



Study of Additives for Self-Healing of Cement Stone Under Hydrocarbon Gas Migration Conditions

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Abstract

This study addresses the long-standing issue of cement sheath integrity in oil and gas wells, a challenge that persists despite over 40 years of research. The focus is on self-healing cement systems, which can repair cracks either autogenously (through unhydrated cement particles reacting with water) or autonomously (using additives like swelling agents or bacteria). However, limitations such as crack size and material compatibility require tailored solutions. Rubber and Natural Rubber (NR) additives, in solid and liquid forms, were tested as modifying agents. The key requirement was their ability to block channels larger than 0.3 mm, with agglomeration and activation prevented by treatment with hydrophobizing agents and dry cement. Building on previous success with oil-swelling additives, the study shifts focus to gas and gas condensate reservoirs. A custom testing stand was developed to simulate downhole conditions, creating artificial channels in cement with diameters of 0.8 mm. The setup demonstrated that certain additives could seal these channels within 3 to 7 days. The findings contribute to understanding additive performance and offer a step toward replicating wellbore conditions in the lab. This technology has the potential to enhance zonal isolation, reduce remediation costs, and improve safety in challenging reservoir environments.

Keywords: Modifying additive; Self-healing cement; Well completion; Zonal isolation; Gas and gas condensate fields.

Received: 17 April 2025; Revised: 05 May 2025; Accepted: 22 May 2025.

Article type: Research article.

1. Introduction

Modern oil and gas well drilling originated in the mid-19th century in Baku. Since then, drilling technologies have made significant advances: well depths have increased dramatically, and so have the complexities of wellbore stabilization and operational integrity. However, despite rapid technological progress in the oil and gas industry, issues related to high-quality well cementing remain unresolved. Complications such as inter-zonal fluid migration and annular pressure buildup can lead to water flooding and contamination of extracted resources.^[1,2] Due to its direct contact with the

reservoir, the cement sheath is particularly vulnerable to various mechanical and thermal stresses. These can result in cracking of the cement matrix and loss of zonal isolation, causing gas, oil, or water inflow, surface seeps, and crossflow between formations. Such failures may lead to severe environmental consequences and, in extreme cases, loss of human life.

Currently, the most common method of restoring well integrity is remedial squeeze cementing. During this process, a plugging slurry is injected into the annular space through pre-perforated casing sections. However, this technique has several disadvantages, including difficulty in precisely locating the leakage zone and high operational costs. Furthermore, the geometry of modern wells prevents direct human access to critical areas, making diagnostics and intervention extremely challenging. Therefore, there is a growing need for innovative technologies that can maintain well integrity autonomously, without human intervention. One of the most promising but underexplored approaches is the development of self-healing cement systems. These materials could not only improve the durability and safety of wells but also significantly reduce repair and maintenance costs.^[3-8]

Self-healing cement slurries have been under investigation for over 40 years by both international and domestic

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companies. The core concept involves sealing cracks that form in the cement sheath using advanced additives—such as chemical agents or composite modifiers—that activate upon exposure to formation fluids. The concept of self-healing polymer-based materials was first introduced in the 1980s. In 1981, Donald Judd demonstrated that polymer-based agents could seal cracks in cement, thereby extending its service life.^[9] There are three major types of self-healing cement systems: encapsulated systems, vessel-contained systems, and unencapsulated systems. The main difference between these lies in their activation mechanisms. The activation process typically involves three stages: a) crack formation due to mechanical overload; b) ingress of formation fluids, which act as a trigger for the additive; c) blockage of the fluid migration path by expansion or chemical reaction of the additive. As a result, fluid movement is halted, and zonal isolation is restored.

