Chitosan- and Alginate-Based Conductive Gels for Electroencephalography: Comparative In Vitro Characterization of Fifteen Formulations

Ruth M. Colón Rodríguez[1]; Víctor J. Ferrer Del Valle[2]; Jairo Rondón[3]
[1,2,3]*Biomedical Engineering Department, Polytechnic University of Puerto Rico, Puerto Rico*
colon_126189@students.pupr.edu, ferrer_157751@students.pupr.edu, jrondon@pupr.edu

Abstract– Electroencephalography (EEG) relies on an effective electrode–skin interface, commonly established using conductive gels that may present limitations related to dehydration, residue, and removal. This work reports an exploratory comparative in vitro screening of conductive gels based on chitosan, sodium alginate, and chitosan–alginate hybrids, including selected formulations containing montmorillonite. Thirty-five formulations were prepared and macroscopically screened, and fifteen homogeneous gels were retained for characterization of apparent viscosity, pH, apparent contact angle, impedance magnitude, and phase angle. Interfacial measurements were performed using silicone artificial skin conditioned with a Kokubo-inspired ionic conditioning solution (SBF-I). Electrical characterization was conducted for both the gel alone and the gel–silicone configuration using frequency sweeps from approximately 20 Hz to 20 MHz, with ~20 Hz and 1 kHz selected as comparative screening points. Apparent viscosity ranged from 9,168 to 35,310 cP, pH from 3.95 to 7.07, and apparent contact angle from 61.96° to 107.91°. Gel-only impedance decreased from 85–230 kΩ at ~20 Hz to 25–80 kΩ at 1 kHz. ALG-6*, a montmorillonite-containing alginate formulation, combined the highest apparent viscosity, lowest apparent contact angle, and favorable interfacial impedance and was provisionally prioritized for further study. Replicated formulation batches, inferential statistical analysis, biological evaluation, and EEG signal-quality testing are required before clinical application.

Keywords - conductive hydrogel; chitosan; alginate; electrode–skin impedance; electroencephalography

Chitosan- and Alginate-Based Conductive Gels for Electroencephalography: Comparative In Vitro Characterization of Fifteen Formulations

Ruth M. Colón Rodríguez[1]; Víctor J. Ferrer Del Valle[2]; Jairo Rondón[3]
[1,2,3]*Biomedical Engineering Department, Polytechnic University of Puerto Rico, Puerto Rico, colon_126189@students.pupr.edu, ferrer_157751@students.pupr.edu, jrondon@pupr.edu*

Abstract*– Electroencephalography (EEG) relies on an effective electrode–skin interface, commonly established using conductive gels that may present limitations related to dehydration, residue, and removal. This work reports an exploratory comparative in vitro screening of conductive gels based on chitosan, sodium alginate, and chitosan–alginate hybrids, including selected formulations containing montmorillonite. Thirty-five formulations were prepared and macroscopically screened, and fifteen homogeneous gels were retained for characterization of apparent viscosity, pH, apparent contact angle, impedance magnitude, and phase angle. Interfacial measurements were performed using silicone artificial skin conditioned with a Kokubo-inspired ionic conditioning solution (SBF-I). Electrical characterization was conducted for both the gel alone and the gel–silicone configuration using frequency sweeps from approximately 20 Hz to 20 MHz, with ~20 Hz and 1 kHz selected as comparative screening points. Apparent viscosity ranged from 9,168 to 35,310 cP, pH from 3.95 to 7.07, and apparent contact angle from 61.96° to 107.91°. Gel-only impedance decreased from 85–230 kΩ at ~20 Hz to 25–80 kΩ at 1 kHz. ALG-6*, a montmorillonite-containing alginate formulation, combined the highest apparent viscosity, lowest apparent contact angle, and favorable interfacial impedance and was provisionally prioritized for further study. Replicated formulation batches, inferential statistical analysis, biological evaluation, and EEG signal-quality testing are required before clinical application.

***Keywords* - conductive hydrogel; chitosan; alginate; electrode–skin impedance; electroencephalography**

I. INTRODUCTION

Electroencephalography (EEG) is a noninvasive technique used to record the electrical activity generated by the cerebral cortex through electrodes positioned on the scalp. Since the first human EEG recordings [2], the technique has become widely used for the diagnosis and monitoring of neurological conditions such as epilepsy, sleep disorders, and encephalopathies, and has also supported applications in neurorehabilitation, cognitive research, and brain–computer interfaces [27]. The quality of the acquired signal depends strongly on the electrode–skin interface, where an electrically conductive medium is commonly used to reduce interfacial impedance, stabilize electrode contact, and provide a continuous ionic pathway between the electrode and the skin [13].

Conventional EEG recordings typically rely on conductive gels or pastes formulated with polymeric matrices, humectants, and ionic salts. Although these materials provide effective electrical coupling, they may pose practical limitations during prolonged monitoring, including progressive dehydration, increasing contact impedance, residue accumulation in hair, and difficulty with removal [29]. Alternative dry and semi-dry electrode technologies have been developed to reduce or eliminate the need for conductive gels; however, these systems can exhibit higher or less stable electrode–skin impedance and greater sensitivity to motion artifacts than conventional wet interfaces [14], [16]. Consequently, the development of conductive materials that maintain adequate electrical coupling while improving handling and material sustainability remains an active area of biomedical engineering research.

Hydrogels based on naturally derived polysaccharides represent a promising platform for this purpose because their hydrated polymer networks can support ionic transport while providing soft and conformable interfaces [9], [28]. Chitosan, a polysaccharide obtained by deacetylation of chitin, exhibits film-forming capability, cationic functionality, and properties that have supported extensive investigation of its use in biomedical hydrogel systems [8], [30]. Sodium alginate, derived primarily from brown algae, forms hydrated networks that can be ionically crosslinked with divalent cations and is widely used in biomedical applications for its water retention and gel-forming properties [11], [19]. When combined, chitosan and alginate can form polyelectrolyte complexes through electrostatic interactions between protonated amino groups and negatively charged carboxylate groups, producing network structures with properties that differ from those of either polymer alone [3]. Both polymers can also be obtained from renewable marine resources and processing by-products, providing additional motivation to investigate them as alternatives to conventional synthetic polymer-based materials [18], [20].

