GREEN SYNTHESIS OF CONDUCTIVE HYDROGELS INCORPORATING SILVER NANOPARTICLES FROM *CURCUMA LONGA* FOR TRANSDERMAL DICLOFENAC DELIVERY

Carmen-Marinela Mihăilescu[1,2,3], Carmen-Elisabeta Manea[1], Andreea Popa[1], Mihaela Savin[2], Vasilica Schiopu[2], Oana Brincoveanu[2], Daniel Cord[1], Mirela Rimbu[1], Horia Sebastian Iliescu[1], Roxana-Colette Sandulovici[1], Adina Turcu-Stiolica[4]

[1]Faculty of Pharmacy, Titu Maiorescu University, Bucharest, Romania
[2]National Institute for Research and Development in Microtechnology (IMT Bucharest), Romania
[3]Doctoral School, University of Medicine and Pharmacy of Craiova, Petru Rares Street, 200349, Craiova, Dolj, Romania
[4] Pharmacoeconomics Department, University of Medicine and Pharmacy of Craiova, Craiova, Faculty of Pharmacy, Petru Rares Street, 200349, Craiova, Dolj, Romania
DOI: https://doi.org/10.66793/titum19proceeding20

Abstract

This work reports the green synthesis and characterization of a conductive hydrogel incorporating silver nanoparticles (AgNPs) biosynthesized from Curcuma longa extract for the transdermal delivery of diclofenac. The hydrogel matrix was based on poly(vinyl alcohol) (PVA) and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), ensuring mechanical stability and electrical conductivity. The biosynthesized AgNPs exhibited a characteristic plasmon resonance around 430 nm, confirming their formation and stability. FTIR and UV–Vis spectroscopy confirmed the interaction between PVA, PEDOT:PSS, and AgNPs, while SEM micrographs revealed a porous, interconnected morphology suitable for drug diffusion. Swelling and rheological analyses demonstrated high water retention and viscoelastic stability, while diclofenac release studies showed a sustained release profile. The resulting hybrid system represents a biocompatible, electrically responsive platform for advanced transdermal drug delivery applications.

1. INTRODUCTION

Hydrogels are three-dimensional polymeric networks capable of absorbing large quantities of water, mimicking the extracellular matrix of biological tissues. Their biocompatibility and ability to respond to external stimuli such as pH, temperature, and electrical fields make them suitable for a wide range of biomedical applications, including drug delivery, wound healing, and biosensing. Conductive hydrogels (CHs) combine the hydration properties of conventional hydrogels with the electrical conductivity of conjugated polymers, such as polyaniline (PANI) or poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS). These hybrid materials enable electro-responsive drug release and facilitate communication with biological tissues. The incorporation of metallic nanoparticles such as silver (AgNPs) can further enhance the functional properties of hydrogels. Green synthesis routes using plant extracts as reducing and stabilizing agents have emerged as sustainable alternatives to conventional chemical synthesis. Curcuma longa (turmeric) contains curcuminoids and phenolic compounds capable of reducing Ag^{+} ions to Ag^{0} nanoparticles, while simultaneously capping and stabilizing them. These AgNPs provide antibacterial and anti-inflammatory properties, making them ideal for topical and transdermal systems. Diclofenac, a non-steroidal anti-inflammatory drug (NSAID), was chosen as the model compound to evaluate controlled drug release from the hydrogel.

2. EXPERIMENTAL DETAILS

2.1 Materials

Poly(vinyl alcohol) (PVA, 98–99% hydrolyzed), PEDOT:PSS (1.3 wt% aqueous dispersion), and diclofenac sodium were obtained from Sigma-Aldrich. Fresh rhizomes of Curcuma longa were used for the biosynthesis of AgNPs. All aqueous solutions were prepared with deionized water.

2.2 Green synthesis of silver nanoparticles (AgNPs)

An aqueous extract of Curcuma longa was prepared by boiling 10 g of finely ground turmeric powder in 100 mL of distilled water for 10 min, followed by filtration through Whatman No. 1 paper. To 80 mL of 1 mM $AgNO_3$ solution maintained at 60 °C under stirring, 5 mL of the turmeric extract was added dropwise. The color change from pale yellow to brown indicated the formation of silver nanoparticles. The colloidal suspension was centrifuged at 12 000 rpm for 15 min and redispersed in water for further use.

