

Article

Epoxy Resin/Ionic Liquid Composite as a New Promising Coating Material with Improved Toughness and Antibiofilm Activity [†]

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Abstract

Long-chain imidazolium-based ionic liquids (ILs) possess a broad-spectrum biological activity and are considered promising antifouling agents for protective coatings. A new hydrophobic IL, 1-dodecyl-3-methylimidazolium dodecylbenzenesulfonate (C₁₂C₁IM-DBS), has been synthesized, and a modified epoxy coating material containing 10, 20, and 30 wt% of this IL was prepared by dissolution of C₁₂C₁IM-DBS in commercial DER 331 epoxy resin, followed by a curing phase with diethylenetriamine. Infrared analysis revealed physicochemical interactions between the hydroxyl groups of the resin and the IL. Spectrophotometric studies showed no release of C₁₂C₁IM-DBS after 30 days of exposure of the modified coatings to water. The plasticizing effect of the IL on the epoxy resin was established by differential scanning calorimetry analysis. The introduction of 10 and 20% C₁₂C₁IM-DBS into DER 331 reduced its glass transition temperature from 122.8 °C to 109.3 and 91.5 °C, respectively. The hardness of epoxy resin decreased by approximately 26% after the introduction of the IL. Moreover, DER 331/C₁₂C₁IM-DBS coatings on steel substrates showed significantly improved impact resistance compared to neat resin. The antibiofilm efficiency of DER 331/C₁₂C₁IM-DBS coatings was evaluated by assessing the capability of two biofilm-forming model strains, *Staphylococcus aureus* ATCC 25923 and *Pseudomonas aeruginosa* PA01, to form attached biofilms on the surface. The IL effectively inhibited *S. aureus* surface-associated biofilm development even at the lowest content of 10%. On the contrary, an approximately 50% inhibition of biofilm metabolic activity was detected for DER 331/C₁₂C₁IM-DBS coatings containing 20% and 30% of the IL. Overall, the results of this study indicate that the hydrophobic IL C₁₂C₁IM-DBS is an efficient modifying additive for epoxy resins, which can significantly improve their operational properties for various industrial applications.



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Keywords: epoxy resin; protective coating; ionic liquid; plasticization; impact resistance; antibiofilm

1. Introduction

Epoxy resins are very convenient and economical materials for the production of protective coatings since they do not require the use of solvents and do not form low-molecular by-products during curing. Epoxy coatings have superior adhesion to various substrates, low shrinkage, excellent corrosion resistance, high mechanical strength, and abrasion and impact resistance. The main application areas of these versatile materials are protective primers and anticorrosive coatings in the marine, automotive, and building industries, as well as pipeline coatings in the oil and gas industry [1–5]. Marine epoxy coatings are widely used in the shipbuilding industry to protect multiple parts of marine vessels, such as hulls, ballast tanks, cargo tanks or holds, decks, etc. Like other solid substrates, epoxy coatings are quickly colonized by microbial biofilms in a moist environment, leading the way for the occurrence of macrofouling by aqueous organisms [6,7]. In the shipping industry, fouling promotes surface corrosion and causes high frictional resistance, leading to higher fuel consumption and speed reduction [8]. Biofouling also damages exposed marine facilities such as oil production platforms, power plants, industrial water inlet systems, or seawater-cooled systems [9–12]. The introduction of both inorganic and organic biocides into protective coatings is a common approach to inhibit biofilm formation on their surface. Thus, immobilization of functional inorganic nanofillers such as TiO_2 , ZnO , and Ag/TiO_2 in the porous epoxy matrix imparts excellent antibiofilm activity to the coatings [6,13,14]. The high surface area of inorganic oxides and the presence of polar hydroxyl groups allow their good dispersion in the polymer matrix due to strong interfacial adhesion. These antibacterial composites can kill both Gram-positive and Gram-negative microorganisms upon contact with the aqueous environment and are promising in designing self-disinfecting surfaces [6]. New antifouling formulations based on epoxy resin and halloysite clay nanotubes loaded with the commercial organic biocide DCOIT have also been reported [15]. The release time of the clay-encapsulated DCOIT was significantly extended compared to that of neat biocide, allowing for long-lasting antifouling activity. In addition to common biocidal epoxy compositions, which release biocides as their mode of action, contact-active antibiofilm systems are becoming increasingly popular as well. These sterile-surface materials contain biocides, which are water-insoluble and can kill microorganisms at the polymer-water interface. A new water-insoluble polymeric antimicrobial agent, poly(m-aminophenol), was synthesized and used as a modifying additive for epoxy coatings [16]. The prepared coatings, containing 5 wt% of poly(m-aminophenol), were found to have excellent antibacterial activity, as well as good antifouling performance by inhibiting the formation of bacterial biofilms. The modified epoxy coatings also demonstrated high corrosion-protected performance for mild steel [16]. Several studies reported the preparation of epoxy coatings containing natural antimicrobial agents such as oregano essential oil (OEO) [17] and *Artemisia annua* (AAP) powder [18]. Both additives imparted antimicrobial activity to epoxy coatings, as well as improved their impact strength and fracture resistance at the content of 5%–15%. It is worth noting that the brittle nature of epoxy resins causes serious drawbacks, such as low impact strength and low fracture resistance [15,19]. The preparation of modified epoxy coatings with enhanced mechanical properties and antifouling activity can therefore significantly improve their durability, as well as expand the areas of their potential applications. The long-chain onium salts, also known as ionic liquids (ILs), are of particular interest as multifunctional

modifying additives for epoxy resins. ILs are known for their wide range of antimicrobial activity against Gram-positive and Gram-negative bacteria and fungi, as well as their antibiofilm activity against a range of pathogenic microorganisms [19–21]. Long-chain 1-alkyl-3-methylimidazolium ILs exhibit antibiofouling activity against barnacle larvae with LC_{50} values less than 1 μ M [22]. However, common water-soluble ILs seem unsuitable as biocides in protective coatings due to their poor leaching resistance. This may also have a negative environmental impact since these compounds possess relatively high toxicity [23,24]. From this point of view, water-immiscible ILs are much more attractive. The results of recent studies revealed that such additives impart antibiofilm and antifouling activity to protective coatings at a sufficiently high content [25,26]. To our knowledge, there is no data in the literature regarding antifouling coatings based on epoxy resin and ILs. Several studies reported the use of imidazolium-based ILs as multifunctional modifying additives for epoxy resins, which can play the role of curing agents and plasticizers [27,28]. Phosphonium-based ILs were also found to have a plasticizing effect on crosslinked epoxy polymer matrix, as well as improve the fire resistance of the material [26].

In this study, a new hydrophobic ionic liquid, 1-dodecyl-3-methylimidazolium dodecylbenzenesulfonate ($C_{12}C_1IM-DBS$), has been synthesized and tested as a potential antifouling additive for epoxy coatings. A composite material, based on commercial epoxy resin DER 331 and $C_{12}C_1IM-DBS$, has been prepared and characterized in terms of antibiofilm activity, as well as with regard to its morphological, surface-related, physicochemical, and thermophysical properties.

