Research Square

Preprints are preliminary reports that have not undergone peer review.
They should not be considered conclusive, used to inform clinical practice,
or referenced by the media as validated information.

Effect of Agarose Content on the Thermal, Rheological, and Electrochemical Properties of Proline: Glycerol-based Eutectogels

Harveen Kaur

khehraharveen@gmail.com

Guru Nanak Dev University https://orcid.org/0009-0002-1796-7835

Research Article

Keywords: Deep eutectic solvent; Proline:Glycerol; Eutectogel; Agarose; Rheology; Thermal stability; Electrochemical properties, Viscoelastic behavior

Posted Date: November 12th, 2026

DOI: https://doi.org/10.21203/rs.3.rs-11718143/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License.
Read Full License

Additional Declarations: The authors declare no competing interests.

EFFECT OF AGAROSE CONTENT ON THE THERMAL, RHEOLOGICAL, AND ELECTROCHEMICAL PROPERTIES OF PROLINE:GLYCEROL-BASED EUTECTOGELS

Author: Harveen Kaur

Department of Chemistry, Guru Nanak Dev University, Amritsar, India

Email: khehraharveen@gmail.com

ABSTRACT

Deep eutectic solvent (DES)-based eutectogels combine the physicochemical characteristics of DESs with the structural properties of polymeric networks. In the present study, a proline DES with a 1:3 molar ratio was incorporated into agarose to prepare eutectogels with different agarose contents. Two formulations were investigated using 50 g and 100 g of agarose per 100 g of DES, respectively. The prepared eutectogels were characterized by thermogravimetric analysis (TGA), rheological measurements, cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS) to investigate their thermal, viscoelastic, flow, and electrochemical behaviour. TGA showed initial mass loss associated with volatile/moisture components followed by major thermal degradation at higher temperatures. Rheological measurements showed that the storage modulus (G′) remained higher than the loss modulus (G″) for both formulations, indicating predominantly elastic and gel-like behaviour. The higher agarose-content formulation exhibited greater G′ and viscosity, while both eutectogels showed shear-thinning behaviour. CV measurements demonstrated measurable electrochemical responses, with the 50% eutectogel showing a higher current at +1 V (3.7 μA) than the 100% eutectogel (0.93 μA). EIS measurements provided complementary information on the resistive and interfacial behaviour of the eutectogels. Overall, the results demonstrate that variation in agarose content can influence the thermal, rheological, and electrochemical behaviour of proline eutectogels, providing a basis for further optimization of DES–biopolymer systems.

Keywords: Deep eutectic solvent; Proline:Glycerol; Eutectogel; Agarose; Rheology; Thermal stability; Electrochemical properties, Viscoelastic behavior.

(1) INTRODUCTION

1.1 DEEP EUTECTIC SOLVENT

Deep eutectic solvents (DESs) are now widely known as a new class of ionic liquid because they show many similar characteristics and properties of ILs. The terms DES and IL have been used interchangeably in the literature but it is necessary to point out that these are actually two different types of solvents. DESs are systems formed from a eutectic mixture of Lewis or Bronsted acids and bases which can contain a variety of anionic and/or cationic species in contrast, ILs are formed from systems composed primarily of one type of discrete anion and cation. It is illustrated here that although the physical properties of DESs are similar to other ILs, their chemical properties are entirely different from the ionic liquids. [1][2][3][4]

Indeed, many reports pointed out the hazardous toxicity and the very poor biodegradability of most ILs. ILs with high purity have certain impurities, even in trace amounts, affect their physical properties. Additionally, their synthesis is far to be environmentally friendly since it generally requires a large number of salts and solvents in order to completely exchange the anions. These drawbacks together with the high price of common ILs unfortunately hamper their industrial emergence and new concepts are now strongly needed in order to utilize these systems in a more rational way. [5][6]

To overcome the high price and toxicity of ILs, a new generation of solvent, named Deep Eutectic Solvents (DES), has emerged at the beginning of this century. Abbott et al. noted an abnormally depression in melting at the eutectic composition (where deep eutectic solvent form) of certain hydrogen bond donors (HBDs) and acceptors (HBAs) are mixed. This behaviour was first observed in 1:2 mole fraction combination of powdered choline chloride [melting point, Tm ≈ 302 °C] with crystalline urea [Tm ≈ 133 °C] which reportedly results in a liquid at room temperature [melting point of mixture at eutectic composition, Eutectic = 12 °C]. [7]

The major components of DESs are Hydrogen Bond Donors (HBDs) and Hydrogen Bond Acceptors (HBAs) which interact with each other through hydrogen bonding. Their melting points are related to hydrogen bonding. The charge delocalization occurs through hydrogen bonding between HBAs and HBDs. **Hydrogen bond donor:** The molecule or functional group that provides the hydrogen atom for the hydrogen bond is called hydrogen bond donor. HBDs are often

amines, alcohols, or carboxylic acids. **Hydrogen bond acceptor:** Hydrogen bond acceptor is an atom or molecule that participate in hydrogen bond by accepting the hydrogen bond from hydrogen bond donor, typically an electronegative atom like nitrogen, oxygen, fluorine with lone pair of electrons. There are some of the examples of hydrogen bond acceptor are given below. HBAs include quaternary ammonium or phosphonium salts. [8][9]

1.2 TYPES OF DES

Type I, are considered to be of an analogous type to the well-studied metal halide/imidazolium salt systems. Examples of type I eutectics include the well-studied chloroaluminate/imidazolium salt melts and less common ionic liquids formed with imidazolium salts and various metal halides including AgCl, CuCl, LiCl, $CdCl_2$, $CuCl_2$, $SnCl_2$, $ZnCl_2$, $LaCl_3$, YCl_3, and $SnCl_4$.

Type II ,Hydrated metal halides and choline chloride is known as type II. The relatively low cost of many hydrated metal salts coupled with their inherent air/moisture insensitivity makes their use in large scale industrial processes viable.

Type III eutectics, DESs are commonly formed by a hydrogen-bond acceptor (HBA) and a hydrogen-bond donor (HBD), without metal salts. They are often composed of quaternary ammonium salts or other HBAs with organic compounds such as alcohols, carboxylic acids, amides, or sugars. Examples include choline chloride:glycerol and choline chloride:urea systems. A range of hydrogen bond donor have been studied to date, with deep eutectic solvents formed using amides, carboxylic acids, and alcohols. These liquids are simple to prepare, and relatively unreactive with water; many are biodegradable and are relatively low cost. derivatives.

Type -V Recently proposed type V DES are prepare only by mixing non-ionic precursor as they show lower viscosities than their ionic counterpart and non-negligible vapour pressure. (May be advantageous for processes evaporation is desirable).[7][10]

1.3 PHYSICAL PROPERTIES OF DEEP EUTECTIC SOLVENT

1.3.1 Viscosity: Viscosity is an important physical property of DESs and is generally higher than that of many conventional solvents. The high viscosity mainly arises from strong hydrogen-bonding and electrostatic interactions between the DES components, which restrict the mobility of the species. Viscosity depends on the chemical nature and molar ratio of the components,

temperature, and water content. Increasing temperature or water content generally reduces viscosity. [11][12]

1.3.2 Ionic Conductivity: DESs generally exhibit relatively low ionic conductivity, which is closely related to their high viscosity and restricted ion mobility. Conductivity usually increases with increasing temperature as viscosity decreases. It is also influenced by the nature and molar ratio of the hydrogen-bond acceptor and donor, as well as the water content of the DES. [13][14]

1.3.3 Surface Tension: Surface tension is influenced by the intermolecular interactions between the hydrogen-bond acceptor and hydrogen-bond donor in DESs. Its value depends on the chemical composition and molar ratio of the components, temperature, and molecular structure. Changes in these factors can alter the strength of interactions within the DES and consequently affect its surface properties. [15][16]

1.4.4 Refractive Index: Refractive index is one of the significant physical properties of DESs, which provides useful information about the composition of deep eutectic solvents. The refractive index values play an important role in the calculation of molar refraction by using the Lorentz–Lorenz equation.

$$R = M/\rho(nD + 1/nD' + 1)$$

Where, R_D, M, ρ, and n_D are molar refraction ($cm^3 \cdot mol^{-1}$), molar mass ($g \cdot mol^{-1}$), density ($g \cdot cm^{-3}$) and refractive index, respectively. The molar refraction shows molecular interaction between HBAs and HBDs of DESs components. Refractive index is inversely proportional to the temperature by increasing temperature, the hydrogen bond interactions between the DES mixture reduces, and DESs become less dense due to decreasing their refractive index. The refractive index values depend on the size of molecules and the molecules with larger sizes have a higher refractive index. [17][18][19]