Despite the growing literature on conductive hydrogels and advanced electrodes for biopotential monitoring, comparatively few studies have systematically evaluated chitosan-, alginate-, and chitosan–alginate-based formulations specifically as conductive couplants for EEG applications. Previous work has often focused on individual formulations or on bulk material response, whereas the behavior of a candidate gel at a skin-like interface can differ substantially from its bulk response [13], [14], [29]. A comparative screening approach that integrates physicochemical properties with interfacial electrical behavior can therefore provide a more appropriate basis for selecting formulations for subsequent biological and electrophysiological

evaluation. Accordingly, this study aimed to conduct a comparative in vitro screening of chitosan-, sodium alginate-, and chitosan–alginate based conductive gels intended for further development as EEG couplants. Thirty-five candidate formulations were prepared across three material families, including selected formulations containing montmorillonite clay, and fifteen macroscopically homogeneous gels were retained for further evaluation. These formulations were compared in terms of apparent viscosity, pH, apparent contact angle, and frequency-dependent electrical response under two experimental configurations: the gel alone and the gel applied to silicone artificial skin conditioned with simulated body fluid. The objective of this screening stage was to identify formulations with the most favorable combination of physicochemical and electrical characteristics for subsequent replicated, biological, and electrophysiological testing, rather than to establish clinical safety or EEG recording performance.

A. Sustainability and Production

Chitosan and alginate provide a renewable-material basis for conductive-gel development. Chitosan can be produced from chitin recovered from crustacean-processing residues, including shrimp-shell waste, while alginate is obtained primarily from brown marine algae [6], [18], [20]. Their use does not by itself demonstrate that a complete formulation is biodegradable or environmentally preferable; extraction, purification, additives, processing energy, shelf life, and end-of-life behavior also influence sustainability. At the production level, chitin and chitosan commercialization still faces challenges associated with raw-material variability, process standardization, extraction efficiency, and scale-up [21]. These considerations are relevant to the present work because a laboratory formulation that performs adequately must also be reproducible and manufacturable before it can progress toward practical use.

B. AI-Driven Data Optimization

The present study did not use artificial intelligence to select formulations or interpret the experimental measurements. However, once replicated datasets are available, data-driven methods could support future formulation optimization by relating polymer concentration, ionic content, humectant fraction, montmorillonite content, pH, viscosity, wetting, and impedance response across multiple objectives. Recent work has described AI-assisted approaches for hydrogel design and optimization [12]. In this project, such methods are therefore considered a future development tool rather than part of the evidence used to rank the current single-batch formulations.

C. Regional Perspective

The marine origin of both polymers gives this work particular relevance to Latin America and the Caribbean, where seafood-processing residues and marine biomass may provide feedstocks for higher-value biomaterials. Shrimp-processing waste can serve as a source of chitin for subsequent conversion to chitosan [6], [20], while alginate is sourced from marine algae [18]. Developing regional purification and formulation capacity could create a pathway for local value addition and, where technically and economically feasible, reduce dependence on fully imported finished materials. The present study did not evaluate regional supply volumes, production costs, or life-cycle impacts, so these points are presented as a technology-development opportunity rather than as a demonstrated economic or environmental benefit.

D. Functional Architecture of a Conductive Gel

A conductive gel for surface biopotential recording is a multicomponent system in which each family of ingredients controls a distinct function: the polymer network defines mechanical integrity, crosslinkers stabilize it, humectants preserve hydration, salts provide mobile charge carriers, viscosity modifiers control spreadability, and additives modulate microbial stability and reinforcement [28], [5]. Montmorillonite was included because layered silicates reinforce polysaccharide networks and can raise ionic mobility along the clay galleries [22].

E. The Electrode–Gel–Skin Interface

Electrically, the recording site behaves as a series–parallel network in which the electrode–electrolyte double layer, the gel bulk, and the stratum corneum each contribute a resistive and a capacitive term. The resulting impedance is frequency dependent and largest in the low-frequency band where most EEG rhythms lie, which is why interfacial impedance and its phase serve as primary screening parameters [14], [7]; impedance mismatch between electrodes additionally converts subject movement into differential voltage, the classical origin of motion artifact [23]. Fig. 1 shows the equivalent circuit used to conceptualize the electrode–gel–skin interface.

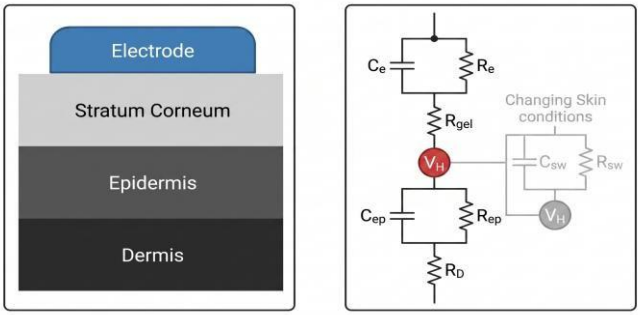


Fig 1. Equivalent circuit of the electrode–gel–skin interface used as the conceptual framework for the impedance measurements. Inspired by the model discussed in [14] and [23].

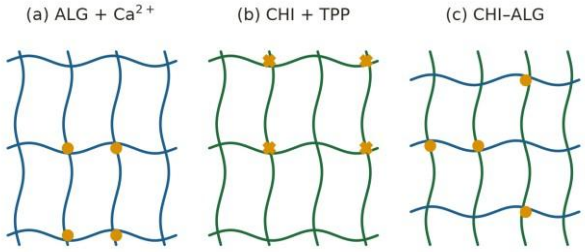


Fig 2. Schematic of the network-forming mechanisms assumed for the three families: (a) egg-box junctions between alginate chains and divalent calcium, (b) bridging of protonated chitosan chains by tripolyphosphate, and (c) electrostatic pairing of chitosan amine groups with alginate carboxylate groups in the hybrid matrix.

Fig. 2 illustrates the crosslinking strategies applied here: calcium-mediated egg-box junctions in alginate,

tripolyphosphate-mediated bridging of protonated chitosan chains, and the polyelectrolyte complex formed when both polymers are combined.

