

Copolymers of Diallyldimethylammonium Chloride and Vinyl Ether of Monoethanolamine: Synthesis, Flocculating, and Antimicrobial Properties

Yerbol Dauletov^{1,2} · Nurxat Nuraje² · Kaldibek Abdiyev¹ · Zhexenbek Toktarbay³  · Maryamgul Zhursumbaeva¹

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Abstract N,N-diallyl-N,N-dimethylammonium chloride (DADMAC) and vinyl ether of monoethanolamine (VEMEA) copolymer P(DADMAC-VEMEA) were synthesized using radical polymerization in water media using ammonium persulfate as an initiator. The synthesis of this copolymer was investigated over a wide composition range at low conversion levels. The viscosity of the copolymers was measured in aqueous 1 M NaCl. Flocculating action and antimicrobial activities of the copolymers P(DADMAC-VEMEA) against microbacteria and sulfate-reducing bacteria (SRB) were studied. Synthesized copolymers had flocculating effects and can be used to purify wastewater from particles of bentonite clays. Also, the copolymer with a molar composition [DADMAC]:[VEMEA] = (80÷20):(20÷80) retarded the population of both bacteria, Gram-positive (*Staphylococcus aureus*) and Gram-negative (*Escherichia coli*) in the diffusion zone. Furthermore, the copolymer with a molar composition [DADMAC]:[VEMEA] = (90÷10):(20÷80) had bactericidal properties against SRB. Minimum inhibitory

concentration (MIC) of the copolymer varied from 0.01% to 0.1% depending on the molar composition of the copolymer.

Keywords Radical copolymerization · Flocculation · Antimicrobial activity · Sulfate-reducing bacteria

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Introduction

In recent years, (co)polymers based on N,N-diallyl-N,N-dimethylammonium chloride (DADMAC) have been widely used in many industrial applications as flocculants in potable water and waste water treatment (Abdiyev et al., 2015a; Wandrey et al., 1999), electrochemical sensors based on gel–gel technology (Cabello-Carramolino and Petit-Dominguez, 2009), cationic polyelectrolytes in multiwalled carbon nanotube production for the extraction of toxic polychlorinated biphenyls (Zeng et al., 2012), coagulants (Verezhnikov et al., 2001), polycomplex formation agents (Nurkeeva et al., 2000; Shaikhutdinov et al., 2007), antibacterial blend films (Lu et al., 2008), hydrogels (Dragalova et al., 1992), and in other fields of industry due to their good solubility in water, nontoxicity, and high positive charge density.

Carbon-13 nuclear magnetic resonance (¹³C-NMR) has shown that during the radical homopolymerization reaction, five-membered rings of DADMAC were exclusively formed (Lancaster et al., 1976; Masterman et al., 1994; Valade et al., 2009; Wandrey et al., 1981) and the rings were connected by ethylene bridges in the 3,4-position (Masterman et al., 1994; Wandrey et al., 1981). According to Wandrey et al., 1999,

✉ Nurxat Nuraje
nurxat.nuraje@ttu.edu

✉ Kaldibek Abdiyev
abdiyev_almaty@rambler.ru

¹ Department of Chemical technology of Organic compounds and Polymers, Kazakh National Technical Research University named after K. I. Satpaev, 22 Satpaev str., Almaty, 050013, Republic of Kazakhstan

² Department of Chemical Engineering, Texas Tech University, Lubbock, TX 79409, USA

³ Department of analytical, colloidal chemistry and technology of rare elements, Faculty of Chemistry and Chemical Technologies, Al-Farabi Kazakh National University, Al-Farabi avenue 71, Almaty, 050012, Republic of Kazakhstan

poly-DADMAC synthesized by free radical polymerization in water media at low conversion using ammonium persulfate as an initiator contains only 0.1–3.0% pendant double bonds.

(Co)polymers based on simple vinyl ethers (SVE) are widely used in the production of paints, microelectronics, adhesives (Valade et al., 2009), (poly)surfactants (Khussain et al., 2012; Lievens and Goethals, 1996), share-memory materials (Reyntjens et al., 1999), thermoresponsive polymers and colloidal stabilizers (Bulychev et al., 2004; Verdonck et al., 2003), artificial leather, lubricants, plasticizers, thickeners, lubricating oils, pastes, antifreezing agents, and coagulants (Zhubanov et al., 1985).