1.4 EUTECTOGEL

Eutectogels have been investigated for a range of potential applications because of their combination of structural integrity and the physicochemical properties of the incorporated solvent. A major limitation of DESs in practical applications is their inherently liquid nature, which can lead

to leakage, limited shape stability, and difficulties in maintaining the solvent within a defined structure. To address these limitations, DESs can be immobilized within three-dimensional polymeric networks to form **eutectogels**. Eutectogels are soft, semi-solid materials in which a DES is retained within a crosslinked or physically associated polymer network. In such systems, the polymer matrix provides structural integrity, while the DES acts as the liquid functional phase, retaining its characteristic hydrogen-bonding environment and ionic or molecular interactions. The resulting combination can provide a balance between the fluidity and solvent functionality of DESs and the mechanical stability of polymeric materials. [20][21]

1.4.1 COMPONENTS OF EUTECTOGELS

Eutectogels generally consist of two principal components: a **deep eutectic solvent (DES) phase** and a **polymeric network** that immobilizes and retains the solvent. The combination of these components determines the structural, physicochemical, rheological, and transport properties of the resulting material. Depending on the intended application, additional components such as crosslinking agents, fillers, nanoparticles, or functional additives may also be incorporated to modify specific properties.

(I) DEEP EUTECTIC SOLVENT PHASE

The DES constitutes the functional liquid phase of a eutectogel. It is typically formed by combining a hydrogen-bond acceptor (HBA) with a hydrogen-bond donor (HBD), although other DES compositions are also possible. The nature and molar ratio of the DES components influence properties such as viscosity, polarity, hydrogen-bonding interactions, thermal behavior, and ionic or molecular transport. When incorporated into a polymer network, the DES can act not only as the liquid phase but also as a plasticizing or functional component, depending on its interactions with the polymer chains.

(II) POLYMERIC NETWORK.

The polymer component provides the three-dimensional framework required to immobilize the DES and maintain the shape and structural integrity of the eutectogel. Both synthetic polymers

and natural or biopolymer-based materials can be used. The polymer concentration, molecular characteristics, crosslinking density, and network architecture influence the mechanical strength, viscoelastic behavior, swelling, solvent retention, and transport properties of the resulting gel. In biopolymer-based eutectogels, interactions between functional groups of the polymer and DES components can further contribute to network formation and stability.

(III) **ADDITIONAL FUNCTIONAL COMPONENTS.**

Depending on the desired application, eutectogels may contain additional components that introduce or enhance particular functionalities. Crosslinking agents can be used to strengthen the polymer network, while nanoparticles, fillers, or other functional molecules may be incorporated to modify mechanical, electrical, thermal, optical, or catalytic properties. However, the specific composition should be selected according to the required characteristics of the final material. [22][23]

Thus, the properties of a eutectogel are governed not simply by the individual properties of the DES and polymer, but by their **interactions within the resulting network**. The choice of DES composition and polymer therefore provides an important means of controlling the structure and performance of eutectogels.

1.4.2 PROPERTIES OF EUTECTOGELS

The incorporation of a deep eutectic solvent into a polymeric network can produce a material with a combination of the physicochemical characteristics of the DES and the structural stability of the polymer matrix. As a result, eutectogels can exhibit a range of properties that are influenced by the composition of the DES, polymer type, polymer concentration, crosslinking or network structure, and interactions between the DES components and polymer chains.

Thermal properties are an important characteristic of eutectogels because the polymer network can influence the thermal behavior of the immobilized DES. Intermolecular interactions between the DES and polymer chains may modify the mobility of the components and affect thermal stability. Thermogravimetric analysis (TGA) is commonly used to investigate mass-loss behavior and identify the temperature ranges associated with solvent loss and decomposition of the gel components.

Rheological and mechanical properties determine the structural integrity and deformation behavior of eutectogels. The formation of a three-dimensional polymer network can transform the liquid-like DES phase into a viscoelastic material capable of maintaining its shape. Parameters such as storage modulus (G′), loss modulus (G″), complex viscosity, and shear-dependent viscosity provide information about the elastic and viscous contributions of the material. The relative magnitude of G′ and G″ can be used to characterize the predominant solid-like or liquid-like response of the gel. Increasing polymer concentration or network strength can generally modify chain mobility and increase the resistance of the material to deformation.

Transport and electrochemical properties are also relevant for eutectogels containing ionic or highly polar DES components. Immobilization of the DES within a polymer network can influence the movement of charge-carrying species through the material. Consequently, the polymer–DES interactions and network density may affect ion transport and electrochemical response. Techniques such as cyclic voltammetry and electrochemical impedance spectroscopy can provide information about the electrochemical behavior and resistance characteristics of the material.

Swelling, solvent retention, and dimensional stability are additional properties that depend on the interaction between the DES and polymer network. The ability of the network to retain the DES while maintaining its structural integrity is particularly important for practical applications. These characteristics can be affected by polymer concentration, crosslinking density, DES composition, and the affinity between the polymer and DES components. [24][25]

Overall, the properties of eutectogels arise from the interplay between the DES phase and the polymeric network. Therefore, controlling the DES composition and polymer network provides a means of tailoring thermal, rheological, mechanical, transport, and electrochemical characteristics for specific applications. A systematic understanding of these structure–property relationships is essential for the development of eutectogels as functional and sustainable materials.

In the present study, a proline–glycerol (Pro) deep eutectic solvent with a 1:3 molar ratio was incorporated into agarose to prepare DES-based eutectogels with different agarose contents. The prepared gels were investigated using thermogravimetric analysis, rheological measurements, cyclic voltammetry, and electrochemical impedance analysis to examine their thermal behavior, viscoelastic properties, shear response, and electrochemical characteristics. Particular attention was given to the effect of increasing agarose content on the properties of the DES–polymer

network. This study provides an experimental basis for understanding the relationship between polymer concentration and the physicochemical behavior of Pro–agarose eutectogels and contributes to the broader development of DES-based sustainable functional materials.

(2) EXPERIMENTAL SECTION

2.1 MATERIALS

Glycerol is purchased from LOBA Chemicals, Mumbai, India. L-Proline and Agarose I are purchased from sigma Aldrich.

2.2 METHODS

2.2.1 SYNTHESIS OF DES

The deep eutectic solvent (DES) was prepared by mixing **L-proline and glycerol in a 1:3 molar ratio** using a thermal mixing method. The components were mixed and heated at approximately **75 °C** with continuous mixing until a clear and homogeneous liquid was obtained. The resulting DES was transferred into sealed vials to minimize contamination and loss of components during storage. The prepared DES was subsequently stored in a **desiccator** until further use.

2.2.2 SYNTHESIS OF EUTECTOGEL

Two agarose-based eutectogel formulations were prepared using proline deep eutectic solvent (DES) at a 1:3 molar ratio. For formulation A50, 50 g of agarose was incorporated into 100 g of DES, whereas formulation A100 contained 100 g of agarose with 100 g of DES. Thus, the agarose-to-DES mass ratios were 1:2 for A**50** and 1:1 for A**100**, respectively as seen in table 2. The mixtures were heated to approximately 75 °C under stirring to facilitate dissolution and homogenization of agarose within the DES. The hot mixtures were subsequently cooled to room temperature, during which gelation occurred, yielding agarose-based eutectogels with different polymer contents.

Table 2. formulation of 2 different DES based eutectogels by weight of agarose.

Formulation	Agarose	DES	Agarose:DES
A50	50 g	100 g	**1:2**
A100	100 g	100 g	**1:1**

(3) RESULTS AND DISCUSSIONS

3.1 THERMAL STABILITY OF GEL

Thermogravimetric analysis (TGA) is a thermal analysis technique used to evaluate the thermal stability and decomposition behaviour of a material by continuously measuring its mass as a function of temperature. In TGA, a small quantity of sample is placed in a crucible and heated at a controlled heatin rate under a specified atmosphere. The instrument continuously records the change in sample mass with increase in temperature. The resulting curve of mass per unit weight loss versus temperature provides information about moisture/ volatile loss, decomposition and amount of thermally stable residue remaining after heating. [26][27]

(I) TGA Analysis of L-proline:Glycerol (1:3) DES and Agarose Eutectogels:

The thermal stability of glycerol, agarose, DES, and corresponding A50 and A100 eutectogels could be clearly seen in figure 1.