Using self-healing cement can therefore reduce both direct and indirect costs related to remediation. Moreover, multiple studies have confirmed the feasibility of autogenous healing, a natural process where cracks are sealed without additional modifiers. This process relies on the hydration of unreacted cement particles when water penetrates the fracture network.^[10] However, autogenous healing has a key limitation—it is effective only for cracks no wider than 0.3 mm.^[11-14]

Gas and gas-condensate fields, such as Urengoy, Kovykta, Chayanda, and Bovanenkovo, represent some of the most challenging environments for well construction in Russia. These sites are characterized by permafrost zones, gas hydrate formations, gas-saturated horizons, shallow losses, and cryopegs (saline frozen formations), which significantly complicate cementing operations. Conventional primary cementing techniques often fail under such conditions, resulting in compromised zonal isolation, annular pressure buildup, and gas or fluid migration. These challenges highlight the critical need for advanced, self-healing cement solutions

tailored for use in complex reservoirs.

2. Materials and methods

After a series of successful studies on the effect of water-oil-swelling additives on the self-healing ability of cement stone, the focus shifted to the issue of the sealing of gas and gas-condensate wells. Therefore, the task was set to develop a new cementing material whose cement stone would operate based on the principle of self-healing, *i.e.*, blocking the influx of reservoir fluids (gas) with the help of modifying additives.^[15,16]

After interacting with the reservoir fluid, swelling particles become activated and close the cracks. The main requirements for the modifying additive are: a significant swelling coefficient to block large channels (greater than 0.3 mm) and no premature activation during the preparation, pumping, and displacement of the cement slurry. Additionally, the effect of the additive on the basic properties of the cement slurry and the resulting cementing material should be minimal.

The following materials were considered as modifying additives: rubber PK23, polymers KP654 and KP785, as well as an additive natural Rubber (NR) in both solid and liquid forms. Rubber PK23 is an elastic, high-molecular-weight material obtained by vulcanizing natural rubber. Its structure consists of a chaotic polymer chain of hydrocarbons connected by sulfur atoms. Polymers, like the above-mentioned materials, are a general term for high-molecular-weight substances made up of large numbers of atomic groupings.^[17-20]

NR is a high-molecular-weight unsaturated polymer of isoprene with many double bonds. A distinctive feature of this reagent is its elastic-plastic properties: NR can restore its original structure after being subjected to external forces; NR can maintain its shape once it has been deformed by external forces. Additionally, NR has low gas permeability and is insoluble in water and alkalis.

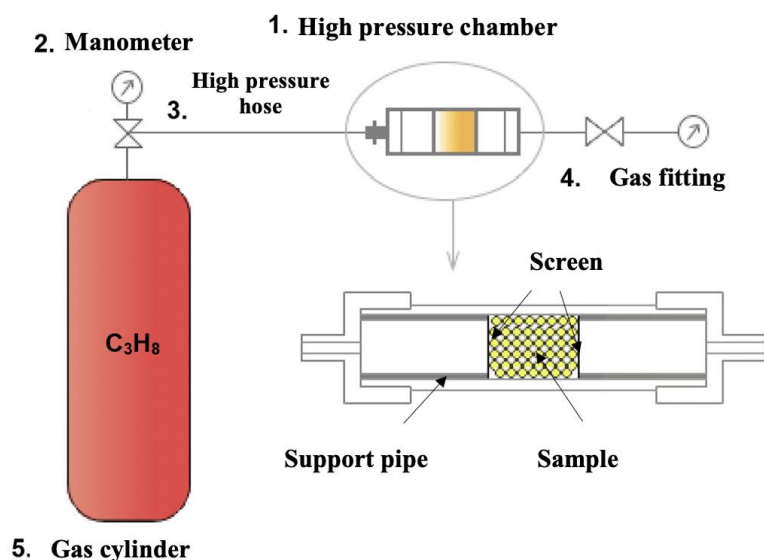


Fig. 1: Schematic representation of the testing stand.