2. Materials and Methods

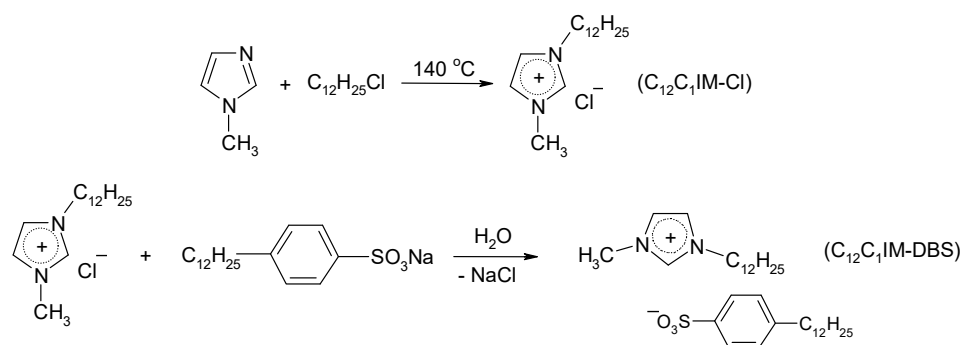
2.1. Materials

1-methylimidazole (for synthesis), 1-chlorododecane (97%), sodium dodecylbenzenesulfonate (technical grade), methylene chloride, hexane, and ethyl acetate (98%) were supplied by Sigma-Aldrich and used as received.

The commercial epoxy resin DER 331 (Dow Chemical, Midland, MI, USA) was used for the preparation of the protective coatings.

2.2. Synthesis of 1-Dodecyl-3-Methylimidazolium Dodecylbenzenesulfonate ($C_{12}C_1IM-DBS$)

1-dodecyl-3-methylimidazolium chloride ($C_{12}C_1IM-Cl$) was synthesized according to Scheme 1. A mixture of 1-methylimidazole (10 g, 0.12 mol) and 1-chlorododecane (31 g, 0.15 mol) was stirred at 140 °C for 24 h. After cooling to room temperature, the solid product was washed with a hexane–ethyl acetate mixture (3:1 (*v/v*), 3 × 50 mL). Residual solvents were removed under vacuum at 60 °C. The ionic liquid $C_{12}C_1IM-Cl$ was obtained as a white solid compound with a melting point of 46–47 °C.



Scheme 1. Synthesis of the ionic liquid $C_{12}C_1IM-DBS$.

An aqueous solution of sodium dodecylbenzenesulfonate (24 g/300 mL) was added to the stirred solution of C₁₂C₁IM-Cl (20 g, 0.07 mol) in 200 mL of water. The mixture was stirred for 1 h, and the product was subsequently extracted with methylene chloride (2 × 250 mL). The combined solution was dried over sodium sulfate. Methylene chloride was distilled off, and the residual solvent was removed under vacuum. The ionic liquid C₁₂C₁IM-DBS was obtained as a light brown semi-solid product.

The structure of synthesized compounds was confirmed by nuclear magnetic resonance (NMR). A ¹H NMR spectrum was recorded in CDCl₃ on a Varian Gemini-2000 (400 MHz) spectrometer (Varian, Inc, Palo Alto, CA, USA).

1-dodecyl-3-methylimidazolium chloride (C₁₂C₁IM-Cl)

¹H NMR (400 MHz, CDCl₃): δ = 0.84 (t, *J* = 6.7 Hz, 3H, CH₃), 1.14–1.36 (m, 18H, CH₃(CH₂)₉), 1.87 (t, *J* = 7.3 Hz, 2H, NCH₂CH₂), 4.10 (s, 3H, NCH₃), 4.23–4.32 (m, 2H, NCH₂), 7.32 (t, *J* = 1.8 Hz, 1H, C₄-H), 7.49 (t, *J* = 1.8 Hz, 1H, C₅-H), 10.58 (d, *J* = 1.6 Hz, 1H, C₂-H).

1-dodecyl-3-methylimidazolium dodecylbenzenesulfonate (C₁₂C₁IM-DBS)

¹H NMR (400 MHz, CDCl₃): δ = 0.68–0.91 (m, 9H, CH₃); 0.96–1.34 (m, 33H, CH, CH₂); 1.36–1.65 (m, 4H, CH₂); 1.79 (p, *J* = 6.9 Hz, 2H, NCH₂CH₂); 4.00 (s, 3H, NCH₃), 4.17 (t, *J* = 7.4 Hz, 2H, NCH₂); 7.11 (dd, *J* = 15.4, 8.2 Hz, 2H, C₃-Ar-H, C₅-Ar-H); 7.28 (t, *J* = 1.8 Hz, 1H, C₄-H), 7.44 (t, *J* = 1.8 Hz, 1H, C₅-H), 7.75–7.82 (m, 2H, C₂-Ar-H, C₆-Ar-H); 9.88 (d, *J* = 1.7 Hz, 1H, C₂-H)

2.3. Preparation of Modified Epoxy Coatings

The curing agent D.E.H.™ (diethylenetriamine) was mixed with liquid epoxy resin (10% (*w/w*)). C₁₂C₁IM-DBS (10, 20, and 30 wt%) was added to this mixture, which was then stirred for 15 min at room temperature. Silicone forms (1 × 1 × 1 cm) were filled with liquid epoxy formulations and cured for 24 h at 25 °C and 4 h at 60 °C. The prepared samples of epoxy composites were further tested for their physicochemical properties, as well as antibiofilm activity. To prepare the modified epoxy coatings, stainless steel bars (20 × 10 × 0.1 cm) were cleaned by wiping them with acetone, drying them in air, and sanding them for 10 min. Liquid epoxy formulations were applied to the surface of the bars using a paintbrush.

Curing of the coatings was carried out for 24 h at 25 °C and 4 h at 60 °C. The thickness of the coatings was 30 ± 5 μm. The control samples were prepared by curing neat epoxy coatings.

2.4. Characterization of the Modified Epoxy Resin

The vibrational properties of the IL and its composites with epoxy resin were studied using a Nicolet 6700 FTIR spectrometer (Thermo Fisher Scientific, Waltham, MA, USA). Polymer samples were placed in contact with a single-reflection diamond ATR (Attenuated Total Reflection) crystal, and spectra were collected over the range of 600–4000 cm^{−1} (50 scans).

Contact angle measurements were performed using a Drop Shape Analyzer DSA25E (Krüss, Hamburg, Germany) by the sessile drop method. All reported contact angles are the average of at least five measurements taken at different locations on the polymer surface. Water absorption of DER 331/C₁₂C₁IM-DBS composites was determined using test specimens of 1 × 1 × 0.2 cm, which were kept in distilled water at 25 °C. The samples were periodically removed, wiped with a filter paper, and weighed on an analytical balance with a precision of ±0.1 mg.

