

Laboratory Researches of the Polymeric Composition in the Pore Space of Bulk Models

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Abstract

This article describes the results of laboratory filtration and microtomography researches of polymeric composition based on hydrolyzed polyacrylonitrile. We detected changes of filtration and rheological properties of the polymeric composition in the pore spaces according to the time and distance and to removal of the model away from the bulk end when simulating the in-situ water shutoff. As a result of the filtration and microtomography researches we proposed a graphic-analytical method of calculation of the relaxation time of the polymeric composition which is a viscoelastic liquid. Later it is used to describe the flow of viscoelastic liquid in the pore space. The results of evaluation of the relaxation time in dynamic tests in the oscillating voltage mode and the relaxation time value obtained with the help of graphic-analytical method, have high precision and reliability, and can be described by the same equations that allows us the use of the experimental data for the calculation of filtration parameters at injection of viscoelastic liquid. The proposed method of processing the rheological, filtration and microtomography researches of hydrolyzed polyacrylonitrile can be recommended as a method for describing the flow of a viscoelastic liquid. This method allows choosing waterproofing composition with optimum properties with regard to its rheological and filtration properties and modes of its injection into productive formation, it allows to estimate the permeated volume of gel, its distribution in the pore space, change of rheological properties during filtration, pressure gradient of injection at different time points and different parts of the pore space.

Keywords: in-situ water shutoff, polymeric composition, filtration researches, microtomography researches, rheological researches.

INTRODUCTION

For a more detailed study of the process of filtration and penetration of the polymeric composition into the pore space, we offered microtomography researches [1] of bulk models after injection of water isolating composition with a radiopaque substance.

MAIN PART MICROTOMOGRAPHY RESEARCH METHODS

During the microtomography research of core-samples [2,3], in this case of bulk model, the presence of a model of formation water in it, of the polymeric composition, and possibly entrained air brings up a problem of the precise and clear separation of these phases when taking the results of experiments. To do this, the radiopaque substance is used. For the water-soluble reagents the iodine-containing compounds are often used (for example, sodium iodide, potassium iodide). We used potassium iodide according to GOST 4232-74 (State Standard) as an X-ray contrast agent. The concentration of KI was chosen experimentally taking into account the absorption coefficient of X-ray produced by bulk model material. To make a clear visualization and to identify the saturating substance in the composition of the model, it is necessary that the X-ray density of the saturating liquid was much larger than the X-ray formation density). So, we determined that adding 0,5 wt%. of potassium iodide to the polymeric composition results in getting a radiopaque image of system "bulk model – polymeric composition – model of formation water".

Microtomography researches were conducted on a computer high-energy microtomograph SkyScan 1173 at the Department of Historical and Dynamic Geology of the Mining University.

Three-dimensional visualization of bulk models is presented in figures 1, 2.

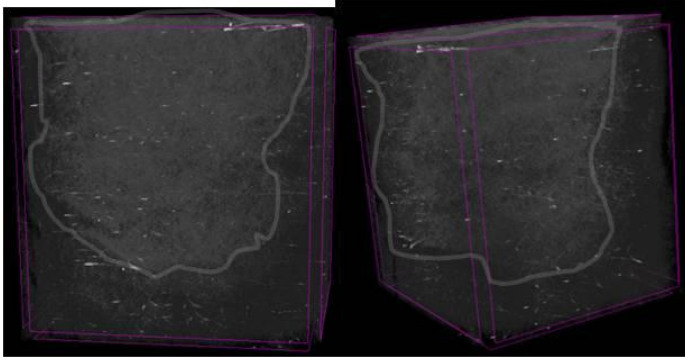


Figure 1. Visualization of bulk model №8 after injection of the polymeric composition in the three-dimensional space ($K_{perm}=8,11 \cdot 10^{-3} \mu m^2$)