2.3 Preparation of PVA/PEDOT:PSS-AgNP hydrogel loaded with diclofenac

A 10 wt% PVA solution was prepared by dissolving PVA in distilled water at 90 °C with constant stirring. After cooling to 40 °C, 5 wt% PEDOT:PSS dispersion and the biosynthesized AgNPs (2 mL, $A_{430} \approx 1.0$) were added and homogenized. Diclofenac sodium (0.1 g per 10 mL of polymer solution) was incorporated, and the mixture was cast into Petri dishes. The hydrogel was formed by three freeze–thaw cycles (–20 °C/4 h followed by thawing at 25 °C/1 h).

2.4 Characterization techniques

UV–Vis spectra were recorded using a Hitachi U-0080D spectrophotometer in the range 200–800 nm. Fourier transform infrared (FTIR) spectra were obtained with a Bruker Tensor 27 spectrometer using ATR mode. Surface morphology was analyzed by scanning electron microscopy (SEM, Nova NanoSEM 630). Swelling experiments were performed by immersing pre-weighed hydrogel discs in phosphate-buffered saline (PBS, pH 7.4) at 37 °C. The swelling ratio was calculated as $(W_t - W_0)/W_0$. Drug release studies were performed in PBS (pH 7.4) at 37 °C and absorbance at 276 nm was monitored spectrophotometrically.

3. RESULTS AND DISCUSSION

3.1 UV–Vis analysis

To confirm the formation of silver nanoparticles (AgNPs) synthesized using turmeric extract, UV–Vis absorption spectra were recorded for several samples during synthesis(Figure 1). The obtained spectra exhibited a characteristic surface plasmon resonance (LSPR) band of AgNPs in the 420–440 nm region, confirming their successful formation. A progressive increase in absorption intensity was observed from the light pink to the dark red sample, indicating a higher concentration of nanoparticles. This enhancement in absorption can be correlated with variations in the reaction parameters (extract:$AgNO_3$ ratio = 1:6, reaction time = 45 min, temperature = 60 °C, etc.), suggesting different efficiencies in silver ion reduction and nanoparticle stabilization under distinct experimental conditions.

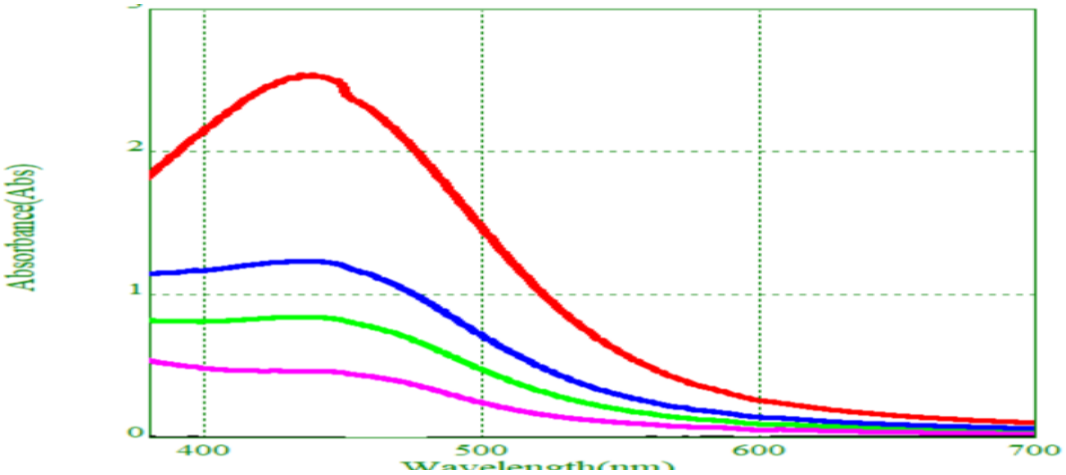


Figure.1 UV–Vis absorption spectra were recorded for several samples during green synthesis