Vickers hardness values of epoxy resin samples were measured using a Vickers hardness indenter (UIT-HVmicro-1, Microtech, Kharkiv, Ukraine). The load on the indenter was 100 g. An average value of five measurements was determined. Impact resistance of epoxy coatings was evaluated according to ISO 6272-1 [29] using the NOVOTEST-V1-53007

(NOVOTEST, Samar, Ukraine) impact tester. In this test, the coated steel bar was placed on a fork with the coating facing outwards (direct impact). A ball with a diameter of 8 mm and a weight of 1 kg was dropped freely down a guide tube on the coating from a variable height. After the impact, the deformed zone of the specimen surface was examined for cracks and flaking. The minimum drop height for which the coating cracks or peels from its substrate was determined.

The adhesive properties of epoxy coatings were studied by the lap shear test [29]. Stainless-steel bars of $10 \times 2 \times 0.5$ cm were used as a basis. The steel surfaces were pretreated with 360-mesh sandpaper to remove surface impurities and metal oxides, then with 500-mesh sandpaper to obtain a smooth surface, and finally degreased by immersion in acetone for 10 min. After the surface treatment, two steel bars were coated with liquid epoxy formulations along the edges and assembled into single-lap shear joints with an overlap length of $1.5 (\pm 0.1)$ cm. The mold was placed under a press, and the epoxy adhesive was cured for 24 h at 25 °C and 4 h at 60 °C at a contact pressure of 2.5 bars. An example of an adhesive joint after curing is shown in Figure 1. The tensile shear strength of the adhesive joint was determined using the universal testing machine Instron 3400 (Instron Ltd, High Wycombe, UK) equipped with a load cell of 500 N. The tensile load was applied at a deformation rate of 2 mm/min. An average value for the tensile strength was obtained from five samples of each coating [30].

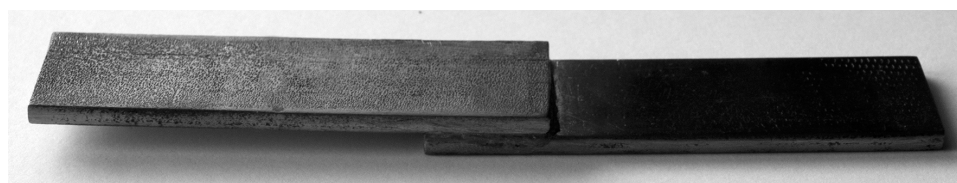


Figure 1. Stainless steel bars joined with a DER 331/C₁₂C₁IM–DBS composite.

Scanning electron microscopy (SEM) images were acquired using a MIRA 3 microscope (Tescan GmbH, Brno, Czech Republic, operating at a 10 kV electron beam energy. To enhance imaging quality and mitigate charging effects, a 6 nm platinum layer was sputtered onto the sample surface using a Kurt J. Leskermagnetron (Jefferson Hills, PA, USA). Chemical composition spectra and mapping analysis were conducted through energy-dispersive X-ray spectroscopy (EDX) employing a Bruker XFlash detector (Bruker Nano GmbH, Berlin, Germany) directly integrated into the SEM.

XPS measurements were performed using an EnviroESCA system (SPECS Surface Nano Analysis, GmbH, Berlin, Germany) with a monochromated Al K_α X-ray source (1486.6 eV). Signal detection was conducted using a Phoibos 150 NAP 1D-DLD (SPECS, GmbH, Berlin, Germany) hemispherical energy analyzer in the FAT (Fixed Analyzer Transmission) mode at 1 mbar of argon. The samples were stuck to the stainless-steel sample holder using indium foil, and the spot size was about 200 μm. During these XPS measurements, the core-level spectra of C 1s, O 1s, N 1s, and S 2p were recorded with a pass energy of 20 eV, a step size of 0.1 eV, and a dwell time of 0.3 s.

Differential scanning calorimetry (DSC) analyses were performed on a Discovery Q 250 calorimeter (TA Instruments, New Castle, DE, USA). The temperature range was set from −90 °C to 200 °C, with a temperature rate of 10 °C/min and an N₂ flow of 50 mL/min. For the analysis, curves were taken from the second heating run.

The release of C₁₂C₁IM–DBS from the coatings in water was studied by UV–visible spectrophotometric analysis using a Jenway 6850 spectrometer (Bibby Scientific Ltd., Stone, UK). The water-soluble precursor (C₁₂C₁IM–Cl) was used for creating a calibrating graph. The absorbance of a series of aqueous solutions was measured at 212 nm (the characteristic peak of the imidazolium cation [31]) in a concentration range of $1 \cdot 10^{-5}$ – $1 \cdot 10^{-4}$ mol/L. Each

painted steel bar (3 samples) was kept in 1 L of deionized water at 25 °C. The solution was periodically analyzed by measuring the absorbance at the mentioned wavelength. Each measurement was repeated three times.

The antibiofilm efficacy of C₁₂C₁IM-DBS was assessed by evaluating the ability of two model biofilm-forming strains, specifically, *Staphylococcus aureus* ATCC 25923 and *Pseudomonas aeruginosa* PA01, to form surface-attached biofilms on epoxy coatings after 48 h of static incubation. Metal substrates (1 cm²) coated with either DER 331 or DER-C12C1IM-DBS were sterilized by autoclaving at 105 °C for 30 min. Each substrate was placed in a well of a sterile 24-well polystyrene plate containing 2 mL of Luria Broth (LB) inoculated with 10 µL of an overnight culture (~10⁹ CFU/mL). Each variant was tested in six replicates. Plates were incubated at 37 °C for 48 h. Controls were included by incubating sterile films in LB (n = 4). After incubation, substrates were washed three times to remove planktonic and loosely attached biofilms. Simultaneously, the supernatant from each well was plated to quantify planktonic CFUs. To assess the total biofilm metabolic activity, an MTT assay was performed. Biofilm-coated substrates (n = 6 per strain) were transferred to fresh sterile 24-well plates and treated with 200 µL of 0.05% MTT (3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide; Sigma-Aldrich, Gillingham, UK) in LB. Plates were incubated at 37 °C for 2 h. The LB-MTT medium was centrifuged at 10,000 g for 10 min, and the resulting formazan pellet was resuspended in 200 µL DMSO. An additional 200 µL of DMSO was added to each sample. From the resulting 400 µL solution, 100 µL was transferred to a 96-well polystyrene plate, and absorbance at 570 nm (A₅₇₀) was measured using a Multiskan FC Microplate Photometer (ThermoFisher Scientific, Waltham, MA, USA).

Biofilms formed on epoxy-coated substrates were further analyzed using Confocal Laser Scanning Microscopy (CLSM), following previously established protocols. For visualization, biofilms were stained with 1 mM ethidium bromide and 5 µg/mL calcofluor white (both from Sigma-Aldrich). To preserve the integrity of the biofilm structure, no additional washing steps were performed after staining. Samples were not chemically fixed; instead, a coverslip was gently placed over the stained surface prior to imaging. CLSM was conducted using a Leica TCS SPE Confocal system equipped with a coded DMi8 inverted microscope (Leica Microsystems GmbH, Wetzlar, Germany), operated via the Leica Application Suite X (LAS X) software, version 3.4.1. Imaging parameters included excitation at 405 nm and emission collection between 450 and 500 nm for calcofluor white, and excitation at 532 nm with emission collected between 537 and 670 nm for ethidium bromide. Pixel quantification was performed using the LAS X software.