F. Commercial Reference Products

Commercial EEG couplants define a practical performance reference for a biopolymer gel. Table I compiles reference values from manufacturer documentation and previous work in the same laboratory line. The commercial products listed in the table are identified as non-biodegradable, whereas the prior HEC laboratory formulation is listed as partially biodegradable [16], [26].

TABLE I
REFERENCE VALUES FOR COMMERCIAL EEG/BIOPOTENTIAL PRODUCTS AND PRIOR LAB HEC FORMULATION

Product	Cond. (mS)	Viscosity (cP)	pH	Biodeg.
Signagel	~121	180,000–260,000	5.4–6.4	No
Spectra 360	~1,116	Low	~7.27	No
Ten20	Moderate	High (paste)	~6.5	No
NuPrep	N/A (prep)	N/A	~4.0	No
HEC 2.5% (prior lab work)	120.3	11,530	6.42	Partial

II. METHODOLOGY

A. Materials

Chitosan from shrimp shells, sodium alginate, montmorillonite K10, sodium chloride, calcium chloride, sodium tripolyphosphate, glacial acetic acid, glycerol, 1,2-propanediol, methyl hydroxyethyl cellulose (MHEC), micronized silica, polysorbate 80, and deionized water were used as received. Xanthan gum was commercially sourced as NOW Real Food Xanthan Gum Pure Powder. Commercial silicone tattoo practice sheets were used as a reproducible, non-biological substrate for comparative interfacial measurements.

A Kokubo-inspired [10] ion-containing conditioning solution was prepared using laboratory-available reagents to provide a reproducible aqueous ionic environment for conditioning the silicone substrate. The solution was not intended to reproduce the standardized Kokubo simulated body fluid- inspired (SBFI) protocol. Each 500 mL batch contained 4.0175 g NaCl, 0.1775 g $NaHCO_3$, 0.1125 g KCl, 0.0710 g Na_2HPO_4, 0.2025 g $MgCl_2 \cdot 6H_2O$, 0.1390 g $CaCl_2$, 0.0355 g Na_2SO_4, and 7.5 mL of 1 M HCl, with deionized water added to bring the final volume to 500 mL. The solution was adjusted to approximately pH 7.4 and used exclusively as an in vitro conditioning medium rather than as a physiological model of human scalp skin.

B. Research Design, Formulation Strategy, and Screening

This work was designed as an exploratory comparative in vitro screening study. Thirty-five formulations were prepared in three families: alginate only (ALG), chitosan only (CHI), and chitosan–alginate hybrids (CHIALG). Within each family, composition varied by polymer concentration, salt load, humectant fraction, presence of a trace crosslinker ($CaCl_2$ or sodium tripolyphosphate at 0.05% w/v), and presence of montmorillonite. Every batch was inspected after resting 12 to 24 h and retained only if homogeneous and free of phase separation and visible aggregates. Fifteen formulations met these criteria: ALG-1, ALG-2*, ALG-4, ALG-6*, CHI-2*, CHI-4, CHI-7, CHIALG-1 to CHIALG-4, CHIALG-6, CHIALG-9*, CHIALG-11*, and CHIALG-12*, with an asterisk denoting a montmorillonite-containing formulation. The remaining 20 batches were discarded due to incomplete dissolution, syneresis, or a gritty texture. The design was intended to prioritize candidates for subsequent replicated testing, not to establish statistically significant differences among formulations.

C. Preparation Procedure

Alginate gels were prepared by dispersing sodium alginate in deionized water under continuous mechanical stirring until a transparent viscous phase formed, followed by sequential addition of the salt solution, humectants, and viscosity modifiers. Chitosan gels were dissolved in dilute acetic acid before the same additives were incorporated, and hybrid gels were obtained by slowly combining the two dispersions under stirring to avoid immediate polyelectrolyte precipitation. When used, montmorillonite was pre-swollen in water before blending, and trace crosslinkers were added last, dropwise. All gels rested 12 to 24 h at room temperature before measurement.

D. Preparation and Conditioning of the Silicone Substrate

A modified simulated body fluid (SBF-I) inspired by the Kokubo protocol and laboratory-adjusted as an ion-containing conditioning solution for the silicone substrate was prepared. A final volume of 500 mL contained 4.0175 g of NaCl, 0.1775 g of $NaHCO_3$, 0.1125 g of KCl, 0.0710 g of Na_2HPO_4, 0.2025 g of $MgCl_2 \cdot 6H_2O$, 0.1390 g of $CaCl_2$, 0.0355 g of Na_2SO_4, and 7.5 mL of 1 M HCl. Deionized water was added to obtain a final volume of 500 mL.

The conditioning solution was used as a reproducible in vitro ionic environment and was not intended to replicate human sweat, extracellular fluid, or the scalp's complete chemical environment.

Silicone artificial-skin samples were immersed in the laboratory-prepared SBF-I at laboratory temperature for 24 h. Following conditioning, excess surface liquid was gently removed before gel deposition and characterization. The original experimental protocol explicitly treats this material as a comparative nonbiological substrate rather than a complete model of human scalp skin.

E. Characterization

Macroscopic evaluation included color, apparent homogeneity, visible aggregates, and phase separation (Fluorescence microscopy was performed using a Nikon Eclipse Ci-L microscope equipped with a Nikon DS-Qi2 digital camera). Apparent viscosity was measured on the gels using an Anton Paar ViscoQC 300-R rotational viscometer equipped with a PTD 100 Cone-Plate system and reported in centipoise (cP).

The purchase documentation confirms the ViscoQC 300R/PTD 100 configuration. The pH of each formulation was measured after preparation and stabilization using a laboratory pH meter.

Electrical characterization was performed using a Keysight E4990A impedance analyzer under two configurations: gel

alone and gel applied to SBF-I-conditioned silicone. Archived frequency sweeps extended from approximately 20 Hz to 20 MHz. Impedance magnitude $|Z|$ and phase angle were analyzed, with approximately 20 Hz and 1 kHz selected as comparative points. The ~20-Hz value represents the lowest frequency available in the archived sweep and replaces the 1Hz designation used in the original laboratory tables. Apparent contact angle (Ramé-hart Model 190) was measured using a goniometer with the gel deposited directly onto SBF-I-conditioned silicone. Ten measurements were obtained per formulation and reported as mean ± standard deviation. The values were interpreted as comparative indices of gel spreading rather than thermodynamic surface-energy measurements. The original experimental records confirm ten measurements per formulation.

