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DEVELOPMENT AND CHARACTERISATION OF POLYVINYL ALCOHOL BASED POLYMERIC SYSTEMS AS POTENTIAL PLATFORMS FOR BIOMEDICAL AND AGRICULTURAL APPLICATIONS

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Abstract

In the context of developing biocompatible materials for medical and agricultural applications, polyvinyl alcohol (PVA) has emerged as a versatile polymer, serving as the core component of the formulations investigated in this study. The work focused on the development of PVA-based gel and film systems for topical drug delivery systems and wound dressings, as well as for soil mulching and controlled-release fertilizer carriers. Preliminary studies were conducted to select appropriate concentrations of polymer, plasticizer and crosslinking agent. The resulting gels underwent rheological characterization, and were subsequently processed into films by casting. The films were evaluated in terms of visual appearance, water absorption capacity, and surface wettability. Based on the gels flow properties and the films hydrophilicity, two formulations were selected for further investigation: 10 and 15% PVA gels containing 15% citric acid relative to dry substance. To improve their physicochemical properties, the gels were then blended with a 3% methylcellulose (MC) gel in various ratios. The newly obtained gels exhibited enhanced pseudoplastic behavior, which facilitates the film-forming process, resulting in films with significant potential for future biomedical and agricultural applications.

Key words*: polyvinyl alcohol, hydrogels, films, methylcellulose, flow behavior, hydrophilicity.*

INTRODUCTION

Polymers are crucial in various industries as the current trend in biomaterials focuses on developing biocompatible polymeric materials, often incorporating natural excipients to improve systemic properties (Kalirajan et al., 2021).
Polyvinyl alcohol (PVA) is a synthetic polymer with thermoplastic behaviour, high biodegradability, and biocompatibility. Unlike most polymers, PVA is not obtained through the polymerization of its component monomers but through the hydrolysis of polyvinyl acetate (Liang et al., 2024).
The first synthesis of PVA dates back to 1924 and is attributed to Dr. Herman Staudinger (Li et al., 2025), who described it as an odourless polymer, soluble in water, acids and polar solvents. The polymer solubility in aqueous solutions is highly dependent on its degree of hydrolysis and molecular weight.
PVA can be processed into various forms, including hydrogels and polymeric films, which are widely used in applications ranging from

medical contact lenses and wound dressings to food packaging, detergent doses, clothing, and agricultural mulch materials (Teodorescu et al., 2018; Yang et al., 2024).

PVA-based hydrogels and films have attracted increasing attention due to their favourable properties, such as soft and elastic structure, superior mechanical features, ease of handling, better barrier properties, stability, loading capacity, and controlled release of various active ingredients (Rahman Khan & Rumon, 2025).

Both forms exhibit adjustable physical, chemical, and biological properties, which can be modified according to the intended application through the incorporation of specific additives (Feldman, 2020).The main methods for adjusting the physico-chemical properties of polymers are crosslinking and plasticizer addition (Liang et al., 2024; Yang et al., 2024).

Crosslinking can be achieved through both physical and chemical methods. Physical crosslinking techniques include freeze-thaw cycles, directional freezing, UV or γ irradiation, and thermal treatments (Rahman Khan & Rumon, 2025). Chemical crosslinking is carried out using various agents, among which the most commonly used are glutaraldehyde, citric acid (Lodi et al., 2024) and boric acid (Lambertini et al., 2024). In the context of applications in biomedical and agricultural fields, the use of a biocompatible and environmentally friendly crosslinking agent, such as citric acid, is preferred (Salihu et al., 2021; Rodrigo & Munaweera, 2025).

Plasticizers are low molecular weight substances that intercalate between polymer chains, reducing intermolecular forces, and increasing molecular mobility, thereby improving the mechanical properties of the material (Hacker & Mikos, 2011). Examples of plasticizers reported in the literature include glycerine (Han et al., 2023), propylene glycol, polyethylene glycol (Shojaee Kang Sofla et al., 2020), caprolactam, and sorbitol (Xu et al., 2020).

To improve the physicochemical properties of the formulations, beyond plasticizers and crosslinking agents, PVA could be blended with various polymers, such as cellulose derivatives (Kida et al., 2020; Abdullah et al., 2021), chitosan (Pugar et al., 2024), sodium alginate (Kumar et al., 2022), starch (Iskalieva et al., 2023), and gelatine (Rivera Hernández et al., 2024) allowing for tailored film properties across a wide range of applications.

Among the cellulose derivatives, methylcellulose was selected for this study. A mixture of PVA and methylcellulose, each at an approximate concentration of 4%, has previously been reported, with radiation used as the crosslinking agent, designed as an oral drug delivery platform (Kenessova et al., 2025). In an another study, MC/PVA composite hydrogels with tunable thermoresponsive properties were obtained through a mild physical preparation method, for potential use as injectable carriers (Geng & Xiao, 2009).

In agriculture, PVA hydrogels and films can be used to minimize resource loss associated with the use of conventional fertilizers, mitigate the risks related to non-biodegradable polymer-coated urea (Wan Anuar et al., 2025) and also contributing to the stabilization and controlled release of nutrients and biocides, thereby supporting sustainable and economically efficient agricultural practices (Malka & Margel, 2023; Jayanudin et al., 2024).

In the biomedical domain, PVA-based hydrogels and films are suitable for delivering drugs to the wound site, exhibit self-adhesion to the injury, and reduce the risk of wound reopening upon dressing removal due to their biodegradable nature (Huang et al., 2023). These properties can also be extended to the development of transdermal patches for drug delivery, where PVA is combined with specific excipients or permeation enhancers to facilitate drug transdermal absorption (Solís et al., 2021).

