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ORIGINAL RESEARCH ARTICLE

Synthesis of hydrophilic antiviral coating using polyethylene glycol (PEG) for glass surfaces

S. Reasmyraj[1,*], A. Syafiq[1], B. Vengadaesvaran[1], M. Shakeel Ahmad[1], Nasrudin A. Rahim[1], Pouya Hassandarvish[2], Sazaly Abubakar[2], A. K. Pandey[3]

[1] *Higher Institution Centre of Excellence (HICoE), UM Power Energy Dedicated Advanced Centre (UMPEDAC), Level 4, Wisma R&D, Universiti Malaya, Jalan Pantai Baharu, Kuala Lumpur 59990, Malaysia*

[2] *Higher Institution Centre of Excellence (HICoE), Tropical Infectious Diseases Research & Education Centre (TIDREC), Level 4, Wisma R&D, Universiti Malaya, Jalan Pantai Baharu, Kuala Lumpur 59990, Malaysia*

[3] *Research Centre for Nano-Materials and Energy Technology (RCNMET), School of Science and Technology, Sunway University, No. 5, Jalan Universiti, Bandar Sunway, Petaling Jaya, Selangor Darul Ehsan 47500, Malaysia*

*** Corresponding author:** S. Reasmyraj, reasmyraj99@gmail.com

ABSTRACT

Disease epidemics may spread quickly and easily throughout nations and continents in our current global environment, having a devastating effect on public health and the world economy. There are over 513 million people worldwide who have been infected, and more than 6.2 million have died due to SARS-CoV-2. There are treatments but no cures for most viruses. Nevertheless, the spread of viruses can be limited by introducing antiviral coatings on public area surfaces and personal protective equipment (e.g., face masks). This work aims to fabricate a polymer-based coating with acrylic resin as a binder that possesses great antiviral activity against the Feline coronavirus (FCov). The chosen polymer, polyethylene glycol (PEG), is used as an antiviral agent because it contains "green" chemistry benefits such as non-toxicity, being inexpensive, readily recyclable, safe, natural, non-flammable, biocompatible, and biodegradable. The PEG/acrylic coating systems of different weight percentages were coated on the glass substrates by the spray-coating method and cured at room temperature for 24 hours. The developed PEG/acrylic coating system that contains 20 wt% of PEG exhibits the highest anti-viral activities (99.9% against FCov) compared to the other weight percentages. From this study, it has been observed that the hydrophilicity of the coating plays an important role in its antiviral activity. The developed coating has a hydrophilic property, in which the contact angle was measured at 83.28 ± 0.5°. The FTIR reveals that there are no existing toxic components or new components contained in the coating samples.

Keywords: polyethylene glycol (PEG); antimicrobial; coatings; FCoV

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1. Introduction

The direct individual-to-individual spread of a viral infection can be prevented by following the rules of physical distancing, using facemasks, and practicing good hygiene[1]. However, indirect contact, another way of viral transmission, poses the greatest risk since it spreads the virus using inanimate things as carriers for the pathogen[2–5]. Electronics, lift buttons, light and fan controls, handrails, taps, and seats are typically contaminated surfaces that might transmit viruses[6]. Since the virus can survive in the atmosphere and on surfaces for several days to weeks, once it has adhered to hands, it spreads to the mouth, nose, and eyes[7].

The indirect spread of human coronaviruses, specifically SARS-CoV-1 and SARS-CoV-2, through aerosol media[8], and from a variety

of surfaces, including cardboard, stainless steel, copper, plastic, and other healthcare-related materials present in hospital environments, is well documented in literature data[7]. The coronavirus was found to have a 72, 72, 3, 24, and 4 h life span on stainless steel, plastic, aerosols, cardboard, and copper, respectively[8]. In addition, fungal infections, especially those acquired in hospitals, are a major problem[9]. This particular category of healthcare-associated infections (HIAs) results in high rates of illness and death and has tremendous extra expenditures[6]. Additionally, immunocompromised individuals, particularly those with AIDS, are especially vulnerable to HIAs[10,11].

As with bacteria and viruses, fungi's adhesion to both abiotic and biotic surfaces, which is difficult to get rid of the biofilm communities, worsens the degree of severity of microbial infections[12]. For instance, 400,000 hospital-acquired bloodstream infections a year are caused by one of the most prevalent pathogen species, Candida, and are frequently linked to implanted medical devices[10,13,14]. Nearly 700,000 fatalities are recorded worldwide each year as a result of diseases with antimicrobial resistance. The world economy will need to spend $100 trillion on these diseases and related problems, which are estimated to cost the lives of over 10 million individuals each year[6], if new medicines are not produced to prevent the increase of antimicrobial resistance by 2050. Infections caused by repeated contact with contaminated objects, such as doorknobs, switches, personal protective equipment, and wooden or plastic surfaces, are one of the ways viruses or any microbial population propagate in healthcare environments[15]. Therefore, methods to stop these surfaces from retaining bacteria are required. One such method is the application of antimicrobial/antiviral coatings.