F. Data Reduction and Reporting Conventions

Conductivity was not measured directly; it was derived from impedance magnitude under an assumed cell geometry held identical for every formulation, so the reported values are internally comparable indices rather than absolute material conductivities. Viscosity, pH, and impedance were determined once per formulation on a single experimental batch. Consequently, no inferential statistics were applied to comparisons among formulations. The ten contact-angle readings represent repeated measurements on the same formulation specimen and were summarized as mean ± standard deviation; they do not constitute independent formulation batches. Independent batch replication remains necessary before statistical differences among formulations can be established.

Two conventions are retained for traceability. The low-frequency columns of Tables III and IV are labeled ~20 Hz because the archived sweeps begin near 2×10^1 Hz; these entries represent the lowest-frequency points available in the archived measurements rather than measurements at 1 Hz. The ordinate labels of the archived impedance traces were corrected to kilohms and degrees in Figs. 6 and 7; the traces themselves were not modified.

G. Ethical and Environmental Considerations

The study was entirely in vitro: no human subjects, animal models, or biological tissues were involved, and no scalp application was performed; all interfacial measurements used synthetic silicone substrates conditioned with the laboratory-prepared ionic solution. Reagents were kept at laboratory scale, and acidic and salt-containing residues were neutralized before disposal according to institutional procedures. Any future biological evaluation for direct skin-contact development should follow an appropriate risk-based framework such as ISO 10993-1 [25].

III. RESULTS

A. Macroscopic Evaluation

The fifteen retained formulations showed visible differences in color, opacity, and consistency. Alginate formulations ranged from translucent to opaque, whereas montmorillonite containing alginate gels showed a light cream appearance. Chitosan and chitosan–alginate formulations ranged from translucent to pale or white gels, with visible pits or dispersed specks in selected samples. No retained formulation showed visible macroscopic phase separation at the time of characterization. Representative formulations are shown in Fig. 3.



Fig 3. Macroscopic appearance of representative retained gels: (a) ALG-6*, (b) CHI-4, and (c) CHIALG-6.

B. Viscosity, pH, and Apparent Contact Angle

Table II collects the physicochemical characteristics of the fifteen retained formulations. apparent viscosity ranged from 9,168 cp for chi-7 to 35,310 cp for ALG-6*. the four highest values were observed for ALG-6*, CHIALG-4, CHI-2*, and CHIALG-9*, three of which contained montmorillonite. because the formulations differed simultaneously in polymer composition and additives, the independent contribution of montmorillonite cannot be established from these data alone.

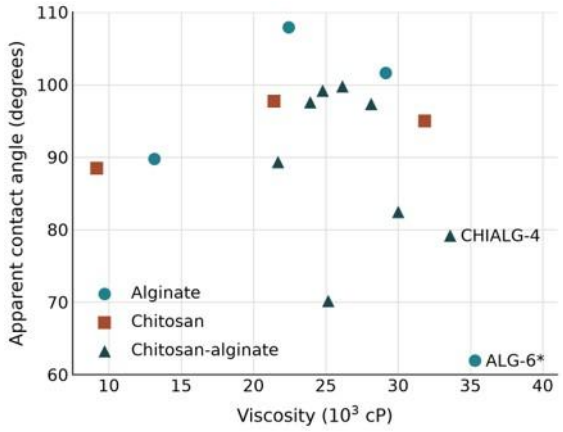


Fig 4. Apparent viscosity versus apparent contact angle for the fifteen retained gels. Points below the dashed line spread more readily on the SBF-I conditioned silicone substrate.

TABLE II
APPARENT VISCOSITY, PH, AND APPARENT CONTACT ANGLE OF THE RETAINED FORMULATIONS

Formulation	Viscosity (cP)	pH	Contact angle (°), mean ± SD, n = 10
ALG-1	22,430	5.91	107.91 ± 0.13
ALG-2*	13,130	4.28	89.80 ± 0.05
ALG-4	29,120	5.72	101.61 ± 0.14
ALG-6*	35,310	4.98	61.96 ± 0.23
CHI-2*	31,840	4.33	95.00 ± 0.00
CHI-4	21,430	4.36	97.73 ± 0.59
CHI-7	9,168	4.58	88.44 ± 0.05
CHIALG-1	21,680	4.00	89.35 ± 0.07
CHIALG-2	24,780	4.34	99.22 ± 0.06
CHIALG-3	25,150	4.16	70.18 ± 0.04

CHIALG-4	33,580	7.02	79.21 ± 0.31
CHIALG-6	26,140	7.07	99.80 ± 0.12
CHIALG-9*	29,980	4.08	82.47 ± 0.24
CHIALG-11*	28,120	4.12	97.37 ± 0.16
CHIALG-12*	23,910	3.95	97.57 ± 0.05

An asterisk denotes a montmorillonite-containing formulation.

The pH ranged from 3.95 for CHIALG-12* to 7.07 for CHIALG-6. Most chitosan-containing formulations were acidic, whereas CHIALG-4 and CHIALG-6 showed nearneutral values of 7.02 and 7.07, respectively. Apparent contact angle ranged from 61.96 ± 0.23° for ALG-6* to 107.91 ± 0.13° for ALG-1. The low standard deviations indicated limited variability among the ten measurements obtained for each formulation. ALG-6*, CHIALG-3, CHIALG4, and CHIALG-9* showed the lowest apparent contact angles and therefore the greatest relative spreading on the SBF-I-conditioned silicone substrate under the experimental conditions. ALG-6* combined the highest apparent viscosity with the lowest apparent contact angle.

C. Electrical Behavior of the Gel Alone

Table III reports impedance magnitude and phase angle for the gel-only configuration at the lowest available frequency of the archived sweeps (~20 Hz) and at 1 kHz. Impedance magnitude decreased from 85–230 kΩ at ~20 Hz to 25–80 kΩ at 1 kHz. Phase angles also shifted toward less negative values as frequency increased, indicating a greater resistive contribution at 1 kHz. At ~20 Hz, CHIALG-12* showed the lowest impedance magnitude (85 kΩ), followed by CHIALG-3, CHIALG-6, and CHIALG-9* at approximately 99 kΩ. At 1 kHz, CHI-2* showed the lowest gel-only impedance (25 kΩ), followed by CHIALG-4 and CHIALG-6 (31 kΩ), CHIALG-12* (34 kΩ), and CHIALG-3 (36 kΩ). The CHIALG-4 value was rechecked against the archived spectrum during revision and corrected from the originally transcribed 21 kΩ to 31 kΩ.