PVA is also widely used in tissue engineering for repair and replacement of damaged tissues (Nathan et al., 2023; Thakare & Hazarika, 2025). Although PVA hydrogels exhibit a high water retention capacity, they lack the needed mechanical strength and flexibility under physiological (homeostatic) conditions, which limits their standalone applicability in such contexts, also requiring additives to fine-tune their characteristics (Han et al., 2023).

Another application in the field of tissue engineering involves the use of polyvinyl alcohol and calcium silicate as a matrix for bone regeneration, as currently, bone fractures are frequently treated using metal implants, which present significant drawbacks, including the risk

of incompatibility, high cost, and the need for multiple surgical interventions (Spaniol et al., 2019; Shendage et al., 2025).

Taking into account the aforementioned aspects, the main objective of this work was the selection and optimization of PVA-based formulations to improve the film-casting process and the physicochemical properties of the resulting polymeric systems. Specifically, this study aimed to: (i) determine the effect of citric acid concentration on the rheological behaviour of the gels; (ii) evaluate the influence of the crosslinking agent and plasticizer on the water absorption capacity and surface wettability of PVA films; (iii) identify the optimal formulation parameters for an adequate pseudoplasticity of the gels, and a balanced hydrophilicity and swelling of the films; (iv) assess the influence of methylcellulose blending on pseudoplasticity to facilitate the film-casting.

The selection of biocompatible and environmentally sustainable components, PVA, glycerine, citric acid, and methylcellulose, minimizes ecological impact while enhancing the safety profile for biomedical applications.

MATERIALS AND METHODS

For the preparation of polyvinyl alcohol-based gels, the reagents consisted of: polyvinyl alcohol with a molecular weight of 70,000-100,000 g/mol (Sigma), anhydrous glycerine 92.10 g/mol (Chemical Company), citric acid 102.13 g/mol (Loba Chemie), methylcellulose 4000 cP (Janssen Chimica) and distilled water.

The equipment used in the preparation stage consisted of: analytical balance (Sartorius), high-precision laboratory electric stirrer 3000RPM, 220V/50Hz 200W (Velp Scientifica), magnetic stirrer with heating DLAB MS-H380 Pro, 200-1500 RPM (DLAB Scientific Co.), water bath with immersion thermostat, and recirculation pump 230V/50-60 Hz 1300W (Falc).

The composition of the prepared gels is presented in Table 1.

In this paper, samples A0, A1, A2 and A3 will be referred to as series A and samples B0, B1, B2, B3 will be referred to as series B.

The preparation of gels for both series was carried out according to the protocol presented in Table 2.

Table 1. Composition of gels based on PVA, anhydrous glycerine and citric acid

Sample code	PVA (%)	Anhydrous glycerine (%)	Citric acid (%)*
A0	10	10	-
A1	10	10	10
A2	10	10	15
A3	10	10	20
B0	15	5	-
B1	15	5	10
B2	15	5	15
B3	15	5	20

*expressed relative to dry PVA weight

Table 2. Gels preparation protocol

Step	Instruction
1	The water bath was heated to a temperature of 90°C. A support was installed to hold the vessel used for preparation, ensuring that the vessel walls remained immersed in water up to the level of the solution inside.
2	The required amount of PVA was weighed, taking into account the desired final concentration.
3	The necessary amount of distilled water for the preparation was placed in a Berzelius beaker.
4	Using a magnetic stirrer, PVA was gradually added to the distilled water at room temperature to ensure proper dispersion and minimize material loss.
5	The Berzelius beaker was then secured in the support and placed in the water bath previously heated to 90°C.
6	The contents of beaker, consisting of PVA dispersed in water, were subjected to mechanical stirring until the PVA was completely dissolved and a homogenous solution was obtained.
7	Subsequently, after cooling the mixture to 50°C, the required amount of anhydrous glycerine was weighed and added to the PVA colloidal solution under continuous mechanical stirring to ensure optimal incorporation.
8	Finally, the pre-weighed citric acid was gradually added under continuous mechanical stirring to obtain the corresponding gels.
9	The mixture was maintained under mechanical stirring at 200 rpm until complete homogenization was achieved, after 35 minutes.

The films were prepared by casting the gels onto glass slides and dried at room temperature for 72 hours. To ensure uniform thickness, 1.5 g of gel was applied to each slide.

The rheological analysis of the gels was performed using a rotational viscometer, model RM 100 PLUS (Lamy Rheology). In this context, a coaxial cylindrical system was used, consisting of a set of accessories, namely the MK-DIN1 spindle with a diameter of Ø = 30 mm, together with an attachment composed of a DIN1-Cap and a DIN1-Tube. The measurements were conducted at room temperature (23°C), and the flow behavior of the gels was evaluated over a rotational speed range between

