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Biocide Coating from Polydiallyldimethylammonium Chloride—What Molecular Weight Should We Choose?

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Abstract: Biocidal compositions based on interpolyelectrolyte complexes and a low molecular weight antibiotic can become a promising material for creating biocidal coatings, as they combine wash-off resistance and dual biocidal action due to the biocide and the polycation. Molecular mass characteristics of polymers play an essential role in the physics and mechanical properties of the coatings. In this work, the properties of polydiallyldimethylammonium chloride (PDADMAC) coatings of various molecular weights are investigated and assumptions are made about the optimal molecular weight needed to create antibacterial compositions. To study the resistance to washing off and moisture saturation of the coatings, the gravimetric method was used, and the adhesive properties of the coatings were studied by dynamometry. It has been established that an increase in molecular weight affects the wash-off resistance of coatings, but does not affect moisture absorption and adhesion mechanics of coatings. All samples of PDADMAC were demonstrated to exhibit the same antibacterial activity. Thus, when developing systems for creating antibacterial coatings, it must be taken into account that in order to create stable coatings, the requirement to use PDADMAC with a high degree of polymerization is necessary for the coating desorption control during wash off-but not mandatory for the control of mechanical and antibacterial properties of the coating.

Keywords: polycation; polydiallyldimethylammonium chloride; polymer coating; antibacterial coatings

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1. Introduction

One of the goals of sustainable development established in 2015 by the United Nations General Assembly is the fight against hunger, which includes ensuring food security [1,2]. An important problem that threatens food safety is the formation of biofilms and the spread of pathogenic bacteria in the premises for the production and storage of food products, as well as on equipment for its transportation [3–7]. Processing of production shops with formulations based on low molecular weight antibiotics may not lead to the desired result, since some biocides have low adhesion to the surfaces being treated and, as a result, require frequent renewal. In addition, bacteria can rapidly develop resistance to low molecular weight biocides, which, in addition, are not always able to fight all types of bacterial cultures inhabiting food shops [8,9]. Polymers have proven themselves as new antibacterial functional coatings. As a rule, in such cases, the polymers are the matrix for low molecular weight biocides, which give the coating durability, but do not have an antibacterial effect by themselves [10]. Also, in most research works, such coatings are the product of a complex and expensive synthesis [11].

It is important to create affordable and cheap antibacterial compositions. In this regard, special attention should be paid to coatings from interpolyelectrolyte complexes (IPECs) [12]. Biocidal compositions based on IPEC with a predominance of cationic groups have regions with freely charged groups capable of providing a biocidal effect and adhesion to hydrophilic surfaces, as well as hydrophobic regions that increase adhesion to hydrophobic surfaces and make it possible to modify such systems with low molecular weight antibacterial agents.

Most of the works aimed at the creation of biocidal coatings from IPEC are devoted to the study of coatings obtained by layer-by-layer deposition [13–15]. However, the most promising from a practical point of view is the method of obtaining a coating by applying a prepared solution of water-soluble IPEC onto a substrate. It has been established that coatings based on polydiallyldimethylammonium chloride (PDADMAC) and sodium polystyrene sulfonate (PSS) coatings can be prepared from solution of the complex on various types of surfaces, both hydrophilic and hydrophobic, and that such coatings are resistant to water wash-off. Such coatings are of great interest, since, as is known from the literature, PDADMAC itself has a high biocidal activity [16,17]. Thus, coatings based on PDADMAC with PSS with an included antibiotic will have both wash-off resistance and dual biocidal action.

For the synthetic polymers, the average molecular weight and molecular weight distributions are the key parameters that determine the physical and mechanical properties of the materials [18–20]. In general, PDADMACs are products of radical polymerization using various initiators [21]. As a result, they have broad polydispersity. The average molecular weights can be controlled by the concentrations of the initiators and monomers and the process temperature. Additional control of the molecular weights and their distribution can be achieved by using more precise and controlled polymerization procedures, which can complicate the process of obtaining PDADMAC on an industrial scale, as well as make it less commercially available [22].

Thus, there is a big question about the optimal degrees of polymerization of polymers required to create such coatings, since the molecular weight can affect the antibacterial and physicochemical properties. In particular, the main task is to establish the role of the molecular weight of the predominant polycation in the complex—PDADMAC.

