Supplementary Information for

Deep Eutectic Solvent-Mediated Medical Dressings: The Evolution from Green Processing Solvents to Multifunctional Integrated Media

Zexin Lin, Caiyue Le, Zhixia Li, Dan Kong, Yani Guo, Yimin Sun*

Hubei Key Laboratory of Plasma Chemistry and Advanced Materials, School of Materials Science and Engineering, Wuhan Institute of Technology, Wuhan, 430205, China

Table S1 Summary of preparation processes for DES-based medical dressings and hydrogels via hydrogen bonding and supramolecular interactions.

Entry	Year	First Author	Brief description of preparation
1	2020.12.10	Qingmiao Zhang	PVA, drug components, and a choline chloride/mannose NADES were electrospun and vacuum-dried to form solid dressings via hydrogen-bonded entanglement.
2	2023.2.15	Xiangyu Sun	Chitosan heated in a lactic acid/choline chloride NADES was hydrated and vacuum-dried to form a biofilm via supramolecular hydrogen bonding.
3	2023.10.20	Xiangyu Sun	Similar to the above process, chitosan heated in a lactic acid/choline chloride DES was hydrated and vacuum-dried to form a supramolecular hydrogen-bonded biofilm.
4	2024.2.26	Z. L. Shaw	A hydrated cellulose pellicle was mixed with a choline chloride/glycerol DES and antimicrobial agents, then freeze-dried to form a eutectogel via hydrogen bonding.
5	2025.1.20	Bin Li	Resveratrol and chitosan were dissolved in a hydrated supramolecular HP-β-CD/levulinic acid DES and crosslinked overnight via supramolecular interactions.
6	2025.6.16	Jia-Yu Yang	A precursor containing a choline chloride/ethylene glycol DES, acrylic acid monomer, and viscosity modifiers (HEC/collagen) was exposed to UV light to form an ionic gel via photo-initiated free-radical polymerization.
7	2025.6.22	Jos´e M. Silva	Xylan and chitosan dissolved in an aqueous choline chloride/lactic acid DES were cast and oven-dried to form films driven by hydrogen bonding and solvent evaporation.

8	2026.3.15	Gleb Dubinenko	A lactic acid/tetraethylammonium chloride DES was directly mixed with PVA or a PVA/PVP blend to construct eutectogels via hydrogen-bonded physical crosslinking.

Table S2 Summary of preparation processes for DES-based medical dressings and hydrogels crosslinked via metal-ligand coordination.

Entry	Year	First Author	Brief description of preparation
1	2024.10.25	Ruigang Zhou	PVA, chitosan, and a natural choline chloride/ethylene glycol DES were physically crosslinked in a one-pot method driven by metal-polyphenol (CuTA) coordination.
2	2025.1.14	Ruigang Zhou	Chitosan, PVA, and a choline chloride/ethylene glycol DES containing microwave-extracted *Moringa* were mixed in one step to form a hydrogel driven by metal-polyphenol (CuTA) coordination.

Table S3 Summary of preparation processes for DES-based medical dressings and hydrogels via ionic crosslinking.

Entry	Year	First Author	Brief description of preparation
1	2024.1.26	Budiono Ujaya Putra	Cellulose nanofibers pretreated with ethylene glycol/citric acid and choline chloride/urea DESs were mixed with chloramphenicol and sodium alginate, cast, and immersed in a $CaCl_2$ solution for ionic crosslinking.
2	2025.6.25	Mohammad Hashem Hashempur	Chitosan, gallic acid-grafted gelatin (GGA), and *Nigella sativa* extracted via an ammonium acetate/lactic acid DES were mixed and dried at room temperature to form a xerogel film via hydrogen bonding.

Table S4 Summary of preparation processes for DES-based medical dressings and hydrogels fabricated via photo-initiated polymerization.