In the chemical structure of vinylalkyl ethers (general structural formula $\text{CH}_2=\text{CH}-\text{OR}$, where R – alkyl radical), the (+M) mesomeric effect from the alkoxy group side appears, which enhances the nucleophilic property of the double bond and increases the stability of the resulting carbocation (Zhubanov et al., 1985). For this reason, SVE readily polymerize by the “living” cationic mechanism in the presence of mineral acids or electron-withdrawing catalysts (Kammiyada et al., 2015; Lievens and Goethals, 1996; Lin et al., 2015, 2016; Reyntjens et al., 1999; Verdonck et al., 2003). In the presence of conventional radical initiators, the homopolymers of SVE are formed and oligomers are mainly produced (Zhubanov et al., 1985). High-molecular homopolymers of SVE can be obtained by the method of radiation polymerization (Goineau et al., 1977; Nurkeeva et al., 2002; Suzuki et al., 1977). Although SVE show low reactivity in radical homopolymerization reactions, they can react with various monomers via a free radical copolymerization mechanism (Dergunov et al., 2003; Sugihara et al., 2016; Vorob'eva et al., 2007; Zhenissova et al., 2005).

One problem arising in the oil and gas industry is the biogenic formation of hydrogen sulfide (H_2S) in the reservoir (Fink, 2003). H_2S causes faster corrosion of engineering units, which causes higher operating costs, formation of iron sulfide, and release of gas itself resulting in serious environmental and health hazards. Biofilms often contribute to the formation of H_2S because of sulfate-reducing bacteria (SRB; <https://www.corrosionpedia.com/definition/1046/sulfate-reducing-bacteria-srb>). SRB can obtain energy by oxidizing organic compounds or molecular hydrogen while reducing sulfate, sulfite, thiosulfate, and elemental sulfur to hydrogen sulfide. SRB populate anaerobically in water, often in the environment of oil and natural gases (Ali, 2007). Microbiologically influenced corrosion (MIC) has been estimated to cause 15% to 30% of all failures induced by corrosion in all spheres of industry (Fink, 2003).

For inhibition of SRB, various biocidal polymers are used (Ahmed et al., 2008; Kenawy et al., 2007; Kenawy and Mahmoud, 2003). However, these polymers are not always effective. According to Battersby et al., 1985,

treatment of SRB with sublethal concentration of biocides can lead to the development of biocide-resistant mutants. For example, the mutant strain (*Desulfovibrio desulfuricans* 19TK) is resistant to all known cationic biocides. Therefore, a mixture of biocides is recommended to prevent the development of mutant strains. Thus, the creation of new effective antimicrobial compounds (biocides) and widening of their assortment are key issues.

To produce a flocculant with antimicrobial activity against SRB, copolymers of DADMAC with vinyl ether of monoethanolamine (VEMEA) were synthesized. These monomers were chosen because DADMAC is a biocide against bacteria, algae, and fungi (Jaeger et al., 1984), and VEMEA has an antibacterial action (Zhubanov et al., 1985).

Experimental

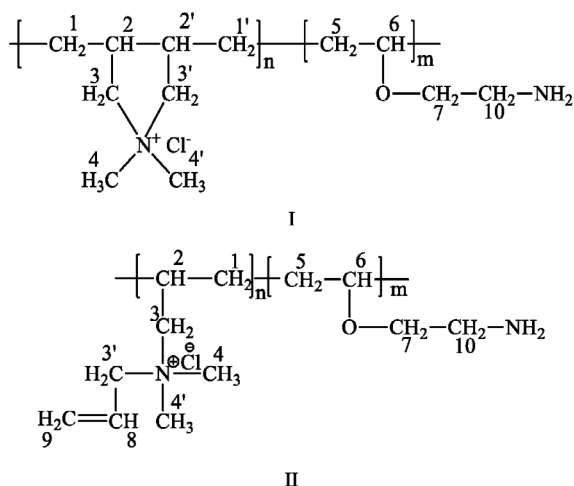
Reagents

The monomer DADMAC (65 wt.% in water, density 1.056 g cm^{-3}) was purchased from Sigma Aldrich (USA) and was used without additional purification. The monomer VEMEA was purchased from JSC “Carbit” (Temirtau city, Kazakhstan) and dehydrated with sodium metal and purified by distillation before usage. The boiling point and refractive index of VEMEA fraction used were 387 K and 1.4382 at 298 K, respectively.

