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A physics-informed core-shell mori-tanaka model for predicting the mechanical reinforcement of Chitosan/PVA hydrogels functionalized with Aloe vera and green-synthesized ZnO nanoparticles

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ABSTRACT

Chitosan/poly(vinyl alcohol) (CS/PVA) hydrogels functionalized with Aloe vera (AV) and green-synthesized ZnO nanoparticles (ZnO-NPs) show promise as multifunctional wound dressings. While experiments reveal a nonlinear increase in Young's modulus with ZnO loading, the underlying reinforcement mechanism remains unclear. We propose a physics-informed, three-phase core-shell Mori-Tanaka model that accounts for AV-induced matrix softening, Zn^{2+} -mediated ionic cross-linking, and a load-transferring interphase around ZnO-NPs. Calibrated against experimental data (0–2 wt.% ZnO), the model predicts Young's modulus with high accuracy (e.g., 1.54 MPa predicted vs. 1.55 MPa measured at 2 wt.%). Monte Carlo analysis shows mechanical performance is governed primarily by the effective matrix and interphase moduli, not the intrinsic stiffness of ZnO, highlighting interfacial chemistry as the key design lever. This work provides a predictive framework for rational design of mechanically tunable hydrogels for wound healing.

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1. Introduction

Not only do skin injuries (burns, tumors, abrasions and chronic wounds), increase the level of difficulty when treating those patients in a basic way, they also require other methods of management. There are several different wound treatments available, such as autologous grafts, cadaveric grafts, topical creams, and tissue engineering methods, in addition to multiple combinations thereof. Currently wound dressings are more frequently used in the treatment of wounds due to their potential benefits to the rate of healing, the effect that the dressing has on the moisture levels of the wound

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environment, and the protection that the dressing provides against microbe exposure [1-3]. Hydrogels have become the preferred material for creating advanced wound dressings based on their ability to absorb fluids and their close resemblance to the composition of native tissue in terms of their physical characteristics (e.g. soft, flexible, and low surface tension in body fluid) [4-7]. Degradable hydrogels can also be used as "smart" delivery systems for controlled release of pharmaceutical products.

Chitosan (CS), an organic polymer made from chitin, has many uses in treating wounds because it is biodegradable, biocompatible, naturally possesses some anti-bacterial properties, has strong

adhesion to wounds [8, 9]. The reason why CS is antibacterial is due largely to the positive charge of amino groups that are part of CS and the negative charge of different elements within the bacterial membrane [10]. However, despite its wide application in the clinic, CS has limited mechanical strength and will decompose quickly when exposed to physiological conditions. Therefore, CS is often mixed with a synthetic polymer like poly(vinyl alcohol)(PVA) to give the resulting hydrogels greater mechanical strength and improved resistance to degradation) while still maintaining some degree of compatibility with living cells [11, 12].

Polyvinyl alcohol (PVA) has many of the same characteristics as CS, including being non-toxic, water soluble, and a highly crystalline polymer, but unlike CS, PVA has excellent film-forming properties and high mechanical strength. This makes PVA ideal for use with CS in formula dressing formulations. [13]. When combined to form a CS/PVA composite, we gain access to CS's antimicrobial properties while also benefitting from PVA's excellent mechanical properties. Together, these create a hydrogel composite that offers excellent performance in dynamic wound healing applications. Techniques that are commonly used to produce these hydrogels include freezing/thawing, casting from solution, and phase separation (PS). The PS method utilizing the coagulation bath with sodium hydroxide (NaOH)/sodium sulfate (Na_2SO_4) in an aqueous bath has been shown to promote highly crystalline structures among PVA-based hydrogels producing a more durable hydrogel by maintain higher moisture retention capacity along with reducing the degradation rates [14].

The most recent developments involved modifying CG/PLA hydrogel systems with biological agents in order to provide some benefits as therapy. Aloe vera has been used as a medicinal plant throughout history and contains polysaccharides, glycoproteins and enzymes, phenolic compounds, along with other pharmacologically active components that show anti-inflammatory, antimicrobial, and angiogenesis potential. These properties of aloe vera also will improve regenerative processes of tissue and enhance wound healing [15-19]. Relatedly, zinc oxide nanoparticles (ZnO-NPs) have increasingly been integrated into wound care due to their strong antibacterial properties, chemical stability and the slow release of Zn^{2+} ions capable of stimulating the proliferation of cells and collagen synthesis [20, 21]. The antibacterial action of ZnO-NPs is based mainly on the production of reactive oxygen species (ROS) and the subsequent disruption of the membrane integrity of bacterial cells, leading to cell lysis and death [22, 23].

