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Low-impedance tissue device interface using homogeneously conductive hydrogels chemically bonded to stretchable bioelectronics

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Intro Young's Modulus (E) hydrogels (hgels) impedance (Z)	Most stretchable devices developed have high E; high E likely means microscale nonconformal contact, which leads to higher impedance Hydrogels are promising, but there are many issues like electrical performance, material homogeneity, and integration reliability We want hgel that has low impedance, appropriate E, & high conductivity
Results (1) PEDOT:PSS (PP) polyacrylamide (PAAm) polyaniline (PANi) polyurethane (PU)	2 strategies: 1) incorporating low conc PEDOT:PSS into PAAm hgel, then decorating with PANi; inside elastomeric PU well and connected to stretchable Cr/Au/Ti electrode 2) adhesion between hgel and stretchable device by modifying PU well w/ chemical anchoring groups Resulting hgel: - conductivity in between metal filler hgels and polymer hgels - very low impedance and E (desireable for tissue)

There are generally two types of hgels: metal filled and polymer-based. Both of these have tradeoffs in terms of E, Z, and conductivity. This paper tries to create a mix of the two

Results (2)	PAAm has good E, but bad conductivity ; mixing it w/ conducting fillers usually leads to poor homogeneity
PEDOT:PSS - PAAm ↳ 2-mode hgel	In PEDOT:PSS - PAAm hgel, PEDOT:PSS particles dispersed thru PAAm (low conc.)
PEDOT:PSS - PANi - PAAm ↳ 3-mode hgel	Adding PANi to composite establishes 3D conducting pathways in hgel matrix
	homogeneous PEDOT:PSS dispersion maintained at 3.3% wt ; jumps from 20 mS/cm to ~24 S/cm w/ PANi functionalization
	Fig 2D-F : 3-mode composite shows much lower and more consistent impedance spectrum across pH & frequency, but higher E than the 2-mode hgel (fig S4)
	low impedance PANi achieved/saturates when precursor ratio of aniline (mL)/ acrylamide (g) is greater than .5 - saturates at ~1.5
	2 conducting mechanisms of 3-mode hgel : (molecular interactions) 1) HCl in aniline solution leads to secondary doping of PP 2.) PANi molecules are synthesized along dope PP network

The hgel uses PP, PAAm, and PANi. The hgel shows a very consistent impedance across frequencies and relatively lower impedance at higher pHs.

Results (3)	secondary doping - adding additional dopants into material that's already been doped
	phase separation between PEDOT & PSS; PEDOT benzoid state changes to quinoid state → 3D fibrous structures ↳ more conductive
	PANi forms uniform layer along 3D PP quinoid structures due to $\pi-\pi$ and columbic interactions
X-ray photoelectron spectroscopy (XPS)	↗ less PSS to act as insulating shell XPS reveals increased PEDOT/PSS ratios
	impedance of 3-mode hgel at higher pHs significantly lower than 2-mode (157Ω vs ~620Ω)
	From 0-300% stretching range, 3-mode resistance increased by ~39%
conductivity (κ)	hydrogel would need to be covered w/ encapsulation layer to prevent dehydration ↳ leads to E↑, stretchability↑, and κ↑

The HCl in the aniline soln leads to secondary doping of the PP, which changes it's state. The quinoid state creates a sort of scaffold for the PANi to go along with. This results in relatively homogeneous 3D conductive pathways in the hgel matrix.

Results (4)	TMSPMA treatment to anchor hydrogel to PU well; much better interfacial toughness ↳ energy needed to propagate a crack
	TA treatment to further increase interfacial toughness
	Fab for stretchable bioelectronics: Si/SiO_2 wafer → Au patterning → epoxy → PI isolation → attach tapes/films → laser cutting → remove → spray mold release agent
	cell viability tests went well
	impedance relatively stable on live rat skin with regular skin, mild burn, and severe burn
Discussion	3-mode hgel to increase conductivity, lower impedance, and lower E
	various treatment methods to increase adhesion

There were further treatments w/ TMSPMA to the PU well and TA to the hydrogel to improve adhesion properties. The resulting product is biocompatible multi-channel sensor array for Z and pH measurement in vivo/vitro