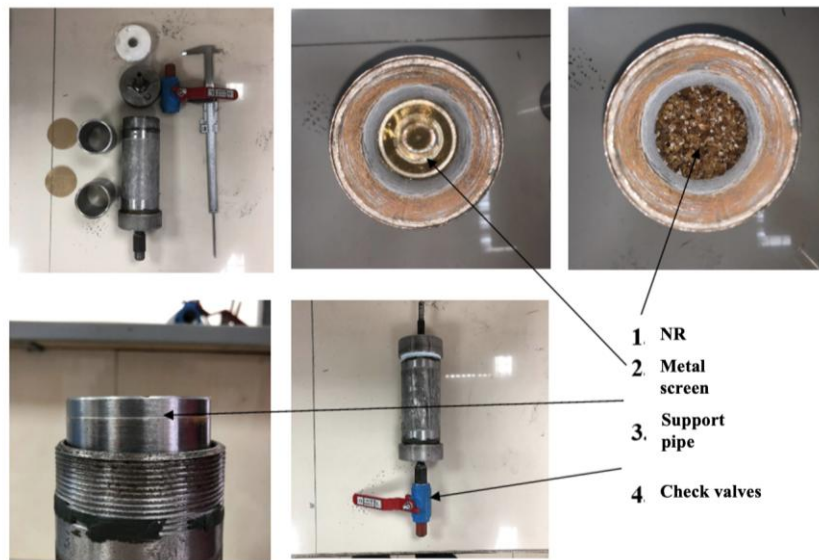


Fig. 2: Preparation of the equipment.

A special testing stand was designed for the experiments to simulate the flow of liquid hydrocarbons through artificial channels in cement stone. The testing stand consists of a sealed base, cylindrical plastic pipes with a diameter of 50 mm, which serve as molds for pouring the cement slurry, wires with cross-sectional diameters ranging from 0.3 to 0.7 mm, a stand to secure the entire structure, a 200 ml graduated cylinder, a timer, and a pipe for generating hydrostatic pressure.

The study of gas permeability in cement samples with artificial cracks was conducted using the stand with gas injection (Figs. 1-2 and Fig. S1). The stand consists of three production tubing pipes with a diameter of 76 mm, plugs with openings for gas hoses, a support for the tubing, laboratory burners, and systems for supplying propane and methane. The purpose of the experiment is to simulate well conditions where the cement sheath loses its integrity. Gas passes through channels in the cement stone, thereby activating the self-

healing additive.

To simulate channels in the cement stone, wires of the same diameter were installed at equal distances from each other inside a 20 cm long cylindrical pipe, thereby modeling channels of a specific size. Wires with diameters of 0.3, 0.5, and 0.7 mm were used in the experiments; they were fixed to a sealed base and positioned strictly vertically, without bends or irregularities. The pipes were filled with cement slurry prepared using Class G cement and a water-to-cement ratio (W/C) of 0.44, with varying amounts of modifying additives. The cement stone samples in the pipes were then cured for 10–12 hours in an air-humid environment. After curing, the wires were removed, leaving behind artificial channels in the cement stone. The samples were stored for an additional period before being tested on the experimental setup. For each test, 3 to 5 parallel experiments were conducted, and the final result was taken as the average value.