TABLE III
ELECTRICAL RESPONSE OF THE GEL ALONE AT 20 HZ AND 1 KHZ

Formulation	|Z| 20 Hz (kΩ)	|Z| 1 kHz (kΩ)	φ 20 Hz (°)	φ 1 kHz (°)
ALG-1	210	38	−48	−10
ALG-2*	210	38	−48	−10
ALG-4	230	40	−51	−21
ALG-6*	157	38	−48	−12
CHI-2*	120	25	−52	−7
CHI-4	167	62	−38	−3
CHI-7	150	80	−31	0
CHIALG-1	105	43–44	−52	−10
CHIALG-2	115	45	−45	−12
CHIALG-3	99	36	−47	−8
CHIALG-4	153	31	−55	−10
CHIALG-6	99	31	−55	−12
CHIALG-9*	99	38	−42	0
CHIALG-11*	108	40	−50	−3
CHIALG-12*	85	34	−38	−2

D. Electrical Behavior at the Artificial-Skin Interface

Table IV reports the same three quantities for the gel applied to SBF-I-conditioned silicone skin. At ~20 Hz, most formulations showed higher impedance than in the gel-only configuration. ALG-1 increased from 210 to 400 kΩ, ALG-4 from 230 to 435 kΩ, and CHIALG-4 from 153 to 350 kΩ. ALG-2* was the principal exception, with its tabulated low-frequency impedance decreasing from 210 to 135 kΩ. CHI-2* showed a smaller increase from 120 to 135 kΩ.

At 1 kHz, the lowest tabulated interfacial impedance was obtained for ALG-6* (25 kΩ), CHIALG-12* (27 kΩ), CHIALG-6 (28 kΩ), and CHIALG-11* (29 kΩ).

Phase angles remained negative at ~20 Hz for all formulations. At 1 kHz, the interface generally produced more negative phase angles than the gel-only condition, consistent with an increased capacitive contribution after introduction of the conditioned silicone substrate. No equivalent-circuit fitting was performed; therefore, individual resistive and capacitive components were not separated.

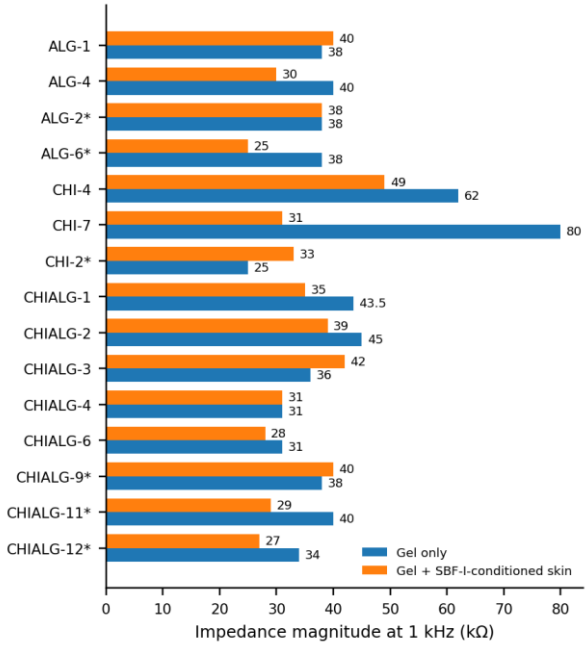


Fig 5. Impedance magnitude at 1 kHz for the gel alone and for the gel applied to SBF-I conditioned silicone artificial skin.

TABLE IV
ELECTRICAL RESPONSE OF THE GEL ON SBF-I-CONDITIONED ARTIFICIAL SKIN

Formulation	|Z| 20 Hz (kΩ)	|Z| 1 kHz (kΩ)	φ 20 Hz (°)	φ 1 kHz (°)
ALG-1	400	40	−56	−30
ALG-2*	135	38	−64	−28
ALG-4	435	30	−57	−35
ALG-6*	170	25	−56	−30
CHI-2*	135	33	−49	−13

CHI-4	259	49	−51	−21
CHI-7	182	31	−56	−28
CHIALG-1	175	35	−46	−13
CHIALG-2	150	39	−44	−12
CHIALG-3	175	42	−58	−12
CHIALG-4	350	31	−58	−40
CHIALG-6	223	28	−56	−36
CHIALG-9*	243	40	−54	−26
CHIALG-11*	187	29	−50	−19
CHIALG-12*	169	27	−52	−23

E. Frequency-Domain Behavior of Selected Formulations

The full impedance spectra provided additional information beyond the two screening frequencies. For the gel-only condition, impedance magnitude decreased steeply across the low-frequency portion of the measured range, reaching a minimum in the approximately 1–2 kHz region before increasing moderately at higher frequencies. ALG-6* decreased from approximately 155 kΩ at the lowest measured frequency to about 38 kΩ near 2 kHz. CHI-2* reached approximately 28 kΩ near 1–2 kHz, whereas the archived CHIALG-4 trace reached approximately 31 kΩ near 1 kHz. The increase observed at higher frequencies may reflect fixture, geometric, or cabling contributions that were not independently modeled.

With the SBF-I-conditioned silicone substrate, the spectra also decreased strongly at low frequency but tended to approach broader plateaus at higher frequencies. ALG-6* and CHIALG4 approached relatively stable impedance values in the tens-of kilohms range above approximately 10 kHz. This behavior is consistent with an increased resistive contribution after introduction of the substrate, although equivalent-circuit fitting would be required to assign individual electrical components. The phase spectra showed strongly negative values at the lower measured frequencies and approached zero as frequency increased. Within the measured low-frequency region, the negative phase values indicate a substantial capacitive contribution to the electrical response.

