before the hydrostatic pressure drops below the formation pressure; reducing the possibility of gas intrusion into the structure of the bonding slurry (Figure 3). Apparatus from Figure 4 was used for measurements.
- Ensuring the shortest possible transition time from 50 Pa to 250 Pa during the build-up of the static structural strength of the slurry (Figure 5). This results in the tightening of possible gas channels in the structure of the bonding slurry [3,9,12,14,18]. The apparatus from Figure 6 was used for measurements.



Figure 3. Diagram of hydrostatic pressure reduction below formation pressure.



Figure 4. Apparatus for testing gas migration during slurry bonding-hydraulic press (OGI-NRI design).



Figure 5. Diagram for the build-up of static structural strength and the formation of a transition period.



Figure 6. Apparatus for testing the build-up of static structural strength in slurry—two-chamber UCA SGSM Ofite model 120-53.

When developing sealing slurry formulations, various additives and admixtures are used in order to reduce the possibility of uncontrolled gas flows in boreholes through the setting slurry and after it has set, i.e., microcements, microsilicas, nanosilicas, which tighten the microstructure, as well as latex and cationic polymers, which shorten the gelling time of the slurry and thus reduce the possibility of natural gas flowing through the hardened cement slurry [2,13,21–24].

4. Research Findings

Laboratory tests conducted by the Oil and Gas Institute, the National Research Institute and AGH University of Science and Technology show significant differences in effectiveness of removing uncontrolled gas outflows as a result of comparing technological parameters of the base slurry, on the one hand, and the slurry after modification with selected additives and admixtures, on the other. Table 1 shows the control slurry formulation (Contr slurry) and the modified formulation (Mdf slurry). The technological parameters of the fresh slurries are summarised in Table 2. There was a significant reduction in the filtration of the slurry from a value of 150 cm³/30 min to just over 2 cm³/30 min. The values of rheological parameters have also decreased. In order to minimise the possibility of uncontrolled gas outflows at the contact between the cement coating—rock formation and the cement coating—casing pipes, the presence of gravitational water was eliminated. The changes made to the slurry formulations result in short thickening times from 30 Bearden Consistency units (Bc) to 100 Bc, thus minimizing the occurrence of gas migration during slurry bonding.

Table 1. Base slurry and slurry with selected additives and admixtures.

Parameter —	Kind of Slurry	
	Contr	Mdf
w/c water	0.54	0.45
Defoaming additive	0.30	0.50
Liquefying additive	0.20	0.20
Anti-filtration additive	0.20	0.20
GS additive		4.0
C ₂ anti-filtration additive		2.0
Latex		6.0
Latex stabilizer		2.0
n-SiO ₂ aqueous solution		6.0
Microcement		10.0
G High Sulphate Resistant (G-HSR) oil well cement	100.0	100.0
Bulking additive	0.30	0.30

Table 2. Technological parameters of the fresh slurries.

Parameter		Kind of Slurry	
		Contr	Mdf
Density (g/cm^3)		1.78	1.81
Flow properties (mm)		250	300
Filtration ($cm^3/30$ min)		150	2.2
Plastic viscosity (mPa·s)		220	61.5
Flow limit (Pa)		23.5	2.1
Structural strength (Pa)		5.8	3.6
Water separation (%)		3.0	0.0
	Bc value	Time: min.	
Thickening time	30 Bc	2–48	2–19
	100 Bc	4–43	2–29
From 30 to 100 Bc (min)		115	10

Figures 7 and 8 show the gas migration test and static structural strength build-up tests for the control slurry. This slurry obtained the end of bonding time after lowering the hydrostatic pressure value below the formation pressure. The consequence of this can be gas intrusion into the structure of the cement coating and the subsequent creation of gas migration paths. Additionally, a large amount of filtrate in the upper part of the measuring



cylinder indicates a low anti-migration efficiency of the slurry in question. During the static structural strength test (Figure 8), no transition period was obtained from a value of 50 Pa to 250 Pa.

Figure 7. Course of the gas migration test during bonding for slurry with low anti-migration resistance.



Figure 8. Static structural strength build-up for slurry with low anti-migration resistance.

Figures 9 and 10 show the results of tests on modified slurry which is guaranteed to reduce the possibility of uncontrolled gas flow. This slurry reaches the end of bonding (EB) before the hydrostatic pressure drops below the formation pressure. Thus, it shows a very good ability to maintain the hydrostatic pressure at a high level (Figure 9). Slurry filtration at the top of the cylinder was low, confirming the effectiveness of this slurry in counteracting gas migration. Furthermore, a short transition time of 52 min from 50 Pa to 250 Pa and a steady increase in structural strength were obtained (Figure 10).



