

INFLUENCE OF THE MOLECULAR PARAMETERS OF ANIONIC ACRYLAMIDE COPOLYMERS ON THE VALUE OF THE THOMS EFFECT IN DIRECT OIL EMULSIONS

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The influence of molecular parameters (molecular weight and polydispersity in molecular weight) on the value of the reduced Thoms effect in 10% direct oil emulsions has been analyzed with the example of the specimens of anionic acrylamide copolymer of different molecular weight, which were obtained by the ultrasonic-destruction method. The prospects of anionic acrylamide copolymers for use as admixtures reducing the drag of turbulent oil-emulsion flows have been noted for the complex heterophase system studied.

Introduction. In recent years, researchers have shown growing interest in methods for reducing the drag of turbulent liquid flows [1]. One of the most original and promising variants of solution of the problem lies in introducing small (at the level of hundredths and thousandths of a percent) additions of high-molecular-weight compounds (Thoms effect) into turbulent flows [1–3]. Quite an extensive amount of data on evaluation of various regularities of the Thoms effect has been obtained at present for individual (co)polymers in relatively simple dispersive media: predominantly in water or in organic solvents [2–7]. In particular, in aqueous media, one has studied the influence of numerous active parameters of the system, i.e., the concentration and characteristics of a polymer admixture (chemical nature, molecular weight, and chemical composition), the properties of a dispersive medium (pH, ionic strength, and nature of the solvent), the velocity of a turbulent liquid flow, the parameters of a limiting circuit, and others, on the value of the Thoms effect. However in many cases actual liquids dealt with in practice represent much more complex multicomponent and heterophase systems, e.g., emulsions or suspensions. One of the most important and interesting examples of such actual systems is provided by direct and invert oil emulsions formed in the stages of production and delivery of oil from highly-drowned oil beds [8]. We emphasize that oil emulsions (both direct and invert ones) have a substantially higher dynamic viscosity than water and dehydrated oil. This circumstance alone involves additional complications (in particular, leads to a sharp increase in energy consumption) in high-speed (in the turbulent regime) delivery of oil emulsions to the site of their separation, since the distances for which they are delivered may attain several tens of kilometers.

Description and Results of the Experiment. With the use of our previous systematic investigations of evaluation of the regularities of turbulent flow of aqueous solutions in the presence of water-soluble polymeric admixtures [3, 9] as the basis ones, in this work, we have analyzed the fundamental possibilities of using anionic acrylamide copolymers as an admixture in a much more compositionally complex actual disperse system — a direct oil emulsion — with the aim of finding methods for intensification of the high-speed delivery of oil and oil products. In the work, the emphasis is on the quantitative evaluation of the influence of the molecular parameters of polymer admixtures on the value of the Thoms effect.

Investigations of the Thoms effect in direct oil emulsions were carried out on a modified turbulent rheometer whose simplified diagram is presented in Fig. 1. The experiment was as follows. We poured 320 cm³ of the liquid under study into a thick-walled steel tank 5 of capacity 350 cm³. Excess pressure monitored by manometer 3 was produced in receivers 4 by means of the gas vessel 1 using reducer 2. At a prescribed instant of time, we switched on the valve's electric drive 7 which in turn opened valve 6. The liquid under pressure flowed, through capillary 8, from

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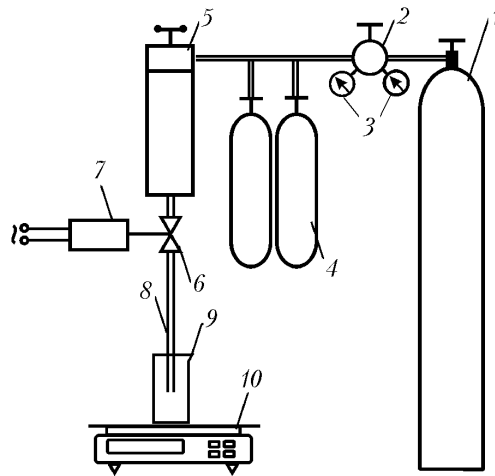


Fig. 1. Simplified diagram of a modified turbulent rheometer: 1) vessel with nitrogen; 2) reducer; 3) manometer; 4) receivers; 5) steel tank with the liquid under study; 6) automatic valve; 7) valve's electric drive; 8) capillary; 9) collector sleeve; 10) electron balance.