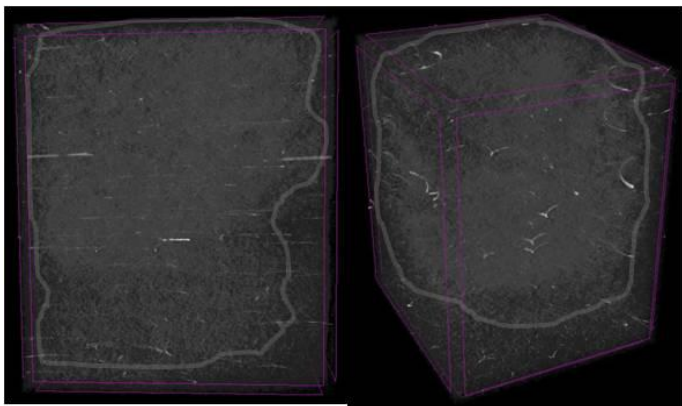


Figure 2. Visualization of bulk model №2 after injection of the polymeric composition in the three-dimensional space ($K_{perm}=20 \cdot 10^{-3} \mu m^2$)

The permeated polymeric composition with potassium iodide has a lighter colour, as it has a higher X-ray density than the surrounding area of the bulk model. To separate one thing from another the area with water isolating composition is marked with a gray line.

The bulk models tomograms show that the polymeric composition has different ways of penetration into the pore space: at the entry the composition is more solid (higher contrast) while in the remote parts the composition is present in smaller amounts (lower contrast).

RESULTS OF THE MICROTOMOGRAPHY RESEARCHES

Interpretation and analysis of the obtained results of microtomography researches were carried out using CT-analyser

program for a full two-dimensional and three-dimensional quantitative image analysis. The following parameters were obtained: pore volume, porosity, specific area, volume equivalent diameter (pore diameter), etc. All these parameters were obtained for each of the pores within the acceptable resolution of the instrument, that conditions a large amount of data.

The most valuable parameter is the pore diameter which determines the pore volume. There are various patterns to describe the pore spaces of the formation.

The most relevant is the representation of the pore space as conducting channels [4]. For a more accurate calculation of the volume of the permeated polymeric composition, the bulk model was provisionally divided into equal cylinders each 2.5 mm high. The calculation of the volume of permeated polymeric composition V_{pol} is reduced to summing volumes of these channels of different diameters filled with waterproofing composition:

$$V_{pol} = l \times \sum_{i=1}^n S_i \quad (1)$$

l – segment length ($l=2,5$ mm), mm;

S_i – area of the i -th channel, mm^2 , which is defined by the formula:

$$S_i = \frac{\rho \times d_i^2}{4} \quad (2)$$

d_i – diameter of the i -th channel, mm.

In case of the spherical shape of the pore, the volume of the permeated polymeric composition V_{pol} can be determined by the formula:

$$V_{pol} = \frac{4}{3} \times \rho \times \sum_{i=1}^n d_i^3 \quad (3)$$

When analyzing the amount of pores, their diameters were grouped according to the following dimensions, mm: 0,01-0,015; 0,015-0,02; 0,02-0,025; 0,025-0,03; 0,03-0,035; 0,035-0,045; 0,045-0,05; 0,05-0,055; 0,055-0,06; 0,06-0,065; 0,065-0,07; 0,07-0,075; 0,075-0,08; 0,08-0,85; 0,085-0,09; 0,09-0,95; 0,095-0,1.

It was determined that the maximum number of pores occupied by the polymeric composition corresponds to the range of pores with a minimum diameter 0,01-0,015 mm (figure 3), which, apparently, are the most sensitive to changes of the rheological properties of the polymeric composition: during the time it takes before the polymeric composition penetrates to a remote part of the bulk model, the effective viscosity of the composition increases.

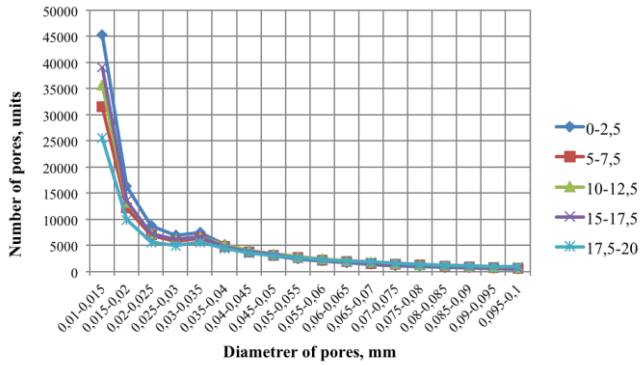


Figure 3. Distribution of the number of pores in the bulk model №8 with the permeated polymeric composition according to their sizes