Figure 9. Course of the gas migration test during setting for slurry with high anti-migration resistance.



Figure 10. Static structural strength build-up for slurry with high anti-migration resistance.

5. Analysis of the Quality and Durability of the Cement Coating with Regard to the Possibility of Uncontrolled Gas Flows

When analysing the problem of gas outflow through the cement coating, corrosion of the hardened slurry in the annular space must also be taken into account. Test results available in the literature indicate that after 30 years, the cement coating has corroded in approx. 60% of the boreholes and deteriorated its tightness, as shown graphically

in Figure 11. The main factors causing corrosion of cement coating in a borehole are formation brine, high temperature and pressure at a given depth [1,11]. The combination of this groups of factors results in chemical compounds with reduced mechanical strength and sometimes no bonding ability [4]. Therefore, in order to design the sealing slurry formulation with improved corrosion resistance, it is necessary to:

- Ensure the lowest possible content of Ca(OH)₂ (portlandite) and C₃A phase in the slurry.
- Provide the highest possible amount of calcium silicate hydrate (CSH) phase building up the cement matrix.
- Ensure that the permeability and porosity of the slurry is as low as possible (taking into account that the proportion of gel pores is greater than the proportion of capillary pores) [7,16,17,19,25].





The destructive effect of ions present in reservoir brines on hardened cement slurries can be represented by the following chemical reactions [11,19,25]:

• Double exchange between magnesium and calcium cations, e.g.,

Magnesium ions (Mg^{2+}) .

• Transformation of portlandite into brucite

$$Ca(OH)_2 + MgCl_2 \rightarrow Mg(OH)_2 + CaCl_2$$

• Extraction of calcium ions Ca²⁺ from the CSH phase and formation of brucite

$$C_{1,75}SH_n + MgCl_2 \rightarrow C_{1,75-x}SH_n + x CaCl_2 + x Mg(OH)_2$$

• Formation of expansive hydrated basic magnesium chloride

 $2Ca(OH)_2 + 3MgCl_2 + 2H_2O \rightarrow Mg_3Cl_2 (OH)_4 \cdot 2H_2O + 2CaCl_2$

$$C_{1.75}SHn + 3MgCl_2 \rightarrow Mg_3Cl_2 (OH)_4 \cdot 2H_2O + C_{1.75-x}SH_n$$

Sulphate ions (SO_4^{2-}) present in the pores of hardened cement slurry cause the formation of salts that are difficult to dissolve. This process is accompanied by a significant increase in the volume of the slurry, and consequently the formation of high tensile stresses in the cement matrix. The swelling compounds formed are gypsum dihydrate and tricalcium aluminate trisulphate hydrate called ettringite.

The hydration products most frequently attacked by sulphate ions are portlandite and C_3A and hydrated calcium aluminates:

$$Ca(OH)_2 + SO_4^{2-} \rightarrow CaSO_4 \cdot 2H_2O + 2OH^-$$

Gypsum dihydrate

 $\label{eq:action} \begin{array}{l} 3\,\text{CaO}\cdot\text{Al}_2\text{O}_3+\text{SO}_4{}^{2-}+n\,\text{H}_2\text{O}+\text{Ca}(\text{OH})_2\rightarrow 3\,\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot3\text{CaSO}_4\cdot32\text{H}_2\text{O}+m\,\text{H}_2\text{O}\\ \\ \text{Ettringite} \end{array}$

On the basis of an analysis of long-term laboratory test results on corrosion of hardened cement slurries, the following conclusions can be drawn [3,9,16,17,25]:

- In order to limit magnesium corrosion, measures are taken to achieve the lowest possible permeability and porosity of the hardened cement slurry, which reduces the penetration of aggressive Mg²⁺ ions into the cement matrix. Good results are obtained by using mineral additives (primarily granulated blast furnace slag containing small amounts of calcium compounds or fly ash). Therefore, increased resistance to magnesium ion attack is achieved by using metallurgical or ash cements.
- Prevention of sulphate corrosion is based on the use of cements with increased resistance to sulphate aggression (mainly High Sulphate Resistant (HSR) type oil well cements). Polymeric additives are also used to create a micro-membrane between the cement grains, as well as fine mineral fillers.