Entry	Year	First Author	Brief description of preparation
1	2024.2.11	Xiangzhen Meng	A lignocellulosic porous skeleton was immersed in a polymerizable acrylic acid/choline chloride DES and solidified via UV photo-initiated polymerization.
2	2024.8.9	Lingzhuang Meng	HEAA monomer and a photoinitiator were dissolved in a ternary choline chloride/glycerol/zinc chloride DES and solidified in a mold via UV photo-initiated free-radical polymerization.
3	2024.10.21	Gorawit Yusakul	An emulgel containing a hydrophobic acetic acid/menthol DES (loaded with extracts) and GelMA was shaped via 3D bioprinting and fully crosslinked via visible-light photo-polymerization.
4	2024.12.24	Shuai Liu	Quaternized chitosan and an acrylic acid/AMPS DES were mixed with acrylamide and cured via UV photo-initiated polymerization to form a polyelectroactive eutectogel.
5	2025.3.22	Huan Liu	An ink containing a polymerizable VP/IA/NIPAM DES, MXene, and mangiferin was formed into microneedle arrays via DLP 3D printing and subsequent UV photo-initiated crosslinking.
6	2025.5.15	Jia-Yu Yang	A precursor of a choline chloride/ethylene glycol DES, HEAA monomer, and $ZnCl_2$ was exposed to UV light to solidify via photo-initiated free-radical polymerization.
7	2025.7.15	Zijie Wang	A crosslinked carboxymethyl cellulose hydrogel was sequentially dehydrated in ethanol and soaked in a choline chloride/glycerol DES to form a smart eutectogel via solvent exchange.
8	2025.7.24	Ruigang Zhou	A sodium alginate solution and a $CaCl_2$/CuTA solution containing a choline chloride/glycerol DES were sprayed together, causing rapid gelation via simultaneous ionic crosslinking and metal-polyphenol coordination.
9	2026.4.24	Phuc Khanh Lam	A polymerizable zinc chloride/NIPAM DES with ethylene glycol and lithium chloride was cast into a mold and solidified via UV photo-initiated polymerization.

Table S5 Summary of preparation processes for DES-based medical dressings and eutectogels via chemical crosslinking and solvent exchange strategies.

Entry	Year	First Author	Brief description of preparation
1	2020.12.8	Tatum A. Bardsley	Bovine gelatin in HFIP and a choline geranate (CAGE) DES were electrospun into nanofiber scaffolds, followed by chemical crosslinking using glutaraldehyde vapor to fix the membrane.
2	2024.6.14	Lei Ma	Chitosan dissolved in a supramolecular β-cyclodextrin/lactic acid DES underwent chemical crosslinking with genipin to form films, sponges, or gels depending on the casting method.
3	2025.6.23	Hui Sun	A polymerizable sulfobetaine methacrylate/carvacrol DES with a crosslinker and photoinitiator was exposed to UV light to solidify via photo-initiated polymerization.
4	2026.1.7	Pingping Liu	Gelatin, polylysine, and waterborne polyurethane rapidly formed a hydrogel, which was then immersed in a choline chloride/glycerol DES to yield a composite eutectogel via solvent exchange.

Table S6 Summary of preparation processes for DES-based medical dressings and hydrogels constructed via dynamic covalent bonds and multi-network strategies.

Entry	Year	First Author	Brief description of preparation
1	2020.8.20	Yilei Wang	A choline chloride/mannose NADES was prepared. Dopamine and HEMA/DMAEMA monomers were added and left at 30°C to form a multi-network hydrogel via self-polymerization and hydrogen bonding.
2	2021.5.29	Yilei Wang	Sodium hyaluronate, chitosan, dopamine, and aloe vera were mixed into a choline chloride/glucose DES at 30°C, undergoing a dopamine-mediated dynamic sol-gel transition to form a hydrogel.
3	2021.9.10	Wen Li	Freeze-dried dopamine-conjugated hyaluronic acid (DASH) was immersed in a choline chloride/glucose DES for a dynamic sol-gel transition, followed by *in situ* reduction of silver nanoparticles.
4	2023.11.22	Yu Tian	A polymerizable acrylamide/choline chloride/glycerol DES and thiolated hyaluronic acid were heated with initiators to form a double-network eutectogel via simultaneous polymerization and crosslinking.
5	2023.12.12	Ting Huang	Sodium hyaluronate, botanical extracts, and polydopamine were mixed into a glucose/choline chloride DES, rapidly forming a hydrogel via a polydopamine-mediated dynamic sol-gel transition.
6	2024.11.6	Shaghayegh Vakili	A host-guest complex of dopamine-grafted gelatin and polycyclodextrin was mixed with oxidized sodium alginate and a polymerizable DADMAC/glycerol DES, followed by heating to form a supramolecular multi-network eutectogel.
7	2024.12.17	Chen Chen	A PVA solution containing a bioactive rosmarinic acid/proanthocyanidins/ethylene glycol DES and a xylitol/borax solution were mixed to form a sprayable hydrogel via rapid dynamic covalent crosslinking.