TABLE 1. Hydrodynamic Indices of a 10% Direct Oil Emulsion in the Presence of Different Doses of Anionic Acrylamide Copolymers with $M = 3.75 \cdot 10^3$ kg/mole

Analyzed system	$\mu \cdot 10^3$, Pa-sec	P , atm	Liquid flow rate, $10^5 \text{ m}^3/\text{sec}$	Average flow velocity, m/sec	$Re \cdot 10^{-3}$	T	γ , m^3/kg
Water	1.007	5	2.91	11.07	20.12	—	—
		10	5.14	19.55	35.54	—	—
		15	6.74	25.62	46.57	—	—
Emulsion	1.197	5	2.90	11.02	16.57	—	—
		10	5.05	19.21	28.90	—	—
		15	6.52	24.82	37.33	—	—
Emulsion +0.0004% of the copolymer	1.345	5	3.35	12.74	17.05	0.156	39.03
		10	5.38	20.45	27.38	0.064	16.10
		15	6.99	26.60	35.61	0.072	17.91
Emulsion +0.0008% of the copolymer	1.378	5	3.54	13.45	17.58	0.221	27.63
		10	5.67	21.57	28.19	0.123	15.34
		15	7.03	26.73	34.94	0.077	9.64
Emulsion +0.002% of the copolymer	1.474	5	4.05	15.39	18.82	0.400	19.91
		10	6.20	23.58	28.83	0.228	11.42
		15	7.34	27.90	34.11	0.125	6.27
Emulsion +0.004% of the copolymer	1.592	5	4.21	16.03	18.14	0.456	11.40
		10	6.70	25.47	28.83	0.327	8.17
		15	8.01	30.47	34.49	0.230	5.72
Emulsion +0.008% of the copolymer	1.806	5	4.23	16.08	16.05	0.461	5.77
		10	7.07	26.88	26.83	0.400	5.00
		15	8.68	33.02	32.96	0.332	4.15
Emulsion +0.012% of the copolymer	1.890	5	4.12	15.66	14.95	0.425	3.54
		10	6.99	26.60	25.40	0.387	3.23
		15	8.88	33.78	32.25	0.364	3.03

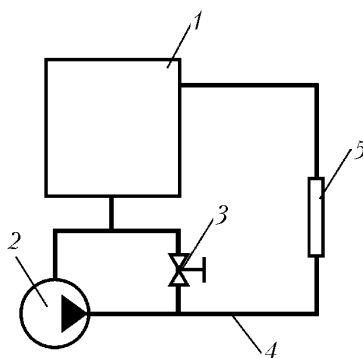


Fig. 2. Diagram of the setup for obtaining direct emulsions: 1) working vessel; 2) guided-vane pump; 3) valve for control of the liquid flow rate; 4) discharge line; 5) set of grid membranes with different dimensions of cells for "crushing" emulsion particles.

the tank 5 to collector 9. After a certain time, the valve automatically closed. The mass of the liquid flowing out of the tank 5 over this period was recorded using a single-arm automatic electron balance 10 (A&D Company, Japan). The quantitative characteristics of the Thoms effect were the dimensionless parameter T and the reduced Thoms effect γ [9, 10]:

$$T = \frac{m - m_0}{m_0}, \quad (1)$$

$$\gamma = \frac{m - m_0}{m_0 C}. \quad (2)$$

The advantages of the modified experimental setup compared to the standard turbulent rheometers are related to the possibility of varying, within wide limits, the geometric parameters of detachable capillaries (length and diameter) and the velocity of outflow of the liquid (with change in the tank pressure) and to the more convenient and reliable (compared to the standard volumetric analysis of samples) monitoring of the masses of liquids transmitted by the capillary. All the experiments have been carried out on a turbulent rheometer with the use of a glass capillary with a diameter of $1.83 \cdot 10^{-3}$ m and a length of 0.6 m for three fixed pressures P in the rheometer's tank: 5, 10, and 15 atm. Table 1 enables one to clearly visualize the basic hydrodynamic indices of turbulent flows of direct oil emulsions in the presence of different doses of a polymeric admixture in one experiment of this run.