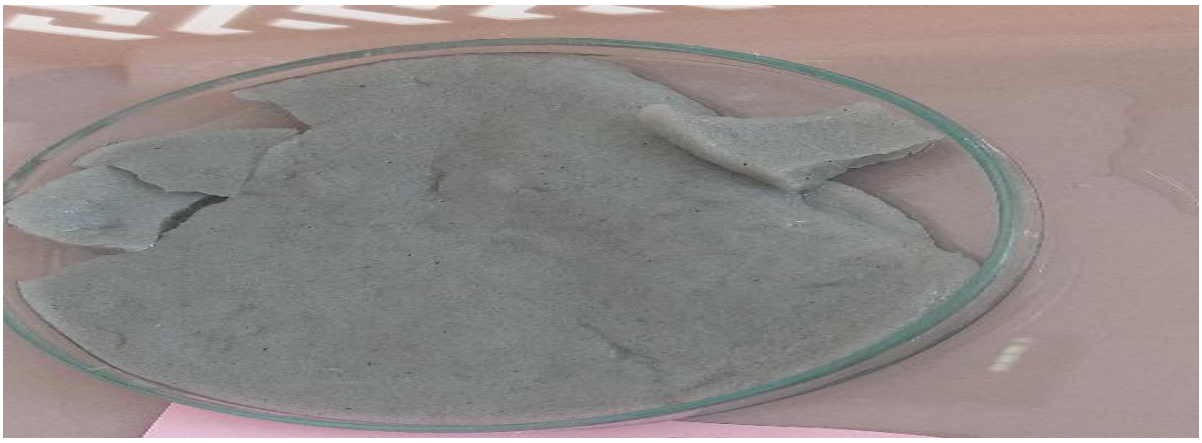
The visual appearance of the reaction mixtures provided qualitative evidence of nanoparticle formation and stability. In Figure 2, five representative samples (1–5) are shown, displaying a clear color shift from pale yellow to dark orange. These color variations reflect both particle size and nanoparticle concentration, as supported by previous reports. The more intensely colored samples corresponded to higher absorbance values in the UV–Vis spectra (Figure 1), confirming a more efficient synthesis process.

Figure 2. Samples numbered 1–5 obtained after the reaction between turmeric extract and $AgNO_3$. The color intensity varies depending on the efficiency of Ag nanoparticle synthesis.

3.2 Preparation and Characterization of PEDOT–PVA–Chitosan–AgNP–Diclofenac Composite Hydrogel

To obtain a functional material with biomedical potential, a composite hydrogel based on PEDOT, PVA, chitosan (CS), silver nanoparticles synthesized from turmeric extract (AgNPs), and sodium diclofenac was prepared using a thermal gelation method followed by casting in Petri dishes. The gelation process and the physical appearance of the samples were visually monitored, as shown in Figure 3 (A–B).

A.



B

Figure 3. Visual representation of the PEDOT–PVA–CS–AgNP–diclofenac hydrogel formation process.

(A) Casting of the homogeneous hydrogel mixture into Petri dishes during preparation.

(B) Appearance of the formed composite hydrogel after gelation and solidification.

Initially, a homogeneous suspension containing the polymers PEDOT (poly(3,4-ethylenedioxythiophene)), PVA (polyvinyl alcohol), and chitosan was prepared in a suitable solvent medium to allow uniform dissolution and mixing. Chitosan, a biopolymer derived from chitin, provides antimicrobial properties and biocompatibility, while PVA acts as a stabilizing and crosslinking agent that enhances mechanical integrity. PEDOT contributes electronic conductivity and electroactivity to the hybrid hydrogel. The reaction mixture was poured into Petri dishes (Figure 3A–B) and allowed to gel under controlled temperature conditions. Gelation was driven by intermolecular interactions among the functional groups of the polymers and physical–chemical crosslinking induced by thermal treatment. The silver nanoparticles were previously synthesized via a green route using turmeric extract, which acts as both a reducing and stabilizing agent. The presence of curcumin contributes to colloidal stability and introduces antioxidant functionality into the system.

The synthesized AgNPs were subsequently incorporated into the polymeric matrix along with sodium diclofenac, a non-steroidal anti-inflammatory drug (NSAID), to produce a composite hydrogel with enhanced therapeutic potential. Upon crosslinking, the hydrogel gradually hardened, forming a self-supporting structure. Surface contraction and minor cracking were observed as water evaporated, indicating network consolidation. The matured hydrogel exhibited satisfactory elasticity, cohesion, and water-retention capacity, features essential for biomedical or pharmaceutical applications.

The final gelled material was portioned into compact discs or small fragments for further use, depending on the intended application. The measured specific conductivity of the composite hydrogel was 2.05 mS cm^{-1} at 25 °C, indicating good ionic transport and suitability for use in controlled drug-delivery systems or electrochemical biosensors.

3.2 FTIR Characterization of Synthesized PEDOT and Composite Hydrogel

The FTIR spectra of the PVA/PEDOT:PSS–AgNP hydrogel showed broad O–H stretching at 3300 cm^{-1}, C=O bands around 1640 cm^{-1}, and peaks corresponding to sulfonate groups from PSS at 1030 cm^{-1}. The shift in these peaks compared to pure PVA confirmed hydrogen bonding and electrostatic interactions among the components. The presence of diclofenac was identified through characteristic C–Cl and aromatic C=C bands at 748 cm^{-1} and 1500 cm^{-1}.