The initial decrease in mass observed at lower temperature can mainly be associated with the removal of physically bound water and other volatile components or impurities. After this initial region, the samples show a more pronounced weight loss at higher temperatures, corresponding to the thermal decomposition of DES components and/or agarose network. The DES shows a major decomposition region approximately between 180-270^{o}C, indicating that DES remains relatively stable before undergoing substantial thermal degradation. Glycerol shows a comparatively sharp mass-loss region at around 200-270^{o}C. for the agarose containing systems, the behavior changes noticeably. The agarose sample maintains a relatively high mass over a wider temperature range

and undergoes its major degradation at higher temperatures, with considerable residue remaining at the end of analysis. This indicates the formation of thermally stable polysaccharide structure. The A50 and A100 eutectogels also show an initial mass loss followed by a major decomposition region. Importantly, both gel samples retain a significant fraction of their original mass even at the higher temperatures, unlike pure DES, which undergoes much more extensive mass loss. The residual mass is approximately 8% for A50 gel and 15% for A100 gel from presented curve.

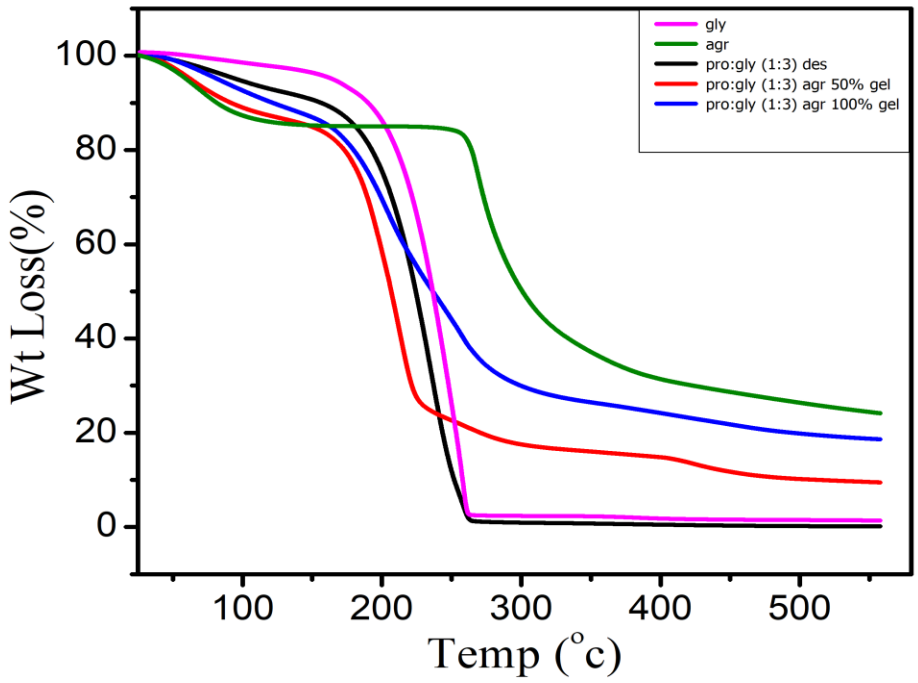


Figure 1 TGA graph of DES component, DES and GELS.

(II) Comparison of A50 and A100 agarose eutectogels:

The second graph (figure 2) shows that the two eutectogels have similar initial behavior, with approximately 13-14% mass loss around 150°C. This region can be attributed primarily to the loss of moisture/ volatile components. The main decomposition occurs at higher temperatures. The graph indicates approximately

59% mass loss for A50 and 60% for A100, over the major degradation region. The curves then gradually decrease, leaving a greater residual mass for A100 agarose formulation.

The higher residual mass observed for the A100 is consistent with its greater agarose content and the formulation of a more substantial polymeric network. Overall, the TGA profiles demonstrate that incorporation of agarose produces a distinct thermal-degradation behavior compared with the neat proline:glycerine DES.

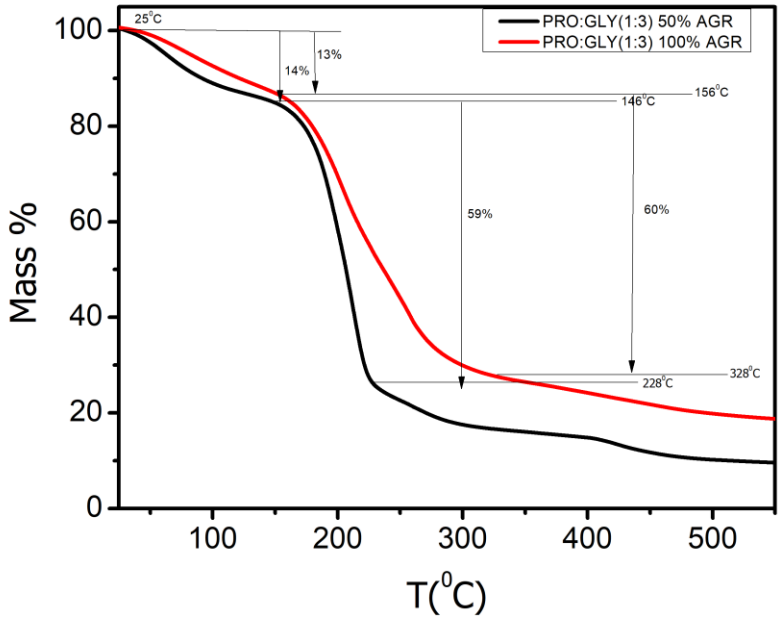


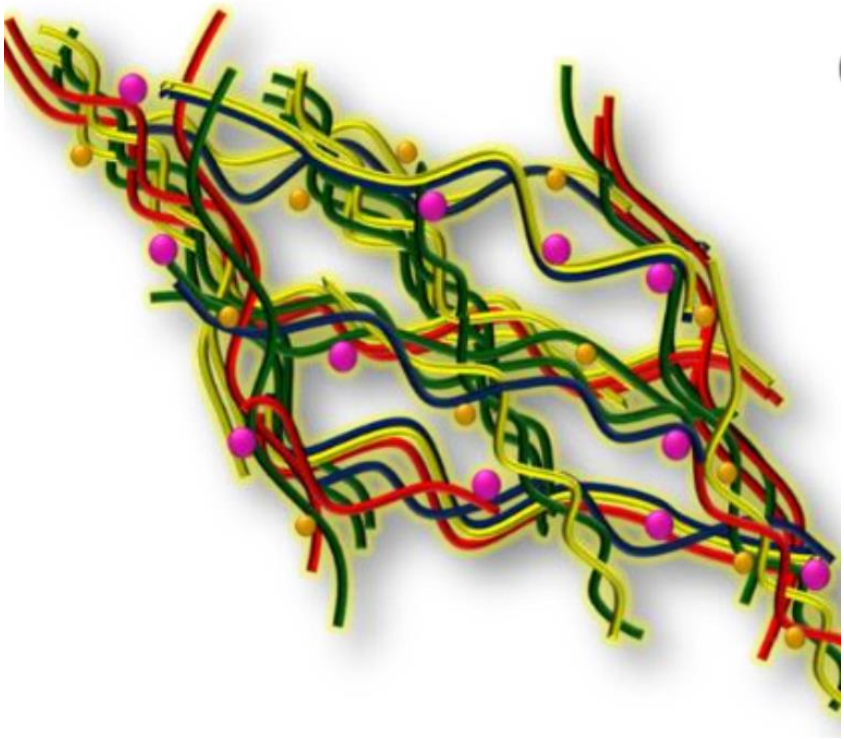
Figure 2 TGA graph of 50% and 100% agrose gel

3.2 STRUCTURAL DESIGN AND RHEOLOGICAL PROPERTIES OF EUTECTOGELS.

(A) STRUCTURAL DESIGN OF THE EUTECTOGELS:

The eutectogels were designed by incorporating proline:glycerol DES within an agarose based three-dimensional polymeric network. Agarose is a polysaccharide capable of forming physically cross-linked network through intramolecular interactions and chain association. The incorporation

of DES provides a hydrogen bond rich liquid phase within the polymeric matrix. The hydroxyl functionalities of glycerol and the polar functional groups of proline can participate in hydrogen bonding interactions with functional groups present on the agarose chains. Such intermolecular interactions contribute to the association between DES phase and agarose network, and facilitate the retention of DES within polymeric matrix. [28][29]



The schematic representation illustrates the possible intermolecular interactions between the agarose chains and the hydrogen-bond donor (HBD) and hydrogen-bond acceptor (HBA) sites associated with the DES. Here, pink circle denotes the hydrogen bond donor, yellow circle represents the hydrogen bond acceptor and different color threads represent the agarose molecule showing attraction with HBD and HBA. The formation of these interactions within the polymer matrix results in a physically interconnected network capable of immobilizing the DES. Consequently, the macroscopic viscoelastic properties of the eutectogels are governed by the combined contributions of the agarose network and the intermolecular interactions between agarose and the Proline:Glycerol DES.