Table S7 Summary of Hydrogen Bond Acceptor (HBA) and Hydrogen Bond Donor (HBD) Constituents of Deep Eutectic Solvents in Representative Literature.

Entry	Year	First Author	Hydrogen Bond Acceptor (HBA)	Hydrogen Bond Donor (HBD)
1	2013.6.28	Sara García-Argüelles	choline chloride	1,8-octanediol
2	2019.8.26	Joana M. Silva	menthol	saturated fatty
3	2020.8.20	Yilei Wang	choline chloride	mannose
4	2020.12.8	Tatum A. Bardsley	choline bicarbonate	geranic acid
5	2020.12.10	Qingmiao Zhang	choline chloride	mannose
6	2021.5.29	Yilei Wang	choline chloride	glucose
7	2021.9.10	Wen Li	choline chloride	glucose
8	2022.9.4	Xin-Yao Ye	chlorocholine chloride	urea
9	2023.2.15	Xiangyu Sun	choline chloride	L-lactic acid
10	2023.2.22	Bayan Alkhawaja	choline chloride	malonic acid
11	2023.10.20	Xiangyu Sun	glycerol	L-lactic acid
12	2023.11.22	Yu Tian	choline chloride and Bet	AM, glucerol
13	2023.12.12	Ting Huang	choline chloride	glucose
14	2023.12.30	Manman Zheng	choline chloride	oxalic acid
15	2024.1.23	Guangrui Ma	choline chloride	chloroacetic acid and urea
16	2024.1.26	Budiono Ujaya Putra	choline chloride	urea
17	2024.2.11	Xiangzhen Meng	choline chloride	Acrylic acid

18	2024.2.26	Z. L. Shaw	choline chloride	glycerol
19	2024.6.14	Lei Ma	β-CD(β-cyclodextrin)	L-lactic acid
20	2024.8.9	Lingzhuang Meng	choline chloride	glycerol and zinc chloride
21	2024.10.21	Gorawit Yusakul	L-menthol	acetic acid
22	2024.10.25	Ruigang Zhou	choline chloride	ethylene glycol
23	2024.11.6	Shaghayegh Vakili	DADMAC	glycerol
24	2024.12.17	Chen Chen	RA and PC	ethylene glycol
25	2024.12.24	Shuai Liu	QCS	Aac and AMPS
26	2025.1.14	Ruigang Zhou	choline chloride	ethylene glycol
27	2025.1.20	Bin Li	HP-β-CD	Lev
28	2025.3.22	Huan Liu	VP	IA
29	2025.5.15	Jia-Yu Yang	choline chloride	ethylene glycol
30	2025.6.22	José M. Silva	choline chloride	lactic acid
31	2025.6.25	Mohammad Hashem Hashempur	Acrylic acid	L-lactic acid
32	2025.7.15	Zijie Wang	choline chloride	glycerol
33	2025.7.24	Ruigang Zhou	choline chloride	glycerol
34	2026.1.7	Pingping Liu	choline chloride	glycerol
35	2026.3.15	Gleb Dubinenko	Tetraethylammonium chloride	L-Lactic acid
36	2026.4.24	Phuc Khanh Lam	Zinc chloride	N-isopropylacrylamide