Alvandi et al. [24] synthesized zinc oxide nanoparticles (ZnO-NPs) via green chemistry by utilizing an aqueous extract of *Matricaria chamomilla* and investigated the simultaneous incorporation of Aloe vera and green-synthesized ZnO-NPs into chitosan/poly(vinyl alcohol) (CS/PVA) hydrogels to evaluate the future wound dressing applications. Furthermore, they systematically evaluated the effect of the new Aloe vera and ZnO-NPs in the nanocomposite hydrogels on both the mechanical materials of interest, including tensile strength, Young's modulus, and elongation at break.

The goal of this study is to devise a physics-informed micromechanical model to predict the Young's modulus of CS/PVA/Aloe vera/ZnO nanocomposite hydrogels that have recently been developed as advanced wound dressings by Alvandi et al. [24]. While the original experimental study systematically characterized the nonlinear mechanical property enhancement as a function of loading ZnO nanoparticles, it did not provide a

mechanistic explanation of the loss of stiffness and critical role of Zn^{2+} ionic cross-linking and interfacial conditions between the polymer matrix and nanoparticles. To address this mechanistic explanation, we introduced a three-phase core-shell Mori-Tanaka model that considers (1) the softening of the matrix and enhanced porosity due to the presence of Aloe vera, (2) the stiffening of the polymer matrix due to ZnO-driven ionic cross-linking with $-\text{NH}_2$ and $-\text{OH}$ groups, and the presence of (3) a different mechanically interphase shell layer around each ZnO nanoparticle. The model is validated against the experimental results from Alvandi et al. [24], and a Monte Carlo uncertainty quantification methodology is utilized to evaluate which pars are most influential on mechanical performance. This work also rationalizes the complex non-monotonic trends and serves as a predictive design tool for optimizing hydrogel mechanics to specified wound types and anatomical sites.

2. Theoretical framework for Young's modulus prediction

Mechanical evaluations were conducted as per ASTM D882-02 standard for thin polymeric films utilizing a universal tensile testing equipment (Santam STM-1, Iran). Hydrogel formulations evaluated include: (1) CS/PVA (base hydrogel), (2) CS/PVA/AV with 5wt.% Aloe vera (AV), (3) CS/PVA/AV (5 wt.%) further loaded with 0.5, 1, and 2 wt.% of green-synthesized ZnO nanoparticles (ZnO-NPs). Hydrogel specimens were cut into rectangular strips of length 4 cm, width 1 cm, and thickness 0.3 cm. All the tests were completed on fully hydrated (wet) hydrogels, at room temperature and a crosshead speed of 5 mm/min [24].

2.1. Aloe vera-induced matrix softening

Aloe vera (AV) acts as both a hydrophilic porogen and a plasticizing agent. Results show that by adding AV to CS/PVA, Young's modulus decreases while porosity and swelling ratio increases [24]. This softening is due to disruption of hydrogen-bonding interactions between CS and PVA chains facilitated by incorporation of AV and by increased water retention which serves to further plasticize the base matrix. The base matrix modulus (i.e., without ZnO) is therefore estimated as:

$$E_0 = E_{CS/PVA} - k_{AV}\varphi_{AV} \quad (1)$$

where $E_{CS/PVA} = 0.69 \text{ MPa}$ is the Young's modulus of pure CS/PVA hydrogel, $\varphi_{AV} = 5 \text{ wt.}\%$ is the Aloe vera content, and $k_{AV} = 0.044 \text{ MPa} \cdot \text{wt.}\%^{-1}$ is the softening coefficient derived from experimental data.