The PEDOT obtained through this method is non-processable (insoluble) but can be dispersed in water using a surfactant such as sodium dodecyl sulfate (SDS) for various applications.

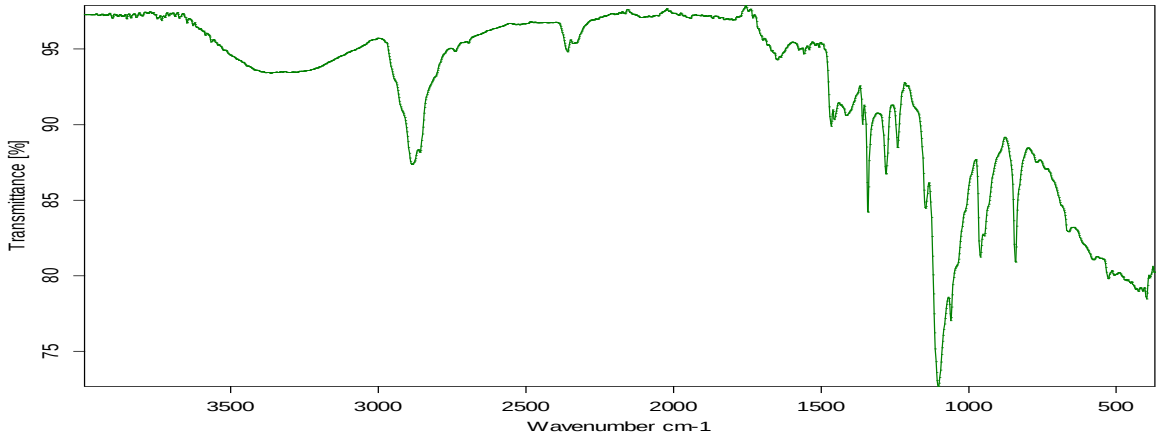


Figure 4. FTIR spectrum of the PEDOT/PVA/chitosan/AgNP composite hydrogel loaded with turmeric and diclofenac. The spectrum shows characteristic absorption bands corresponding to functional groups O–H, N–H, C=O, and C–O–C, indicating chemical interactions between the matrix components and the active

The FTIR spectrum of the synthesized PEDOT polymer displays characteristic absorption bands confirming the formation of the conjugated polymer structure. The broad band observed at 1510–1550 cm^{-1} corresponds to the C=C stretching vibrations of the thiophene ring, indicating the presence of conjugated aromatic segments. In the region of 1300–1350 cm^{-1}, a distinct band is attributed to inter-ring C–C stretching, typical of the rigid and planar PEDOT backbone. Another significant band appearing in the 1130–1200 cm^{-1} region is assigned to C–O–C stretching vibrations, characteristic of the ethylenedioxy groups within the EDOT unit. The presence of this band confirms successful polymerization from the EDOT monomer.
A sharp band located near 980–990 cm^{-1} corresponds to C–S–C deformation of the thiophene ring, while the signal around 840–870 cm^{-1} is associated with C–H out-of-plane vibrations. A weak band between 700–750 cm^{-1} represents the ring deformation mode of the thiophene unit. These spectral features confirm the formation of doped PEDOT with possible contributions from polaronic and bipolaronic states, as later verified by UV–Vis spectroscopy. Therefore, FTIR analysis confirms the successful synthesis of PEDOT with preservation of its conjugated structure, essential for achieving the desired electrical properties.

3.3 SEM morphology

SEM micrographs revealed a porous, interconnected structure with uniformly distributed silver nanoparticles embedded within the polymeric network. The pore size (20–50 µm) facilitates efficient water absorption and drug diffusion. The incorporation of PEDOT:PSS imparted a smooth, conductive texture (Figure 5).