0.3 and 60 rpm, corresponding to shear rates between 0.4 and 78 s^{-1}.
The Ostwald-de Waele model (Eq. 1) was applied to the experimental data:

$$\tau = K \cdot \dot{\gamma}^n \quad (1)$$

Where: τ represents the shear stress (Pa); $\dot{\gamma}^n$ - shear rate (s^{-1}); K - consistency index ($Pa \cdot s^n$); n - flow behaviour index (dimensionless).
The goniometric analysis of the films was performed using a KSV Cam 101 Scientific Instrument goniometer (Helsinki, Finland), equipped with a digital camera. The contact angle method was applied, using distilled water as the testing liquid. Briefly, a small droplet of water (~3 µL) was deposited onto the surface of the film using a Hamilton microsyringe. The digital camera, positioned perpendicular to the polymeric film surface, captured the image of the droplet in contact with the film (Huang et al., 2023).
The droplet image was analysed to determine its shape, and the contact point between the water and the polymeric film surface. Both the left and right contact angles were measured to obtain an average value.
All measurements were carried out in triplicate, and the reported values represent the mean of three determinations.
This parameter provides insight into the hydrophilic behavior of the film in different applications, and its equilibrium value is determined using Young equation (Eq. 2):

$$\gamma_{SG} = \gamma_{SL} + \gamma_{LG} \cdot \cos\theta \quad (2)$$

Where: θ defines the contact angle, and γSG, γSL, γLG - the interfacial surface tensions at the solid-gas, solid-liquid, and liquid-gas interfaces, respectively.
The water absorption capacity of the films was evaluated using a gravimetric method, with distilled water as the swelling medium. The analysis was performed at room temperature (23°C). Initially, pieces of approximately 0.25cm^2 were cut from the polymeric films and weighed to record their initial dry mass. The weighed samples were then placed in 2.5 mL of distilled water (Tudoroiu et al., 2023).
At predetermined time intervals, the samples were removed from the water and weighed to determine their hydrated mass. The water absorption capacity (g/g) was calculated using the equation found below (Eq. 3):

$$\text{Absorption capacity (g/g)} = \frac{W_t - W_i}{W_i} \quad (3)$$

Where: W_t represents the mass of the films after hydration at different time intervals, and W_i - the initial mass of the films in their dry form.
Taking into consideration the results obtained during the evaluation stage of the previously prepared polymeric systems, correlated with the desired properties of the final products – moderate water absorption capacity and an adequate degree of hydrophilicity – two representative compositions were selected, one from each developed series, the compositions corresponding to samples A2 and B2.
To improve the rheological characteristics of the gels, and film-casting process, a 3% methylcellulose (MC) gel was prepared (Tudoroiu et al., 2023). The composition of the PVA-MC gels is presented in Table 3.

Table 3. Percentage composition of polymeric systems based on PVA, anhydrous glycerine, citric acid and methylcellulose

Sample code	A2 gel (%)*	B2 gel (%)*	3% MC gel (%)*
MC1	80	-	20
MC2	70	-	30
MC3	-	80	20
MC4	-	70	30

*expressed to 100 g of final gel

Further, the rheological behaviour of the MC1-MC4 gels was evaluated as previously described.

RESULTS AND DISCUSSIONS

The rheological behaviour of the gels based on polyvinyl alcohol, anhydrous glycerine, and citric acid was evaluated from rheograms of shear stress versus shear rate, presented in Figure 1 for the series A, and Figure 2 for the series B.
Analysis of Figures 1 and 2 reveals similar profiles, showing an increase in shear stress with increasing shear rate for all samples in series A (10% PVA) and series B (15% PVA). For both series, the smallest increase in shear stress was observed for uncrosslinked samples A0 and B0,

while the most pronounced increase was noticed for samples A2 and B2, containing 15% concentration of citric acid reported to dry mass of PVA.

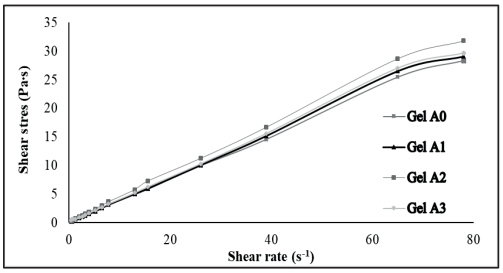

Figure 1. Shear stress vs. shear rate rheograms for series A gels (10% PVA) containing 0-20% citric acid

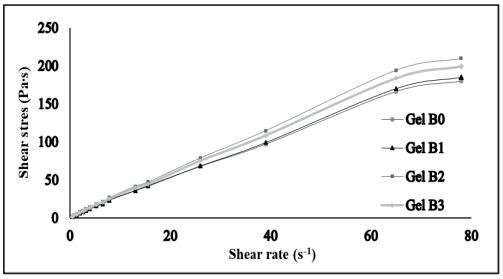

Figure 2 Shear stress vs. shear rate rheograms for series B gels (15% PVA) containing 0-20% citric acid

To quantify the rheological behaviour of the designed polymeric systems, the Ostwald-de Waele model (Eq. 1) was applied, describing the variation of shear stress as a function of shear rate. This allowed the assessment of the specific rheological parameters for this model, namely the flow behaviour index and the consistency index, presented in Table 4, along with determination coefficient (R^2) values.

Table 4. Rheological parameters (K, n) and determination coefficient (R^2) according to Ostwald-de Waele model for the gels of series A and B

Gel	Consistency index (K-Pa·s^n)	Flow behavior index (n)	Determination coefficient (R^2)
A0	0.400	0.99	0.9984
A1	0.420	0.98	0.9976
A2	0.610	0.89	0.9978
A3	0.560	0.85	0.9930
B0	3.530	0.95	0.9976
B1	3.640	0.92	0.9977
B2	4.250	0.89	0.9975
B3	3.970	0.88	0.9977

Values obtained for determination coefficient higher than 0.9900 indicate that the data for all gels fit well the Ostwald-de Waele model.

The flow index (n) suggested rheological behaviour ranging from near-Newtonian to weakly pseudoplastic.

As the crosslinker concentration increased, a progressive decrease in n values was observed, with A3, respectively B3 showing the most noticeable pseudoplastic behaviour in their corresponding series.