3. Results

3.1. FT-IR Analysis

In the IR spectrum of C₁₂C₁IM-DBS (Figure 2, spectrum 1), the broad band in the frequency range between 3300 and 3700 cm^{−1} is assigned to the stretching vibrations of H₂O molecules, which are associated with the IL. The well-defined bands at 3151 and 3101 cm^{−1} are attributed to the ring C(4/5)-H and C(2)-H stretching modes of the imidazole ring, respectively [32]. The strong bands at 2852 and 2922 cm^{−1} are assigned to the symmetric and asymmetric stretching vibrations of the methylene groups, and the two weaker bands observed at 2872 and 2955 cm^{−1} are assigned, respectively, to the symmetric and asymmetric stretching vibrations of the methyl group of the alkyl chains [33]. The weak bands at 1601, 1494, and 1406 cm^{−1} correspond to aromatic C=C stretching vibrations of benzene rings. The band at 1575 cm^{−1} is assigned to both the ring in-plane symmetric CH₃(N) and CH₂(N) stretching modes. The band at 1464 cm^{−1} is associated with the CH₂ scissoring mode, and the small-intensity one at 1379 cm^{−1} with the CH₃ bending

mode of the alkyl chain [33]. The strong band at 1188 cm^{-1} corresponds to the C–N–C symmetric stretching vibrations of the imidazole ring, and the shoulder at 1213 cm^{-1} to the asymmetric stretching vibrations of the S=O group. The strong band at 1033 cm^{-1} is assigned to the symmetric stretching vibrations of this group. The asymmetric band at 1124 cm^{-1} is attributed to a combination of the benzene ring breathing vibration and SO_3 stretching mode [26,34].

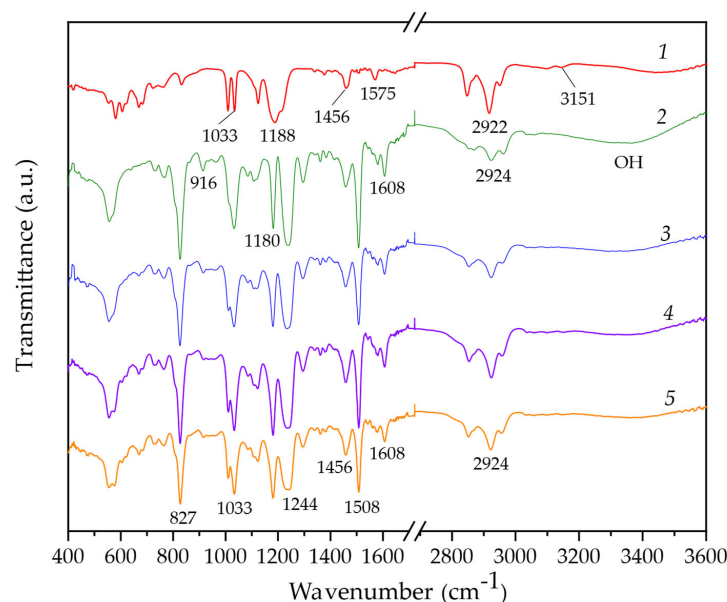
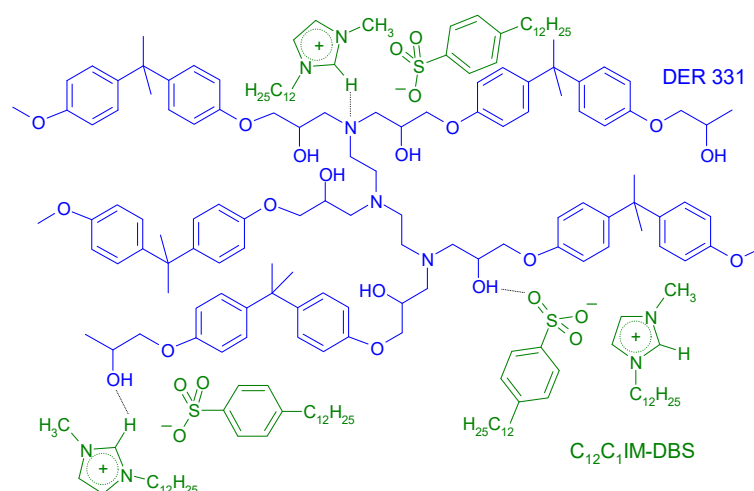


Figure 2. IR spectra: 1— $\text{C}_{12}\text{C}_1\text{IM-DBS}$, 2—DER 331, 3—DER 331/ $\text{C}_{12}\text{C}_1\text{IM-DBS}$ (10%), 4—DER 331/ $\text{C}_{12}\text{C}_1\text{IM-DBS}$ (20%), and 5—DER 331/ $\text{C}_{12}\text{C}_1\text{IM-DBS}$ (30%).

The IR spectrum of cured DER 331 epoxy resin (Figure 2, spectrum 2) contains a broad band in the $3100\text{--}3700\text{ cm}^{-1}$ region, which is assigned to OH and NH stretching vibrations. The weak band at 3035 cm^{-1} corresponds to the stretching vibrations of aromatic C–H bonds. The bands observed at 2854 and 2924 cm^{-1} are assigned to the symmetric and asymmetric stretching vibrations of the methylene groups, and the bands at 2870 and 2962 cm^{-1} are assigned, respectively, to the symmetric and asymmetric stretching vibrations of the methyl groups. The strong band at 1508 cm^{-1} , as well as weaker bands at 1608 and 1577 cm^{-1} are attributed to the stretching vibrations of aromatic C=C bonds [17,35]. The spectrum also reveals the presence of characteristic adsorption bands for the bending vibrations of the CH_2 and CH_3 groups at 1456 , 1384 , 1362 , and 1296 cm^{-1} [35]. The strong bands at 1244 and 1180 cm^{-1} are assigned to the stretching vibrations of C–O groups and the band at 1032 cm^{-1} to the C–O–C (aromatic ether) stretching mode [35,36]. The medium band at 1107 cm^{-1} and the weak band at 1083 cm^{-1} correspond to the stretching vibrations of the C–OH (alcohol) groups [36]. In the low-frequency region, the weak band at 916 cm^{-1} is assigned to the symmetric stretching vibrations of C–O–C bonds of the residual oxirane rings. The strong band at 825 cm^{-1} corresponds to the out-of-plane bending vibrations of aromatic C–H bonds, and the weak band at 769 cm^{-1} to the rocking vibrations of CH_2 groups [35,36].

In the IR spectra of DER 331/ $\text{C}_{12}\text{C}_1\text{IM-DBS}$ composites, the absorption band of oxirane rings at 916 cm^{-1} significantly decreased, which confirms successful curing of epoxy resin in the presence of the IL (Figure 2, spectra 3–5). A significant reduction in the intensity of the band in the $3100\text{--}3700\text{ cm}^{-1}$ region is also observed, even at low IL content (Figure 2, spectra 3–5). This may indicate the formation of hydrogen bonds between the hydroxyl groups of the cured epoxy resin and the polar groups of $\text{C}_{12}\text{C}_1\text{IM-DBS}$, in particular the slightly acidic C–H groups of the imidazolium cation, as well as the SO_3 groups of the

dodecylbenzenesulfonate anion. Moreover, the tertiary amine groups of the curing agent diethylenetriamine can also form hydrogen bonds with imidazolium cations (Scheme 2). Overall, this physicochemical interaction may lead to a good compatibility between the IL and the polymer matrix.