To prepare direct emulsions we took oil from the Makarovskoye oil field (Zainskneft' Oil and Gas Production Department, Tatarstan, $\nu = 46.3 \cdot 10^{-6}$ m²/sec and $\rho = 913$ kg/m³). A nonionic surfactant — Neonol AF9-10 — was used for stabilization of the emulsions. A setup whose diagram is given in Fig. 2 was used for obtaining stable 10% direct oil emulsions with stable characteristics of dispersed-phase-particle size. Calculated amounts of oil, distilled water, and stabilizer (in an amount of 2% of the oil volume) were poured in the working vessel 1; thereafter, the mixture produced was transferred, in a circulation manner, through the discharge line 4 by pump 2. The oil-emulsion particles were reduced to the necessary size by crushing in unit 5 with a set of metal grids with successively decreasing cell diameters. The necessary flow rate of the liquid was controlled using valve 3. Such a method of preparation of direct oil emulsions guaranteed their high aggregative and sedimentation stability and virtual identity in dispersed-phase-particle size. According to the data of optical microscopy, the oil-particle size ranged from 2 to 7.5 μ m.

We used specimens of anionic acrylamide copolymers produced by the ultrasonic-destruction method from the initial commercial copolymer DP9-8177 (with molecular weight $M = 3.75 \cdot 10^3$ kg/mole and a content of ionic (acrylate) groups $\beta = 3.7\%$) as polymeric objects with high indices of the parameters T and γ in aqueous media, which are convenient for comparable analysis. Ultrasonic destruction of the copolymer was carried out in aqueous solutions of con-

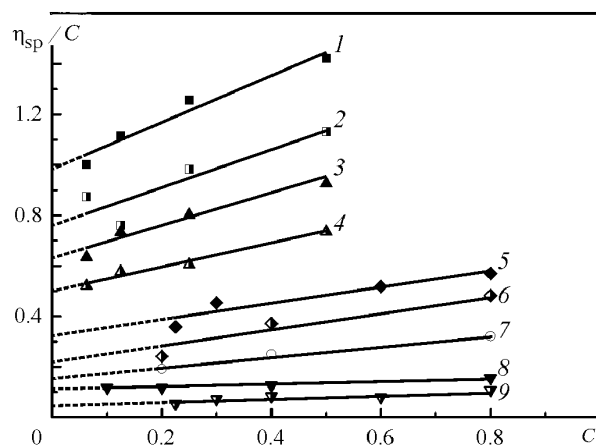


Fig. 3. Concentration dependence of the viscosity numbers (the solvent is a 0.5 normal NaCl) for specimens of anionic acrylamide copolymer of different molecular weight $M \cdot 10^{-3}$: 1) 3.75, 2) 2.72, 3) 2.12, 4) 1.59, 5) 0.89, 6) 0.53, 7) 0.36, 8) 0.22, and 9) 0.07 kg/mole. C , kg/m³.

TABLE 2. Basic Characteristics of the Specimens of Anionic Acrylamide Copolymers ($\beta = 3.7$ wt. %), Obtained by the Ultrasonic-Destruction Method

Specimen No.	Q , W/cm ²	t , min	$[\eta]$, m ³ /kg	$M \cdot 10^{-3}$, kg/mole
1	—	—	0.984	3.75
2	1.02	6	0.765	2.72
3	4.4	3	0.628	2.12
4	4.4	4	0.500	1.59
5	4.4	7.3	0.319	0.89
6	8.0	5	0.213	0.53
7	11.8	4.3	0.155	0.36
8	15.2	5	0.106	0.22
9	18.9	4	0.043	0.070

