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Takayanagi model and Monte Carlo simulation for predicting the Young's modulus of PVC/AgNP nanocomposites: Towards self-disinfecting materials to reduce SARS-CoV-2 transmission in healthcare settings

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A B S T R A C T

Poly (vinyl chloride) (PVC) is widely utilized in medical tools and structure applications owing to its biocompatibility, stability, and cost-efficiency. Nevertheless, conventional PVC is susceptible to microbial attachment, indicating the need for a self-disinfecting material. The present study analyzed PVC nanocomposites bio-filled with 0–0.5 wt% silver nanoparticles (AgNPs) to investigate their mechanical behavior using the Takayanagi model and Monte Carlo simulation approach. The Takayanagi model calculates the effective Young's modulus based on a series-parallel arrangement of polymer matrices and the filler, while Monte Carlo simulation simulates uncertainties in the mechanical properties of PVC and AgNPs. Model predictions agreed with experimental data closely, with Young's modulus decreasing as the nanoparticle content increased. Monte Carlo generated confidence intervals further confirming the efficacy of the approach. Evaluation outcomes indicate that the Takayanagi model in combination with stochastic simulations accurately predicts the mechanical properties of PVC/AgNP nanocomposites, which supports design of self-disinfection materials for healthcare applications in which both antimicrobial activity and mechanical performance is required.

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1. Introduction

For over 30 years, poly (vinyl chloride) (PVC) has been used in the medical field. It remains among the most widely used materials because of its ability to resist microbial growth, ease of cleaning, suitability for sterilization, and its use in single-use applications, which can help reduce healthcare-associated infections [1, 2]. PVC has also been commonplace in bottles, electrical cables, household appliances, pipelines, and food packaging films [3]. PVC's extensive range of technological applications across fields (healthcare and packaging) is driven by its unique properties, including flexibility, transparency, chemical resistance, biocompatibility, mechanical strength, processability, cost-effectiveness, market availability, and recyclability [4].

To avoid thermal decomposition of the composite during manufacturing, which can compromise the mechanical properties of biomedical devices and structural components, stabilizers and processing additives are important [5, 6]. Although virgin PVC exhibits some bacteriostatic activity against certain strains, both stabilized and plasticized PVC lack sufficient antimicrobial activity to prevent biofilm formation [7, 8]. Through the leaching of phthalate ester plasticizers, surface microcracks, and UV-induced flaws, enhanced activity has made additive-containing PVC more prone to microbial colonization [9]. Because the material builds up biofilms through multilayered bacterial colonies embedded in extracellular polysaccharides, it is important to add biocides to PVC formulations [10].

Even with ongoing discussions and the continuing concern about the potential health effects of stabilizers and processing additives under both short- and long-term exposure, PVC continues to be a beneficial material. Its attributes of biocompatibility, chemical stability, resistance to sterilants, and cost have made it among the most common polymers used in medicine. Thus, PVC is a common component of many clinical devices, such as flexible blood bags, urine ostomy pouches, tubing systems, inhalation and oxygen masks, and various types of personal protective equipment [11, 12]. Since late 2019, Severe Acute Respiratory Syndrome Coronavirus 2 (SARS-CoV-2) and its variants have caused large-scale health crises and significant strain on healthcare systems worldwide [13]. These viruses are

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highly contagious and are transmitted mainly through droplets or aerosols [14, 15]. Importantly, research has shown that SARS-CoV-2 can remain infectious for up to 24 hours on polymeric surfaces [16]. Therefore, the development of self-disinfecting polyvinyl chloride (PVC) is significant for the production of structural and medical products (e.g., wall guards, handrails, and similar items) which could help lessen epidemiological risk in hospital and clinical environments [17, 18].