In this paper, we will consider the main properties of coatings based on PDADMAC with different molecular weights and make a recommendation on the choice of the degree of polymerization for creating stable antibacterial coatings.

2. Materials and Methods

2.1. Materials

The three samples of polydiallyldimethylammonium chloride (PDADMAC) with average molecular mass M = 400–500 kDa (PDADMAC400), M = 200–300 kDa (PDADMAC200) and M < 100 kDa (PDADMAC100) from Sigma-Aldrich (St. Louis, MO, USA) were used as received without additional purification.

Glass cover slips with an area of 3.24 cm^2 were used in the experiments on washing-off polymeric coatings. Glass slides with an area of 19.76 cm^2 were used to study the moisture saturation and adhesive properties of films. Before all the experiments, glass substrates were subjected to the sample preparation stage. Cleaning and degreasing of glass surfaces were carried out as follows: the substrate was dipped in methanol and vigorously shaken for a minute. After that, the glass substrate was activated: the coverslip was treated with 1 M KOH solution, then washed with bidistilled water and dried in an air atmosphere. Bidistilled water with a conductivity of 0.05 μS/cm was used in all the experiments.

2.2. Methods

2.2.1. PDADMAC Coatings Wash-Off Procedure

The freshly cleaned substrate (square glass cover slip) with 3.24 cm^2 area was weighed. The 200 μL aliquot of the 20 mg/mL polymer solution was applied to the substrate so that the entire glass surface was covered with the solution. The sample was left to dry overnight in air atmosphere. The prepared sample was once again weighed and the mass of the film was calculated as the difference between the masses of substrate with film and substrate without the coating. Each cycle of wash-off was as follows: 200 μL of water was applied to the glass with coating, so that it completely covered the surface of the film. After two minutes of incubation the liquid was removed and the sample was left to dry. The sample was weighed and the mass loss was calculated. The experiments were carried out at a relative humidity of 15–20%.

2.2.2. PDADMAC Coatings Moisture Saturation Analysis

Freshly cleaned substrate (glass slide) with 19.76 cm^2 area was weighed. The 1220 µL aliquot of the 20 mg/mL solution of polymer was applied to the substrate so that all the area was covered with the solution. The sample was left to dry overnight in an oven with 5% relative humidity. The prepared sample was weighed again, the mass of the film was calculated as the difference between the masses of substrate with film and the substrate without coating, and this value was used as a reference. Then the samples were kept for a day in a chamber in which a certain relative humidity was maintained. After that, the samples were weighed again. In total, several intervals of relative humidity values were obtained: 5–6%, 13%, 19%, 41–44%, 61%, 68%, 88–90% (to set the humidity in chamber the saturated solutions of simple salts were used). The weight gain of the coating after incubation in a controlled humidity environment was used to evaluate the ability of the polymer coating to absorb water from the air. The control of the humidity was performed using ASTM standardized Temperature and Humidity Datalogger DT-172 by CEM Test Instrument (Moscow, Russia).

In addition to gravimetry, the moisture content of the samples was controlled by thermogravimetric analysis (TGA) using synchronous thermal analysis instrument STA 449 F3 Jupiter by Netzsch (Selb, Germany). Lyophilized samples of PDADMAC100, PDADMAC200 and PDADMAC400 were used for the experiment, the sample weighed was 1–5 mg. The heating rate was 10 K/min. The sample was heated in a controlled atmosphere chamber (relative humidity 40%) and the change in mass was simultaneously recorded [23]. The thermal analysis was performed using the equipment purchased in the scope of the Program for Development of Lomonosov Moscow State University.

The gravimetry analyses were made using precise balances VLA-120 M by Gosmetr (Saint Petersburg, Russia).

2.2.3. PDADMAC Coatings Adhesive Properties Analysis

Freshly cleaned substrate (glass slide) with 19.76 cm^2 area was weighed. The aliquot of the 20 mg/mL polymer solution was applied to the substrate so that the entire surface was covered with the solution. Two minutes later, the polymer solution was removed and the substrate was washed with bidistilled water. Then, the glass slide was covered from above with previously cleaned glass and dried for 24 h. After drying, the adhesive properties of the polycations were evaluated by the stress required to separate the two glass substrates. The experiments were carried out at room temperature and relative humidity of 15–20%. The adhesive properties were evaluated by dynamometry on a tensile testing machine by Metrotest (Moscow, Russia). The experiments were carried out at constant rate of the traverse 5 mm/min. The resulted data was collected using the software supplied by manufacturer.