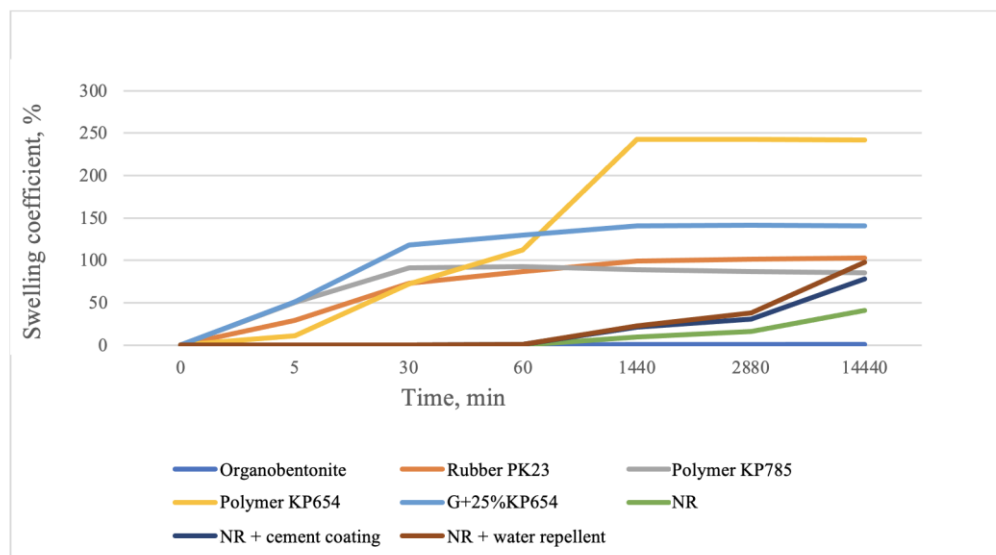


Fig. 3: Swelling coefficient of the additives.

Table 1: Swelling coefficient of the additives.

Additive	Time, min						
	0	5	30	60	1440	2880	14440
Swelling coefficient, %							
Organobentonite	0	0	0	0	1	1	1
Rubber PK23	0	30	73	87	99	102	103
Polymer KP785	0	51	92	93	89	87	85
Polymer KP654	0	11	72	112	242	242	242
G + 25% KP654	0	51	118	130	141	142	141
NR	0	0	0	0.50	10	16	41
NR + cement coating	0	0	0	0.87	21	30	78
NR + water repellent	0	0	0	1.00	23	38	98

3. Results and discussion

At the initial stage of the study, the swelling kinetics of the additives were examined using a portable network graphics (PNG) device according to the Zhigach-Yarov method, which demonstrated their inertness to water. Subsequently, the swelling of the additives was investigated in a gas condensate environment. Table 1 and Fig. 3 present the results of the swelling of the examined additives.

The polymers KP654 and KP785, as well as the rubber PK23, reach their maximum values within the first few hours of measurements, which does not meet the required specifications. Therefore, it was decided to conduct further experiments only with NR as the modifying additive. The main issue when working with NR is the re-agglomeration (clumping) of the particles after grinding. This problem was

solved by treating the NR particles with a water repellent agent and applying a dry cement coating to prevent particle clumping. The swelling graphs of NR treated with a water repellent agent and dry cement are shown in Fig. 4.

According to Fig. 4, NR treated with cement and a water repellent agent swells more actively than pure NR. This means that the treated additive has a larger contact area with the condensate. As an alternative, an additive in the liquid form of NR, as well as in powder form, was developed. Figs. S2a and b show micro-particles of cement with 20% liquid NR content. Further examination of the stone revealed elastic NR films that bind the cement particles together (Fig. S3). Next, a study on the swelling properties of the powder in a hydrocarbon environment was conducted. The results of the swelling of the powdered NR are presented in Fig. 5.