centration 0.1% on a UZDN-1 setup with an operating frequency of 22 kHz with variation of the radiation power Q and the exposure time t . In accordance with [11, 12], the regime of ultrasonic treatment eliminated side chemical processes and ensured the identity of all the resulting copolymer specimens not only in composition but also in chemical heterogeneity (compositional polydispersity). As a result of this run of experiments we obtained nine specimens of anionic copolymers differing by more than one and a half order of magnitude in molecular weight. The molecular weights M of the specimens obtained were calculated from the Staudinger–Mark–Houwink formula [13]

$$[\eta] = Km^a, \quad K = 0.0068, \quad a = 0.785. \quad (3)$$

The values of the limiting viscosity numbers $[\eta]$ were determined by extrapolation of the dependences $\eta_{sp}/C = f(C)$ in a 0.5 normal NaCl solution to $C = 0$ (Fig. 3). As is clear from the figure and particularly from Table 2, the copolymer specimens obtained were characterized by the large difference in the values of molecular weights, which contributes to a more objective and reliable evaluation of the influence of the molecular characteristics of a polymeric admixture on the values of the parameters T and γ .

An analysis of experimental data will start with a discussion of the concentration dependences of the value of the Thoms effect for specimens of anionic acrylamide copolymers differing in molecular weight (Fig. 4). In Fig. 4, we should primarily note one very important regularity unpredictable in advance: in such a complex multicomponent and heterophase system as direct oil emulsions, we have the Thoms effect and it is quite appreciable — the concentration

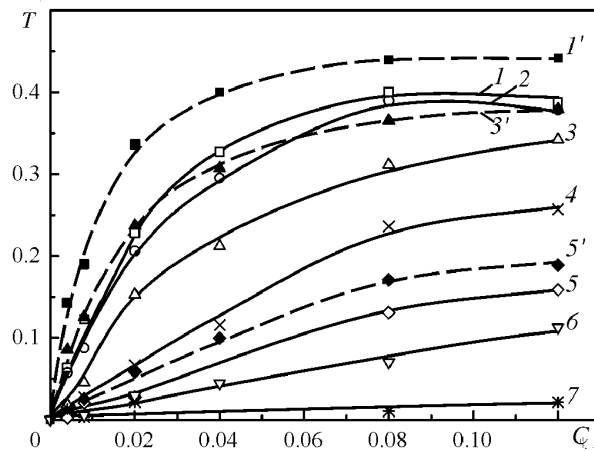


Fig. 4. Concentration dependences of the value of the Thoms effect for specimens of anionic acrylamide copolymers with the following molecular weight $M \cdot 10^{-3}$: 1 and 1') 3.75, 2) 2.72, 3 and 3') 2.12, 4) 0.89, 5 and 5') 0.53, 6) 0.36, and 7) 0.22 kg/mole; rheometer pressure $P = 10$ atm; 1', 3', and 5') dependences for the aqueous copolymer solutions. C , kg/m^3 .

dependences $T = f(C)$ manifest themselves for all the copolymer specimens studied. The behavior of the concentration dependences for the polymeric admixtures studied can be understood if the concept that the optimum concentration of a polymeric admixture (C_{opt}) exists in the most active wall zone of the turbulent flow [14–16] holds for direct oil emulsions too. The length of such a zone may vary within wide limits and is dependent on numerous parameters: the average velocity of the liquid flow, the linear dimensions, the nature of materials, the geometry and roughness of the limiting-circuit surface, the value of electrokinetic potential and the length of individual elements of the electric double layer occurring at the phase boundary, the ionic strength, the local concentrations of active components [3], and others. Deliberately avoiding controversial issues of particularization of the mechanism of the Thoms effect, we only note that for multicomponent systems like direct oil emulsions, the local concentrations of individual active components (primarily macromolecular and dispersed-phase particles) in the wall zone may differ significantly from the analogous macroscopic indices on an entire delivered-liquid volume basis. Certain but much smaller differences in the concentrations of active components (compared to the flow core) occur in the transition zone of the flow. As the concentration of the polymer in the flow generally increases, its content in the wall zone first increases too, but to a certain limit, up to the attainment of C_{opt} , and thereafter is maintained at a constant level without changing, in practice [17]. The latter circumstance is the main reason for the value of the Thoms effect "going beyond the limit" in Fig. 4.