Recently, antiviral polymers have gained interest as antiviral agents in coatings. PEG is a network of three-dimensional hydrophilic polymers that can absorb a lot of water while retaining its three-dimensional structure[16]. The hydrophilic nature of PEG makes the polymer categorized as a passive material that inhibits the adhesion of microbes to surfaces[17,18] and protein adsorption on its surface[19]. PEG is recognized as a human-friendly material since it is non-toxic, safe, non-flammable, and affordable[20]. Therefore, PEG is widely used in a variety of daily items, such as food packaging[21], cosmetics[22], and more than 10 types of drugs[23]. PEG is also known for encapsulating the nanoparticles, which enhances the antiviral effects with reduced cytotoxicity as well[24].

Tavakoli et al.[25] and Ghaffari et al.[26] reported a more potent antiviral effect of PEGylated zinc oxide nanoparticles (ZnO NPs) than only ZnO NPs, against HSV-1 and H1N1 influenza viruses. They have reported that PEGlyated ZnO exhibits more antiviral reduction compared to bare ZnO, with 94.6% and 52.2%, respectively. Nalawade et al.[27], have also evaluated the bactericidal activity of PEG against three different strains. From the study, PEG 1000 was effective against S. mutans and E. coli at 25% and did not show any bactericidal activity against E. faecalis. Chirife J et al.[28] studied the in vitro antibacterial activity of PEG with molecular weight 400 against Klebsiella pneumonia, S. aureus, and E. coli, where the bacterial cells showed clumping and morphological changes, resulting in cell death within 2–4 h of incubation. Additionally, HSV-1 and influenza viruses were also mildly inhibited by PEG, with inhibition rates of 13.8% and 13.5%, respectively, for both viruses[29,30]. The antibacterial efficacy of PEG-modified with ascorbic acid was evaluated against two bacterial species, one gram-positive: Staphylococcus aureus (S. aureus) and the other gram-negative: Escherichia coli (E.coli)[24]. The antiviral studies were performed against bacteriophage lambda. The antimicrobial activity against bacteria, fungi, and viruses showed significant inhibition.

In this study, we investigated a simple and eco-friendly, potent antimicrobial coating that was developed against viruses. The biocompatible FDA-approved polymer (Food and Drug Administration, USA), PEG, has been employed in the coating system as an antimicrobial agent without any modification or fillers incorporated. Acrylic resin was used as a binder to improve the adhesion of PEG to the glass surface. The coating system was characterized to study the compatibility between PEG and acrylic and the wettability properties of the coating system. A study on the relationship between the weight percentage of antiviral polymers and the

efficiency of coating has been discussed further. The antimicrobial efficiency of the polymer coating was evaluated against feline coronavirus.

2. Experimental method

2.1. Substrate preparation

The substrate used in this study is glass slides with a dimension of 5 cm × 5 cm. Ethanol was used to thoroughly clean the glass slides of any impurities. Glass slides were then dried at room temperature after being cleaned.

2.2. Preparation of PEG/Acrylic coating system

Polyethylene glycol (PEG) with a density of 1.006 g/mL was used as an antiviral agent and was purchased from Sigma Aldrich, Malaysia. Water-based acrylic with a 1.01 g/mL density was purchased from Ufon Nano-Chemical Corp., Taiwan, which was employed as a binder. Water was employed as a solvent in this study. **Figure 1** shows the preparation of the sample, including the synthesis of the antiviral coating system and the fabrication process on the glass substrate. Antiviral coating systems have been prepared using an acrylic binder and PEG resin. Firstly, 2 grams of the acrylic binder have been incorporated with PEG at different weight percentages of 0 wt.%, 5 wt.%, 10 wt.%, 15 wt.%, 20 wt.%, and 25 wt.%, namely S0, S1, S2, S3, S4, and S5 resin systems. Then, the prepared mixtures have been dissolved in 10 mL of water using a magnetic stirrer for 2 hours at room temperature. The solution was stirred at 400 rpm. All prepared resins were fabricated on glass substrates using the spray-coating technique and then left to dry at ambient temperature for 24 hours. The final product has been presented in **Figure 2**.