AgNO_3 (purity 99.8 wt.%) and ammonium persulfate $(\text{NH}_4)_2\text{S}_2\text{O}_8$ (purity 99.7 wt.%) were provided by “LaborPharma” Ltd. (Kazakhstan). Acetone (purity 99.9 wt.%) was purchased from “Labchimprom” Ltd. (Kazakhstan). Argon gas with a purity of 99.995 wt.% was purchased from “Ikhsan” Ltd. (Kazakhstan). Distilled water with a conductivity of $2.4 \mu\text{S cm}^{-1}$ (20°C) was used in all the experiments.

Clay

Bentonite clay from the Tagan deposit (East Kazakhstan) was used. Before use, the clay was grounded in a ball mill for 15 min and then sieved through a sieve with a pore diameter of 0.01 mm. For this study, a sample with a particle diameter of not more than 0.01 mm was used. Chemical analysis of the used bentonite sample was carried out by X-ray fluorescence spectroscopy (XRF) using PANalytical Axios PW400. The surface area of the bentonite clay was determined in liquid N_2 by the Brunauer–Emmett–Teller (BET) approach using a Micromeritics ASAP 2020 instrument at -196°C (77 K). The approximate total pore volume of clay was calculated from the amount of adsorbed liquid at 0.99 relative pressures. Distribution of pore sizes was calculated from the



Scheme 1 Suggested molecular structures of the copolymer P(DADMAC-VEMEA)

desorption part of the N_2 adsorption/desorption isotherms, using the Barrett–Joyner–Halenda (BJH) method.

Synthesis

The copolymerization reaction of DADMAC with VEMEA was carried out at different molar ratios of the initial monomers. After purging with argon for 15 min, the ampoule containing monomers and initiator solution was sealed and placed in a water bath heated at 338 K for 5 h. Afterward, the copolymers were precipitated and washed with fresh acetone. This operation was repeated three times to remove unreacted monomers. The precipitated copolymers were then dried under vacuum at 313 K until a constant weight was achieved. The composition of the copolymers was investigated using fourier-transform infrared spectroscopy, hydrogen-1 nuclear magnetic resonance and ^{13}C NMR spectroscopy, and conductometric titration (Abdiyev et al., 2015b; Dauletov et al., 2018).

Conductometric and Viscometric Titrations

The DADMAC portion in the copolymers was determined using the conductometric titration method where copolymer

solution was titrated with AgNO_3 solution using a conductometer (conductivity module 856, Metrohm) (Erbil et al., 2000). The intrinsic viscosity $[\eta]$ of the polymers was measured in aqueous 1 M NaCl at (298 ± 0.1) K using an Ubbelohde viscometer (Frolov and Grodsky, 1986).

Flocculation Measurements

Flocculation was carried out with bentonite suspensions. An amount of 5 g of a ground and sieved bentonite sample with largest dimension of 0.01 mm was dispersed in the 1-L beaker containing distilled water. Then, the suspension was left for 24 h to let the bentonite completely swell. Before treatment with the flocculant, the suspension was prepared by stirring at 300 rpm for 10 min in water (EKROS 8000). Then, the suspension was treated with different dosages of flocculant solutions. After the mixture was further stirred at 100 rpm for 3 min, the suspensions were left to settle; the turbidity of the suspensions was measured after each trial. Optical density of the suspensions was measured at 740 nm (D) using the spectrometer KFK-3-01 (Russia) (Frolov and Grodsky, 1986). Distilled water was used as a reference. The residual turbidity (RT) was calculated according to the following equation:

$$\text{RT} = \frac{\text{OD}_{740s}}{\text{OD}_{740i}} * 100\%, \quad (1)$$

where OD_{740s} denotes OD_{740} after the addition of the flocculant and OD_{740i} denotes OD_{740} of the suspension in the absence of the flocculant under the same experimental conditions (Abdiyev et al., 2015a; Frolov and Grodsky, 1986).