In analyzing Fig. 4, it is necessary to note that the value of the Thoms effect for one and the same concentration of the polymeric admixture in the aqueous medium (curves 1', 3', and 5') is somewhat higher than the analogous indices for the direct oil emulsion (curves 1, 3, and 5). The above differences in the values of the parameter T in water and in the direct oil emulsions analyzed are related to the processes of adsorption of the polymer on oil particles. Part of the polymer macromolecules goes from the aqueous dispersive medium to the surface layer of dispersed-phase particles and becomes less active (and in the limit — "inoperative") as far as the Thoms effect is concerned.

In Fig. 4, we also easily track the dependence of the Thoms effect on the molecular weight of a polymeric admixture. Figure 5 enables us to more clearly visualize this. It is clear from the figure that the explanation for the natural growth in the value of the reduced Thoms effect γ with molecular weight is quite simple and logical: the standard size of macromolecules and consequently the efficiency of the polymeric admixture as the agent reducing the drag of turbulent liquid flows grows with polymeric molecular weight [3, 9].

In evaluating more accurately the influence of the molecular characteristics of polymeric admixtures on the values of the parameters T and γ , in addition to M , we should allow for such an important parameter as polydispersity in copolymer molecular weight. We showed earlier that the dependence of the Thoms effect on the polydispersity in molecular weight is of a fairly complex and difficult-to-predict character for aqueous solutions [9]. It is logical to as-

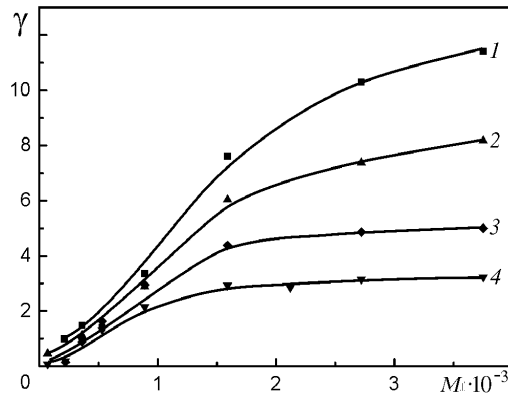


Fig. 5. Dependence of the value of the reduced Thoms effect γ on the molecular weight of polyacrylamide for the following concentrations C of the polymer: 1) 0.02, 2) 0.04, 3) 0.08, and 4) 0.12 kg/m³. $P = 10$ atm. γ , m³/kg; M , kg/mole.

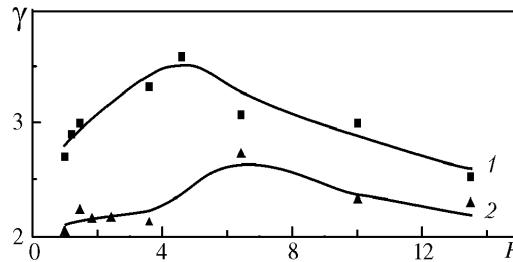


Fig. 6. Reduced Thoms effect γ vs. Shultz parameter F for blends of anionic acrylamide copolymers with $M = 2.3 \cdot 10^3$ kg/mole at different P : 1) 5 and 2) 15 atm. $C = 0.12$ kg/m³; γ , m³/kg.

sume that the appearance of a new phase (oil) in the analyzed system that is much more compositionally complex, i.e., a direct oil emulsion, may involve additional complications in the behavior of the resulting dependence of the Thoms effect on the polydispersity in M for the polymeric admixture.

The assumption made needs experimental verification. A tentative analysis has shown that γ as a function of the polydispersity in M can be studied more thoroughly if we take copolymer specimens with average molecular weight $\bar{M} = 1.59 \cdot 10^3$ kg/mole as reference for comparison. To prepare a series of copolymer specimens with such a fixed \bar{M} value we used a procedure analogous to [9]. The quantitative characteristic of polydispersity of a polymeric admixture in molecular weight is the value of the Shultz parameter [18]

$$F = \left[\frac{\sum_{i=1}^k \omega_i M_i}{\sum_{i=1}^k \frac{\omega_i}{M_i}} \right] - 1, \quad (4)$$

where ω_i is the mass fraction of the i th component of the mixture with $M = M_i$. The Shultz parameter is a quantitative characteristic of polydispersity in M , since there is a slope-similar dependence between these parameters: increase in the polydispersity in M is accompanied by the natural growth in the parameter F .