Scheme 2. Possible physicochemical interactions between epoxy resin and $C_{12}C_1IM-DBS$.

3.2. Morphological and Surface Properties of DER-331/ $C_{12}C_1IM-DBS$ Coatings

The distribution of the IL in the coatings was evaluated by means of energy-dispersive X-ray spectroscopy (EDX). Figure 3 illustrates EDX spectra of pure epoxy resin and epoxy resin modified with varying quantities of IL. Table 1 provides the weight percentages of the elemental composition in the samples.

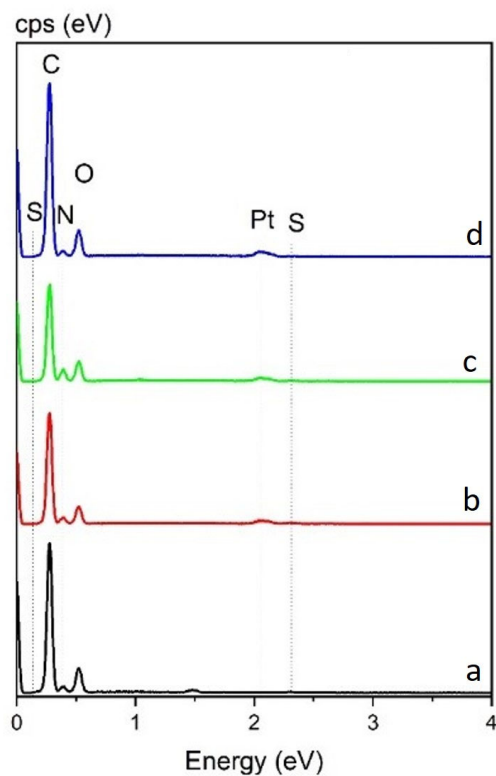
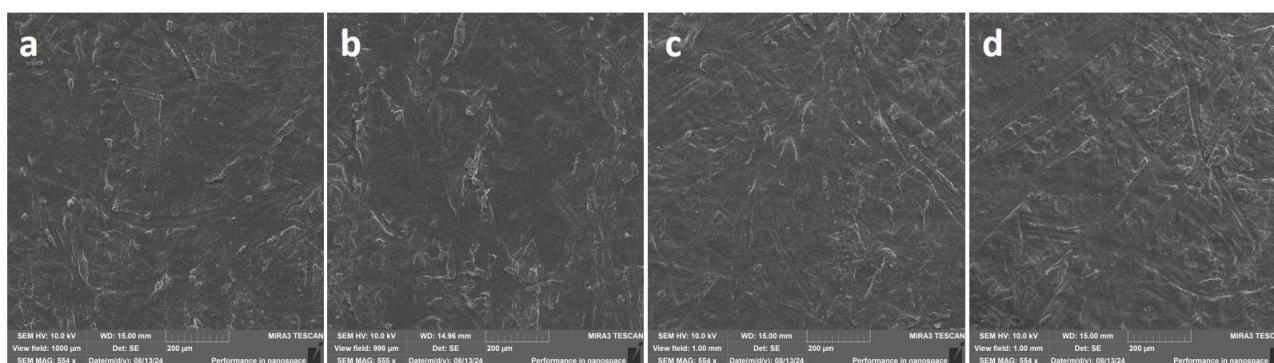


Figure 3. EDX spectra of DER 331 (a) and DER 331/ $C_{12}C_1IM-DBS$ composites containing 10% (b), 20% (c), and 30% (d) $C_{12}C_1IM-DBS$.

Table 1. Composition of the samples according to EDX analysis.

	DER 331 (Control)	DER 331/ C ₁₂ C ₁ IM-DBS (10%)	DER 331/ C ₁₂ C ₁ IM-DBS (20%)	DER 331/ C ₁₂ C ₁ IM-DBS (30%)
Element	Weight %			
C	73.7	74.3	73.5	74.30
O	20.7	19.8	20.0	19.20
N	5.5	5.4	5.8	5.20
S	0.1	0.6	0.8	1.30

The analysis of the SEM images of the DER331/C₁₂C₁IM-DBS composites (Figure 4) indicates their homogeneity. Complementary EDX mapping (Figure S1) reveals the presence and uniform distribution of nitrogen (N) and sulfur (S) atoms throughout the bulk of the samples, indicating effective incorporation of the IL.

**Figure 4.** SEM images of the composites' surfaces for DER 331 (a) and DER 331/C₁₂C₁IM-DBS composites containing 10% (b), 20% (c), and 30% (d) of IL.

Surface analysis by XPS (Figures 5, S1 and S3) further supports these findings. In particular, Figure 5 demonstrates an increase in sulfur content at the composite surface with increasing amounts of the IL. Notably, the sulfur content at the surface shows only a slight increase between 10 wt% and 20 wt% IL. However, at 30 wt%, a more pronounced increase in the sulfur peak is observed. This may suggest that at higher C₁₂C₁IM-DBS content, the epoxy matrix becomes less capable of fully incorporating the additive, leading to partial migration of IL to the surface.

The results of the water contact angle measurements on epoxy coatings are illustrated in Figure 6. The surface of neat DER 331 resin has a water contact angle (θ_w) of 74°, which indicates its hydrophilic properties [14,37]. The introduction of 20 and 30 wt% of C₁₂C₁IM-DBS significantly increased the wettability of epoxy coatings, which is manifested by a strong decrease in θ_w values (Figure 6). This effect is probably due to the increased content of IL on the surface of the polymer matrix. Thus, the formation of a hydration layer between water molecules and imidazolium cations can make the surface more hydrophilic.

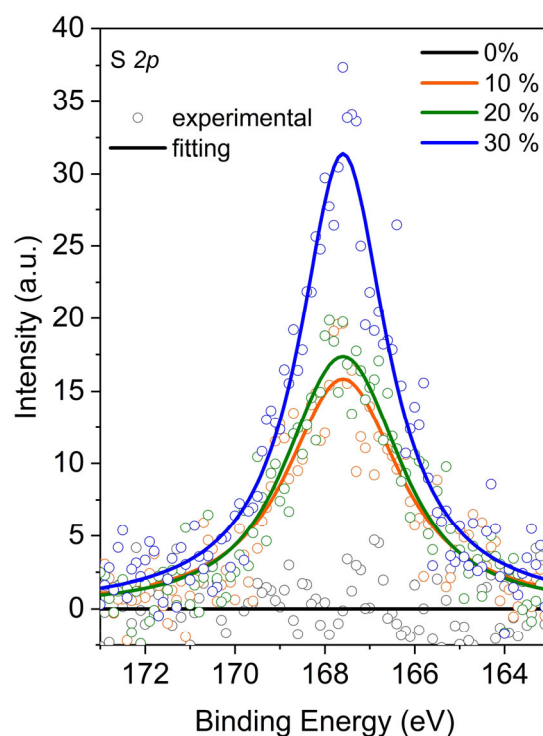


Figure 5. XPS S 2p core level spectra for DER 331 (black) and DER 331/C₁₂C₁IM-DBS composites containing 10% (red), 20% (green), and 30% (blue) of IL.