Regarding the consistency index, similar values were observed for sample A0 (uncrosslinked) and A1 (crosslinked with 10% concentration of citric acid). Maximum value was registered for sample A2 (15% concentration of citric acid), with an increase of consistency index of about 1.45 times relative to samples A0 and A1.

A similar trend is noticed for series B, noting that the initial values of K parameter were higher for all samples compared to those obtained for series A, which may be attributed to a higher PVA concentration.

For this series, an increase in the citric acid content to 15%, corresponding to sample B2, led to a 1.20-fold increase in the consistency index compared to B0 and B1.

In summary, for both series a 10% addition of citric acid did not significantly change the rheological behavior, while the maximum viscosity is reached for a 15% concentration of citric acid. A further increase in the crosslinking agent concentration to 20% did not result in an additional increase in this parameter, suggesting a limit in the crosslinking efficiency was reached at 15%.

Regarding the PVA-to-glycerine ratio, polymer concentration was found to play a determining role in the consistency of gels across for both series. Notably, gels formulated with 15% PVA (series B) exhibited values of the consistency parameter *K* approximately 7-9 times higher than those recorded for 10% PVA gels (series A), for the same concentration of citric acid relative to PVA dry mass, demonstrating that increasing polymer content leads to a considerably denser and more robust gel network.

Regarding other studies on PVA-based gels, Teodorescu et al. (2016) reported gels containing 15% PVA/PVP at various ratios,

crosslinked through freeze-thaw cycles, which exhibited viscoelastic behaviour.

Another approach, described by Hapipi et al. (2020), involved blending PVA with magneto-rheological plastomers. These materials exhibited distinctive rheological behaviour, characterized by shear thickening at low shear rates and shear thinning at high shear rates. The content of carbonyl iron particles was found to directly correlate with rheological performance (Hapipi et al., 2020).

The rheological behaviour of PVA and hyaluronic acid (HA) mixture was investigated by Lewandowska (2020). Pure PVA solutions exhibited shear-thinning behaviour regardless of solvent type, with no significant solvent-dependent differences in apparent viscosity. For HA/PVA gels the flow behaviour was governed by composition and solvent. A synergistic viscosity enhancement was observed in acidic blends at low shear rates, resulting from attractive intermolecular forces between polymeric components, though this effect diminished at higher shear rates due to hydrogen bond disruption. The consistency index (k) values also decreased with increasing temperature, consistent with the expected thermal behaviour of solution viscosity (Lewandowska, 2020).

Following formation by casting method, films obtained from the corresponding gels were clear, homogeneous, exhibiting transparency, absence of air bubbles, ease of peeling, and good elasticity.

The surface hydrophilicity of films was assessed by contact angle measurements. Several factors influence this assessment, including surface tension (higher surface tension leads to a larger contact angle due to reduced interaction with the film surface), film roughness (a rough surface provides more contact points for water, resulting in a lower contact angle, while a smooth surface increases the contact angle), physicochemical properties of the film and the liquid (molecular interactions between the two phases determine whether the liquid spreads over the film), and ambient temperature and pressure (variations in temperature and pressure can modify the contact angle, thereby affecting the equilibrium between the three phases) (Quéré, 2008; Drelich, 2019; Owusu et al., 2025).

A low contact angle (< 90°) indicates strong interaction between the film and the liquid (Giovambattista et al., 2007). All samples exhibited contact angles lower than 40°, indicating a pronounced hydrophilic character, adequate for water absorption (Figure 3).

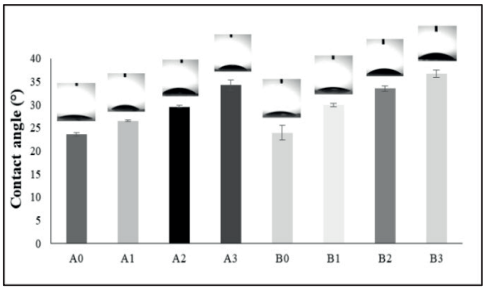

Figure 3. Images of the water droplets in contact with PVA-based films for series A and B, corresponding to the contact angles; values are mean ± SD (n = 3)

For series A, the average contact angle values ranged from 23.61 ± 0.31 to 34.30 ± 1.05°. The lowest contact angle was observed for sample A0, which did not contain a crosslinking agent. It appears that increasing the concentration of the citric acid led to a progressive increase in the contact angle values, with the highest value recorded for sample A3, containing the maximum crosslinker concentration. These results can be explained by the increase in polymer network density associated with higher concentrations of the crosslinking agent in the samples, leading to an increase in contact angle of 1.45 times.

Regarding the samples from series B, a similar trend was observed, with an increase in contact angle values as the concentration of the crosslinking agent in the samples varies from 0 to 20%. Within this series, the average contact angle values ranged from 23.94 ± 1.39 (B0) to 36.73 ± 0.76° (B3), translating into an increase by a factor of 1.53.

Based on our knowledge there is no previous study regarding contact angle measurement for a formulation of PVA/MC films. However, there is a study measuring the contact angle using another cellulose derivate, namely carboxymethyl cellulose blended with PVA and *Calendula officinalis* extract. The study reported an average angle of 14.6°, indicating a very hydrophilic film was obtained (Huang et al., 2023). Simple MC films are less hydrophilic,

with a contact angle of 45° being reported (Tudoroiu et al., 2023).
Overall, for our study a compositional balance between 10% PVA/10% glycerine and 15% PVA/5% glycerine resulted in a similar surface hydrophilicity of the films.
To further characterize the films, the absorption capacity was evaluated. This represents a critical parameter in designing systems that achieve an optimal balance between mechanical stability, water retention, and controlled permeability (Priya et al., 2024).