2.2.4. Estimation of Minimal Inhibitory Concentrations (MIC) of PDADMAC

The estimation of MIC values was made for Gram-positive bacteria *B. subtilis* in Lysogeny broth (LB) medium and in Tryptic Soy Broth (TSB) medium. This last medium is favorable for biofilm formation. The MICs in LB and TSB medium were determined using a broth microdilution assay [24]. The cell concentration was adjusted to approximately 5×10^5 cells/mL. The three solutions of PDADMACs with an initial concentration of 20 mg/mL were used as the test compound. The polycations solutions were serially diluted two-fold in a 96-well microplate (100 µL per well). The microplates were covered and incubated at 37 °C with shaking. The OD600 of each well was measured, and the MIC was assigned as the lowest concentration of the tested compound that resulted in no growth after 16–20 h. Bacterial cell growth was measured at 590 nm using a microplate reader (VICTOR X5 Light Plate Reader, PerkinElmer, Waltham, MA, USA).

In the statistical analyses, the average results of at least five experiments are presented as mean values.

3. Results

3.1. Wash-Off Resistance Study

The resistance of the polymer films towards wash-off with water was controlled by the weight loss of the sample. The results are presented in Figure 1 as the dependence of the residual mass of the film upon the number of the wash-off cycle. For PDADMAC200 and PDADMAC400, about 65% of weight loss was observed after the first wash-off cycle and almost all of the polycation was removed after four cycles of the wash-off. At the same time, PDADMAC100 lost about 80% of its coating mass after the first wash-off cycle.

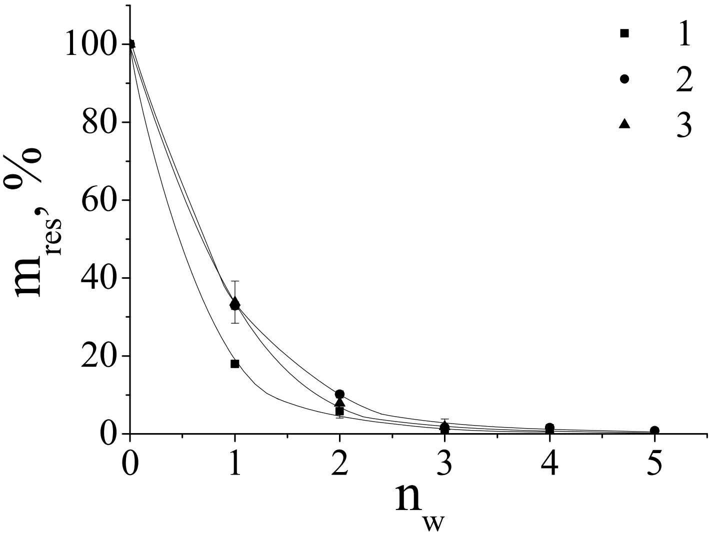

Figure 1. Dependence of the percentage of residual mass of coatings (m_{res}) formed from PDADMAC 100 (1), PDADMAC 200 (2) and PDADMAC 400 (3) on the number of wash-off cycles (n_w).

3.2. Estimation of Moisture Saturation of Coatings

On the next step, the ability of the PDADMAC coating to absorb water from environment was tested. The results are presented in Figure 2 as dependence of the relative growth of the weights of the films upon the relative humidity of the environment. It has been established that an increase in humidity from 15% to 70% leads to a gradual increase in the weight of the coating from 5% to 40%. No influence of the molecular weight of the PDADMACs upon the ability of the films to absorb water was detected—the curves completely coincided for the PDADMAC100, PDADMAC200 and PDADMAC400. The polymer films adsorbed on the glass surfaces did not change their shapes during the experiment.