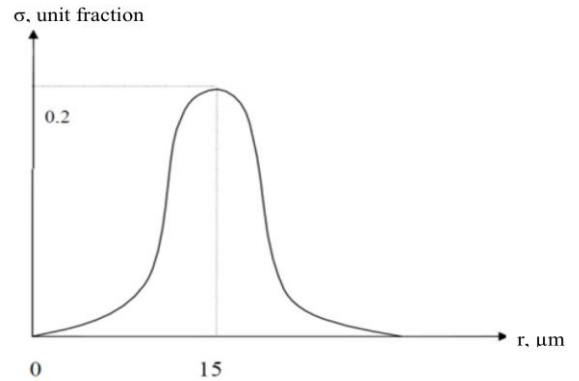


Figure 5. Distribution of pores according to their sizes

An important property of the porous body is the distribution of the volume of pores according to their radii. This relation is described by the integral and differential curves (Picture 4).

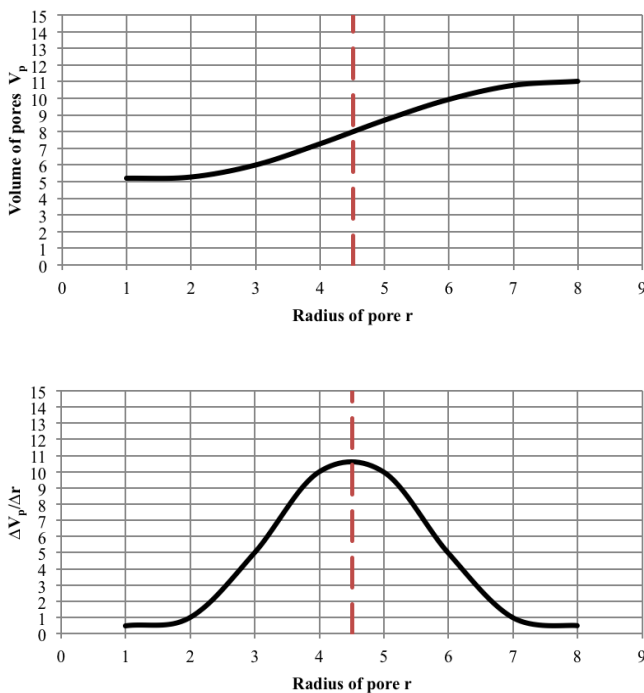


Figure 4. Integral (top) and differential (bottom) curves of pore distribution according to their radii (the numbers are conditional)

The red line shows the maximum change of the pore volume per change unit of their radii that is a sign of the maximum pore volume in which a liquid can be contained. Figure 5 shows a normal pore distribution (differential curve) in terrigenous reservoir according to their sizes, where pores with the sizes $d_{V_{maxpore}}=0,015$ mm [5] prevail.

We built integral curves depending on the distance to the input end of the bulk model. These curves describe the distribution of the volume of the permeated polymeric composition according to pore sizes (figure 6). The red line corresponds to the largest possible volume of the pores, wherein the pore diameter is 0,03 mm (the range of pore diameters is 0,01-0,1 mm), which indicated the reliability of the results.

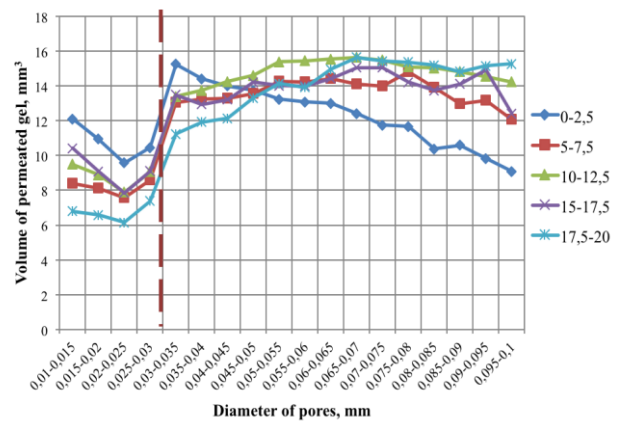


Figure 6. Distribution of pore volume of the bulk model №8, that contains polymeric composition, according to their sizes (the range of pore diameters is 0,01-0,1 mm)

If we analyze the entire range of pore sizes containing the polymer composition, then the maximum amount of permeated water resistant composition is in pores with a diameter equal to $d_{V_{maxgel}}=0,035-0,11$ mm (figure 7).