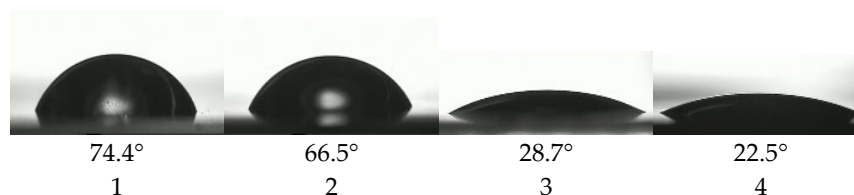


Figure 6. Photographs of water droplets on the surface of epoxy coatings: 1—DER 331 Control, 2—DER 331/C₁₂C₁IM-DBS (10%), 3—DER 331/C₁₂C₁IM-DBS (20%), and 4—DER 331/C₁₂C₁IM-DBS (30%).

The results of the water absorption measurements confirmed the higher hydrophilicity of modified epoxy coatings. The maximum water absorption for control DER 331 coatings was 0.3% (after 15 days of exposure). For DER 331/C₁₂C₁IM-DBS samples containing 20 and 30% of IL, this value was 1.1% and 1.3%, respectively.

UV–visible absorption spectroscopy is a convenient method to determine the concentration of imidazolium ILs in water. Thus, the UV spectrum of 1-dodecyl-3-methylimidazolium chloride (C₁₂C₁IM-Cl), which is a water-soluble precursor for the synthesis of C₁₂C₁IM-DBS, contains two intensive peaks at 191 and 212 nm (Figure 7, curve 1). These peaks are assigned to electronic absorption of imidazolium cations [31]. The calibrating graph was linear in the concentration range of $1.5 \cdot 10^{-4}$ – $8 \cdot 10^{-4}$ mol/L at $\lambda_{\max}$ 212 nm. As one can see from Figure 7, the spectra of water solutions after their contact with neat epoxy resin and DER 331/C₁₂C₁IM-DBS (30%) composite are very similar (Figure 7, curves 2 and 3). This indicates a high resistance of the IL to leaching from epoxy coating, which eliminates its potential negative environmental impact [26].

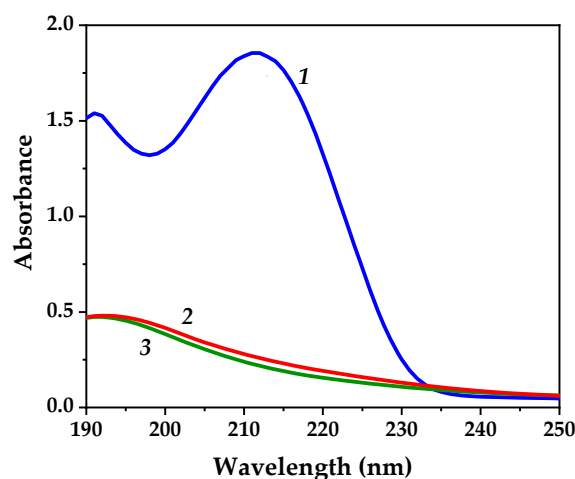


Figure 7. UV spectra of $C_{12}C_1IM-Cl$ ($C = 5 \cdot 10^{-4}$ mol/L) (1) and water solutions after 30 days of contact with DER 331 (2) and DER 331/ $C_{12}C_1IM-DBS$ (30%) (3).

3.3. Thermal and Mechanical Properties of DER 331/ $C_{12}C_1IM-DBS$ Composites

DSC analysis was performed to evaluate the impact of $C_{12}C_1IM-DBS$ on the glass transition temperature (T_g) of cured epoxy resin. On the thermogram of the IL (Figure 8a), the glass transition is observed at -54 °C. The neat DER 331 has a T_g around 122 °C (Figure 8b, curve 1), which is consistent with data in the literature [38]. The introduction of 10 and 20% $C_{12}C_1IM-DBS$ to the epoxy matrix reduced its T_g value by 13.5 and 31.3 °C, respectively (Figure 8b, curves 2 and 3). These results indicate the plasticizing effect of the IL on epoxy resin. Physicochemical interactions between bulk IL ions and hydroxyl groups of epoxy resin (Scheme 2) probably increase the free volume of the system, allowing greater polymer chain mobility. Further increase in $C_{12}C_1IM-DBS$ content to 30% did not have a noticeable effect on the T_g value of DER 331, which may be due to its cross-linked structure.

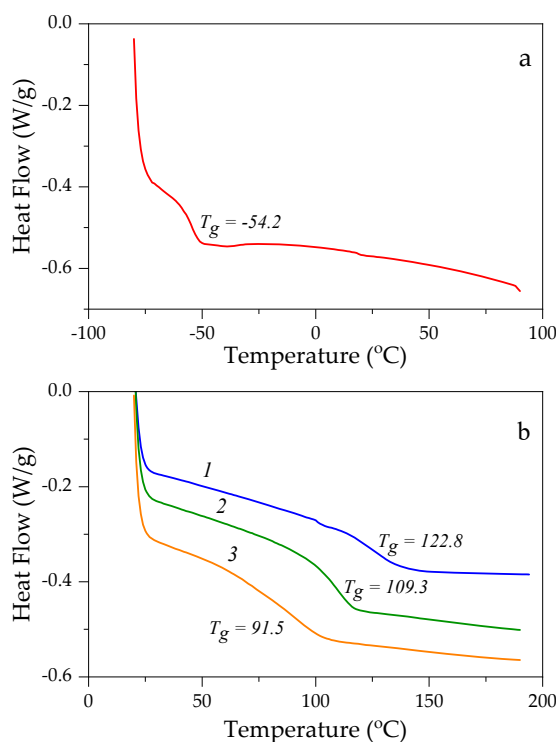


Figure 8. DSC curves: (a) $C_{12}C_1IM-DBS$, (b) 1—DER 331 (control), 2—DER 331/ $C_{12}C_1IM-DBS$ (10%), and 3—DER 331/ $C_{12}C_1IM-DBS$ (20%).

The results of the Vickers hardness studies of epoxy samples are shown in Figure 9. The hardness decreased sharply from 0.215 GPa for neat DER 331 to 0.16 GPa for epoxy composite containing 10 wt% of $C_{12}C_1IM-DBS$. This effect may be caused by the plasticization of epoxy resin, as well as a certain decrease in its cross-linking density. A further increase in the content of IL to 20 and 30% had no noticeable effect on the hardness values, which were 0.158 and 0.157 GPa, respectively (Figure 8). Thus, despite the high content of IL in epoxy composites, they maintain sufficient hardness, which is comparable to those for DER 331 resin filled with diamond nanoparticles [39] or multi-walled carbon nanotubes [40].