Tests to determine the durability of hardened sealing slurries are based on the following procedures [9]:

- 1. Selection of suitable cement slurry formulations for testing, taking into account borehole conditions.
- 2. Preparation of corrosive environments depending on the drilling area.
- 3. Preparation of sealing slurry and making of samples of hardened slurries (hardening of samples in autoclaves, Figure 12).
- 4. Exposing the samples to the selected corrosive environments for a defined exposure time.
- 5. Periodic visual assessment of samples, photographic documentation and testing of strength, porosity and gas permeability, etc.
- 6. Periodic replacement of reservoir brines (every six months).

Samples of hardened slurries are deposited under corrosive conditions for a defined period of time. In autoclaves the temperature reaches 120 °C and the maximum pressure can reach 20 MPa. A reference sample is stored in one chamber of the autoclave in tap water, which represents the reference environment.

Figure 13 shows the values of parameters describing the pore microstructure, while Figure 14 compares the mechanical parameters of samples 1 and 2. Sample 1 is a hardened cement slurry with very low corrosion resistance. This sample has a porosity factor (ratio of the volume of space between the grains to the volume of the entire sample) of 40.8%, a large proportion of the largest pore spaces, and its mechanical parameters decrease significantly during deposition. This sample deteriorated almost completely after 18 months of deposition under conditions of increased corrosivity. Sample no. 2, which was modified to achieve improved corrosion resistance, has a low share of macropores (Figure 13), a low porosity index of 28.7% and a high compressive strength, which is almost constant over time.



Figure 12. Autoclaves for storing samples of cement slurries.



Figure 13. Porosity coefficient values and shares of individual pores for samples 1 and 2.



Figure 14. Compressive strength values for samples 1 and 2.

6. Conclusions

When developing sealing slurries to minimise uncontrolled gas leaks, it is important to modify them at the design stage with additives such as microcement, microsilica, latex, high molecular weight cationic polymers, nanosilica or expansive additives.

The modified formulations discussed in this publication are characterised by appropriate densification times for the given conditions. Such values for the time of thickening allow the slurry to be pumped efficiently into the annular space to be sealed, within the time necessary to carry out the procedure. This time is increased by a safety buffer in the event of complications arising in the preparation of the slurry in the field. The designed cement slurry is characterised by a rapid transition from a consistency value of 30 Bc to a value of 100 Bc during the thickening time test. This short transition time of 10 min goes a long way towards minimising the possibility of gas migration through the binding cement slurry. After injection, the slurry begins to gel and a gas-impermeable cement sheath is formed. The designed slurry with anti-migration properties also has the required rheological parameters, which allow it to be pumped efficiently into the cemented annular space, and to efficiently displace the drilling fluid previously present in the borehole. The newly developed slurry for cementing boreholes with an increased risk of gas migration has a low filtration value and does not show any water retention. As a result, the setting and thickening time is maintained at the required level, because the slurry does not "give off" water in the absorption zones that are often found in the area where gas migration occurs. In addition, a slurry with low filtration and no water retention has a homogeneous structure in the liquid state and does not fractionate. The slurry designed for sealing gas wells is resistant to gas migration during setting, as confirmed by the Static Gel Strength Measurement (SGSM) test. With a transition time of 52 min, it can be concluded that the gas will not be able to penetrate the structure of the cement slurry once it is injected.

When analysing the results of tests on the structure of the cement sheath made of slurry with increased resistance to gas migration, it is found that such a formulation has a significantly lower porosity than standard slurries, and a smaller proportion of pore spaces with the largest diameters. Such conditions confirm the required sealing of the borehole and the elimination of microflows through the cement sheath structure. The mechanical strength values obtained for 18 months of deposition under borehole conditions confirm the erosion resistance of the designed slurry. The samples have comparable strength throughout the storage period, with strengths of over 25 MPa. In contrast, the strength of the control sample decreased due to erosion. The strength of this control sample decreases until, after a final period of exposure to erosive conditions, the sample is destroyed.

Long-term tests of changes in mechanical strength, gas permeability, porosity and microstructure are very important in designing an anti-migration slurry. Such tests allow an objective assessment of the course of destructive processes occurring in the cement sheath during long-term exposure in borehole conditions. Research into the durability of hardened cement slurries is required to improve formulas and develop new slurries for sealing boreholes in various geological and technical conditions.

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