The extent of network formation and the strength of these intermolecular interactions are expected to influence the elastic modulus, viscous response, deformation resistance and flow behavior of the resulting eutectogels. [30]

(B) RHEOLOGICAL PROPERTIES

The rheological properties of eutectogels are important for evaluating their structural stability, flexibility, viscoelastic behaviour, and suitability for pharmaceutical and biomedical applications. These properties are strongly influenced by the type and concentration of polymer, the composition of the deep eutectic solvent (DES), and the interactions between the polymeric network and DES components.

Rheological analysis is generally performed by measuring parameters such as storage modulus (**G′**), loss modulus (**G″**), viscosity, and the response of the gel to changes in frequency or shear rate. G′ represents the **elastic or solid-like contribution**, whereas G″ represents the **viscous or liquid-like contribution**. When G′ is higher than G″ over a broad frequency range, the material exhibits predominantly gel-like or elastic behaviour, indicating the presence of a stable three-dimensional network. [31][32] [33]

The viscoelastic behavior of the eutectogels were investigated using a rotational rheometer. Oscillatory rheological measurements provide the storage modulus (G′) and loss modulus (G″), which represent the elastic and viscous components of the material response, respectively. The storage modulus reflects the energy stored elastically during oscillatory deformation and is therefore associated with the strength and integrity of the gel network, whereas the loss modulus represents the energy dissipated through viscous processes. [34][35][36]

(I) Strain-dependent viscoelastic behavior

Strain-dependent viscoelastic behavior describes how the rheological properties of a eutectogel change when the **amplitude of deformation (strain)** is gradually increased. At very low strain, the deformation is small and the internal polymeric network remains largely undisturbed. This region is called the **Linear Viscoelastic Region (LVR)**. Within the LVR, G′ and G″ generally remain relatively constant. When the applied strain becomes sufficiently high, the interactions

maintaining the gel network begin to be disrupted. Consequently, **G′ may decrease**, indicating a loss of elastic structure. At still higher strain, the gel may undergo significant structural breakdown and eventually behave more like a flowing material. This test therefore helps determine the **mechanical strength and structural stability of the eutectogel** and indicates how much deformation it can tolerate before its internal network is substantially damaged. [37][38]

The strain-amplitude dependence of G′ and G″ for the A50 and A100 eutectogels was investigated at a fixed angular frequency of 10 rad s^{-1}, as presented in Figure 3. It could be clearly seen from figure 3 that at low strain amplitudes, both eutectogels exhibited a nearly strain-independent storage modulus, with G′ remaining higher than G″. The predominance of G′ over G″ indicates that the response of both materials is predominantly elastic within the small-deformation regime, which is characteristic of a well-developed gel network. The region over which the moduli remain approximately independent of strain corresponds to the linear viscoelastic region (LVR), within which the internal structure of the eutectogel is not substantially altered by the applied deformation.

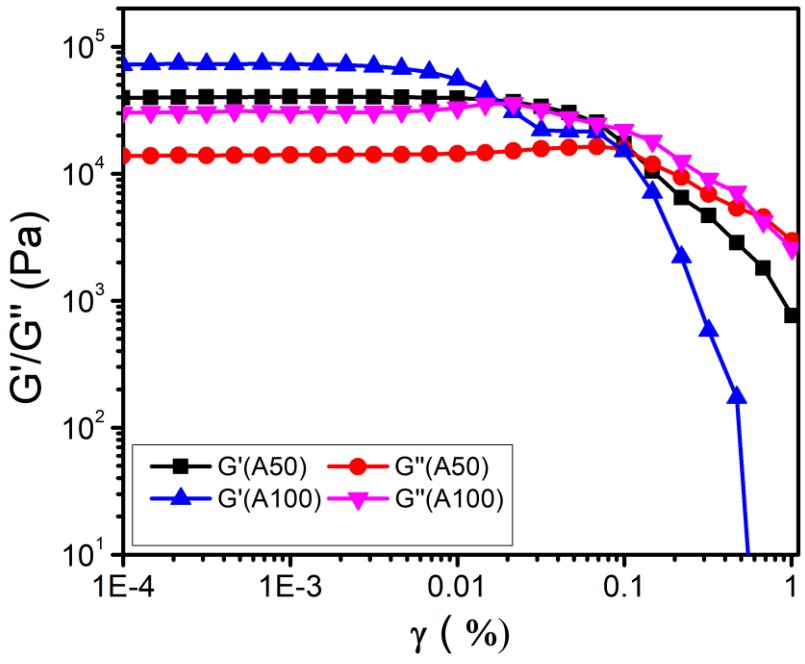


Figure3. Variation of storage (G') and loss modulus (G") as a function of strain (γ %) at a fixed angular frequency (ω=10 rad/s).

The A100 eutectogel exhibited a higher initial G′ than A50, indicating a greater elastic contribution and higher resistance to small-amplitude deformation under the experimental conditions. This difference suggests that the network structure in A100 provides a greater degree of elastic resistance. The higher modulus may arise from differences in the extent of agarose network formation and/or the interactions between the agarose chains and the DES phase. With increasing strain amplitude, a progressive decrease in G′ was observed, particularly beyond the linear viscoelastic region. This reduction in storage modulus indicates that the applied deformation increasingly perturbs the agarose–DES network. At sufficiently high strain, the physical interactions and network junctions are progressively disrupted or rearranged, resulting in a reduction in the elastic response of the material. The decrease in G′ therefore represents the transition from the intact network structure towards a more structurally disturbed state. The strain sweep thus demonstrates that the proline:glycerol–agarose eutectogels possess a distinct elastic network at low deformation but undergo structural weakening when subjected to sufficiently large strains.

(II) Frequency-dependent viscoelastic behavior

Frequency-dependent viscoelastic behavior describes the response of a eutectogel when it is subjected to **oscillatory deformation at different frequencies**. A rheometer applies a small repeated deformation to the gel and measures two important parameters:

- **Storage modulus (G′):** represents the **elastic or solid-like behavior** of the material. It reflects the energy stored during deformation.
- **Loss modulus (G″):** represents the **viscous or liquid-like behavior**. It reflects the energy dissipated as heat.

The relationship between G′ and G″ gives information about the nature of the gel. If **G′ > G″**, the elastic contribution dominates, suggesting a predominantly **gel-like/solid-like network**. If **G″ > G′**, viscous behavior is dominant. Frequency dependence is useful because a material may respond differently to **slow deformation (low frequency)** and **rapid deformation (high frequency)**. If G′ remains relatively higher than G″ over a broad frequency range, it indicates that the three-dimensional network is relatively stable over those testing conditions. [39][40]

The frequency dependence of the storage and loss moduli was investigated at a fixed strain amplitude of 0.1%, and the results are presented in Figure 4. The low strain employed for the measurement ensures that the material is investigated predominantly within its linear viscoelastic regime, thereby minimizing structural damage during the frequency sweep.

For both A50 and A100, G′ remained higher than G″ over the investigated angular-frequency range of 0.1–100 rad s^{-1}. The absence of a crossover between G′ and G″ and the persistent dominance of G′ demonstrate that the elastic component remains greater than the viscous component throughout the experimental frequency range. This behaviour confirms the predominantly solid-like or gel-like character of the eutectogels. Both G′ and G″ showed a gradual increase with increasing angular frequency. The frequency dependence indicates that the viscoelastic response of the eutectogels is influenced by the timescale of deformation. At higher frequencies, the polymer chains and associated intermolecular interactions have less time to undergo large-scale rearrangement during each deformation cycle, resulting in a greater contribution from the elastic network response.