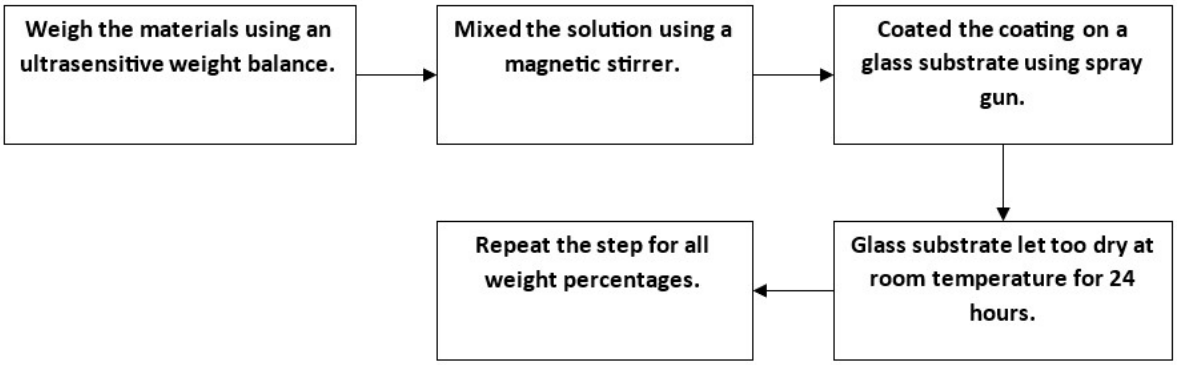

Figure 1. Preparation method of the antiviral coating system.

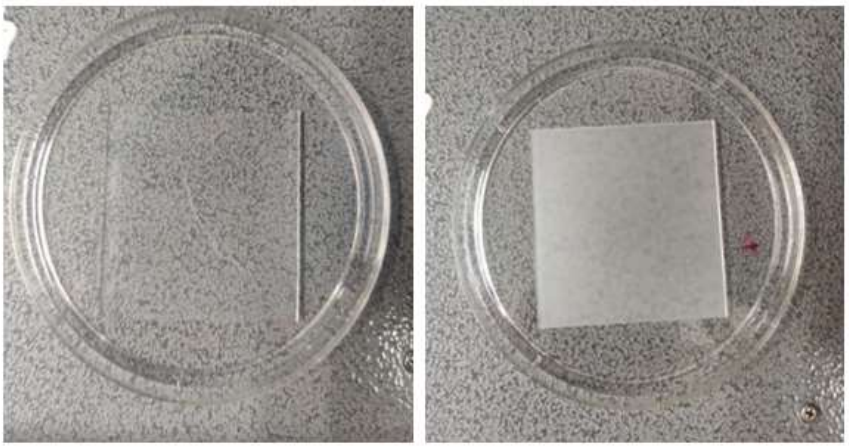

Figure 2. Bare glass sample and coated glass sample.

2.3. Antiviral test

Testing for prolonged efficacy of the samples' virucidal activity was done according to the ASTM E1053 standard protocol. Virus inoculum was added to the slides at 24 hours post-coating. After 30 minutes of contact time, the activity of the test product was immediately neutralized by serial dilution in Dulbecco's Modified Eagles Medium with 2% fetal bovine serum. The mixture was then added to Crandall Feline. Kidney cells were maintained in tissue culture plates, and the formation of virus cytopathic effects was monitored daily. The infected cells were fixed and stained using a paraformaldehyde and crystal violet solution. The virus titers were determined using the Spearman-Karber method and expressed as a tissue culture infectious dose at 50% cytopathic effect ($TCID_{50}$/ml). The virucidal activity was determined by the difference between the logarithmic titer of the virus control and the logarithmic titer of the test samples ($\Delta \log_{10} TCID_{50}$/ml). A reduction in virus titer of 3 $\log_{10}$ (corresponding to an inactivation of $\geq$ 99.9%) was necessary for claiming virucidal activity of the product, according to the U.S. Environmental Protection Agency (EPA).

2.4. Characterization

Fourier transform infrared spectroscopy (FTIR) was used to study chemical cross-linking and bonding in coating systems using an ATR-RXI spectrometer (Perkin Elmer, USA) instrument. The hydrophobicity of the films is analysed by measuring the water contact angle with an Optical Contact Angle 15EC instrument.

3. Results and discussion

3.1. FTIR analysis

Figure 3 shows FTIR spectra of pure acrylic, PEG, and acrylic/PEG blends in different weight percentages.