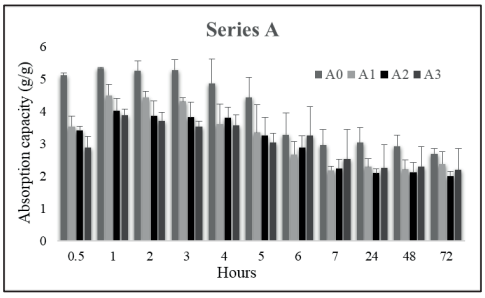

Figure 4. Absorption capacity for the film series A

It is observable that for all the samples from series A, the maximum absorption capacity was reached one hour after the start of the experiment (Figure 4). This behaviour may be explained by the hygroscopic nature of glycerine, which is present in higher concentration in this series. The maximum absorption capacity values for the samples in series A ranged between 3.88 ± 0.20 (A3) and 5.34 ± 0.02 g/g (A0), indicating 1.37 times difference between the two samples. The obtained results may be explained by the effect of the crosslinking agent, which promotes the formation of a denser polymeric network, resulting in a reduced absorption capacity.

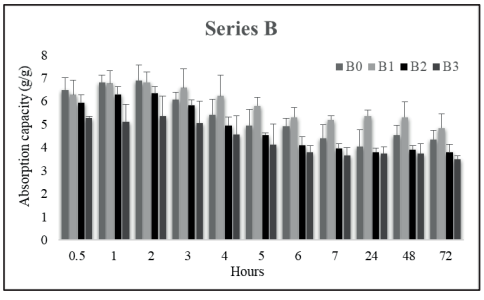

Figure 5. Absorption capacity for the film series B

Regarding the samples from series B (Figure 5), a similar trend was observed, with a decrease in absorption capacity as the concentration of the crosslinking agent increased in the samples. Within this series, the maximum absorption capacity was reached two hours after the start of the experiment. The maximum absorption capacity values for series B ranged between 5.34 ± 0.75 (B3) and 6.90 ± 0.56 g/g (B0), showing a 1.30 times difference between the uncrosslinked sample and the one crosslinked with the maximum amount of citric acid.
An excessive crosslinking may impede solvent penetration and potentially result in the delayed or incomplete release of incorporated bioactive agents.
Comparing series A and series B, it appears that the samples containing a higher PVA concentration exhibited a superior absorption capacity, which may be attributed both to the increased availability of hydrophilic groups and to the higher amount of polymer capable of swelling.
When comparing PVA/MC gel to other systems found in literature, a similar absorption profile was observed, with the highest concentration of PVA corresponding to the highest swelling degree observed between samples (Kenessova et al., 2025).
Taking into account the results of all the previous analysis, two representative compositions, namely A2 and B2, were selected. To further improve their physicochemical properties and to optimize the film-forming process as well, these formulations were mixed with a 3% methylcellulose gel in different proportions, as described in the Materials and Methods section.
All gels of the series bellow are based on polyvinyl alcohol, anhydrous glycerine, citric acid, and methylcellulose. Shear stress - shear rate rheograms were plotted (Figure 6). An increase in shear stress with increasing shear rate was observed.
To assess the effect of incorporating 3% methylcellulose gel at varying ratios into PVA-based gels A2 and B2 on their flow behaviour, the rheological parameters of the Ostwald-de Waele model were determined (Table 5).

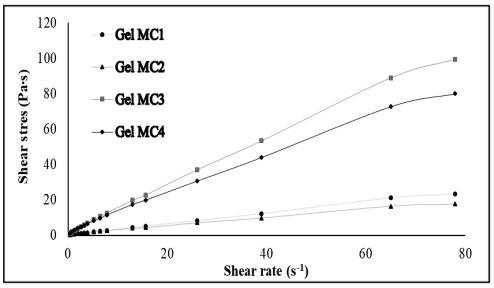

Figure 6. Comparative representation of the shear stress-shear rate rheograms for MC1-MC4 gels

Table 5. Rheological parameters (K, n) and determination coefficient (R^2) according to Ostwald-de Waele model for the gels A2, B2, MC, and MC1-MC4

Gel	Consistency index (K-Pa·s^n)	Flow behavior index (n)	Determination coefficient (R^2)
A2	0.610	0.89	0.9978
B2	4.250	0.89	0.9975
MC	53.160	0.48	0.9938
MC1	0.580	0.79	0.9925
MC2	0.670	0.72	0.9907
MC3	2.220	0.86	0.9985
MC4	2.250	0.81	0.9979

It can be observed that the n value obtained for the 3% methylcellulose gel was 0.48, indicating a pronounced pseudoplastic character. For gels MC1 and MC2, consisting of A2 and methylcellulose gels combined in different proportions (Table 3), lower values of the flow behaviour index were observed compared to that of the A2 gel, indicating an increase in pseudoplasticity related to the incorporation of methylcellulose gel. The lowest n value within this group (0.72) was recorded for the gel MC2, which contained a higher proportion of methylcellulose gel compared to MC1.