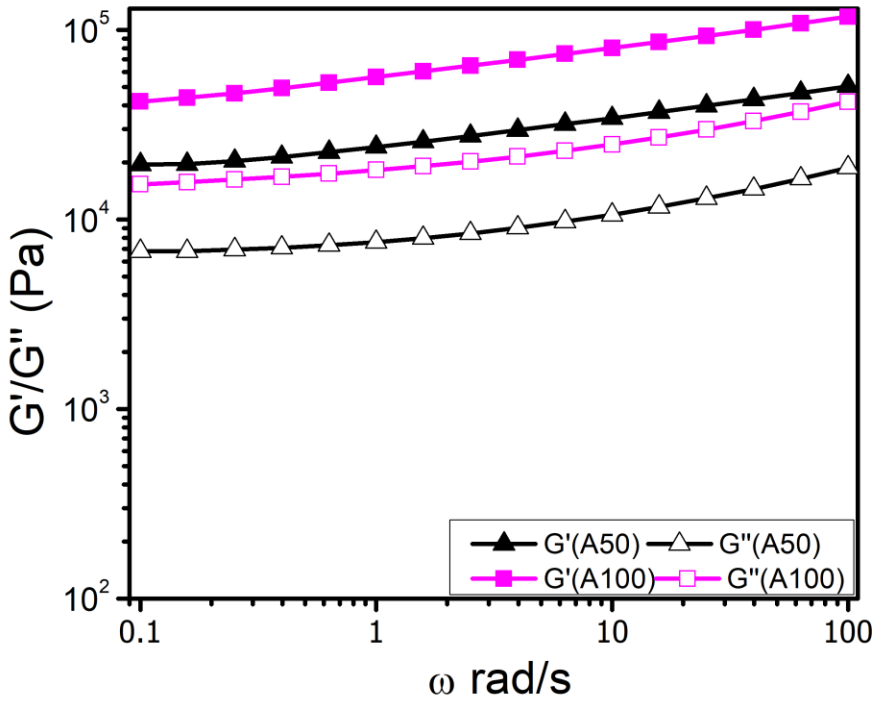


Figure 4 Variation of storage (G') and loss modulus (G") as a function of angular frequency (ω) at a fixed shear strain (γ=0.1%)

The A100 agarose sample exhibited higher values of both G′ and G″ compared with A50 agarose throughout the frequency range. The higher G′ indicates a greater elastic contribution, whereas the simultaneous increase in G″ indicates that viscous dissipation also contributes to the overall response. Nevertheless, the persistent G′ > G″ relationship confirms that the elastic network response dominates for both formulations.

(III) Shear-rate-dependent viscosity

Shear-rate-dependent viscosity describes the change in the **viscosity of a eutectogel with increasing shear rate**. Shear rate represents how rapidly adjacent layers of the material move relative to one another. When shear is applied to a gel, its internal polymeric network can become temporarily rearranged or disrupted. As a result, the resistance of the material to flow may change. For example, if the viscosity **decreases with increasing shear rate**, the material exhibits **shear-thinning (pseudoplastic) behaviour**. This is commonly observed in polymeric gels and is useful for formulations that need to be easily spread, injected, or processed under applied force. After the shear is removed, some gels can partially or completely recover their original structure. This behaviour is related to the ability of the network to **reorganize after deformation**. [41][42][43]

The steady-shear viscosity of the A50 and A100 eutectogels was investigated as a function of shear rate, as shown in Figure 5. A pronounced decrease in viscosity with increasing shear rate was observed for both formulations. This behaviour is characteristic of shear-thinning or pseudoplastic flow. At low shear rates, the eutectogel exhibits relatively high viscosity because the agarose chains and associated DES-containing network provide considerable resistance to flow. As the applied shear rate increases, the polymer chains progressively orient in the direction of flow, while intermolecular associations within the network undergo rearrangement. These changes reduce the resistance of the network to flow, resulting in a decrease in the apparent viscosity.

The A100 eutectogel exhibited a higher viscosity than A50 over much of the low- and intermediate-shear-rate region, consistent with its higher storage modulus observed in the oscillatory measurements. This indicates that A100 possesses a comparatively stronger resistance to deformation and flow under these experimental conditions. At higher shear rates, however, a pronounced reduction in the viscosity of A100 was observed, indicating substantial shear-induced structural rearrangement or breakdown of the network.

The shear-thinning behaviour of both eutectogels is particularly relevant to their processing and potential application because it indicates that the materials can maintain a structured, highly viscous state under low-shear conditions while becoming progressively easier to process or spread under applied shear.

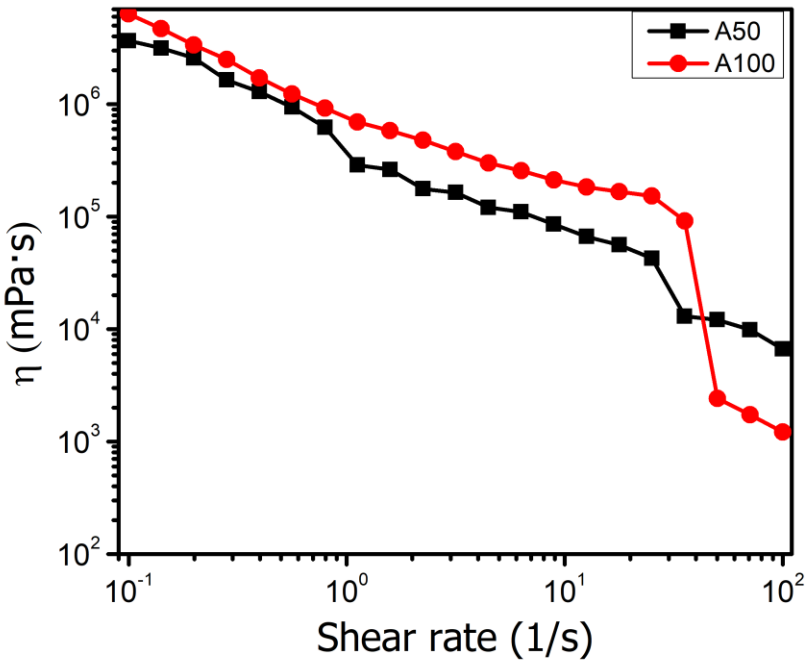


Figure 5 Viscosity (η) ver sus shear rate ($\dot{\gamma}$) profile

3.3 ELECTROCHEMICAL PROPERTIES OF EUTECTOGELS

Cyclic voltammetry (CV) is a widely used electrochemical technique for investigating the electrochemical response, redox behaviour, and charge-transfer processes of electroactive materials. In CV, the potential applied to an electrode is varied over a defined range at a constant scan rate, while the resulting current is recorded. The resulting current–potential curve provides information about oxidation and reduction processes, electrochemical stability, and the reversibility and kinetics of electrochemical reactions. [44][45][46]

Figure 6 presents the cyclic voltammetry curves of the A50 and A100 eutectogels recorded at a scan rate of 200 mV s^{-1} within a potential window of −1 to +1 V at 25 °C using aluminium

electrodes. Both eutectogels exhibited measurable electrochemical responses, indicating the presence of mobile ionic species within the DES-based gel network.

At a positive potential of +1 V, the A50 eutectogel exhibited a maximum current of approximately **3.7 µA**, whereas the A100 eutectogel showed a lower current of approximately **0.93 µA**. This decrease in current with increasing agarose content can be associated with the formation of a denser polymeric network. The increased agarose content may restrict the movement of the DES components and mobile ionic species through the gel matrix. In addition, hydrogen-bonding interactions between agarose and the proline (1:3) DES may further influence the mobility of the ionic species, thereby reducing the measured current.

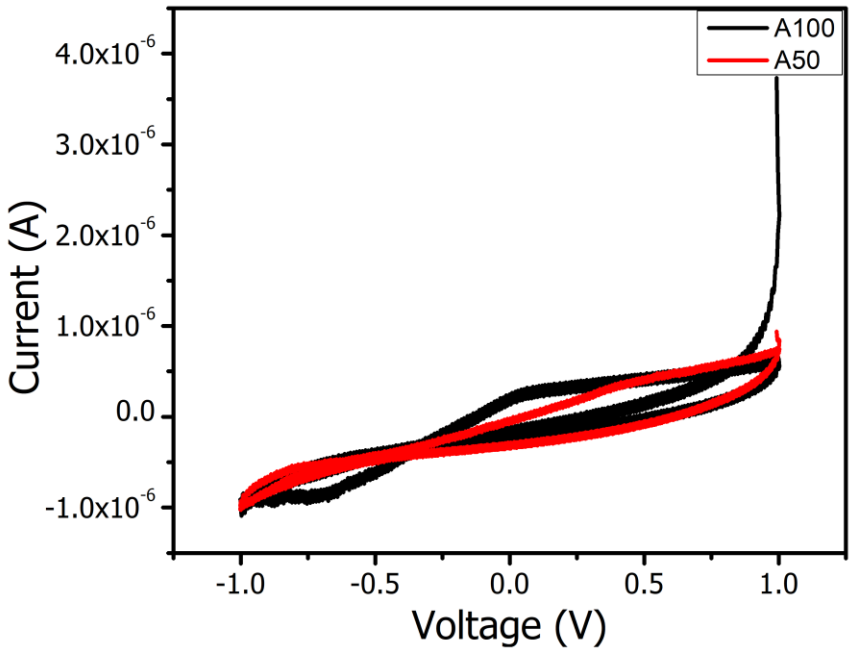


Figure 6 Cyclic Voltammetry (CV) curves at a scan rate of 200 mVs^{-1} for the eutectogels at T = 25°C.