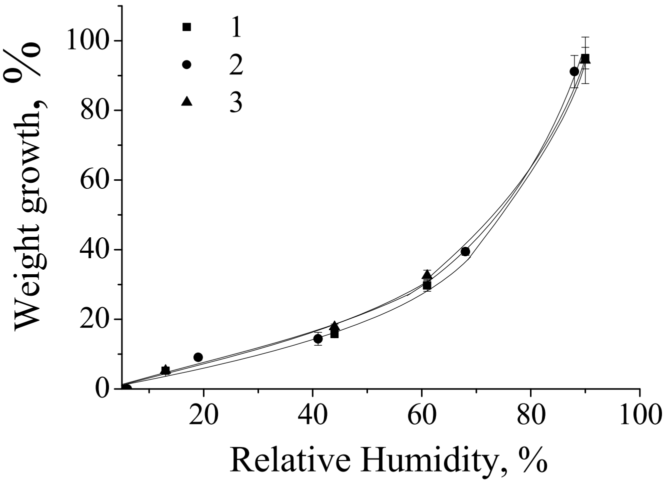

Figure 2. Dependence of the coating formed from PDADMAC 100 (1), PDADMAC 200 (2) and PDADMAC 400 (3) weight on the relative humidity.

The control thermogravimetric experiment was performed for the solid polycations in atmosphere with relative humidity 40%. The TGA experiments with lyophilized samples of PDADMACs has demonstrated that the samples weight loss corresponding to the loss of absorbed water was in interval 26–28% of their initial weight. Thus, these results confirm the experimental data presented in Figure 2.

3.3. Study of Adhesive Properties

To evaluate the adhesive properties of coatings formed from PDADMAC of different molecular weights, the dynamometry method was used. Figure 3 shows a typical stress versus time curve for the polycation under study. The maximum value of the applied stress (peak value) was taken as the characteristics of the adhesive properties of the PDADMAC film.

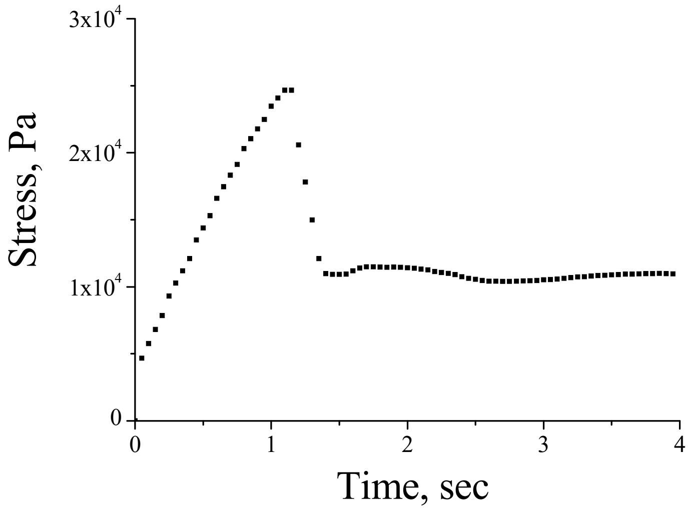

Figure 3. Typical stress versus time curve for PDADMAC100. Traverse rate 5 mm/min.

It was found that no significant difference in adhesive properties could be detected for coatings prepared from all the studied polycations. Thus, the mean value of the stress measurements was 26,600 Pa, 30,870 Pa, and 31,500 Pa for PDADMAC100, PDADMAC200, and PDADMAC400, respectively (please see the Supplementary Materials Figure S1 for the details and typical stress versus time curves for the PDADMACs of different molecular weights).

3.4. Evaluation of the Antibacterial Activity of Polycations

Minimum Inhibitory Concentration (MIC) (the minimum concentration of PDADMAC of different molecular weights at which bacterial growth is completely inhibited) was measured. The results are presented in Table 1. It has been established that the polycations of the presented degrees of polymerization exhibit the same antibacterial activity.

Table 1. MICs for polycations of various molecular weights.

MIC (μg mL^{-1})	PDADMAC100	PDADMAC200	PDADMAC400
TSB medium	0.025	0.025	0.025
LB medium	0.1	0.05	0.1

4. Discussion

The application of an aqueous solution of PDADMAC to the surface of a hydrophilic glass leads to the formation of a polymer coating. It can be seen that for PDADMACs with masses of 200 kDa and more, the profile of the mass loss curve upon washing off remains almost identical, while for the oligomeric fraction with a mass less than 100 kDa, the bulk of the polymer film is removed during the first 2 wash-off cycles. Apparently, this is due to the large diffusion coefficients of small macromolecules, which are able to both dissolve faster in water and move faster in the bulk of the swollen film. However, for all three PDADMAC fractions, the main weight loss is observed after three washing cycles. Thus, for the preparation of formulations based on PDADMAC, it is desirable to use polymers with a molecular weight above 200 kDa. It has also been established by dynamometry that a change in the molecular weight of PDADMAC has practically no effect on the adhesive properties.