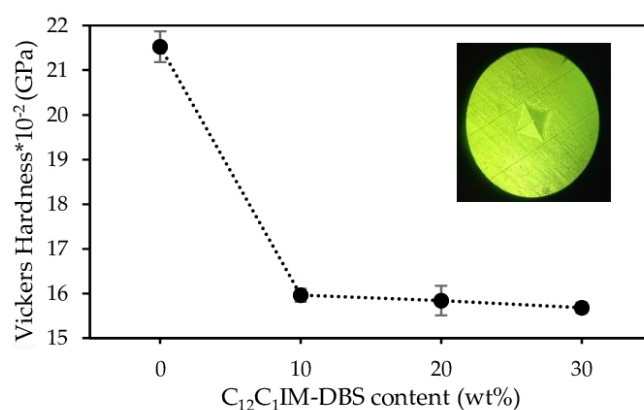


Figure 9. Effect of $C_{12}C_1IM-DBS$ content on the Vickers hardness of DER 331/ $C_{12}C_1IM-DBS$ composites and a microphotograph of an indentation on the surface of a material.

The photos of the epoxy resin-coated steel substrates after the impact resistance tests are shown in Figure 10. The control coatings based on DER 331 resin contain a significant number of concentric cracks and peeling, which indicate their poor resistance to rapid deformation. The defects were significantly reduced on the surface of samples containing 10% IL. Further increases in IL content to 20 and 30% impart excellent impact resistance to the coatings, which did not contain visible mechanical defects (Figure 10). These results clearly demonstrate that the hydrophobic ionic liquid $C_{12}C_1IM-DBS$ is an efficient modifier for epoxy resin, enhancing its flexibility and toughness. Similar effects were observed in other studies when the epoxy resin was modified with oregano essential oil [17], hyperbranched poly(methylacrylate-diethanolamine), and poly(methylacrylate-ethanolamine) [41].

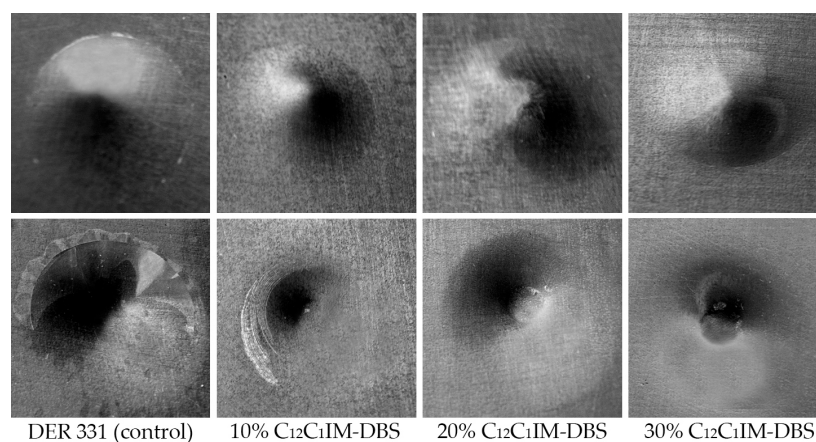


Figure 10. Epoxy coatings on the steel surface after the falling ball impact test: concave surface (top) and convex surface (bottom).

The adhesion strength of epoxy-based coatings and structural adhesives is an important operational property, which directly affects their performance [42,43]. Due to their polar structure, epoxy resins are one of the best adhesive materials [30]. According to mechanical testing data, the shear strength of adhesive joints for neat DER 331 resin is 12 ± 0.7 MPa, which is close to the values reported in other studies [30,42,43]. The incorporation of 10% and 20% of C₁₂C₁IM-DBS into epoxy composites slightly reduces their shear strength by 7%–10%. For DER 331/C₁₂C₁IM-DBS (30%) composites, this effect is more pronounced, and the shear strength of adhesive joints between steel plates is 10 ± 0.3 MPa. The high content of polar ether and hydroxyl groups in epoxy resin is one of the key factors that determines high interfacial adhesion between the coating and the substrate [42]. Probably, the reduction in the adhesion strength of the epoxy coating may be caused by physicochemical interaction between hydroxyl groups with polar ions of IL. However, this effect seems to be negligible even at a high IL content.

3.4. Antibiofilm Activity of DER 331/C₁₂C₁IM-DBS Composites

The antibiofilm properties of DER 331 resin supplemented with 10%, 20%, and 30% of C₁₂C₁IM-DBS were evaluated as it was done before [25,26]. Interestingly, C₁₂C₁IM-DBS proved to be effective against the development of *S. aureus* ATCC25923 surface-associated biofilms in all three concentrations (Figure 11). It also effectively inhibited the growth of planktonic subpopulations of *S. aureus* ATCC25923 in the microcosms despite the high resistance of C₁₂C₁IM-DBS to leaching. On the other hand, 10% DER 331/C₁₂C₁IM-DBS did not inhibit the biofilm development or the planktonic growth in *P. aeruginosa* PA01 microcosms. DER 331/C₁₂C₁IM-DBS (20%) and (30%) approximately inhibited the biofilm metabolic activity by 50%, and there was a one- to two-order-of-magnitude decrease in CFU of planktonic subpopulation compared to the control (Figure 11). In other words, *P. aeruginosa* PA01 proved to be more resistant to immobilized C₁₂C₁IM-DBS compared to the highly sensitive *S. aureus* ATCC25923.

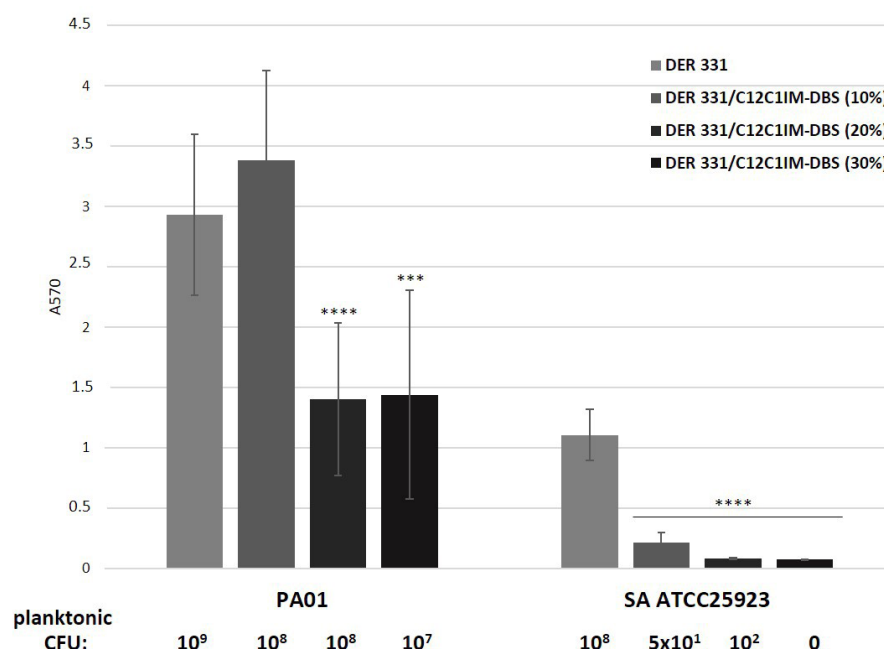


Figure 11. The level of metabolic activity of *P. aeruginosa* PA01 (left) and *S. aureus* ATCC 25923 (right) biofilms formed onto epoxy resin films containing 10%, 20%, and 30% of C₁₂C₁IM-DBS following 72 h of incubation was determined by MTT staining and measured as absorbance at 570 nm (A570). Statistical significance compared to the control. *** $p < 0.005$, **** $p < 0.001$.