At −1 V, the corresponding current values were approximately **−0.84 µA for A50** and **−0.86 µA for A100**. The relatively similar cathodic currents suggest that the electrochemical response at the negative end of the potential window is less strongly affected by the change in agarose content than the positive-potential response. The differences in anodic and cathodic current may arise from differences in ion mobility, electrode–electrolyte interactions, and the contribution of the individual DES components to the electrochemical response.

The appearance of current responses and redox features indicates that the eutectogels possess electrochemically active/mobile species. However, the presence and intensity of CV peaks alone should not be used as a direct measure of ionic conductivity. Quantitative determination of ionic conductivity requires complementary electrochemical impedance spectroscopy (EIS) or conductivity measurements. The CV results nevertheless provide useful information about the electrochemical behaviour of the eutectogels within the investigated potential window.

3.3.1 Electrochemical Impedance Spectroscopy

Figure 7 shows the Nyquist plots obtained for the A50 and A100 eutectogels. A Nyquist plot represents the imaginary component of impedance (Z″) as a function of the real component (Z′) over a range of frequencies and is commonly used to investigate the resistive and charge-transfer characteristics of electrochemical systems. [49]

The high-frequency intercept of the Nyquist plot with the real impedance axis can be associated with the bulk or solution resistance (Rs) of the electrochemical cell, including contributions from the electrolyte and other components of the measurement system. The semicircular portion, when present, is generally associated with the combined effects of bulk resistance and interfacial charge-transfer processes and can be used to estimate the charge-transfer resistance (Rct) with an appropriate equivalent-circuit model. [47][48]

The lower-frequency region, represented by a more inclined or approximately straight line, is commonly related to ion diffusion and interfacial polarization processes. The appearance of this region suggests that ion transport within the eutectogel and at the electrode–electrolyte interface contributes to the overall impedance response.

For the present proline (1:3)-based eutectogels, the EIS results therefore provide complementary evidence regarding ionic transport and interfacial electrochemical behaviour. Differences between the A50 and A100 Nyquist plots can be related to changes in the agarose concentration and the resulting polymeric network. A higher agarose content can restrict the movement of DES-associated ionic species, potentially increasing the resistance to ionic transport. The exact contribution of bulk resistance, charge-transfer resistance, and diffusion should, however, be determined from the fitted equivalent circuit and numerical impedance parameters rather than from the Nyquist plot alone.

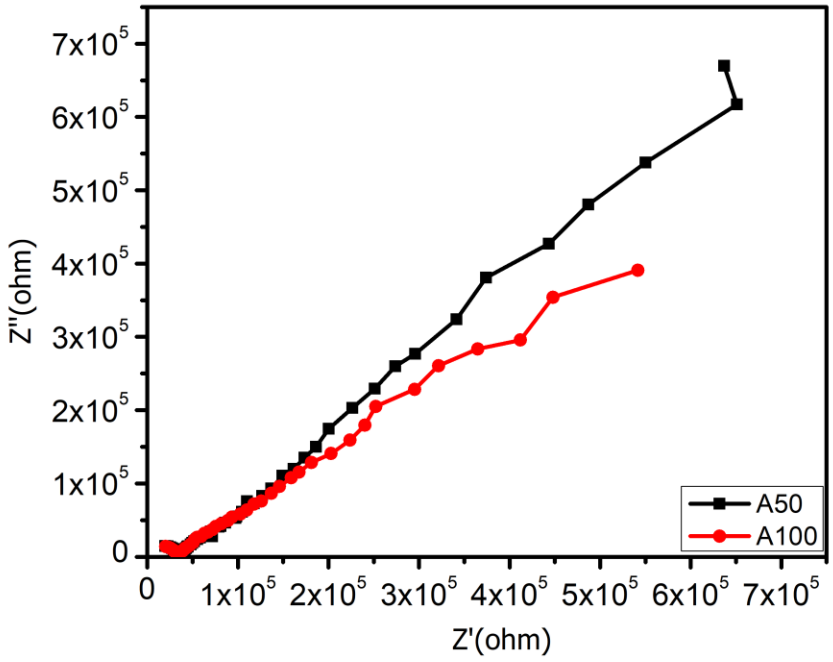


Figure 7 Nyquist Plots of A50 and A100 Eutectic gel.

(4) CHALLENGES

Despite the promising combination of agarose and proline DES, several challenges remain in establishing clear composition–property relationships and translating the observed properties of these eutectogels into practical applications.

1. Optimization of the agarose-to-DES ratio

The properties of agarose–DES eutectogels are strongly influenced by the relative amounts of polymer and DES. In the present system, 50% contained 50 g agarose per 100 g DES, corresponding to an agarose mass ratio of 1:2, whereas A100 contained 100 g agarose per 100 g DES, corresponding to a ratio of 1:1. Increasing the agarose content was associated with changes in the rheological and electrochemical behaviour of the gels. A100 exhibited higher storage modulus and viscosity, indicating greater resistance to deformation and flow, while A50 showed a higher current response at +1 V. These observations suggest a trade-off between strengthening the

polymeric network and maintaining the mobility of DES-associated ionic species. However, only two compositions were investigated in the present study, which limits the ability to establish a detailed composition–property relationship. A broader range of agarose ratios would therefore be required to identify an optimum composition for a particular application.

2. Understanding polymer–DES interactions

The formation and properties of the eutectogels are expected to depend on interactions between agarose and the DES. Hydrogen bonding involving the hydroxyl groups of agaroses and the functional groups of the DES components may contribute to network formation, retention of the DES phase, and the observed thermal and rheological behavior. However, the present characterization does not directly establish the molecular-level nature or strength of these interactions. Techniques such as Fourier-transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC), X-ray diffraction (XRD), and microscopic analysis could provide additional information regarding hydrogen bonding, thermal transitions, crystallinity, and changes in the gel microstructure. Molecular simulation approaches could further complement experimental measurements by providing insight into the organization and interactions of the DES components within the agarose network.

3. Balance between mechanical strength and ionic mobility

An important challenge in this system is achieving an appropriate balance between the mechanical integrity of the polymeric network and the mobility of electrochemically active ionic species. Increasing agarose content from A50 to A100 produced a more concentrated polymeric network and was accompanied by a reduction in the positive current response observed by cyclic voltammetry. This behavior may indicate that the more concentrated agarose network restricts the movement of DES-associated mobile species. At the same time, the higher modulus and viscosity of A100 demonstrate the advantage of increased agarose content in improving mechanical and flow resistance. Thus, increasing polymer content cannot necessarily be expected to improve all properties simultaneously. Future optimization should therefore consider the specific balance of mechanical, rheological, and electrochemical requirements for the intended application.

4. Further quantification of electrochemical properties

The cyclic voltammetry results demonstrate measurable electrochemical responses from both formulations, but CV response alone is insufficient to quantitatively determine ionic conductivity. The EIS measurements provide complementary information regarding resistance and ion-transport processes; however, quantitative interpretation would benefit from equivalent-circuit modelling and systematic comparison of parameters such as bulk resistance and interfacial resistance. Independent ionic-conductivity measurements under controlled temperature and humidity conditions would also help establish the relationship between DES composition, agarose content, and ion transport. Such measurements would allow the electrochemical behaviour of A50 and A100 to be compared more quantitatively.