For gels MC3 and MC4, consisting of B2 and methylcellulose gels, a decrease in the flow behaviour index was also observed compared to the value obtained for B2 gel, showing an increase in pseudoplasticity within this group as well, with the minimum value recorded of 0.81, corresponding to MC 4, the gel with the higher concentration of methylcellulose from its series. To the best of our knowledge, the flow behaviour index of PVA-MC gel formulations has not been specifically investigated in previous studies. Regarding simple MC gels, they present a documented strong pseudoplastic behaviour (Tudoroiu et al., 2023)

Shi et al. described the rheological behavior of PVA, gelatine and carboxymethyl celllulose mixture gels using the Ostwald-de Waele model, confirming its non-Newtonian character. These findings were consistent with the pseudoplastic nature reported for PVA-based gels containing cellulose derivatives (Shi et al., 2024).

Analysis of the consistency index across both groups (MC1-MC2 and MC3-MC4) revealed a slight decrease in consistency index, correlated with increasing proportions of methylcellulose gel within the samples. Additionally, it was observed that in A2, MC1 and MC2 series, the consistency index values were relatively similar, whereas for the B2, MC3 and MC4 series, K values decreased by almost 2 times. This may be explained by the methylcellulose interaction with hydrogen bonds of PVA chains, also resulting in spatial separation of those chains (Kopač, Ručigaj, & Krajnc, 2022).

Overall, the incorporation of methylcellulose did not significantly alter the general rheological behaviour in terms of viscosity, with the polyvinyl alcohol gel remaining the main component. This is favourable, as a large increase in viscosity could hinder the film casting process. However, an improvement in pseudoplastic behaviour was observed, which will have a positive impact on the film-casting process of newly obtained gels.

CONCLUSIONS

The present work reports the design and optimization of polyvinyl alcohol-based polymeric system, using citric acid as a crosslinking agent, glycerine as a plasticizer, and methylcellulose as a rheological modulator. Rheological characterization demonstrated that the consistency index of the PVA-glycerine-citric acid-based gels was primarily increased by higher polymer concentration and, to a lesser extent, by crosslinker content. An optimal crosslinking was achieved at 15% citric acid (reported to dry PVA), beyond which no further improvement in flow resistance was observed.

Hydrophilic properties of the resulting films were evaluated through wettability and water absorption studies. The results indicate that an increase in the crosslinking degree inversely correlates with the hydrophilic character of the film surface, evidenced by higher contact angle values. Similarly, water absorption capacity decreased with advanced crosslinking for both

series. Concerning the polymer and plasticizer concentration, their ratio influences the water swelling behaviour of the films, whereas the surface hydrophilicity remains similar.

Comparative analysis showed that Series B performed superiorly to Series A, exhibiting higher water uptake and reaching swelling equilibrium after two hours, compared to one hour.

To refine the processing characteristics, methylcellulose was incorporated as a rheology enhancer before the casting stage. This addition modulated the flow behaviour by increasing pseudoplasticity, ensuring the formation of uniform and structurally stable films.

A limitation of this study was that the PVA/methylcellulose mixed gels were not cast and analysed in film form. Future studies will address this by casting the gels into films and conducting additional analyses to evaluate the mechanical properties (tensile strength, elongation at break) and incorporating active compounds to develop advanced delivery systems for wound healing and the controlled release of agrochemicals, extending the functional scope of these materials across biomedical and agricultural applications.

REFERENCES

Abdullah, A., B. Aziz, S., & Raza Saeed, S. (2021). Structural, and Electrical Properties of Polyvinyl alcohol (PVA):Methyl Cellulose (MC) Based Solid Polymer Blend Electrolytes Inserted with Sodium Iodide (NaI) salt. *Arabian Journal of Chemistry, 14*, 103388. doi:10.1016/j.arabjc.2021.103388

Drelich, J. W. (2019). Contact angles: From past mistakes to new developments through liquid-solid adhesion measurements. *Advances in Colloid and Interface Science, 267*, 1-14. doi:https://doi.org/10.1016/j.cis.2019.02.002

Feldman, D. (2020). Poly(Vinyl Alcohol) Recent Contributions to Engineering and Medicine. *Journal of Composites Science, 4*(4), 175. doi:10.3390/jcs4040175

Geng, N. N., & Xiao, C. M. (2009). Facile preparation of methylcellulose/poly(vinyl alcohol) physical complex hydrogels with tunable thermosensitivity. *Chinese Chemical Letters, 20*(1), 111-114. doi:https://doi.org/10.1016/j.cclet.2008.09.003

Giovambattista, N., Debenedetti, P. G., & Rossky, P. J. (2007). Effect of Surface Polarity on Water Contact Angle and Interfacial Hydration Structure. *The Journal of Physical Chemistry B, 111*(32), 9581-9587. doi:10.1021/jp071957s

Hacker, M. C., & Mikos, A. G. (2011). Chapter 33 - Synthetic Polymers. In A. Atala, R. Lanza, J. A. Thomson, & R. Nerem (Eds.), *Principles of Regenerative Medicine (Second Edition)* (pp. 587-622). San Diego: Academic Press.