Figure 6 gives the dependences of the reduced Thoms effect on the Shultz parameter for two values of the rheometer pressure; the complex extremum (with a maximum) behavior of the dependence $\gamma = f(F)$ is pronounced for the 10% direct oil emulsions analyzed. Undeniably, in this case we are dealing with the violated principle of additivity of the contribution of individual fractions of copolymers with different M values to the resulting Thoms effect [19]. According to the data of Fig. 6, we also note the shift of the maximum on the curves toward higher values of the argument and a less pronounced character of the dependence $\gamma = f(F)$ with increase in the average velocity (propor-

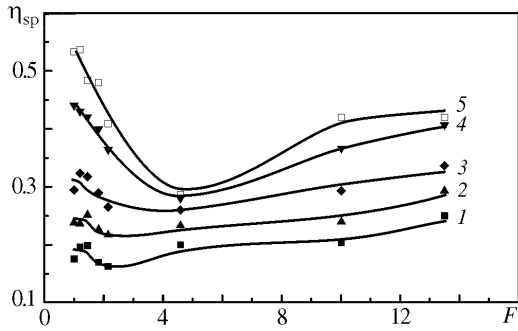


Fig. 7. Specific viscosity of direct oil emulsions vs. Shultz parameter F for different polymer concentrations C : 1) 0.008, 2) 0.02, 3) 0.04, 4) 0.08, and 5) 0.12 kg/m^3 .

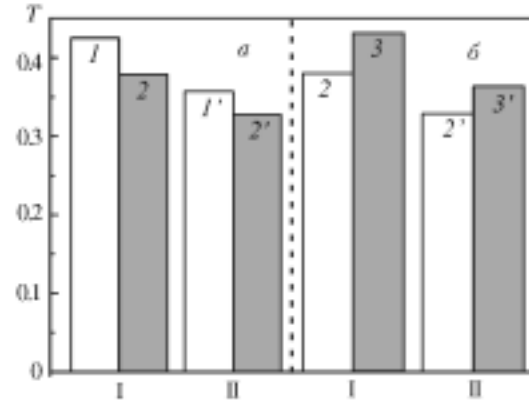


Fig. 8. Histogram of the value of the Thoms effect in emulsions prepared with distilled (1, 2, and 3) and saline (1', 2', and 3') water for: a) anionic acrylamide copolymers with $M = 3.75 \cdot 10^3 \text{ kg/mole}$ (1 and 1') and $M = 1.59 \cdot 10^3 \text{ kg/mole}$ (2 and 2'); b) blends of anionic acrylamide copolymers with $M = 1.59 \cdot 10^3 \text{ kg/mole}$ and different values of the parameter F : 2 and 2') 1; 3 and 3') 3.59.

tional to the Reynolds number) of the turbulent flow. A comparison of the curves of different pressures in the tank (from $P = 5 \text{ atm}$ to $P = 15 \text{ atm}$) enables us to clearly visualize this.

One of the main reasons for the complex character of the dependences $\gamma = f(F)$ may be the manifested dependence of the standard size of coiled macromolecules $(\overline{h^2})^{0.5}$ on the polydispersity M . The presence of such dependence for the anionic acrylamide copolymers studied at a qualitative level is demonstrated by the data of a viscosimetric analysis (Fig. 7), since the well-known Flory relation [20] may be written in the first approximation as

$$(\overline{h^2})^{1.5} \Phi = M \left(\frac{\eta_{sp}}{C} \right) \Big|_{C=\text{const}} \quad (5)$$

The presence of functional dependences between γ , $(\overline{h^2})^{0.5}$, and η_{sp}/C , apart from the dependence $\left(\frac{\eta_{sp}}{C} \right) \Big|_{C=\text{const}} = f(F)$ obtained experimentally (Fig. 7), is mainly responsible for the manifestation of the complex extreme

character of the dependence of the reduced Thoms effect on the polydispersity in M for the polymeric admixture. In oil emulsions, a certain influence on the character of the dependence $\gamma = f(F)$ may also be exerted by the selective processes of macromolecular adsorption on dispersed-phase particles and by the change in the shape of dispersed-phase particles in the transition zone and in the core of the turbulent emulsion flow.