Antimicrobial assessment of the copolymers P(DADMAC-VEMEA)

The antimicrobial activity of the copolymers P(DADMAC-VEMEA) was studied at the Institute of Microbiology and Virology of the Ministry of Education and Science of the Republic of Kazakhstan (IMB&V of ME&Sc RK). The antimicrobial activity of the DADMAC-VEMEA

Table 1 Influence of the monomer concentration in the feed on copolymerization

Monomer concentration in the feed, mol. %		Monomer content in the copolymer, mol. %		C (initiator), wt. %	Reaction time, h	t, °C	[η] in 1 M NaCl solution at 25 °C, dL g ⁻¹
DADMAC	VEMEA	DADMAC	VEMEA				
90	10	95	5	0.10	5	65	0.33
75	25	89	11				0.20
60	40	83	17				0.16
50	50	79	21				0.14
40	60	78	22				0.12
25	75	77	23				0.10

Table 2 Chemical analysis of the bentonite clay sample

Component	%
Na ₂ O	1.00
MgO	3.59
Al ₂ O ₃	22.46
SiO ₂	58.78
SO ₃	0.06
Cl	0.02
K ₂ O	0.59
CaO	3.99
TiO ₂	0.79
Cr ₂ O ₃	0.01
Fe ₂ O ₃	8.60
Total	99.88

copolymer with different molar compositions was carried out on nutrient agar. For this purpose, the copolymers were first dissolved in the sterile distilled water:

- 1- sample (100 mg mL⁻¹) has gel-like consistency;
- 2- sample (100 mg mL⁻¹) is a viscous liquid;
- 3- sample (25 mg mL⁻¹) has gel-like consistency.

Gram-positive bacteria (*Staphylococcus aureus*) and Gram-negative bacteria (*Escherichia coli*) were used as test organisms. Gram-positive bacteria have the crystal violet color because of a thick layer of peptidoglycan while Gram-negative bacteria are colored red or pink (Gram-negative Bacteria vs. Gram-positive Bacteria; https://www.diffen.com/difference/Gram-negative_Bacteria_vs_Gram-positive_Bacteria). *Staphylococcus aureus* causes sepsis (infection of the blood), pneumonia, and other serious infections. Most *E. coli* strains are harmless, but some strains can cause severe food poisoning.

The composition of the nutritional agar was as follows (%): meat extract—0.15; sodium chloride—0.5, yeast extract—0.15; peptone—0.5; agar—20.0; pH 7.4–7.6. To

assess the antimicrobial activity of the copolymers in the Petri dishes, seeded with test cultures (Colony Forming Unit 10⁶ mL⁻¹), the wells were made using a standard drill ($d = 7$ mm), and then copolymers were added to these wells in the form of a solution (0.1 mL, Sample No. 2) or in the form of a gel-like paste (samples No. 1 and 3). Incubation of test organisms with applied copolymers was carried out in the thermostat (Binder) at 37°C for 24 h. Antimicrobial activity of the copolymers was determined by measuring the diameter (in mm) of their inhibition zones to microorganisms' growth (Aitkeldiyeva et al., 2018; Xu et al., 2011). The results of the studies were confirmed by photo documents.

Antimicrobial Action of the Copolymers P(DADMAC-VEMEA) against SRB Investigation

The antimicrobial action of the copolymer P(DADMAC-VEMEA) with molar composition [DADMAC]: [VEMEA] = 90:10 (1), 80:20 (2), and 20:80 (3) on SRB was also tested at IMB&V of ME&Sc RK. The effect of these copolymers on the growth of SRB was studied at three concentrations (wt.%): 0.01, 0.05, and 0.1. In the control variant, SRB was cultured without copolymer. When SRB grows, a black film with a metallic outflow on the inner surface of the tube is observed.