2.2. ZnO-induced matrix re-stiffening via ionic cross-linking

In contrast to inert fillers, ZnO nanoparticles engage in the formation of a network through coordination of Zn^{2+} ions and electron-donating groups ($-\text{NH}_2$ of chitosan and $-\text{OH}$ of PVA) [24]. Ionic cross-linking contributes to the stiffening of the polymer matrix, varying according to concentration. Consequently, the effective matrix modulus with ZnO is written as:

$$E_m^{eff}(w) = E_0 + \alpha w + \beta w^2 \quad (2)$$

where w is the ZnO weight percentage, and $\alpha=0.2772 \text{ MPa wt.}\%^{-1}$, $\beta=0.0984 \text{ MPa wt.}\%^{-2}$ are empirical coefficients calibrated to

reproduce the experimental Young's modulus values across all ZnO loadings (0–2 wt.%).

2.3. Core-shell reinforcement via interphase formation

Each ZnO nanoparticle has an interphase layer around it due to the strong chemical attachments with the polymer chains. The interphase layer has properties that are different in composition and mechanics from both the nanoparticle and bulk matrix and aids in load transfer. Therefore, the composite can be represented as a three-phase system: (1) Core - a ZnO nanoparticle with a radius $a = 13$ nm and a Young's modulus $E_m = 100$ GPa. (2) Shell - the interphase layer, which is $t = 9$ nm thick with the effective modulus of $E_i = 80$ MPa, and this represents a polymer-rich zone for cross-linking. (3) Matrix - the CS/PVA/AV hydrogel with effective modulus $E_m^{eff}(w)$. The equivalent modulus of the core-shell inclusion is:

$$E_{eff} = \frac{E_f a^3 + E_i (b^3 - a^3)}{b^3} \quad (3)$$

where $b = a + t$ is the outer radius of the shell.

The effective volume fraction of the core-shell inclusion is then:

$$V_{eff} = \varphi_f \left(\frac{b}{a}\right)^3 \quad (4)$$

φ_f is the volume fraction of ZnO.

2.4. Mori-Tanaka homogenization

For a shallow concentration of spherical inclusions in an incompressible matrix ($\nu \approx 0.5$) the Mori-Tanaka estimate [25] for the composite Young's modulus is:

$$E_c = E_m^{eff} \left[1 + \frac{V_{eff} \left(\frac{E_{eff}}{E_m^{eff}} - 1 \right)}{1 + \frac{1}{3} \left(\frac{E_{eff}}{E_m^{eff}} - 1 \right)} \right] \quad (5)$$

The model has been fully calibrated against the mechanical data in [24] and is a predictive tool for the design of hydrogel stiffness for wound dressing applications.

3. Results and discussions

3.1. Model prediction of Young's modulus in CS/PVA/Aloe vera/ZnO hydrogels

The mechanical reinforcement of CS/PVA/Aloe vera hydrogels with the addition of green-synthesized ZnO nanoparticles was quantitatively described by using a three-phase core-shell Mori-Tanaka model [24], which was experimentally verified by Alvandi et al. as illustrated in Fig. 1. The model captured the nonlinear evolution of Young's modulus (E) as a function of ZnO loading (0–2 wt.%) well.

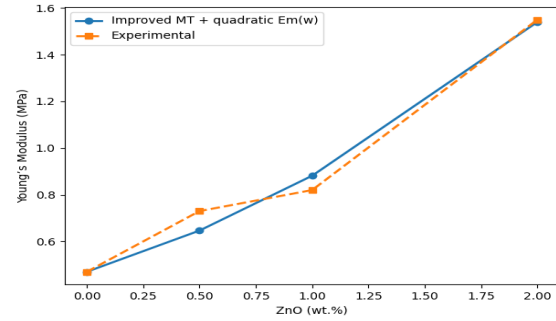


Fig. 1. Comparison of experimental [24] and predicted Young's modulus for CS/PVA/Aloe vera/ZnO nanocomposite hydrogels.