F. Comparative Evaluation and Candidate Selection

Considering the physicochemical and electrical screening together, ALG-6* showed the most balanced overall profile and was provisionally prioritized for further study. It presented the highest apparent viscosity (35,310 cP), the lowest apparent contact angle (61.96 ± 0.23°), an interfacial impedance of 25 kΩ at 1 kHz, and a pH of 4.98. Because each formulation was evaluated from a single experimental batch, this prioritization should not be interpreted as statistically demonstrated superiority.

Several secondary candidates also showed favorable characteristics. ALG-2* presented comparatively low viscosity (13,130 cP) and an interfacial impedance of 38 kΩ at 1 kHz after the archived spectrum was rechecked. CHI-2* combined high apparent viscosity with relatively low gel-only impedance, although its acidic pH would require optimization before future skin-contact evaluation. CHIALG-4 combined high viscosity, low apparent contact angle, a near-neutral pH of 7.02, and a corrected gel-only impedance of 31 kΩ at 1 kHz. CHIALG-6 and CHIALG-12* also showed comparatively low interfacial impedance values at 1 kHz but higher apparent contact angles than the best-spreading candidates.

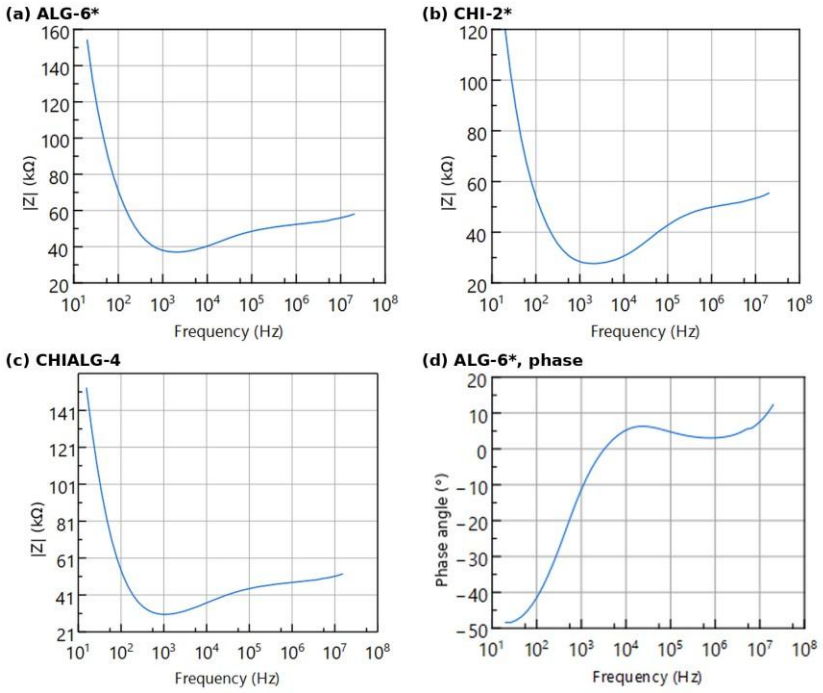


Fig 6. Impedance spectra of the gel alone, reproduced from the archived analyzer sweeps: magnitude for (a) ALG-6*, (b) CHI-2*, and (c) CHIALG-4, and (d) phase angle for ALG-6*. Ordinate labels were corrected to kilohms and degrees; the traces are unmodified.

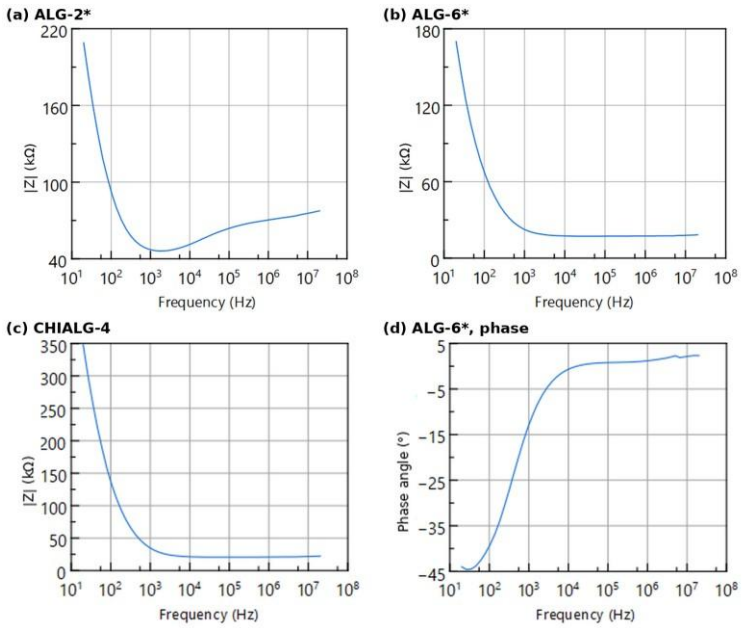


Fig 7. Impedance spectra of the gel applied to SBF-I-conditioned silicone artificial skin: magnitude for (a) ALG-2*, (b) ALG-6*, and (c) CHIALG-4, and (d) phase angle for ALG-6*. Reproduced from the archived analyzer sweeps with corrected ordinate labels.

Overall, the results demonstrate that formulation ranking depends on the combined consideration of physicochemical properties and electrical behavior under both bulk and interfacial conditions rather than on a single performance parameter.

IV. ANALYSIS AND DISCUSSION OF RESULTS

A. Principal Findings

Three main observations emerged from this screening study. First, montmorillonite-containing formulations were frequently represented among the higher-viscosity candidates, and the two montmorillonite-containing alginate gels showed favorable electrical behavior. However, because formulation components varied simultaneously, the independent contribution of montmorillonite cannot be isolated from the present data. Controlled paired formulations would be required to establish its specific effect. Second, gel-only and interfacial measurements produced different formulation rankings. For example, CHIALG-4 exhibited favorable bulk electrical behavior but a substantially higher low-frequency impedance after the introduction of the conditioned silicone substrate. These results support evaluating candidate EEG couplants under an interfacial configuration rather than relying exclusively on bulk measurements [13], [4]. Third, chitosan–alginate hybridization did not produce a uniform improvement across all properties. Nevertheless, CHIALG-4 and CHIALG-6 reached near-neutral pH values, demonstrating that selected hybrid compositions can provide physicochemical characteristics distinct from those of the individual polymer systems.