Results and Discussion

Determination of Copolymer Composition

According to Lancaster et al. (1976); Wandrey et al. (1981); and Masterman et al. (1994), in radical polymerization reaction, monomer DADMAC reacts by forming five-membered cyclic structures in a macrochain. However, poly-DADMAC obtained by free radical polymerization in

Table 3 Isotherm mesopore reports for the bentonite clay particles

Method	Surface area, m ² g ⁻¹	Method	Pore volume, cm ³ g ⁻¹	Method	Pore size, nm
BET surface area	59	Single point adsorption total volume of pores with diameter less than 141 nm at P/P ₀ = 0.99	0.39	Adsorption average pore width (4 V/A by BET)	26
Langmuir surface area	74	<i>t</i> -Plot micropore volume	0.0003	BJH desorption average pore diameter (4 V/A)	27
<i>t</i> -Plot micropore area	2	BJH Desorption cumulative volume of pores with diameter 2–100 nm	0.38		
<i>t</i> -Plot external surface area	57				
BJH desorption cumulative surface area of pores with diameter 2–100 nm	57				

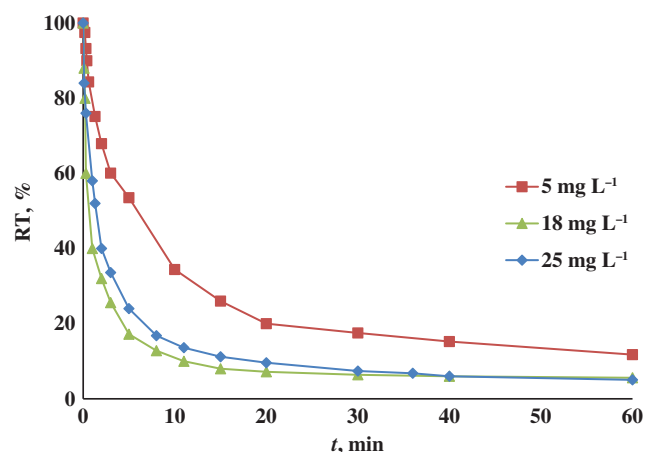


Fig. 1 Dependence of the turbidity of bentonite suspension on the time in the presence of the P(DADMAC-VEMEA) copolymer (molar composition 40:60) with different concentrations

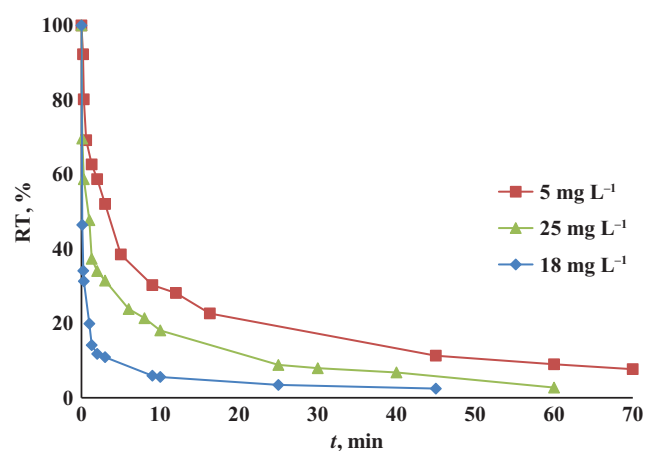


Fig. 2 Dependence of the turbidity of bentonite suspension on the time in the presence of the P(DADMAC-VEMEA) copolymer (molar composition 60:40) with different concentrations

water can contain up to 3% unreacted double bonds (Wandrey et al., 1981, 1999). Hence, we can predict two possible structures of macromolecules of the synthesized copolymer (Scheme 1):

The composition of the synthesized copolymer was determined from the conductometric titration of aqueous solutions of polymers with AgNO_3 (Erbil et al., 2000). Conductometric titration data indicated that DADMAC was more active (Table 1) than VEMEA in the radical polymerization reaction. Therefore, the DADMAC portion in copolymers is higher than in the initial monomer mixture. As the rate of the polymerization itself and intrinsic viscosity of polymers depend on the composition of the initial monomer mixture, with increasing molar concentration of DADMAC in the product, the copolymer viscosity in aqueous 1 M NaCl solution is also increased (Table 1).