The mechanical reinforcement of CS/PVA/Aloe vera/ZnO nanocomposite hydrogels is effectively described using a physics-informed core-shell Mori-Tanaka model (shown in Fig. 1). The experimental results of Alvandi et al. [24] show that with the addition of 2 wt.% of green-synthesized ZnO nanoparticles, the Young's modulus increases non-linearly before stabilizing afterward (0.47 MPa from CS/PVA/AV to 1.55 MPa with ZnO), due to ionic cross-linking and interfacial chemical reactions between Zn^{2+} cations and chains of chitosan and PVA. Our model correctly describes the trend across the ZnO concentration. The simulations belong at low concentrations (< 2 wt.%) to a quadratic matrix stiffening term $E_m^{eff}(w) = 0.47 + \alpha w + \beta w^2$, with w =weight fraction of ZnO, and at high concentrations to a 9 nm seamless interphase layer surrounding the ZnO. The derivative parameters indicate that chemical cross-linking, at least at low concentrations, outperformed conventional filler reinforcement. The model converges very well to the 2 wt.% point (from the model, 1.540 MPa and the measured 1.55 MPa), though again, the 0.5 wt.% concentration was slightly under-predicted but agrees well with the SEM images at 0.5 wt.% indicating limited porosity, and have reduced ductility/brittleness for this material [24]. This shows an important link between experimental results and mechanistic inference and illustrates models that can predict and explain hydrogel mechanics in advanced wound dressing.

3.2. Quantifying model robustness via Monte Carlo uncertainty analysis

To assess the reliability of the proposed core-shell Mori-Tanaka model within the expected range of parameters found in practice, a complete Monte Carlo simulation was performed with all key input parameters experiencing $\pm 10\%$ uniform uncertainty: matrix base modulus (E_m), stiffening coefficients (α, β), densities ($\rho_{ZnO}, \rho_{matrix}$), nanoparticle radius (a), interphase thickness (t), filler modulus (E_f), and interphase modulus (E_i). Our threshold for success was defined as reaching a composite Young's modulus $E_c > 0.82$ MPa, which is the experimental value for the CS/PVA/AV/ZnO hydrogel at 1 wt.% ZnO loading [24].

Fig. 2 illustrates the probability of success ($P(E_c > 0.82)$) exhibiting near-zero to near-one transition behavior over the range of loading of ZnO particles starting at 0.75 wt.% ZnO loading and approaching 1.0 wt.%. This transition behavior is aligned with experimental observations, where below 0.75 wt.% ZnO loading, the system was dominated by AV-induced plasticization while reducing porosity, leading to brittle failure, compared to above 1.0 wt.% ZnO loading, wherein ionic cross-linking and interphase

stiffness surpassed these effects and resulted in mechanical performance.

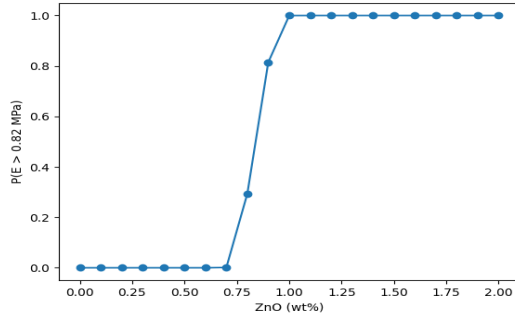


Fig. 2. Probability of achieving a composite Young's modulus greater than 0.82 MPa as a function of the ZnO content.

The conditional probability graphs illustrate the relative effect of each parameter on the probability of achieving the target modulus.

As illustrated in Fig. 3, the probability of meeting the target is near zero when E_m is less than about 0.8 MPa, then quickly rises to 1.0 beyond that value. This indicates that the matrix stiffening caused by Zn^{2+} crosslinking has a stronger effect on mechanical performance than other factors discussed. In practical terms, even if all other parameters are at their optimal settings, formulations with $E_m < 0.8$ MPa will not meet the target.

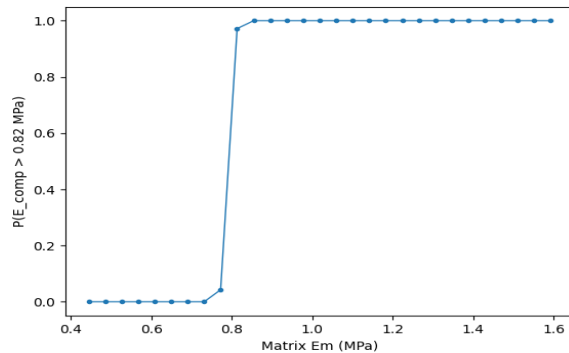


Fig. 3. Probability of achieving a composite Young's modulus greater than 0.82 MPa as a function of the effective matrix modulus E_m .