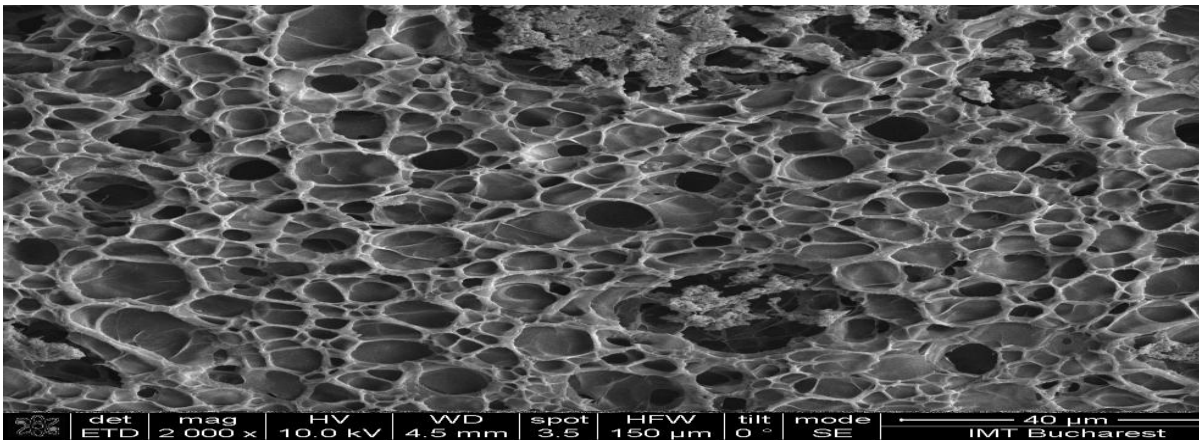


Figure 5. SEM of PEDOT/PVA/chitosan/AgNP composite hydrogel

3.4 Swelling behavior

The swelling ratio reached equilibrium after 4 h, with a maximum of 420%, indicating high hydrophilicity. Rheological analysis demonstrated a dominant elastic response ($G' > G''$), suggesting strong physical cross-linking and mechanical stability suitable for skin application.

3.5 Diclofenac release study

The release profile showed an initial burst within the first hour, followed by sustained diffusion over 24 h, consistent with Fickian diffusion kinetics (Figure 6). The presence of AgNPs slightly slowed the release due to reduced mesh size and possible drug–nanoparticle interactions. This indicates the potential for prolonged anti-inflammatory action in transdermal systems.

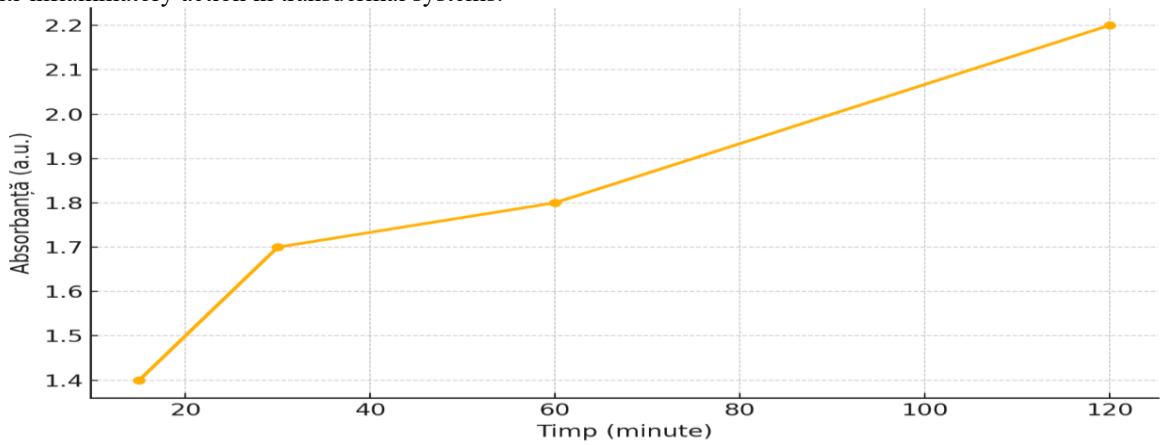


Figure 6. Diclofenac release profile from the PEDOT/PVA/chitosan/AgNP composite hydrogel over time. The progressive increase in absorbance at 276 nm indicates sustained drug diffusion from the polymeric matrix up to 120 minutes.

4. CONCLUSIONS

A conductive PVA/PEDOT:PSS hydrogel incorporating green-synthesized AgNPs from Curcuma longa was successfully prepared and characterized. The hybrid hydrogel exhibited favorable structural, swelling, and mechanical properties, along with sustained diclofenac release. Its combination of biocompatibility, conductivity, and antibacterial potential highlights its suitability for next-generation transdermal delivery platforms and wearable therapeutic systems.

REFERENCES

[1] Raveendran, P., Fu, J., & Wallen, S. L. (2003). Completely green synthesis and stabilization of metal nanoparticles. J. Am. Chem. Soc.

[2] Ahmed, S., Ahmad, M., Swami, B. L., & Ikram, S. (2016). A review on plants extract mediated synthesis of silver nanoparticles. J. Adv. Res.

[3] Chen, S. et al. (2024). Conductive hydrogels for biomedical applications. Biosensors.

[4] Gu, D. et al. (2016). Structural and mechanical properties of PVA/PEDOT:PSS hydrogels. Polymers.