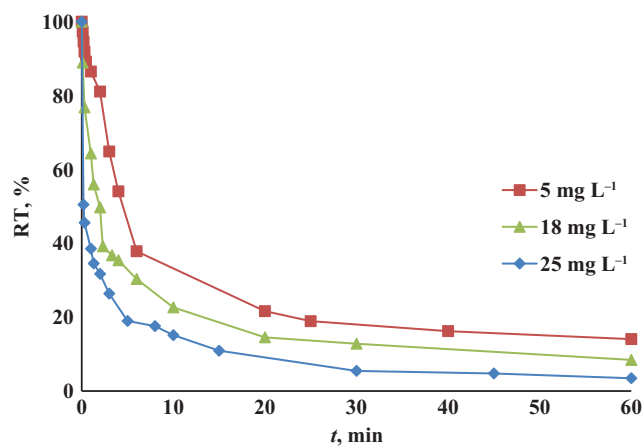


Fig. 3 Dependence of the turbidity of bentonite suspension on the time in the presence of the P(DADMAC-VEMEA) copolymer (molar composition 90:10) with different concentrations

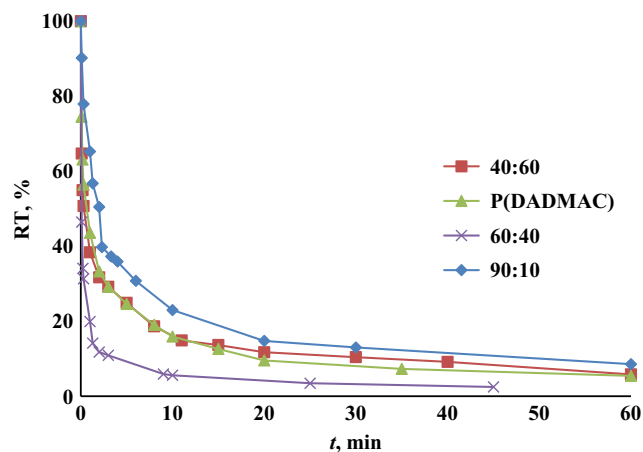


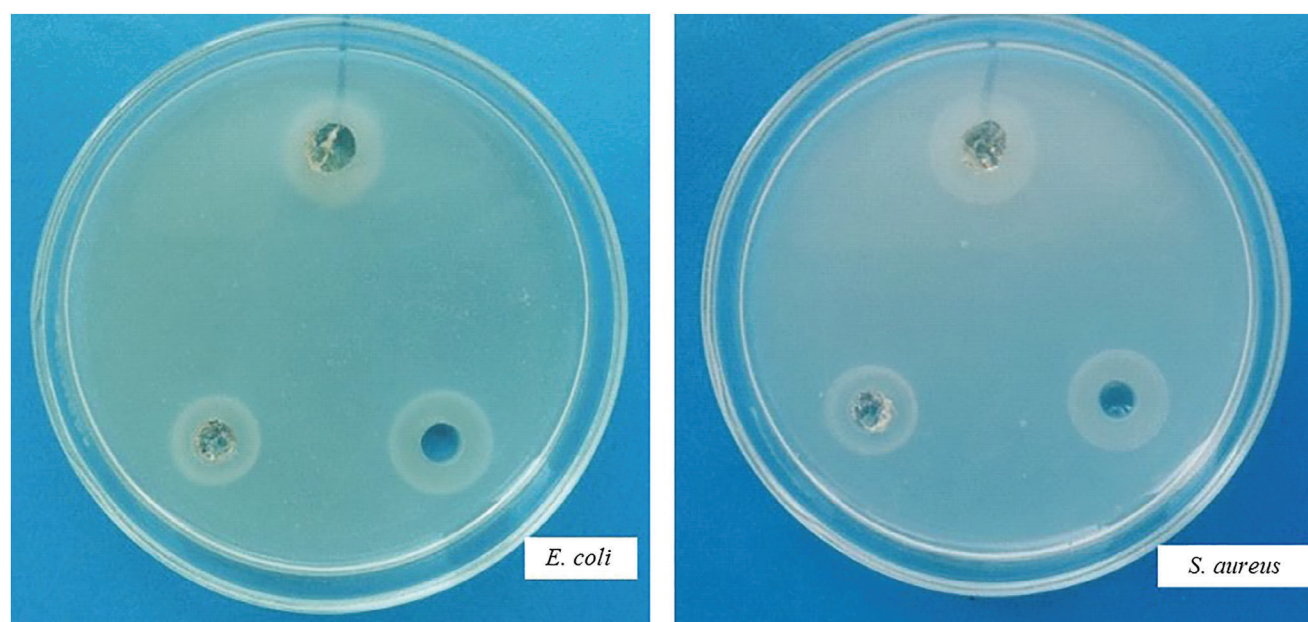
Fig. 4 Dependence of the turbidity of bentonite suspension on the time in the presence of poly-DADMAC and P(DADMAC-VEMEA) copolymers with different molar composition. $C(\text{polymer}) = 18 \text{ mg L}^{-1}$