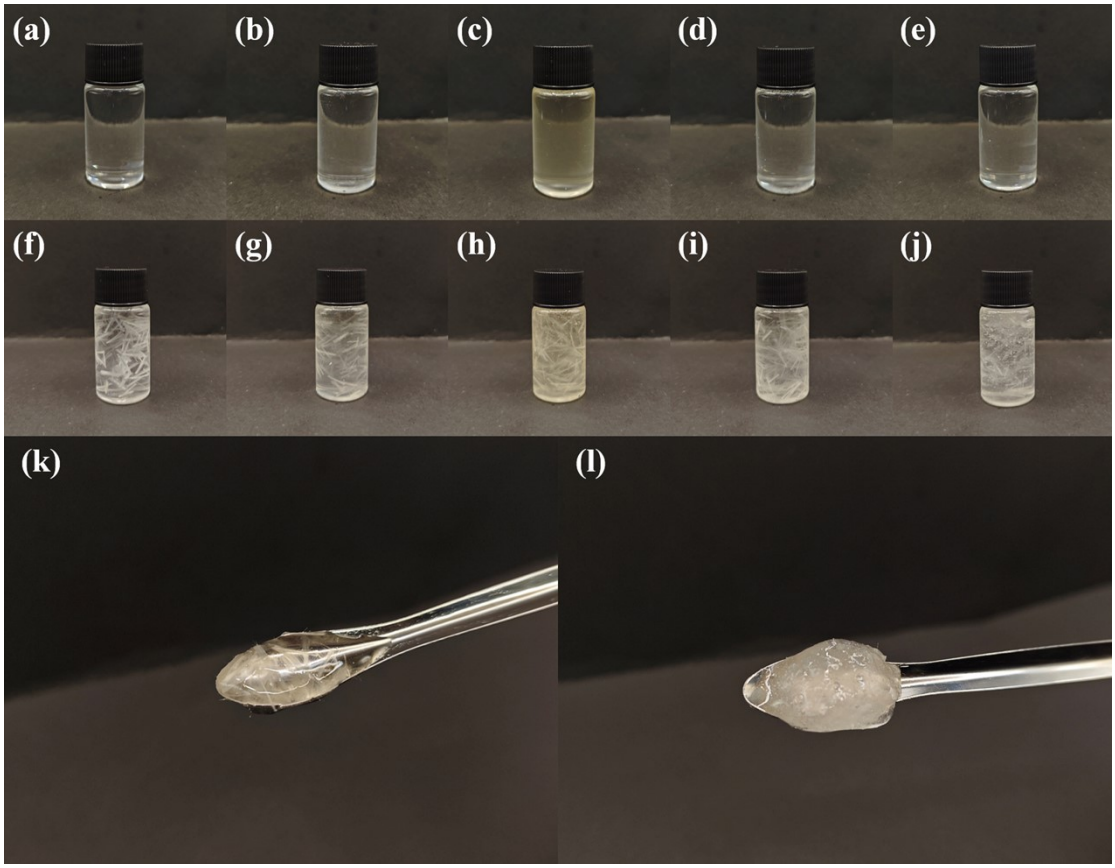

Fig. S1 Dissolution behavior and macroscopic viscous states of Lyocell fibers in distinct Deep Eutectic Solvent (DES) systems: (a–e) Optical images of pure solvent systems at 25 °C: (a) Pure glycerol (Control); (b) ChCl:Urea (1:2); (c) Betaine:Glycerol (1:2); (d) ChCl:Chloroacetic acid (1:2); and (e) ChCl:Citric acid (2:1). (f–j) Macroscopic dissolution states of 0.10 g Lyocell fibers in the corresponding solvents (a–e). (k, i) Magnified optical close-ups of representative systems: (k) The glycerol group (corresponding to f) displaying intact fiber bundles without swelling; (i) The ChCl:Citric acid group (corresponding to j) exhibiting significant swelling and transition to a transparent gel-like state, indicating macroscopic structural disintegration.