It is known that polyelectrolytes are very hygroscopic and coatings obtained from them have high water absorption [25]. Evaluation of the ability of PDADMAC coatings to absorb water from the environment is necessary to assess the adequacy of the perception of the results of gravimetric analysis of wash-off resistance. It has been shown by gravimetry that PDADMAC is adsorbed on the glass surface with the formation of a polymer film capable of absorbing up to 40% of water on an initial weight basis from an environment with a relative humidity of up to 70%. This absorption did not change the integrity and shape of the PDADMAC coating. This result was observed for samples of all presented molecular weights. It should be noted that at a relative humidity of more than 70%, water drops were visible on the polymer coatings obtained from PDADMAC100, PDADMAC200 and PDADMAC400. However, after being placed in a chamber with low humidity, the film then acquired its initial form and integrity. Since experiments on the analysis of adhesion properties and wash-off resistance of polymer coatings are carried out at room humidity in the laboratory (about 15–20%), it seems important to evaluate the behavior of polymer films under these conditions. It has been established that with an increase in relative humidity from 5% to 20%, the mass of all the samples increases up to 10%. With a further increase in relative humidity up to 40%, the films gained up to 20% of the mass from the initial one. It should be noted that for PDADMAC100, PDADMAC200 and PDADMAc400 no difference was observed in the results obtained. Gravimetry results are consistent with the results of TGA, during which it was shown that at a relative humidity of 40%, up to 26–27% of water is present in the samples. Since the gravimetric evaluation of the resistance to wash-off was carried out at a relative humidity in the room below 20%, we can state the reliability of the results obtained. We suppose that the absorption of the water depends on structure of the film and the hydrophilic/hydrophobic balance in macromolecules. The second parameter is the same for all of the samples. It seems that for the flexible PDADMAC even molecular weight 100 kDa is sufficient to ensure formation of the continuous film without a reasonable number of defects that could affect porosity. As a result, the porosity of the studied films did not depend on molecular weights of PDADMACs.

It is known that increase of molecular weight of the polymers of the same structure could decrease or increase their biocidal activity [26–28]. In this work we discuss the behavior of PDADMAC with relatively high degrees of polymerizations. Minimum inhibitory concentrations of PDADMAC of different molecular weights were determined. It was found that the same concentration of inhibition of bacterial growth was observed for all the samples. It is important to stress that for the large-scale application of the polymers as biocide coatings it is assumed to use commercial available polymers. For polymers produced for the flocculation and oil production processes typical molecular weights are hundreds of kDA [25,29]. So, the findings about role of the molecular weights of PDADMAC molecules under investigation in this paper could be applied to the industrial samples of polymers.

5. Conclusions

It can be concluded that polycations can be used to create biocidal compositions based on IPEC in a wide range of PDADMAC molecular weights. However, it is desirable that the molecular weight of PDADMAC should exceed 100 kDa—otherwise the polycation forms coatings that are weakly resistant to washing off. At the same time, it was shown that the adhesive properties of coatings and the antibacterial properties of PDADMAC are practically independent of the molecular weight of the polymers.

Thus, when choosing a polycation for creating antibacterial coatings, the best option from a practical and functional point of view is to choose polymers with a higher degree of polymerization, but this requirement is not mandatory.

It should be taken into account that commercial available PDADMACs, widely used on the market as flocculants and components of oil drilling compositions, have relatively high molecular weights and, as a result, high viscosity of concentrated solutions. Therefore, the production of biocidal coatings using these polymers can be difficult due to the use of unprofitable production lines. At the same time, it was previously shown that, in order to form a stable positively charged water soluble IPEC, it is necessary to use a poly-cation with molecular masses exceeding those of the polyanionic component [30,31]. Therefore, the choice of the PDADMAC molecular weight for the component of the biocidal composition should be conditioned by several competing parameters: wash-off resistance, cost of production, balance of molecular weights between polycation and polyanion.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/physchem3010011/s1, Figure S1: Typical stress versus time curve for PDADMAC coatings on glass-PDADMAC100 (a); PDADMAC200 (b) and PDADMAC400 (c).

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