Characterization of the Bentonite Clay Sample

Table 2 shows the chemical analysis of the bentonite clay sample from Tagansk deposit (East Kazakhstan). The $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio, from chemical analysis, was 1:2.6 revealing the montmorillonite nature of the bentonite sample. In addition, the ratio of $\text{Na}_2\text{O}/\text{CaO}$ of 1:3.98 indicated that the sample was Ca-bentonite rather than Na-bentonite (Veblen et al., 1990). Table 3 illustrates the surface area, pore volume, and pore size of the crushed bentonite particles determined using different methods.

Flocculation of the Particles of Bentonite Suspension with Copolymer P(DADMAC-VEMEA)

The data of flocculant investigations are shown in Figs 1–4. As seen, the minimum value of RT of the suspension (the



Molar composition of the copolymer P(DADMAC-VEMEA) ([DADMAC]:[VEMEA]): (1) 20:80; (2) 80:20, and (3) 40:60.]

Fig. 5 Antimicrobial activity of the copolymer P(DADMAC-VEMEA) against *E. coli* and *S. aureus* (from the top down clockwise direction Number 1, 2 and 3)

maximum degree of purification of the suspension from particles) is reached after 20–60 min, and the degree of purification of the suspension depends on the molar composition of the copolymer. For example, the optimum flocculation concentration of the copolymer with composition 60:40 and 40:60 is 18 mg L, while for composition 90:10, it is 25 mg L. At these concentrations, clean (transparent) water is formed at the top of the suspension. This can be considered as a result of flocculation, under the action of flocculant's macromolecules, the suspension particles precipitated at the bottom of the container (Abdiyev et al., 2015a; Harif et al., 2012).

From figures, we can conclude that copolymer P(DADMAC-VEMEA) with a molar composition of 60:40 has better flocculating ability than homopolymer (poly-DADMAC) and copolymers with other composition (Fig. 4).

Results of the Investigations of the Antimicrobial Properties of the Copolymer P(DADMAC-VEMEA)

From Fig. 5, copolymers were moderately diffused into the agar medium. All copolymer samples in the region of the

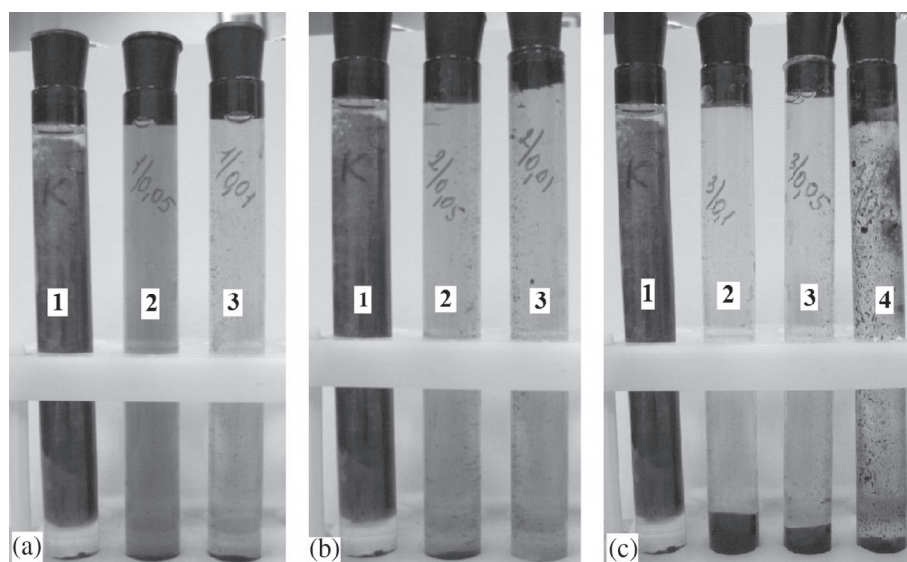
diffusion zone exhibited antimicrobial action equally against Gram-positive and Gram-negative micro bacteria in both microbacterial tests. The results of the test are shown in Table 4. The data show that copolymers of P(DADMAC-VEMEA) with a molar composition (80–20): (20–80) retard the population of Gram-positive and Gram-negative bacteria in the diffusion zone.