CLSM analysis indicated that $C_{12}C_1IM$ -DBS did not affect the structure of the *P. aeruginosa* PA01 biofilms, which retained similar relative amounts of biofilm-associated functional amyloids and carbohydrates, but reduced the total biofilm biomass (Figure 12). Both water-soluble and water-immiscible ILs, comprising the 1-dodecyl-3-methylimidazolium cation, are known to possess high activity against both Gram-positive and Gram-negative bacteria, including *P. aeruginosa* strains [44]. However, the results of current microbiological studies indicate that a high content of $C_{12}C_1IM$ -DBS (up to 20%) is required to impart epoxy resin antibiofilm activity. It can be assumed that the strong physicochemical interaction between the IL and the cross-linked epoxy matrix is the factor that prevents sufficient contents of antibacterial additive on the polymer surface. This assumption is supported by the results of recent studies on the antibiofilm activity of protective coatings based on alkyd resin and hydrophobic IL 1-dodecylpyridinium dodecylbenzenesulfonate (Pyr C_{12} -DBS) [26]. A significant decrease in *P. aeruginosa* biofilm metabolic activity, as well as in cell biomass, was determined for coatings containing 16 wt% of Pyr C_{12} -DBS. At higher IL content, phase separation occurred, which indicated its limited compatibility with the polymer matrix [26]. Hence, so-called contact-active antimicrobial materials, which do not release biocides into aqueous medium, can kill microorganisms at the polymer–water interface in ways similar to free biocide molecules in water solutions. However, the high biocide content on the surface of contact-active material is required to impart a resistance to microbial attack [45,46].

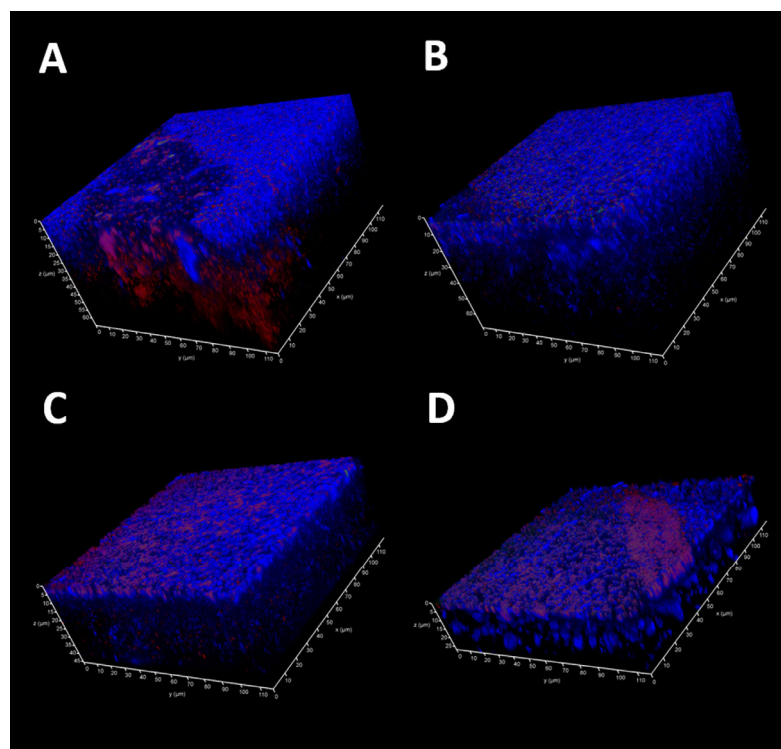


Figure 12. CLSM imaging of *P. aeruginosa* PA 01 biofilms formed onto DER 331 (A) and DER 331 containing 10% (B), 20% (C), or 30% (D) of $C_{12}C_1IM$ -DBS following 48 h of incubation. Ethidium bromide (red signal) was used to visualize cells, and calcofluor (blue signal) was used to visualize biofilm-associated carbohydrates.

4. Conclusions

A new hydrophobic ionic liquid, 1-dodecyl-3-methylimidazolium dodecylbenzenesulfonate ($C_{12}C_1IM$ -DBS), has been synthesized. Modified epoxy coatings containing 10, 20, and 30 wt% of $C_{12}C_1IM$ -DBS were prepared by dissolution of $C_{12}C_1IM$ -DBS in DER 331 followed by curing the resin on the surface of steel substrates. Infrared analysis revealed

physicochemical interactions between the IL and hydroxyl groups of the resin. According to spectrophotometric analysis data, C₁₂C₁IM-DBS has excellent resistance to leaching from epoxy coating in water.

The plasticizing effect of the IL on the epoxy resin was established by differential scanning calorimetry analysis. Thus, DER 331/C₁₂C₁IM-DBS composites containing 10 and 20% of the IL have reduced glass transition temperature (T_g) by 13.5 and 31.3 °C, respectively, compared to neat resin. Impact testing results demonstrated significant improvement in the toughness of the epoxy coatings when modified with 20% and 30% of C₁₂C₁IM-DBS. The hardness of the epoxy resin was reduced by approximately 26% after the introduction of 10% IL and did not change after further increase in the IL content. The tensile shear strength of the adhesive joints based on DER 331/C₁₂C₁IM-DBS composites was reduced by 7%–10% at IL concentrations of 10% and 20%.

The antibiofilm efficiency of the DER 331/C₁₂C₁IM-DBS coatings was evaluated by assessing the capability of two biofilm-forming model strains, *Staphylococcus aureus* ATCC 25923 and *Pseudomonas aeruginosa* PA01, to form attached biofilms on the surface. The modified epoxy coatings effectively inhibited the surface-associated biofilm development of *S. aureus* at the lowest IL content of 10%. However, epoxy composites were much less active against the *P. aeruginosa* strain and showed approximately 50% inhibition of biofilm metabolic activity even at the highest content of C₁₂C₁IM-DBS of 30%. Overall, the results of this research indicate that hydrophobic ionic liquid C₁₂C₁IM-DBS is an efficient modifying additive for epoxy coatings, improving their toughness and resistance to microbial attack. However, further research is needed to assess the resistance of such material to biofouling by aquatic organisms in natural conditions.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/coatings15070821/s1>.

Author Contributions: Conceptualization, S.R. and G.P.; formal analysis, O.M. and G.P.; investigation, S.R., O.M., Y.L., A.M.D., A.H., Y.R., O.D. and O.T.; writing—original draft preparation, S.R.; writing—review and editing, G.P. All authors have read and agreed to the published version of the manuscript.

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