Surface modification and the addition of inorganic agents with intrinsic bactericidal and fungicidal properties are the primary methods for imparting self-disinfecting properties to PVC [19]. Alternatively, blending with cationic polymers or applying cationic functional groups through cationization has been shown to promote the antimicrobial performance of PVC [20]. The benefit of incorporating the antimicrobial agent into the polymer matrix is compatibility with conventional polymer processing (e.g., extrusion, injection molding) and longer-lasting antimicrobial activity beyond surface treatment. However, high concentrations of antimicrobial additives are often needed to achieve bactericidal (not bacteriostatic) performance. To overcome this, polymer nanocomposites have emerged that feature polymeric matrices containing nanoscale antimicrobial agents. With the emergence of COVID-19, self-sanitizing PVC materials are becoming increasingly relevant for reducing secondary infections through surfaces in healthcare environments, where surface contamination leads to hand contact and ultimately virus spread [21, 22].

Common examples of organic antimicrobial agents studied include the addition of natural essential oils, such as clove oil [23], eugenol [24], trachyspermum copticum, mentha pulegium, satureja hortensis [25], geraniol, cuminum cyminum [26], ziziphora clinopodioides, and tarragon [27], which exhibit antibiofilm activity against microorganisms and are used in food, medicine, and various industries due to their antioxidant, antibacterial, and anti-inflammatory properties [28]. Common examples of inorganic antimicrobial agents studied extensively in the production of antimicrobial materials include silver (Ag), copper (Cu), titanium dioxide (TiO_2), zinc oxide (ZnO), cuprous oxide (Cu_2O), and cupric oxide (CuO) [29-35]. Antimicrobial activity is often due to reactive oxygen species (ROS) and the release of metal ions, which cause irreversible damage to viruses, bacteria, and fungal cells [36-38]. Research suggests that silver nanoparticles (AgNPs) or silver-based nanomaterials are better than other agents in polymeric nanocomposites [39-45], some of which were able to inactivate SARS-CoV-2 [46]. While a couple of studies have documented the potential for self-disinfecting behavior of PVC/AgNP nanocomposites [47], no reports have been made on the antiviral activity against SARS-CoV-2. Additionally, it should be noted that in most of these studies, neither Ag nor PVC was used in the melt blending of PVC and AgNPs [48-50].

Da Silva et al. processed PVC nanocomposites with AgNPs using melt processing and, presumably due to the well-documented antimicrobial effect of AgNPs in polymeric systems, demonstrated that melting) ca. 0.1-0.5 wt% AgNPs resulted in an effective reduction in Young's modulus and ultimate tensile strength of PVC in the nanocomposite. Presumably, the microstructural defects introduced by processing did not affect the impact strength. In addition, the authors reported that the yellowness index (YI) was higher and the optical bandgap was lower than those of neat PVC. Interestingly, PVC/AgNP nanocomposites (0.3 wt% and higher) exhibited virucidal activity against SARS-CoV-2 B.1.1.28 within 48 hours, indicating potential use for hospital furniture and equipment that require self-disinfecting properties to reduce secondary transmission of COVID-19 [51]. In the present research, we modeled the Young's modulus of PVC with AgNPs using the Takayanagi model, based on the experimental measurements of Da Silver et al. [51]. We compared the theoretical predictions of the Takayanagi model with experimental data and found good agreement between the model and the measurements.

Lastly, we performed a Monte Carlo simulation to model uncertainties in the material properties of both the matrix and the filler, specifically the moduli of PVC (Em) and AgNPs (E_f). This method provides estimates of the uncertainty and confidence intervals for the predicted composite modulus. Overall, this provided us with a better understanding of the nanocomposite's mechanical properties.

2. Materials and methods

The mechanical properties of polymer nanocomposites depend strongly on the properties of the matrix and the dispersed nanoparticles, as well as on their distribution and interactions. In this research, the Young's modulus of PVC containing AgNPs is analyzed using the Takayanagi model, which treats the composite as a series-parallel combination of matrix and filler phases.