Han, S., Wu, Q., Zhu, J., Zhang, J., Chen, A., Su, S.,Guan, L. (2023). Tough hydrogel with high water content and ordered fibrous structures as an artificial human ligament. *Mater. Horiz., 10*(3), 1012-1019. doi:10.1039/D2MH01299E

Hapipi, N. M., Mazlan, S. A., Ubaidillah, U., Homma, K., Aziz, S. A., Nordin, N. A.,Nazmi, N. (2020). The Rheological Studies on Poly(vinyl) Alcohol-Based Hydrogel Magnetorheological Plastomer. *Polymers, 12*(10), 2332. doi:10.3390/polym12102332

Huang, W.-H., Hung, C.-Y., Chiang, P.-C., Lee, H., Lin, I. T., Lai, P.-C.,Feng, S.-W. (2023). Physicochemical Characterization, Biocompatibility, and Antibacterial Properties of CMC/PVA/Calendula officinalis Films for Biomedical Applications. *Polymers, 15*(6), 1454. doi:10.3390/polym15061454

Iskalieva, A., Yesmurat, M., Al Azzam, K. M., Ainakulova, D., Yerbolat, Y., Negim, E.-S.,Gulzhakhan, Y. (2023). Effect of Polyethylene Glycol Methyl Ether Methacrylate on the Biodegradability of Polyvinyl Alcohol/Starch Blend Films. *Polymers, 15*(15), 3165. doi:10.3390/polym15153165

Jayanudin, Lestari, R. S. D., Barleany, D. R., Pitaloka, A. B., Yulvianti, M., Lumbantobing, P. I., & Prasetyo, Z. E. (2024). Synthesis of polyvinyl alcohol-based polymer hydrogel as water holding in sandy soil using gamma radiation technique and its application for urea loading. *Case Studies in Chemical and Environmental Engineering, 9*, 100634. doi:https://doi.org/10.1016/j.cscee.2024.100634

Kalirajan, C., Dukle, A., Nathanael, A. J., Oh, T.-H., & Manivasagam, G. (2021). A Critical Review on Polymeric Biomaterials for Biomedical Applications. *Polymers, 13*(17), 3015. doi:10.3390/polym13173015

Kenessova, Z. A., Mun, G. A., Urkimbayeva, P. I., Toktabayeva, A. K., Rakhmetullayeva, R. K., Yermukhambetova, B. B.,Alikulov, A. Z. (2025). Development and Characterization of Polymer Blends Based on Polyvinyl Alcohol for Application as Pharmaceutical Dosage Form. *Polymers, 17*(16), 2203. doi:10.3390/polym17162203

Kida, D., Gładysz, O., Szulc, M., Zborowski, J., Junka, A., Janeczek, M.,Karolewicz, B. (2020). Development and Evaluation of a Polyvinylalcohol -Cellulose Derivative-Based Film with Povidone-Iodine Predicted for Wound Treatment. *Polymers, 12*(6), 1271. doi:10.3390/polym12061271

Kopač, T., Ručigaj, A., & Krajnc, M. (2022). Effect of polymer-polymer interactions on the flow behavior of some polysaccharide-based hydrogel blends. *Carbohydrate Polymers, 287*, 119352. doi:https://doi.org/10.1016/j.carbpol.2022.119352

Kumar, A., Sood, A., & Han, S. S. (2022). Poly(vinyl alcohol)-alginate as potential matrix for various applications: A focused review. *Carbohydrate Polymers, 277*, 118881. doi:https://doi.org/10.1016/j.carbpol.2021.118881

Lambertini, L., Coccarelli, G., Toto, E., Santonicola, M. G., & Laurenzi, S. (2024). Poly(vinyl alcohol) gels cross-linked by boric acid for radiation protection of

astronauts. *Acta Astronautica, 221*, 142-154. doi:10.1016/j.actaastro.2024.05.020

Lewandowska, K. (2020). Miscibility Studies of Hyaluronic Acid and Poly(Vinyl Alcohol) Blends in Various Solvents. *Materials, 13*(21), 4750. doi:10.3390/ma13214750

Li, Y., Xie, J., Cheng, H., Wei, X., Chen, J., You, L., & Chen, W. (2025). Polyvinyl alcohol-based polarizers for new displays: molecules, processing and properties. *Soft Matter, 21*(17), 3148-3167. doi:10.1039/D4SM01530D

Liang, X., Zhong, H.-J., Ding, H., Yu, B., Ma, X., Liu, X.,He, J. (2024). Polyvinyl Alcohol (PVA)-Based Hydrogels: Recent Progress in Fabrication, Properties, and Multifunctional Applications. *Polymers, 16*(19), 2755. doi:10.3390/polym16192755

Lodi, L. A., Borges, R., Bortoletto-Santos, R., Oliveira-Paiva, C. A. d., Ribeiro, C., & Farinas, C. S. (2024). Biodegradable PVA-based films for delivery of Bacillus megaterium as seed coating. *Journal of Environmental Chemical Engineering, 12*(6), 114539. doi:https://doi.org/10.1016/j.jece.2024.114539

Malka, E., & Margel, S. (2023). Engineering of PVA/PVP Hydrogels for Agricultural Applications. *Gels, 9*(11), 895. doi:10.3390/gels9110895

Nathan, K. G., Genasan, K., & Kamarul, T. (2023). Polyvinyl Alcohol-Chitosan Scaffold for Tissue Engineering and Regenerative Medicine Application: A Review. *Marine Drugs, 21*(5), 304. doi:10.3390/md21050304

Owusu, E. K., Sinzinkayo, D., Wang, Y., Liu, N., & Yue, T. (2025). Quantitative Characterization of Surface Wettability and Interfacial Energy of Newtonian Fluids on PDMS with Engineered Roughness. *Langmuir, 41*(35), 23867-23874. doi:10.1021/acs.langmuir.5c03164

Priya, S., Choudhari, M., Tomar, Y., Desai, V. M., Innani, S., Dubey, S. K., & Singhvi, G. (2024). Exploring polysaccharide-based bio-adhesive topical film as a potential platform for wound dressing application: A review. *Carbohydrate Polymers, 327*, 121655. doi:https://doi.org/10.1016/j.carbpol.2023.121655