Under actual conditions, water extracted from the earth's interior together with oil has a high salinity (mineralization) (up to 250–300 g/liter) [21]. In this connection, we additionally (qualitatively) evaluated the applicability of the analyzed polymeric admixtures of anionic acrylamide copolymers to acceleration of the process of delivery of actual emulsions with high-salinity water. We selected, for investigations, brine water taken with sour crude in one oil field of the Zainskneft' Oil and Gas Production Department. As is seen in Fig. 8, the values of the Thoms effect for such a system are quite comparable to the analogous indices for emulsions prepared with distilled water. These results demonstrate the potential possibility of using acrylamide copolymers as agents reducing the drag of flows not only in purely aqueous media but also in the actual processes of delivery of high-salinity direct oil emulsions in the turbulent regime. Such a result is in principle clear, since the standard size of macromolecules of polyacrylamide and anionic

acrylamide copolymers with a low degree of hydrolysis (they also include the basis specimen of DP9-8177 copolymer) are dependent on the ionic strength only slightly [22].

In closing, we note that the use of a modified turbulent rheometer and the given scheme of analysis with finding the parameters T and γ makes it possible in principle to primarily test specific (co)polymer specimens as anti-turbulent admixtures in different systems including water-oil media. To do this requires a comprehensive comparison, in identical media, of the values of the parameters T and γ of the basis (standard) (co)polymer used in the actual processes of delivery of turbulent liquid flows by pipelines (including main lines) to the analogous indices for the (co)polymer analyzed (tested) and the conclusion, on this basis, on the potential prospects of specific (co)polymer specimens for use as agents reducing the drag of turbulent liquid flows in pipelines.

CONCLUSIONS

1. Nine specimens of anionic acrylamide copolymer with a wide difference in the values of molecular weights have been obtained under soft conditions by the ultrasonic-destruction method; this enabled us to correctly (in the absence of complications due to the compositional variability of copolymers) evaluate the influence of the molecular weight on the value of the Thoms effect in direct oil emulsions.

2. An analysis of the concentration dependences of the value of the Thoms effect for the copolymers indicated has shown their potential prospects as admixtures reducing the drag of turbulent flows of such complex multicomponent media as direct oil emulsions. For these oil emulsions prepared with distilled and high-salinity (brine) water, we have analyzed the dependences of the value of the reduced Thoms effect on the polydispersity in molecular weight in anionic acrylamide copolymers.

NOTATION

a , K , constants in the Staudinger–Mark–Houwink equation; C , concentration of the polymer in the solution, kg/m^3 ; F , Shultz parameter; $(h^2)^{0.5}$, standard size of macromolecules, m ; k , total number of reference copolymer specimens in the blend; M , molecular weight of the polymer, kg/mole ; \bar{M} , average molecular weight of the polymer, kg/mole ; m and m_0 , mass of the liquid transmitted by the capillary with the added polymer and without it, kg ; P , rheometer pressure, atm ; Q , radiation power in ultrasonic treatment of the copolymer solutions, W/cm^2 ; Re , Reynolds number; T , parameter characterizing the value of the Thoms effect; t , time of ultrasonic treatment of the copolymer solution, min ; Φ , Flory constant; β , weight content of ionic groups in the polymer macromolecules, $\%$; γ , reduced Thoms effect, m^3/kg ; η_{sp} , specific viscosity; η_{sp}/C , viscosity number, m^3/kg ; $[\eta]$, limiting viscosity number, m^3/kg ; μ , dynamic viscosity, $\text{Pa}\cdot\text{sec}$; ν , kinematic viscosity, m^2/sec ; ρ , density, kg/m^3 ; ω , mass fraction. Subscripts: opt, optimum; sp, specific; 0, zero.

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