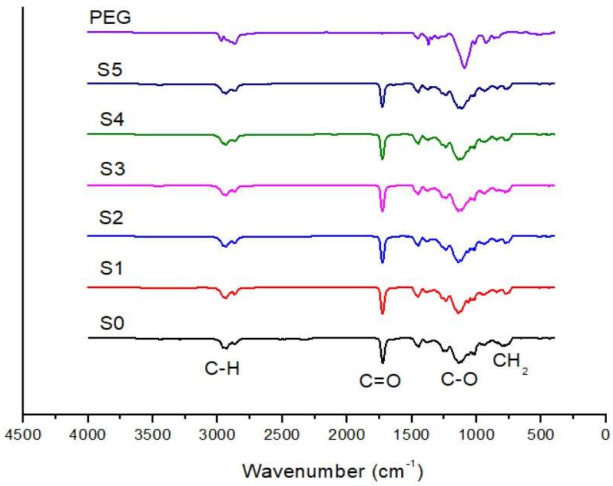

Figure 3. FTIR spectra of S0 (0 wt%), S1 (5 wt%), S2 (10 wt%), S3 (15 wt%), S4 (20 wt%), S5 (25 wt%) and pure PEG samples.

FTIR spectra of pure acrylic and PEG are observed at 3443.67 and 3486.48 cm^{-1} respectively, which are attributed to O-H stretching[31]. As the PEG polymer has been incorporated into acrylic binder, the FTIR spectra shift to a higher wavenumber at 3445 cm^{-1}. Peaks around 2800 cm^{-1}–3000 cm^{-1} are attributed to CH stretching, indicating the presence of an alkyl chain in polymer[32]. Pure acrylic resin showed a sharp peak around 1727.28 cm^{-1}, corresponding to the carbonyl group (C=O)[33,34]. However, for PEG/acrylic blends, it has been observed that the C=O peak becomes broad when the weight percentage of PEG increases in the coating system, indicating a covalent interaction between the C=O bond in an acrylic binder and pure PEG[35,36]. PEG showed C-O stretching at 1296.89 and 1095 cm^{-1}, and pure acrylic resin showed stretching at 1141 and 1119 cm^{-1}, respectively[31]. On the other hand, the combination of PEG and acrylic blends showed the C-O stretching at

1140 and 1116 cm^{-1}. The CH_2 rocking bond of PEG and pure acrylic can be observed at 866.26 and 843.05 cm^{-1} respectively[37]. The increase in weight percentage of PEG in acrylic/PEG blends shows almost similar absorption peaks, which indicates that there is no formation of a new bond or major chemical interaction or changes within the blend[38]. These shifts in wavenumber are caused by changes in the relative contributions of two overlapping bands, instead of a single band gradually changing in frequency in response to modifications in the strength of molecular interactions[39]. Therefore, it has been observed that peak shifting towards pristine PEG wavenumber indicates that there are molecular interactions of PEG in the composite coating system.

3.2. Wettability analysis

Figure 4 represents the water contact angle measurement of all the samples at different weight percentages of PEG. From the contact angle measurement conducted on coated glass surfaces, the average angle obtained is 75.83 ± 0.5°, indicating that the antiviral coating possesses a hydrophilic property as the measured contact angle is below 90° due to the high surface energy of acrylic and PEG[40,41]. A coating with high surface energy tends to have a low contact angle, classifying it as a hydrophilic coating[42].

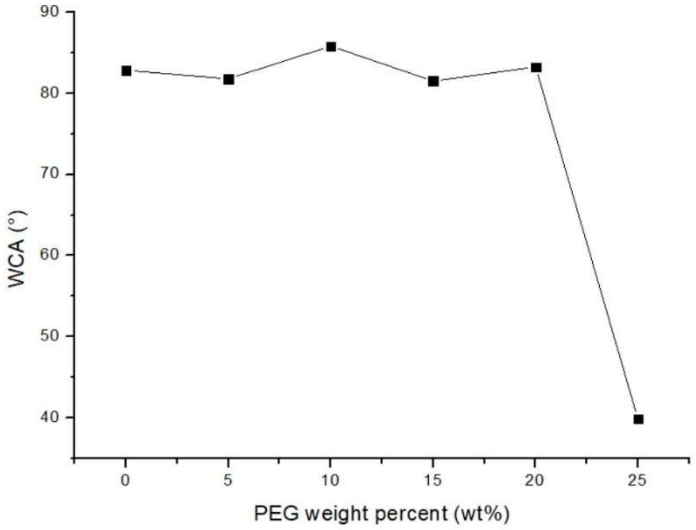

Figure 4. Water contact angle analysis of S0 (0 wt%), S1 (5 wt%), S2 (10 wt%), S3 (15 wt%), S4 (20 wt%) and S5 (25 wt%) samples.