This fact indicates that not the entire maximum possible pore volume is occupied by the polymeric composition. Thus, we can make a conclusion that when choosing a polymeric composition for in-situ water shut off it is important that the composition was able to permeate into the pores with a diameter which corresponds to the maximum pore volume (in this case $d_{V_{maxpore}}=0,03$ mm):

$$d_{V_{maxgel}} \in d_{V_{maxpore}} \quad (4)$$

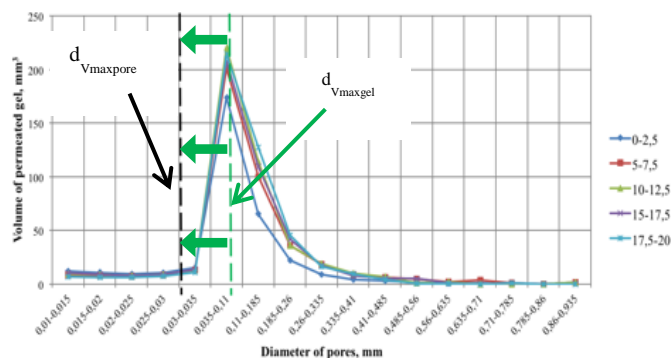


Figure 7. Distribution of the pore volume of the bulk model №8, containing polymeric composition according to their sizes (the range of pore diameters is 0,01-0,935mm)

In case this inequality is not satisfied, the polymeric composition does not fill the pore volume completely due to its high effective viscosity and elastic properties. During the working process [6] we noted that together with a decrease in clearance that shapes the

pore channel, the effective viscosity of the polymeric composition increases.

The Table 1 shows the results of the microtomography researches and calculations.

Table 1. Results of radiography researches, the pore diameter range is 0,01-0,1 mm

№	Parameter		Pore quantity n, units	Pore channel area S, mm ²	Polymer volume V _{cyl} , mm ³	Polymer volume V _{spher} , mm ³
1	Pore sizes, MM		0,01-0,015			
2	Distance to the input end of the bulk model	0-2,5	45362	4,84	12,09	0,30
3		5-7,5	31591	3,37	8,42	0,21
4		10-12,5	35663	3,80	9,50	0,24
5		15-17,5	39068	4,16	10,41	0,26
6		17,5-20	25510	2,72	6,80	0,17
7		20-22,5	48984	5,22	13,05	0,32
8		22,5-25	21257	2,27	5,66	0,14
9	General		247435	26,38	65,94	1,64
...						
10	Pore sizes, MM		0,095-0,1			
11	Distance to the input end of the bulk model	0-2,5	487	3,63	9,07	1,88
12		5-7,5	649	4,84	12,11	2,52
13		10-12,5	763	5,69	14,24	2,96
14		15-17,5	666	4,97	12,41	2,58
15		17,5-20	817	6,10	15,26	3,18
16		20-22,5	778	5,80	14,51	3,02
17		22,5-25	711	5,31	13,27	2,76
18	General		4871	36,35	90,86	18,90

THE RESULTS OF FILTRATION AND RHEOLOGICAL RESEARCHES

To make an in-situ waterproofing it is necessary to know the changes of filtration and rheological properties of the polymeric composition at different moments of time and in different areas of

the pore space (figure 8). The results of complex filtration, rheological and radiography researches are presented in table 2.