5. Thermal and long-term stability

Although thermal analysis provides information about the mass-loss behavior of the eutectogels, long-term stability under practical storage and operating conditions remains to be established. In particular, the presence of moisture can be important for DES-containing materials because water uptake may alter hydrogen-bonding interactions, viscosity, ionic mobility, and the overall gel structure. Future studies should therefore investigate storage stability under controlled temperature and relative humidity conditions. Repeated heating–cooling cycles and prolonged mechanical or electrochemical operation could also be used to determine whether the material retains its structure and functional properties over time.

(5) FUTURE INSIGHTS

Future research on agarose–proline eutectogels can focus on systematically connecting formulation, structure, and performance. First, intermediate agarose ratios between and beyond the A50 and A100 compositions could be investigated to determine whether a specific composition provides an optimal combination of mechanical strength, viscosity, thermal stability, and ionic mobility. The effect of DES composition could also be examined by varying the hydrogen-bond donor or acceptor components while maintaining comparable preparation conditions.

Second, a combination of spectroscopic, thermal, microscopic, and computational techniques could be used to clarify the molecular and structural basis of gel formation. FTIR and DSC could help examine polymer–DES interactions and thermal transitions, while SEM or related imaging techniques could provide information about the morphology of the polymeric network. Molecular

dynamics simulations could further help explain how proline and glycerol interact with agarose chains and how these interactions may influence the transport of mobile ionic species.

Third, the practical performance of these eutectogels should be evaluated for application-specific requirements. For pharmaceutical applications, future studies could investigate drug loading, drug–DES compatibility, release behavior, biocompatibility, and stability. For electrochemical applications, ionic conductivity, electrochemical stability, cycling behavior, and electrode compatibility would require systematic investigation. The biodegradability and environmental profile of the complete agarose–DES formulation should also be considered rather than evaluating the DES or polymer independently.

Overall, the A50 and A100 results demonstrate that changing the agarose ratio provides a practical route for tuning the properties of proline eutectogels. The contrasting rheological and electrochemical responses observed between the 1:2 and 1:1 agarose formulation highlight the importance of balancing polymer-network strength with ionic mobility. Further systematic optimization and structural characterization could provide a stronger basis for designing agarose–DES eutectogels with properties tailored to specific pharmaceutical, sensing, or electrochemical applications.

(7) CONCLUSION

In the present study, agarose-based eutectogels were successfully prepared using a proline (1:3) deep eutectic solvent (DES) as the liquid phase. Eutectogel formation was achieved by heating the agarose–DES mixtures to approximately 75 °C followed by cooling to room temperature, resulting in stable gel networks. The incorporation of DES into the agarose matrix combines the physicochemical characteristics of DESs with the structural properties of polymeric gels.

Thermal analysis indicated characteristic mass-loss regions associated with the removal of volatile/moisture components followed by degradation of the DES–polymer system. Rheological and mechanical characterization demonstrated the viscoelastic nature of the eutectogels, with both elastic and viscous contributions to their behavior. The shear- and strain-dependent responses further indicated the influence of agarose concentration and agarose–DES interactions on the structural stability and resistance of the gels to deformation.

Electrochemical studies demonstrated measurable electrochemical responses associated with mobile ionic species within the polymeric network. Cyclic voltammetry showed that the A50 eutectogel exhibited a higher current at +1 V (3.7 μA) than the A100 eutectogel (0.93 μA), suggesting comparatively greater mobility of electrochemically active ionic species in the lower agarose-content formulation. The reduction in current with increasing agarose content may be associated with a denser polymeric network and stronger agarose–DES interactions that restrict ionic movement. EIS measurements provided complementary information on the resistive, interfacial, and diffusion-related processes within the eutectogels.

Overall, the prepared agarose–DES eutectogels exhibited a combination of thermal stability, viscoelasticity, structural integrity, and electrochemical response. Variation in agarose concentration provides a means of tuning these properties, with the A50 eutectogel showing comparatively higher electrochemical transport, while the higher agarose content of the A100 eutectogel contributes to a more constrained polymeric network. These findings provide a basis for further investigation of agarose-based eutectogels for potential applications in drug delivery, sensing, and other pharmaceutical or electrochemical technologies.

REFERENCES

1. Prabhune, A., & Dey, R. (2023). Green and sustainable solvents of the future: Deep eutectic solvents. *Journal of Molecular Liquids*, *379*, 121676.
2. Tang, B., & Row, K. H. (2013). Recent developments in deep eutectic solvents in chemical sciences. *Monatshefte für Chemie-Chemical Monthly*, *144*(10), 1427-1454.
3. Lei, Z., Chen, B., Koo, Y. M., & MacFarlane, D. R. (2017). Introduction: ionic liquids. *Chemical Reviews*, *117*(10), 6633-6635.
4. Holbrey, J. D., & Seddon, K. R. (1999). Ionic liquids. *Clean products and processes*, *1*(4), 223-236.
5. Vanda, H., Dai, Y., Wilson, E. G., Verpoorte, R., & Choi, Y. H. (2018). Green solvents from ionic liquids and deep eutectic solvents to natural deep eutectic solvents. *Comptes Rendus. Chimie*, *21*(6), 628-638.
6. Dai, Y., Van Spronsen, J., Witkamp, G. J., Verpoorte, R., & Choi, Y. H. (2013). Ionic liquids and deep eutectic solvents in natural products research: mixtures of solids as extraction solvents. *Journal of natural products*, *76*(11), 2162-2173.
7. Smith, E. L., Abbott, A. P., & Ryder, K. S. (2014). Deep eutectic solvents (DESs) and their applications. *Chemical reviews*, *114*(21), 11060-11082.
8. Hansen, B. B., Spittle, S., Chen, B., Poe, D., Zhang, Y., Klein, J. M., ... & Sangoro, J. R. (2021). Deep eutectic solvents: a review of fundamentals and applications. *Chemical reviews*, *121*(3), 1232-1285.
9. Ferreira, C., & Sarraguça, M. (2024). A comprehensive review on deep eutectic solvents and its use to extract bioactive compounds of pharmaceutical interest. *Pharmaceuticals*, *17*(1), 124.
10. Sekharan, T. R., Chandira, R. M., Tamilvanan, S., Rajesh, S. C., & Venkateswarlu, B. S. (2022). Deep eutectic solvents as an alternate to other harmful solvents. *Biointerface Res. Appl. Chem*, *12*(1), 847-860.
11. Gajardo-Parra, N. F., Cotroneo-Figueroa, V. P., Aravena, P., Vesovic, V., & Canales, R. I. (2020). Viscosity of choline chloride-based deep eutectic solvents: experiments and modeling. *Journal of Chemical & Engineering Data*, *65*(11), 5581-5592.

12. Haghbakhsh, R., Parvaneh, K., Raeissi, S., & Shariati, A. (2018). A general viscosity model for deep eutectic solvents: The free volume theory coupled with association equations of state. *Fluid Phase Equilibria*, *470*, 193-202.
13. Reuter, D., Binder, C., Lunkenheimer, P., & Loidl, A. (2019). Ionic conductivity of deep eutectic solvents: the role of orientational dynamics and glassy freezing. *Physical Chemistry Chemical Physics*, *21*(13), 6801-6809.
14. Jurić, T., Uka, D., Holló, B. B., Jović, B., Kordić, B., & Popović, B. M. (2021). Comprehensive physicochemical evaluation of choline chloride-based natural deep eutectic solvents. *Journal of Molecular Liquids*, *343*, 116968.
15. Chen, Y., Chen, W., Fu, L., Yang, Y., Wang, Y., Hu, X., ... & Mu, T. (2019). Surface tension of 50 deep eutectic solvents: effect of hydrogen-bonding donors, hydrogen-bonding acceptors, other solvents, and temperature. *Industrial & Engineering Chemistry Research*, *58*(28), 12741-12750.
16. Kovács, A., Neyts, E. C., Cornet, I., Wijnants, M., & Billen, P. (2020). Modeling the physicochemical properties of natural deep eutectic solvents. *ChemSusChem*, *13*(15), 3789-3804.
17. Chen, Y., Zhao, D., Bai, Y., Duan, Y., Liu, C., Gu, J., ... & Zhang, L. (2022). Tuning refractive index of deep eutectic solvents. *Journal of Molecular Liquids*, *348*, 118031.
18. Taherzadeh, M., Haghbakhsh, R., Duarte, A. R. C., & Raeissi, S. (2020). Generalized model to estimate the refractive indices of deep eutectic solvents. *Journal of Chemical & Engineering Data*, *65*(8), 3965-3976.
19. Wang, J., Ma, C., Zhang, Z., Ge, X., Zhang, Q., & Wei, Y. (2024). Thermodynamic properties, electrochemical properties, and interaction behaviors of quaternary ammonium salts-based deep eutectic solvents. *Journal of Molecular Liquids*, *408*, 125384.
20. Hu, Y., Yang, W., Zhan, J., Pu, C., Zhong, L., & Hou, H. (2025). Eutectogels: recent advances and emerging biological applications. *Advanced Functional Materials*, *35*(36), 2425778.
21. Wang, J., Zhang, S., Ma, Z., & Yan, L. (2021). Deep eutectic solvents eutectogels: progress and challenges. *Green Chemical Engineering*, *2*(4), 359-367.