From the figure, it has been observed that the WCA of the S0 system is 82.86 ± 0.5° without the addition of PEG. When the PEG is added to the system, the WCA of the S1 system decreases to 81.82 ± 0.5°. This increased hydrophilicity of the coating is conferred by terminal diol and polyether chains along the PEG structure, which favor hydrogen bonding to water molecules[18]. The S2, S3, and S4 coating systems have demonstrated WCA values around 85.84 ± 0.5°, 81.55 ± 0.5°, and 83.28 ± 0.5°, respectively. It can be noticed that the contact angle of S5 shows a significant drop to 39.85 ± 0.5°. This significant drop occurs because of the resin network's insufficient cross-link density, leading to a slow curing process[43]. In addition, the excessive amount of the OH functional group in the PEG polymeric chain leads to the formation of intermolecular hydrogen bonds at the solid-liquid-air interphase[17].

3.3. Antiviral activity of the samples

The antiviral activity of prepared samples is shown in **Figure 5**. When exposed to 5% organic soil (5% FBS) for 10 minutes, the S0 coating system demonstrated a 0 log_{10} unit reduction against FCoV. It should be noted that acrylic resin without PEG content merely functions as a binder since it lacks antiviral characteristics. After 10 minutes, the S1 coating system demonstrated a 1 log_{10} unit reduction. The same results, which are 2

log_{10} unit reductions, were observed by the S2, S3, and S5 coating systems. Additionally, S4 had the highest viral titer drop with a 3 log_{10} unit reduction after the incubation period, which is equal to a 99.9% reduction of virus.

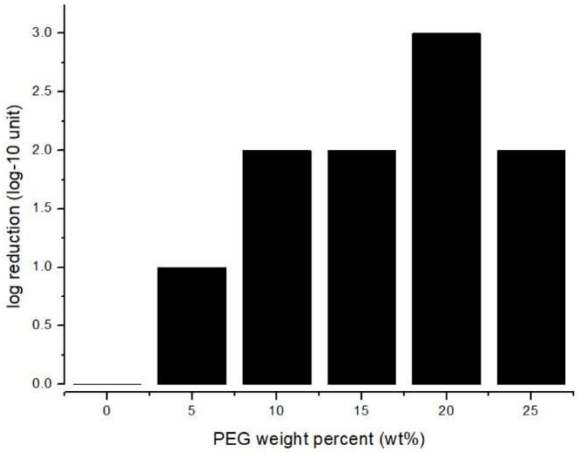

Figure 5. Antiviral activity of S0 (0 wt%), S1 (5 wt%), S2 (10 wt%), S3 (15 wt%), S4 (20 wt%) and S5 (25 wt%) samples.

From the figure, it can be observed that the antiviral activity of the samples continuously rose from S0 to S4, then significantly decreased at S5. This is because of the poor curing of the S5 coating system due to the inadequate cross-link density in resin networks[43]. The adherence of microorganisms to the coating system will eventually grow when the hydrophilic polymer PEG addition increases. Therefore, the antiviral property of S5 will become weaker because the dead virus cells and the internal components that were released after dying will adhere to the surface[44].

4. Conclusion

We have developed an antiviral PEG coating with acrylic binder using a simple mixing method and successfully coated it on glass substrates. The effects of the addition of PEG to acrylic binder in various weight percentages have been studied. A few conclusions have been derived from the experimental observation. The FTIR observations confirmed efficient cross-linking between the acrylic and PEG polymers. The water contact angle test proves that this PEG/acrylic coating is hydrophilic in nature. This antiviral PEG coating has shown great potential for preventing the spread of viruses, especially COVID-19. This coating is designed to repel viruses and other pathogens, making it an effective solution for high-contact surfaces and PPE. The use of 20 wt% PEG coating has been shown to reduce the risk of infection by up to 99.9%, making it an important breakthrough in the fight against viral diseases.

Author contributions

Conceptualization, SR and MSA; methodology, MSA, PH and SA; validation, SR, AS and MSA; formal analysis, SR and AS; investigation, SR; resources, MSA, SA and AKP; data curation, SR and PH; writing—original draft preparation, SR; writing—review and editing, AS and MSA; visualization, SR and AS; supervision, AS and BV; project administration, AS, BV and MSA; funding acquisition, NAR. All authors have read and agreed to the published version of the manuscript.

Conflict of interest

The authors declare no conflict of interest.

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