Table 2. Results of complex filtration and radiography researches

№	Distance to the input end of the bulk model	Time t, s	Effective viscosity, mPa *s	Injection pressure gradient, MPa/m	Volume of the permeated gel, mm ³
1	0-2,5	41	1410	3,47	58,30
2	5-7,5	93	1820	4,49	45,80
3	10-12,5	148	2170	5,65	48,75
4	15-17,5	201	1910	7,32	49,94
5	17,5-20	256	2570	9,76	38,12
6	20-22,5	318	2840	15,15	-
7	22,5-25	360	3040	22,47	33,16

The volume of the permeated polymeric composition was calculated according to pore diameters 0-0,015; 0,15-0,02; 0,02-0,025; 0,025-0,03; 0,03-0,035, which were the most sensitive to the changes of rheological properties of gel.

It is obvious that the injection pressure gradient of viscoelastic liquid is not described with the linear law of filtration (figure 8): with an increase in the effective viscosity of the polymeric composition the pressure drop increases exponentially.

- k – permeability, μm²;
- gradP – injection pressure gradient, Pa/m;
- A – constant factor (A=10) (Wisler E. M., 1970);
- θ – relaxation time, s.;
- d – referential pore size, mm.

Based on the fact that the parameters of viscoelasticity of the polymeric composition depend on the size of pore area, we assumed that the thickness of the polymer layer affects the relaxation time of gel. For this purpose we assessed the relaxation time of the initial polymeric composition for different clearance sizes: 0.275 mm, 0.975 mm, 5.225 mm (Table 3). As the relaxation time we took the time necessary to make the voltage e (2,72) times lower.

Table 3. Results of determination of gel relaxation time for polymer layers of different thickness during the dynamic tests

№	Parameters	Value		
1	Polymer layer thickness h, mm	0,275	0,975	5,225
2	Relaxation time θ, s	17	71	400

At the same time we assessed the polymeric composition relaxation time based on the results of filtration and microtomography researches, i.e. the inverse problem was solved. We used the formula (5) to calculate the relaxation time with different pore diameters d_i (based on results of radiography researches) and filtration speed w_i through these pores with the known values of injection

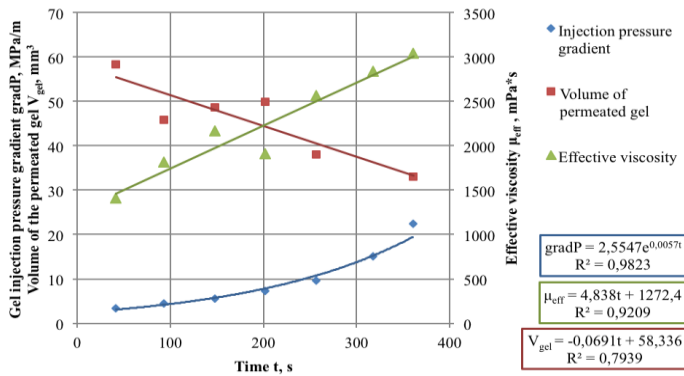


Figure 8. Relation of injection pressure gradient, effective viscosity and volume of the permeated polymeric composition to the time

To describe the flow of viscoelastic liquid in a porous medium we use a formula that takes into account the relaxation time θ of a referential pore size d [7]:

$$\vec{\omega} = -\frac{\mu}{k} \cdot gradP \cdot \left(1 + A \left(\frac{\omega \cdot \theta}{d} \right)^2 \right) \quad (5)$$

where ω – filtration speed, m/s;
 μ – viscosity, mPa*s;

pressure gradient gradP, mobility of polymeric (k/μ) composition at the given moment of time:

$$\theta_i = \frac{d_i}{\omega_i} \cdot \sqrt{\frac{1 - \frac{k \cdot \omega_i}{\mu \cdot \text{gradP}}}{A}} \quad (6)$$

Herewith we calculated the filtration speed w_i for each diameter using the following formula:

$$\omega_i = \frac{Q_i}{S_i} \quad (7)$$

Q_i – fluid flow rate (polymeric composition) through i-th channel, mm^3/s ;

S_i – cross sectional area of i-th channel (based on results of radiography researches), mm^2 .

We determined fluid flow rate according to a certain volume (based on results of radiography researches) in each filtration channel V_i and time t_i :

$$Q_i = \frac{V_i}{t_i} \quad (8)$$

Time t_i spent on a filtration of polymeric composition into pore space is determined by boundary conditions, more specifically by the values of injection pressure gradient at which gel filtration takes place. We used graphical method to determine boundary conditions for polymeric composition filtration: $0,18 \text{ MPa/m} \leq \text{gradP} \leq 3,27 \text{ MPa/m}$.