22. Mercadal, P. A., González, A., Beloqui, A., Tomé, L. C., Mecerreyes, D., Calderón, M., & Picchio, M. L. (2024). Eutectogels: the multifaceted soft ionic materials of tomorrow. *JACS Au*, *4*(10), 3744.
23. Tang, N., Jiang, Y., Zhang, H., & Hu, J. (2025). Innovative eutectogels: Designs, principles, and applications. *Nano Research*.
24. Ding, K., Ning, H., Liu, H., Zhang, X., Zhao, Y., & Zhang, S. (2025). Preparation, Properties, and Applications of Eutectogels. *Zeitschrift für anorganische und allgemeine Chemie*, *651*(10), e202500039.
25. Zhang, J., Hao, S., Yang, J., Zhang, J., Wen, J., Ren, W., ... & Sun, R. (2026). Eutectogels as versatile platforms: design strategies and application prospects. *Green Chemistry*, *28*(3), 1467-1490.
26. Joos, B., Volders, J., da Cruz, R. R., Baeten, E., Safari, M., Van Bael, M. K., & Hardy, A. T. (2020). Polymeric backbone eutectogels as a new generation of hybrid solid-state electrolytes. *Chemistry of Materials*, *32*(9), 3783-3793.
27. Sethi, O., Singla, A., Dutt, R., Singh, G., Sharma, K., Singh, I., ... & Kang, T. S. (2025). Mechanically robust, thermally stable, bio-sourced and flexible eutectogel electrolytes for MXene-based symmetric supercapacitors. *Journal of Materials Chemistry C*, *13*(37), 19271-19284.
28. Nicolau, A., Mutch, A. L., & Thickett, S. C. (2024). Applications of functional polymeric eutectogels. *Macromolecular Rapid Communications*, *45*(21), 2400405.
29. Bryant, S. J., Bryant, G., Drummond, C. J., & Greaves, T. L. (2025). Physico-chemical characterization of amino acid-based deep eutectic solvents. *Molecules*, *30*(4), 818.
30. Jiménez-Ortega, L. A., Kumar-Patra, J., Kerry, R. G., Das, G., Mota-Morales, J. D., & Heredia, J. B. (2024). Synergistic antioxidant activity in deep eutectic solvents: Extracting and enhancing natural products. *ACS Food Science & Technology*, *4*(12), 2776-2798.
31. Xia, H., Ren, M., Zou, Y., Qin, S., & Zeng, C. (2020). Novel biocompatible polysaccharide-based eutectogels with tunable rheological, thermal, and mechanical properties: the role of water. *Molecules*, *25*(15), 3314.
32. Wei, Y., Li, G., & Zhu, F. (2026). Structural, rheological, and physicochemical properties of starch-based eutectogels from diverse botanical sources. *Carbohydrate Polymers*, 125039.

33. Huo, X., Wang, J., Cong, Z., Liu, C., Cai, C., Wang, Y., ... & Niu, J. (2025). Strong and tough eutectogels with broad-range tunable mechanical properties via the hydrogen bond network-specific effect. *Advanced Functional Materials*, *35*(27), 2422464.
34. Li, S., Wu, Y., Zhang, X., Chen, D., Ma, Y., & Yang, W. (2024). Eutectogel-based strain and temperature sensor with widely tunable mechanical properties regulated by copolymerization-induced phase separation and electrostatic interaction. *Materials Today Nano*, *25*, 100440.
35. Yuan, J., Liu, Z., Yin, T., & Meng, S. (2025). A carrier-free therapeutic eutectogel: construction, physical properties, and anti-bacterial ability. *RSC advances*, *15*(34), 27652-27660.
36. Afifah, N., Sarifudin, A., Krisanti, E. A., Purwanto, W. W., & Mulia, K. (2026, February). Insights into the swelling behavior of glucomannan-based eutectogels. In *AIP Conference Proceedings* (Vol. 3380, No. 1, p. 060001). AIP Publishing LLC.
37. Li, G., Zhou, G., Jiang, R., Wang, Y., Zhang, J., Shen, J., ... & Yu, X. (2026). Engineered Graphene Nanointerfaces in Rapidly Gelable Nanocomposite Eutectogel Fibers for Wearable Strain and Glucose Sensing. *Composites Communications*, 102907.
38. Vo, T. H., Lam, P. K., Sheng, Y. J., & Tsao, H. K. (2023). Jammed microgels in deep eutectic solvents as a green and low-cost ink for 3D printing of reliable auxetic strain sensors. *ACS Applied Materials & Interfaces*, *15*(27), 33109-33118.
39. Zhang, H., Wang, P., Xu, Y., Xu, Z., Cai, L., Chen, J., ... & Li, J. (2026). Bamboo-Inspired Ultra-Strong, Recyclable, and 3D Printable Supramolecular Eutectogels for Impact Protection and Stimuli-Responsive Actuation. *Advanced Functional Materials*, *36*(48), e75646.
40. Gong, J., Chen, Y., Liu, D., Cai, H., Jiang, L., & Wang, H. (2025). Multifunctional Ag-PDA@ PCM composite eutectogel based electronic skin for personal thermal management and a machine learning assisted wearable sensor. *Journal of Materials Chemistry B*, *13*(43), 14047-14062.
41. Chiu, S. Y., Vo, T. H., Sheng, Y. J., & Tsao, H. K. (2025). Physical Eutectogel based on synergistic interactions between PEGylated DES and SiO_2 nanoparticles. *Journal of Molecular Liquids*, 128860.

42. Ho, H. P., Vo, T. H., Nguyen, T. M. T., Sheng, Y. J., & Tsao, H. K. (2026). 3D-Printed Wearable Strain Sensors Utilizing Deep Eutectic Solvents and Fumed Silica Nanoparticles. *Langmuir*, *42*(34), 25321-25333.
43. Kassahun, G., Attar, R. E., Abbas, M., & Thuau, D. (2026). Unprocessed Aloe Vera Gel as Quasi-Solid-State Electrolyte for High-Performance Organic Electrochemical Transistors. *ACS Applied Materials & Interfaces*, *18*(5), 8520-8528.
44. Zhong, Y., Lopez-Larrea, N., Alvarez-Tirado, M., Casado, N., Koklu, A., Marks, A., ... & Inal, S. (2024). Eutectogels as a semisolid electrolyte for organic electrochemical transistors. *Chemistry of Materials*, *36*(4), 1841.
45. Kelchtermans, A. S., Joos, B., De Sloovere, D., Mercken, J., Derveaux, E., Adriaensens, P., ... & Hardy, A. (2024). How polymer influences the electrochemical behavior of eutectogels for lithium batteries. *ACS Applied Polymer Materials*, *6*(1), 90-101.
46. Ruan, Y., Peng, M., Li, W., Chen, C., Qin, Z., Su, T., ... & Xie, X. (2026). High-performance conductive eutectogel synthesized by carboxymethyl cellulose-enhanced physicochemical dual network for supercapacitors and wearable electronic devices. *Carbohydrate Polymers*, 125435.
47. Li, H., Jin, Z., Zhang, M., Fu, L., Lin, B., Xu, C., & Huang, B. (2026). An antifreeze polyzwitterionic eutectogel with high conductivity and sensitivity at arctic-like conditions for multifunctional sensors and supercapacitors. *Journal of Colloid and Interface Science*, 139942.
48. Shu, L., Zhang, X. F., Miu, J., & Yao, J. (2024). Robust and highly conductive cellulose-based eutectogel for flexible electronics. *ACS Applied Polymer Materials*, *6*(22), 13785-13794.
49. Chen, W., Ma, J., Yu, D., Li, N., & Ji, X. (2024). Transparent, super stretchable, freezing− tolerant, self− healing ionic conductive cellulose based eutectogel for multi− functional sensors. *International Journal of Biological Macromolecules*, *266*, 131129.