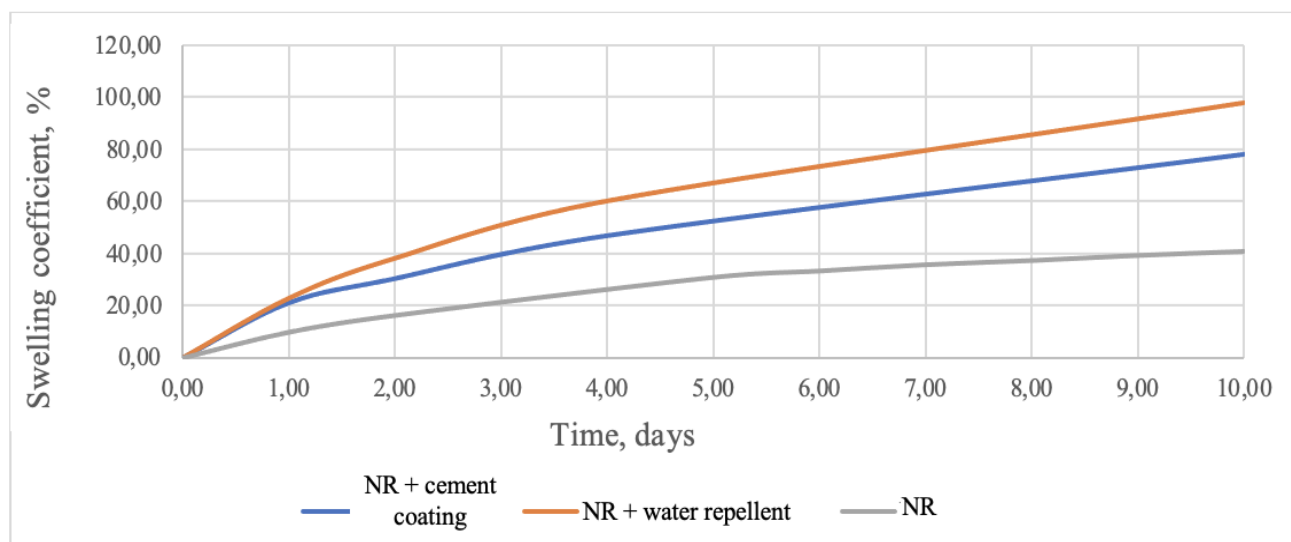


Fig. 4: Effect of surface treatment of additives on the kinetics of their swelling in gas condensate.

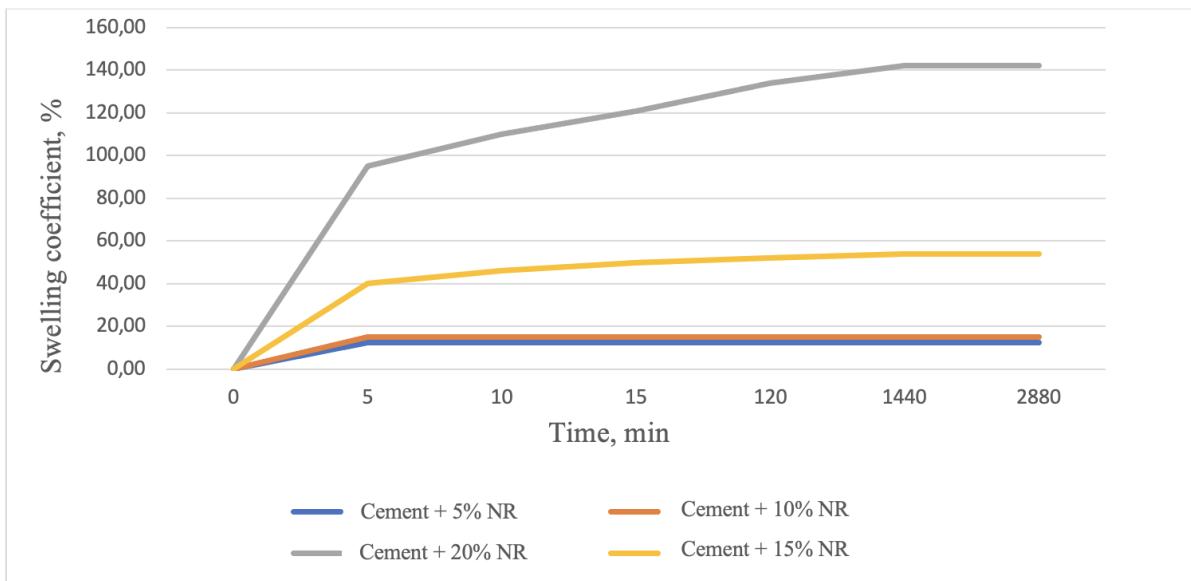


Fig. 5: Results of swelling of cement powder with the addition of NR.