B. Position Against Published and Clinical Benchmarks

Table V compares the present interfacial results with representative values reported for EEG electrode systems. These comparisons should be interpreted with caution because the present measurements were obtained using SBF-I-conditioned silicone rather than human scalp and are therefore not directly equivalent to in vivo electrode–skin impedance.

Among the values supported consistently by the tabulated data and archived spectra, ALG-6* showed an interfacial impedance of approximately 25 kΩ at 1 kHz. This value is of the same order of magnitude as average impedances reported for conventional gel-based EEG caps, while remaining substantially lower than values reported for dry multipin systems [16]. However, it remains above the approximately 5–10 kΩ range commonly associated with conventional wet electrode–skin interfaces [14] and above impedance targets recommended for clinical EEG recording [1], [17].

Overall, the present measurements indicate that selected formulations warrant further optimization, but they should not yet be interpreted as meeting clinical electrode–skin impedance requirements.

TABLE V
INTERFACIAL IMPEDANCE OF THIS WORK AGAINST PUBLISHED VALUES AND CLINICAL REQUIREMENTS

System and configuration	|Z| (kΩ)	Source
This work: ALG-2* on SBF-I conditioned silicone skin	38 (1 kHz)	In vitro, single batch
This work: ALG-6* on SBF-I conditioned silicone skin	25 (1 kHz)	In vitro, single batch
Gel-based EEG cap, tentwenty layout, in vivo scalp	20 ± 106	[16]
Gel-based EEG cap, equidistant layout, in vivo scalp	31 ± 108	[16]
Dry multipin caps, in vivo scalp	464 ± 394; 685 ± 396	[16]
Commercial wet gel, electrode–skin	5–10	[14]

Bare skin before gel application	150–200	[14]
Double-layer conductive hydrogel electrode, 0.5–50 Hz	< 5	[29]
IFCN/ILAE minimum recording standard	< 5 preferred; < 10 acceptable	[17]
ACNS Guideline 1 upper limit	≤ 5	[1]

Published scalp values are in vivo; the present values were obtained on SBF-I conditioned silicone artificial skin and are therefore not directly equivalent.

C. Interpretation of the Frequency Response

The frequency-dependent spectra provide information not captured by the two selected screening frequencies. In the gel only configuration, the decrease in impedance magnitude and strongly negative phase at lower frequencies are consistent with a substantial capacitive contribution. The impedance minima observed near approximately 1–2 kHz indicate a frequency region in which the response becomes increasingly resistive and formulation-dependent. When the conditioned silicone substrate was introduced, several spectra developed broader impedance plateaus at higher frequencies. This behavior is consistent with an additional interfacial contribution to the overall electrical response. However, because equivalent-circuit fitting was not performed, individual resistive and capacitive elements cannot be assigned quantitatively. These observations reinforce the principal experimental finding that bulk gel measurements and gel–substrate measurements provide complementary information and should not be treated as interchangeable representations of candidate EEG-couplant performance.

D. Translation and Sustainability Considerations

The next stage of formulation development should focus on reducing interfacial impedance while preserving adequate viscosity and spreading behavior. Optimizing ionic content, polymer composition, montmorillonite dispersion, and pH is a practical route to improving the selected candidates. Technology transfer from laboratory screening to a reproducible product would also require independent batch replication, control of raw-material variability, defined mixing and crosslinking procedures, scale-up of processing, packaging and shelf-life studies, and quality specifications that can be maintained during manufacturing. Scale-up is particularly relevant for chitin/chitosan systems, for which extraction, standardization, and commercialization barriers have been reported [21].

Before direct scalp application, the selected formulations would require biological evaluation, including cytotoxicity and appropriate irritation and sensitization assessment, within a risk-based framework such as ISO 10993-1 [25]. Development toward a recording electrode or electrode-accessory application would also require consideration of applicable device performance and regulatory requirements [24]. Economic feasibility should be evaluated against commercial conductive gels only after the formulation and manufacturing process are stabilized; raw-material sourcing, purification, processing, quality control, packaging, and shelf life would all contribute to cost. No formal cost analysis was performed in this study, so economic advantage is not claimed. Likewise, although chitosan and alginate provide renewable feedstocks, biodegradability and life-cycle impact of the complete formulations were not measured. Sustainability and regional production therefore remain motivations for continued development rather than demonstrated outcomes.

E. Limitations

This study has several limitations. All measurements were performed in vitro using silicone conditioned with a Kokubo-inspired ionic conditioning solution; therefore, the substrate does not reproduce the electrical, structural, or biological complexity of human scalp skin. Although frequency sweeps from approximately 20 Hz to 20 MHz were available, the comparative analysis focused primarily on ~20 Hz and 1 kHz, and no equivalent-circuit fitting was performed. In addition, the archived sweeps do not provide measurements at 1 Hz.

Each formulation was characterized using a single experimental batch. Consequently, batch-to-batch variability and statistically significant differences among formulations could not be established. The ten apparent contact-angle measurements describe within-sample variability rather than independent formulation replicates. Additional batch replication requested during peer review could not be completed within the revision period because the materials required to reproduce the selected formulations were not available through the institution and the remaining revision time did not permit reacquisition, preparation, conditioning, and repeat characterization. Accordingly, the present work remains an exploratory single-batch screening study, and independent batch replication with inferential statistical analysis will be required in subsequent work once the necessary materials and experimental time are available.

During revision, the electrical data were re-examined against the archived impedance spectra, and two transcription errors in the originally tabulated 1-kHz impedance values were identified and corrected. For gel-only CHIALG-4, the originally reported value of 21 kΩ was corrected to 31 kΩ, consistent with the archived spectrum. For ALG-2* on SBF-I-conditioned silicone, the originally reported value of 18 kΩ was corrected to 38 kΩ, also consistent with the archived spectrum. These corrected values are used throughout the revised manuscript. Because the measurements were obtained from single experimental batches and independent batch replication could not be completed, the corrected values should still be interpreted as single-batch measurements rather than statistically validated formulation performance.

Finally, cytotoxicity, sensitization, biodegradation, drying kinetics, shelf life, and EEG signal quality were not evaluated. Consequently, the present results support candidate prioritization only and do not establish clinical safety, scalp compatibility, or EEG recording performance.