$P(E_c > 0.82)$, as illustrated in Figure 4, is very sensitive to E_i , varying from 0.57 to 0.585 within its $\pm 10\%$ range (72–88 MPa). This sensitivity emphasizes the critical role of the polymer–nanoparticle interface: even modest differences in interphase stiffness might affect load transfer efficiency. The optimal value for E_i appears to be approximately 80 MPa, which corresponds with our calibrated model.

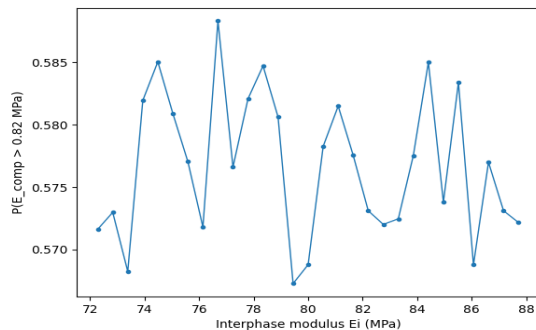


Fig. 4. Probability of achieving a composite Young's modulus greater than 0.82 MPa as a function of the interphase modulus, E_i .

Conversely, Fig. 5 shows little variation in success probability across a range of E_f (90,000 – 110,000 MPa). This means that the reinforcement mechanism is not dependent on the intrinsic stiffness of the ZnO nanoparticles but rather on the chemical interactions that can create a mechanically effective interphase. So, varying E_f by $\pm 10\%$ will not affect the total mechanical response—this is an affirmation of a useful factor in material design.

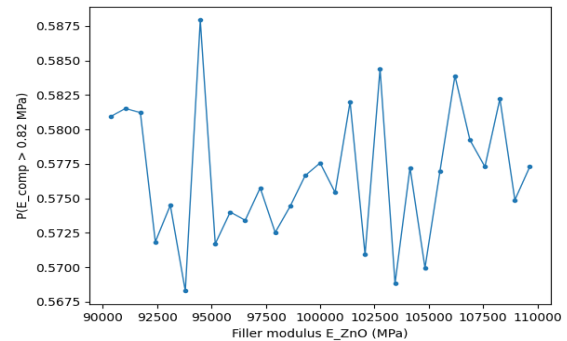


Fig. 5. probability of achieving a composite Young's modulus greater than 0.82 MPa as a function of the ZnO modulus E_{ZnO} .

Collectively, these conclusions indicate that interfacial chemistry, not filler geometry or intrinsic stiffness, is the dominant factor enhancing the mechanical performance of CS/PVA/Aloe vera/ZnO nanocomposite hydrogels.

4. Conclusion

In this study, we introduce a physics-informed, three-phase core-shell Mori–Tanaka micromechanical model for explaining and predicting the nonlinear reinforcement of CS/PVA/Aloe vera/ZnO nanocomposite hydrogels. Examination of the model's predictions reveals that by combining Aloe vera-induced matrix softening, Zn^{2+} -mediated ionic cross-linking and a mechanically active interphase surrounding ZnO nanoparticles was able to capture Young's modulus trends experimentally observed as a function of ZnO loading (0 – 2 wt. %), with high accuracy (for example, predicted Young's modulus of 1.54 MPa at 2 wt. %, versus measured Young's modulus of 1.55 MPa). Monte Carlo uncertainty quantification indicates, as anticipated, that mechanical performance was dictated by the effective matrix modulus and interphase stiffness, and did not depend on the intrinsic rigidity of the ZnO nanoparticles. These findings shift the design framework from filler-based reinforcement concepts to interfacial engineering concepts because chemistry and mechanical properties at the nanoscale dictate mechanics at the macroscale. The proposed model provides a predictive, tunable platform for improving hydrogel stiffness in advanced wound dressing applications that could be further leveraged in other bioactive nanocomposite systems.

Author contributions

Fatemeh Heidari: Investigation, Conceptualization, Writing – original draft, Writing – review & editing. **Sahar Madani:** Investigation, Writing – original draft, Writing – review & editing.

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Conflict of interest

The authors declare no conflict of interest.

Data availability

No data is available.

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