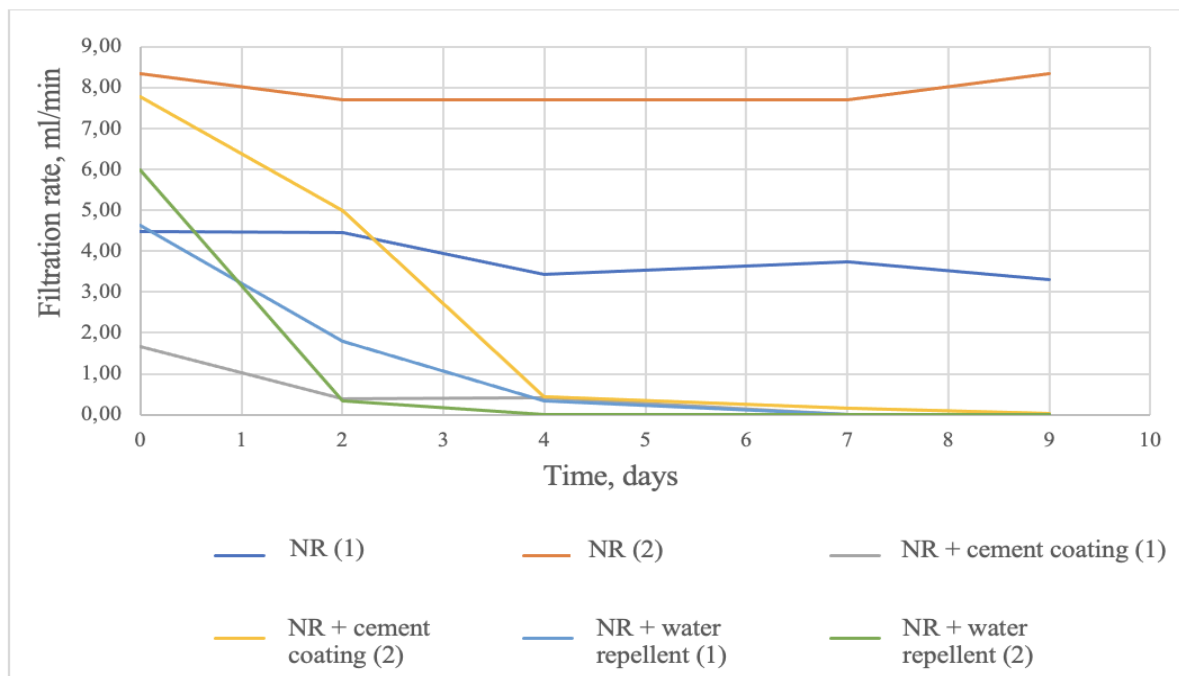


Fig. 6: Filtration rate of condensate through cement stone.

As can be seen from Fig. 6, the best swelling values are achieved with the addition of 20% NR. The next stage of the research was the evaluation of the self-healing capacity of cement stone containing NR. The experiments used Portland cement of the grade PCT-I-G-50 and a specially designed testing stand.^[13,14] Artificial channels with a diameter of 0.8 mm were created in the cement stone. Then, samples of cement stone treated with a hydrophobic agent and cement powder, as well as a control sample with normal density and a water-to-cement ratio of 0.5, were tested on the apparatus. To assess the self-healing ability, the time for 50 mL of gas condensate to flow through the cement stone samples was measured.

Self-healing performance was also evaluated for samples containing liquid NR and NR powder. The results are shown in Fig. 7. As the data demonstrates, the NR additive is effective in both liquid and powdered forms. Within five days of testing, the condensate filtration rate through artificial channels decreased by up to 60%. Fig. 7 shows that samples treated with a surface coating exhibited a more rapid decrease in condensate flow rate compared to the untreated samples.^[21,22] The obtained results prove that the NR additive blocked all filtration channels in the cement stone, which suggests the successful completion of the experiment.