Investigations of the Biocidal Properties of the Copolymer P(DADMAC-VEMEA) against the Growth of SRB

The results of the studies are shown in Fig. 6. The effective copolymer is one in which bacteria (black color) are killed and found at the bottom of the test tube rather than distributed along the length of the test-tube. P(DADMAC-VEMEA) copolymers with a molar composition between 90:10 and 20:80 were tested, and all copolymers showed inhibition of the bacteria. However, for 0.01% concentration, the inhibition was not as effective as evidenced by the darker color.

Table 4 Inhibition zone diameters after treatment of different bacteria that were kept in nutrient agar after 24 h by the copolymer P(DADMAC-VEMEA)

Number of sample (mole fraction composition)	Copolymer condition	Inhibition zone diameter, mm	
		<i>Staphylococcus aureus</i>	<i>Escherichia coli</i>
1. Copolymer DADMAC-VEMEA (20:80)	Gel-like consistency	17	16
2. Copolymer DADMAC-VEMEA (80:20)	Viscous liquid	17	16
3. Copolymer DADMAC-VEMEA (40:60)	Gel-like consistency	14	13



Composition of the copolymer P(DADMAC-VEMEA) (mol. %): (a) 90:10; (b) 80:20; and (c) 20:80.
Concentration of the copolymer P(DADMAC-VEMEA) (wt.%): for (a) and (b) 1—without a copolymer; 2—0.05 and 3—0.01; for (c) 1—without a copolymer; 2—0.1; 3—0.05, and 4—0.01.]

Fig. 6 Growth of SRB in the presence of copolymer P(DADMAC-VEMEA)

SRB growth is suppressed as follows: DADMAC-VEMEA copolymer macromolecules are adsorbed on the surface of microorganisms using their functional groups (Alamri et al., 2012). As a result, a dense adsorption layer is formed on the surface of microorganisms, which limits their access of oxygen and nutrients, which ultimately leads to the death of microorganisms. At the same time, the penetration of biocides into the cell cannot be excluded. When biocides penetrate inside the cell, they can inhibit the respiratory chain enzymes and disrupt oxidative phosphorylation, causing the cell to die. In addition, coagulation of cytoplasmic proteins is possible when exposed to biocides (Kolesnikova et al., 2013; Panteleva et al., 2008).

Thus, on the basis of experimental data, we can conclude that copolymers P(DADMAC-VEMEA) with a molar composition [DADMAC]:[VEMEA] = (90–10):(10–90) have antimicrobial activity against SRB growth at a copolymer concentration of 0.01–0.1% and can be used to destroy SRB.

Conclusion

In this work, P(DADMAC-VEMEA) copolymers with different molar compositions were synthesized by free radical copolymerization in an aqueous solution in the presence of ammonium persulfate as an initiator.

The flocculation ability of the P(DADMAC-VEMEA) copolymer for bentonite clay depends on the concentration

and composition of the copolymer. Copolymer P(DADMAC-VEMEA) with a composition of 60:40 has better flocculating action than poly-DADMAC and copolymers with other compositions and the best flocculation action was revealed within 18–25 mg L.

P(DADMAC-VEMEA) copolymers with a molar composition (80–20):(20–80) retard the population of Gram-positive and Gram-negative bacteria in the diffusion zone. P(DADMAC-VEMEA) copolymers with a molar composition [DADMAC]:[VEMEA] = (90–10):(10–90) have antimicrobial activity against SRB and can be used to inhibit the growth of these bacteria.

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Conflict of Interest The authors declare that they have no conflict of interest.

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