2.1. Takayanagi model

As a specialized model, Takayanagi predicts the effective modulus of a two-phase composite by combining both the matrix and filler contributions in a weighted series-parallel structure [52]. The effective Young's modulus E_c of a composite is given by matrix modulus E_m, filler modulus E_f, and connectivity (or efficiency) of filler networks b, is:

$$E_c = E_m(1-b) + \frac{b}{(1-b) + b\frac{E_f}{E_m}} \quad (1)$$

In this study, the parameter b can be correlated to the volume fraction of the nanoparticles (V_f) as follows:

$$b = \sqrt{V_f} \quad (2)$$

2.2. Monte Carlo simulation

To address uncertainties in the mechanical properties of both the matrix and filler, a Monte Carlo simulation was used. Specifically, Em and Ef are treated as random variables with defined mean values and standard deviations to represent experimental or material variability.

For each weight fraction of nanoparticles, 10000 random samples of Em and Ef are generated, and for each sample, E_c is calculated using the Takayanagi model.

The subsequent distribution of E_c provides estimates of the mean, standard deviation, and confidence intervals for the modulus prediction, enabling robust assessment of composite mechanical behavior. Thus, the combination of the Takayanagi model and Monte Carlo simulation enables the prediction of the Young's modulus of PVC/AgNPs composites with reasonable accuracy, in agreement with experimental results.

3. Results and discussion

Da Silva and colleagues measured the Young's modulus (a measure of stiffness) of PVC/AgNP nanocomposites in the laboratory. Young's modulus was measured in the study for PVC and PVC/silver nanoparticle (AgNP) nanocomposites using a uniaxial tensile test, as described in ASTM D1708. Sample preparation consisted of hot-pressing one mm-thick films for 3 minutes at 190°C under 6 tons of pressure, followed by cutting the samples into microtensile specimens. Uniaxial tensile testing of samples was performed on an Instron Universal Testing Machine equipped with a 50 kN load cell at a crosshead speed of 1.5 mm/min. Young's modulus was derived from the initial linear portion of the stress-strain curve.

Values reported in this study reflect the mean of 3 to 6 replicates. Statistical significance of the reported values was determined using a one-way ANOVA with Tukey's post hoc test (95% confidence level, $p < 0.05$). This mechanical testing standardization protocol established a reliable means for comparing stiffness between pure PVC and nanocomposite polymers containing 0.1, 0.3, and 0.5 wt% AgNPs [51].

In this work, the mechanical properties of PVC reinforced with AgNPs were modeled using the Takayanagi model, and the predictions were compared with the experimental measurements reported by Da Silver et al. The studied weight fraction range of AgNPs was 0-0.5 wt%.

3.1. Takayanagi model predictions

Using the Takayanagi model, the effective Young's modulus of the composites was calculated at each nanoparticle loading. The parameter b was assumed to be the square root of the volume fraction ($b = \sqrt{V_f}$) [53], as is commonly done for random dispersions of fillers.

Fig. 1 shows the effect of AgNP contents on the effective Young's modulus of the composite materials. The data show that Young's modulus decreases with AgNP content. This behavior is in line with the series-dominant result from the Takayanagi model that accounts for the effect of the soft polymer matrix and the stiff filler partially produced in a series-parallel arrangement. This result is confirmed by Da Silver et al. [51].

3.2. Comparison with experimental data

In Fig. 2, the predicted modulus values are compared with experimental measurements [51].

The Takayanagi model captures the general trend of the experimental data and shows appropriate agreement, particularly for low filler loadings. Minor departures from higher weight fractions are expected due to particle aggregation, imperfect dispersion, and interfacial phenomena not accounted for in the relatively simple Takayanagi model. Overall, the coefficient variation is 3%.

3.3. *Monte Carlo simulation*

A Monte Carlo simulation was conducted to account for uncertainties in the mechanical properties of the PVC matrix (E_m) and the silver nanoparticles (Ef), based on engineering data. E_m and E_f are considered to be normally-distributed random variables with mean values of 2.1 GPa and 70 GPa, respectively, and standard deviations of 0.05 GPa for E_m and 5 GPa for E_f. For each weight fraction, 10,000 E_c samples were computed. The mean predicted modulus, along with error bars for the standard deviation, is shown in Fig. 3. Monte Carlo results show the ranges of predicted composite moduli resulting from material property variability. The predicted distributions were relatively narrow, indicating that the model is robust and small uncertainties in E_m and E_f lead to limited variation in E_c.