The next stage of the study involved evaluating the swelling behavior of samples in condensate vapor and gas

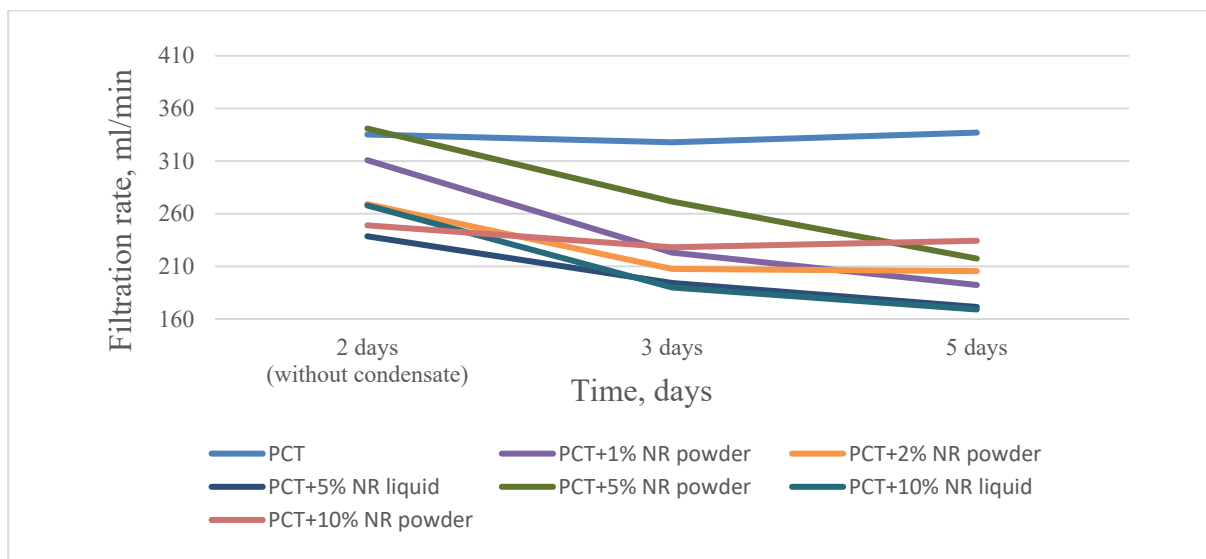


Fig. 7: Filtration rate of condensate through cement stone with artificial channels.

environments. The experiment used gas condensate obtained from a field located in the Republic of Bashkortostan. The NR samples were exposed to condensate vapor for 15 days. No visible changes were observed during this period, so it was decided to continue the experiment using gas, specifically propane. A tubing pipe was prepared for this purpose. The NR additive was placed inside the pipe and secured with a screen to prevent chaotic movement and ejection during gas injection. After seven days of exposure, no noticeable swelling of the additive particles was recorded. This was attributed to insufficient pressure in the system.

After upgrading the equipment, the experiment was

repeated under higher pressure to better simulate reservoir conditions. A gas cylinder with a maximum pressure of 3 atm was used. To evaluate the swelling of the reagents, two parameters were monitored: the mass and volume of the additive before and after gas exposure. According to the results, the sample mass increased by 6.17%, and the volume increased by 12%. Visual inspection showed that the additive particles became softer, more elastic, and began to adhere to each other, forming a cohesive structure. A microscopic analysis was then performed to examine external and structural changes at magnifications of 500:1 and 3000:1 (Figs. 8 and 9).

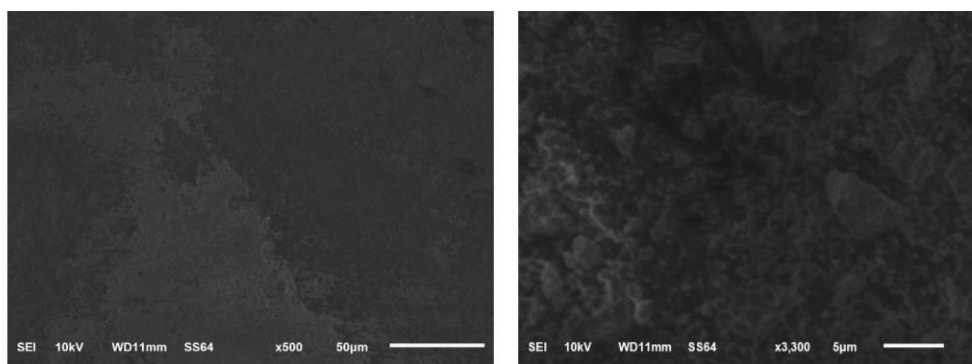


Fig. 8: Structure of NR under the microscope before gas exposure.