Pugar, D., Haramina, T., Leskovac, M., & Ćurković, L. (2024). Preparation and Characterization of Poly(vinyl-alcohol)/Chitosan Polymer Blend Films Chemically Crosslinked with Glutaraldehyde: Mechanical and Thermal Investigations. *Molecules, 29*(24), 5914. doi:10.3390/molecules29245914

Quéré, D. (2008). Wetting and Roughness. *38*(Volume 38, 2008), 71-99. doi:https://doi.org/10.1146/annurev.matsci.38.060407.132434

Rahman Khan, M. M., & Rumon, M. M. (2025). Synthesis of PVA-Based Hydrogels for Biomedical Applications: Recent Trends and Advances. *Gels, 11*(2), 88. doi:10.3390/gels11020088

Rivera Hernández, G., Valdez, H. A., Arango-Ospina, M., Delgado, J. F., Aguilar-Rabiela, A. E., Gorgojo, J. P.,Sánchez, M. L. (2024). PVA-gelatine based hydrogel loaded with astaxanthin and mesoporous bioactive glass nanoparticles for wound healing. *Journal of Drug Delivery Science and Technology, 101*, 106235. doi:https://doi.org/10.1016/j.jddst.2024.106235

Rodrigo, L., & Munaweera, I. (2025). Employing sustainable agriculture practices using eco-friendly and advanced hydrogels. *RSC Adv, 15*(26), 21212-21228. doi:10.1039/d5ra03035h

Salihu, R., Abd Razak, S. I., Ahmad Zawawi, N., Rafiq Abdul Kadir, M., Izzah Ismail, N., Jusoh, N.,Hasraf Mat Nayan, N. (2021). Citric acid: A green cross-linker of biomaterials for biomedical applications. *European Polymer Journal, 146*, 110271. doi:https://doi.org/10.1016/j.eurpolymj.2021.110271

Shendage, S. S., Kachare, K., Gaikwad, K., Kashte, S., Mai, F.-D., & Ghule, A. V. (2025). Cellulose-based bioactive material and turmeric-impregnated flexible and biocompatible scaffold for bone tissue engineering applications. *J. Mater. Chem. B, 13*(15), 4570-4581. doi:10.1039/D4TB02028F

Shi, X., Wu, B., Dong, X., Zhang, Q., Lu, W., & Chen, X. (2024). Preparation and Rheological Properties of the PVA/CMC/Gel Composite for Mining. *ACS Omega, 9*(26), 28253-28267. doi:10.1021/acsomega.4c01502

Shojaee Kang Sofla, M., Mortazavi, S., & Seyfi, J. (2020). Preparation and characterization of polyvinyl alcohol/chitosan blends plasticized and compatibilized by glycerol/polyethylene glycol. *Carbohydrate Polymers, 232*, 115784. doi:https://doi.org/10.1016/j.carbpol.2019.115784

Solís, A. C., Bento, D., Nunes, S., Valente, A., Pais, A., & Vitorino, C. (2021). Rethinking transdermal drug delivery using PVA-NLC based films. *Polymer, 230*, 124032. doi:https://doi.org/10.1016/j.polymer.2021.124032

Spaniol, K. G., Caldas, S. C., Peres, A. P. S., Dos Santos, E. A., & Acchar, W. (2019). β-TCP/PVA sheets crosslinked with citric acid produced via aqueous tape casting for bone regeneration. *Ceramics International, 45*(9), 12417-12422. doi:https://doi.org/10.1016/j.ceramint.2019.03.173

Teodorescu, M., Bercea, M., & Morariu, S. (2018). Biomaterials of Poly(vinyl alcohol) and Natural Polymers. *Polymer Reviews, 58*(2), 247-287. doi:10.1080/15583724.2017.1403928

Teodorescu, M., Morariu, S., Bercea, M., & Săcărescu, L. (2016). Viscoelastic and structural properties of poly(vinyl alcohol)/poly(vinylpyrrolidone) hydrogels. *RSC Advances, 6*(46), 39718-39727. doi:10.1039/C6RA04319D

Thakare, N. R., & Hazarika, S. (2025). Eco-friendly biopolymers and their biomedical applications: A review. *International Journal of Biological Macromolecules, 327*, 147225. doi:https://doi.org/10.1016/j.ijbiomac.2025.147225

Tudoroiu, E.-E., Albu Kaya, M. G., Titorencu, I., Dinu-Pîrvu, C. E., Marin, M. M., Roşca, A.-M.,Ghica, M. V. (2023). Design and evaluation of new wound dressings based on collagen-cellulose derivatives. *Materials & Design, 236*, 112469. doi:https://doi.org/10.1016/j.matdes.2023.112469

Wan Anuar, W. A. N., Ramli, R. A., El-Sayed, M. M., & Warkar, S. G. (2025). Recent study on biodegradable hydrogels for agriculture application: A review. *Journal of Environmental Chemical Engineering, 13*(2), 115679. doi:https://doi.org/10.1016/j.jece.2025.115679

Xu, Y., Xu, Y., Sun, C., Zou, L., & He, J. (2020). The preparation and characterization of plasticized PVA fibres by a novel Glycerol/Pseudo Ionic Liquids system with melt spinning method. *European Polymer Journal, 133*, 109768. doi:https://doi.org/10.1016/j.eurpolymj.2020.109768

Yang, Y., Liang, Z., Zhang, R., Zhou, S., Yang, H., Chen, Y.,Yu, D. (2024). Research Advances in Superabsorbent Polymers. *Polymers, 16*(4), 501. doi:10.3390/polym16040501