For each channel with diameter d_i we calculated the values of injection pressure based on the results of the filtration researches. As an example, Table 4 shows the calculation results for gel injection pressure gradient into filtration channels with average diameters range 0,01-0,015mm, 0,015-0,02mm, 0,02-0,025mm, 0,03-0,035mm.

Table 4. Results of calculations of gel injection pressure gradient for different pore diameters

№	Time t, s	Average pore diameters d_i , mm			
		0,0125	0,0175	0,0225	0,0275
2		gradP, MPa/m			
3	2	1,11	0,70	0,53	0,27
...					
52	100	3,32	2,11	1,58	0,82
...					
102	200	6,65	4,21	3,16	1,64
...					
152	300	11,08	7,02	5,27	2,73
...					
202	400	16,62	10,53	7,90	4,09

We used the above mentioned formulas to calculate the gel relaxation time for different diameters of pore channel. (Table 5).

Table 5. Results of calculations of polymeric composition relaxation time based on the results of the filtration and radiography researches

№	Parameters	Values				
1	Average pore diameter d, mm	0,0125	0,0225	0,0325	0,0725	0,2225
2	Volume of permeated gel V, mm^3	65,94	53,31	90,97	1444,4	252,03
3	Filtartion time t, s	90	216	524	402	402
4	Volume flow rate Q, mm^3/s	0,7327	0,2468	0,1736	3,5929	0,6269
5	Cross sectional area of pore channel S, mm^2	26,38	21,32	36,39	577,74	100,81
6	Filtration speed w, mm/s	0,0278	0,0116	0,0048	0,0062	0,0062
7	Relaxation time θ , s	0,14	0,59	2,08	3,56	10,94
8	Average pore diameter d, mm	0,2975	0,4475	0,5975	0,6725	0,8975
9	Volume of permeated gel V, mm	109,89	33,69	9,17	5,39	4,73
10	Filtartion time t, s	416	470	604	572	588
11	Volume flow rate Q, mm^3/s	0,2642	0,0717	0,0152	0,0094	0,0080
12	Cross sectional area of pore channel S, mm^2	43,96	13,48	3,67	2,16	1,89
13	Filtration speed w, mm/s	0,0060	0,0053	0,0041	0,0044	0,0043
14	Relaxation time θ , s	15,13	25,72	44,13	47,04	64,53

RELATION BETWEEN CALCULATED DATA AND LABORATORY RESEARCHES RESULTS

Then we compared the values of the relaxation time of the polymeric composition, which we calculated based on the results of microtomography and filtration researches, and the ones we determined in dynamic tests in gel oscillating voltages mode (figure 9).

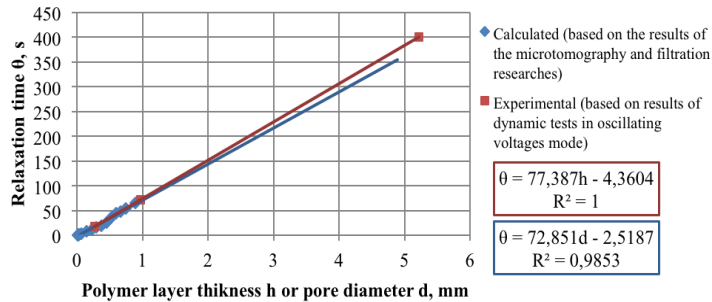


Figure 9. Relation of relaxation time and pore diameter (polymer layer thickness)

The calculated values and experimental results of determination of the relaxation time of the initial polymeric composition are practically equal and are to be described by nearly the same equations. This allows us to use the experimental data to calculate the filtration parameters at injection of viscoelastic liquid.

CONCLUSION

The above described approaches to the complex laboratory researches of polymeric compositions allow us to find the composition with optimum properties for the in-situ water shut off taking into account its rheological and filtration characteristics and the modes of its injection into the productive formation. They also allow us to evaluate the volume of the permeated composition, its distribution in pore space, the changes of rheological properties before and after filtration, injection pressure gradient at different moments of time and in different parts of pore space.

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