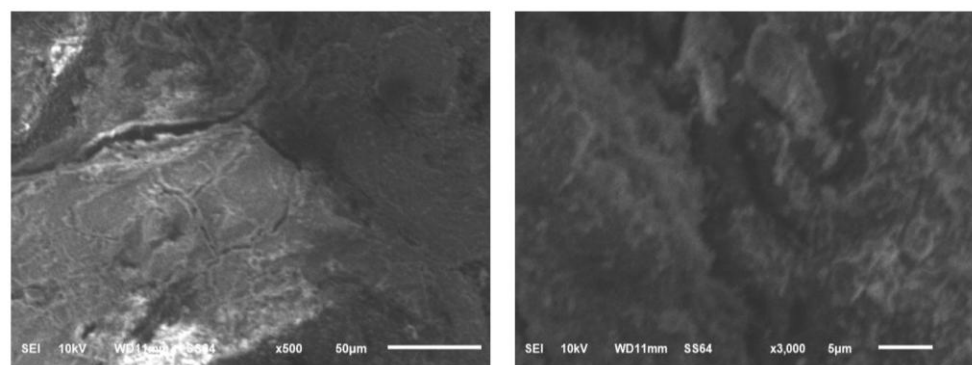


Fig. 9: Structure of NR under the microscope after gas exposure.

Table 2: Results of gas filtration through samples.

Sample	Time, h		
	24	72	168
PCT	Gas-permeable	Gas-permeable	Gas-permeable
PCT + KP654 (5%)	Gas-permeable	Gas-permeable	Gas-permeable
PCT + rubber PK23 (5%)	Gas-permeable	Gas-tight	Gas-tight
PCT + NR (5%)	Gas-permeable	Gas-permeable	Gas-permeable

As shown in Fig. S3, exposure to propane caused the formation of microcracks on the surface of the NR particles. A distinct white coating appeared, and the overall appearance of the particles changed from transparent to milky. This indicates that the gas penetrated the NR's structural matrix. Therefore, increased system pressure has a positive effect on the swelling dynamics of the modifying additive. The results of propane gas filtration through samples containing modifying additives and artificial channels are shown in Table 2. These results demonstrate that complete sealing was achieved using PK23 rubber. At this stage, the NR additive is not capable of blocking gas migration due to the large particle size. The next step will be extended testing using fine-dispersed liquid and powdered forms of NR.

4. Conclusion

The following conclusions were drawn from the conducted research:

- The selection of modifying additives capable of forming self-healing cement systems resistant to hydrocarbon environments was substantiated.
- Formulations for two-layer coatings were developed and tested under hydrocarbon exposure, demonstrating their effectiveness in restoring sealing integrity.
- For the first time, a testing stand was designed, constructed, and validated to simulate hydrocarbon gas migration in the event of zonal isolation failure. The design concept of the stand can be applied for testing well cement systems under realistic field conditions involving gas, gas condensate, or crude oil.
- A method for the preparation and mechanical grinding of a natural rubber-based modifying additive was developed.
- The inclusion of this additive enabled the sealing of channels up to 0.8 mm in diameter within the cement stone within 7 days and completely eliminated its permeability during gas condensate flow.
- The gas condensate used in the experiments was obtained from an underground gas storage facility located in the Republic of Bashkortostan.
- The experiments revealed that hydrocarbon gas pressure has a significant impact on the ability of the additive to seal microchannels within the cement matrix—an important factor to consider when designing cement compositions for specific

field conditions.

The results obtained in this study can be used for the development and optimization of self-healing cementing materials tailored for operation in oil and gas fields with various types of formation fluids.

Acknowledgements

This article was carried out with the financial support of the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan (№AP 25793601).

Conflicts of Interest

There is no conflict of interest.

Supporting Information

Applicable.

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