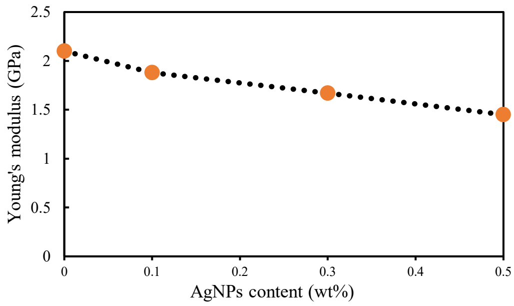

Fig. 1. The effect of AgNPs on the Young's modulus of PVC nanocomposite.

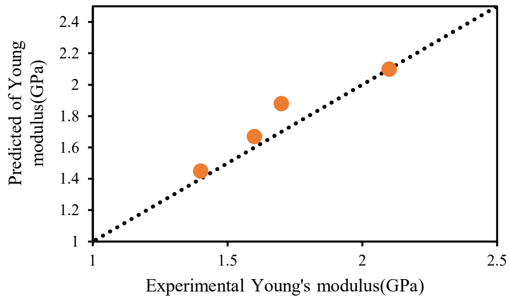

Fig. 2. Comparison of predicted values of Young's modulus with the experimental ones [51].

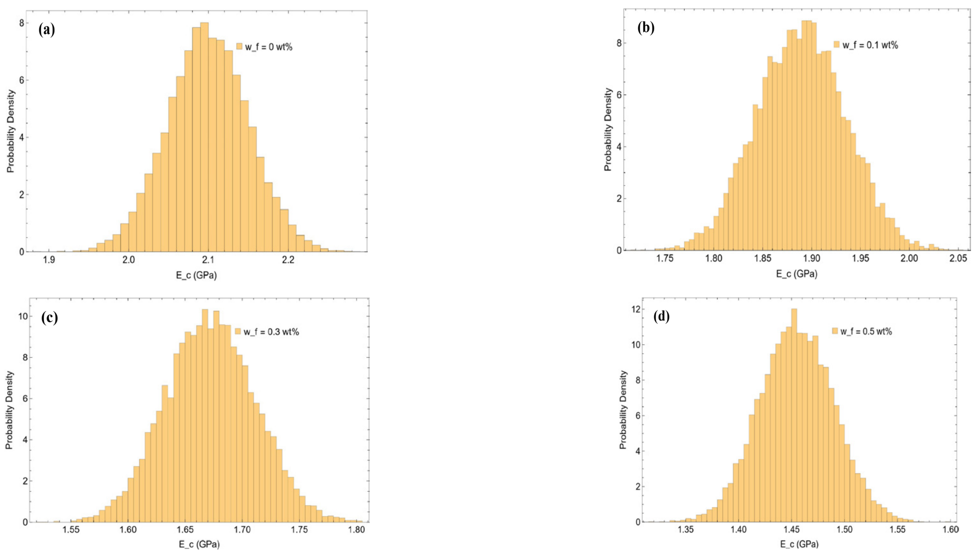

Fig. 3. Estimated probability density function of the effective composite modulus *Ec* at (a) 0, (b) 0.1, (c) 0.3, (d) 0.5 wt% AgNPs.

4. Conclusion

Using the Takayanagi model in conjunction with a Monte Carlo simulation enables a detailed study of the overall mechanical behavior of PVC/AgNPs composites. The Takayanagi model can reasonably predict the overall change in Young's modulus with increasing filler content, while the Monte Carlo component provides a quantifiable framework to account for uncertainties in material properties. So, this approach allows composite design to progress to the next level, as the predicted modulus and its associated confidence intervals can be derived from the uncertainties in constituent properties.

Thus, the study concludes that the Takayanagi model, including a Monte Carlo simulation, can be a valuable tool for predicting and assessing the mechanical properties of metallic nanoparticles reinforced polymer nanocomposites.

Author contributions

Mohammad Hassan Shahavi: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing.

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Conflict of interest

The author declares no conflict of interest.

Data availability

No data is available.

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