V. CONCLUSIONS, RECOMMENDATIONS, AND FUTURE WORK

Fifteen chitosan-, alginate-, and chitosan–alginate-based conductive gels, including selected montmorillonite-containing formulations, were comparatively characterized in vitro for apparent viscosity, pH, apparent contact angle, impedance

magnitude, and phase response. Apparent viscosity ranged from 9,168 to 35,310 cP, pH from 3.95 to 7.07, and apparent contact angle from 61.96° to 107.91°. Electrical measurements showed a strong frequency dependence and demonstrated that introduction of the SBF-I-conditioned silicone substrate altered both impedance magnitude and formulation ranking. Based on the combined screening criteria, ALG-6* was provisionally prioritized as the most balanced candidate, combining the highest apparent viscosity, the lowest apparent contact angle, favorable interfacial impedance, and a pH of 4.98. ALG-2*, CHI-2*, CHIALG-4, CHIALG-6, and CHIALG-12* also showed characteristics that justify further optimization.

The archived spectra showed impedance minima near approximately 1–2 kHz for the gel-only configuration and broader interfacial responses after introduction of the conditioned silicone substrate. During revision, the discrepant 1-kHz values for CHIALG-4 and ALG-2* were rechecked against the archived spectra and corrected in the manuscript; independent experimental confirmation remains pending because new batches could not be prepared within the available materials and revision time. Future work should therefore reproduce selected formulations in independent batches, apply inferential statistical analysis, extend measurements to lower frequencies including 1 Hz, perform equivalent-circuit analysis, evaluate dehydration and storage stability, optimize formulation pH, complete biological testing, and assess scale-up and comparative economic feasibility before direct scalp application or EEG signal-quality comparisons.

ACKNOWLEDGMENT

The authors acknowledge the Undergraduate Research Program for Honor and Outstanding Students (URP-HOS) at the Polytechnic University of Puerto Rico (PUPR), supported by HSI-STEM Grant P031C210139 (SAFRA III), for supporting this work.

REFERENCES

[1] American Clinical Neurophysiology Society, "Guideline 1: Minimum technical requirements for performing clinical EEG," 2008. [Online]. Available: https://www.acns.org/pdf/guidelines/Guideline-1.pdf

[2] H. Berger, "Über das Elektrenkephalogramm des Menschen," Archiv für Psychiatrie und Nervenkrankheiten, vol. 87, pp. 527–570, 1929.

[3] Y. Bu et al., "A conductive sodium alginate and carboxymethyl chitosan hydrogel doped with polypyrrole for peripheral nerve regeneration," RSC Advances, vol. 8, no. 20, pp. 10806–10817, 2018, doi: 10.1039/C8RA01059E.

[4] D. Buxi et al., "Correlation between electrode-tissue impedance and motion artifact in biopotential recordings," IEEE Sensors Journal, vol. 12, no. 12, pp. 3373–3383, 2012, doi: 10.1109/JSEN.2012.2221163.

[5] E. Caló and V. V. Khutoryanskiy, "Biomedical applications of hydrogels: a review of patents and commercial products," European Polymer Journal, vol. 65, pp. 252–267, 2015, doi: 10.1016/j.eurpolymj.2014.11.024.

[6] M. Gao, H. Tang, and H. Zhu, "Advances in extraction, utilization, and development of chitin/chitosan and its derivatives from shrimp shell waste," Comprehensive Reviews in Food Science and Food Safety, vol. 23, no. 5, e70008, 2024, doi: 10.1111/1541-4337.70008.

[7] S. Grassini, "Electrochemical impedance spectroscopy (EIS) for the in-situ analysis of metallic heritage artefacts," in Corrosion and Conservation of Cultural Heritage Metallic Artefacts, Woodhead Publishing, 2013, pp. 347–367, doi: 10.1533/9781782421573.4.347.

[8] H. Hamedi, S. Moradi, S. M. Hudson, and A. E. Tonelli, "Chitosan based hydrogels and their applications for drug delivery in wound dressings: a review," Carbohydrate Polymers, vol. 199, pp. 445–460, 2018, doi: 10.1016/j.carbpol.2018.06.114.

[9] M. Khodadadi Yazdi et al., "Polysaccharide-based electroconductive hydrogels: structure, properties and biomedical applications," Carbohydrate Polymers, vol. 278, 118998, 2022, doi: 10.1016/j.carbpol.2021.118998.

[10] T. Kokubo and H. Takadama, "How useful is SBF in predicting in vivo bone bioactivity?," Biomaterials, vol. 27, no. 15, pp. 2907–2915, 2006, doi: 10.1016/j.biomaterials.2006.01.017.

[11] K. Y. Lee and D. J. Mooney, "Alginate: properties and biomedical applications," Progress in Polymer Science, vol. 37, no. 1, pp. 106–126, 2012, doi: 10.1016/j.progpolymsci.2011.06.003.

[12] Z. Li, P. Song, G. Li, Y. Han, X. Ren, L. Bai, and J. Su, "AI energized hydrogel design, optimization and application in biomedicine," Materials Today Bio, vol. 25, 101014, 2024, doi: 10.1016/j.mtbio.2024.101014.

[13] Q. Liu, L. Yang, Z. Zhang, H. Yang, Y. Zhang, and J. Wu, "The feature, performance, and prospect of advanced electrodes for electroencephalogram," Biosensors, vol. 13, no. 1, 101, 2023, doi: 10.3390/bios13010101.

[14] M. A. Lopez-Gordo, D. Sanchez-Morillo, and F. Pelayo Valle, "Dry EEG electrodes," Sensors, vol. 14, no. 7, pp. 12847–12870, 2014, doi: 10.3390/s140712847.

[15] N. A. Morreno Sierra, "Formulación química y caracterización de un gel para electroencefalografía," M.S. thesis, Univ. de los Andes, Bogotá, 2016. [Online]. Available: http://hdl.handle.net/1992/18909

[16] C. R. Ng et al., "Multi-center evaluation of gel-based and dry multipin EEG caps," Sensors, vol. 22, no. 20, 8079, 2022, doi: 10.3390/s22208079.

[17] M. E. Peltola et al., "Routine and sleep EEG: Minimum recording standards of the IFCN and the ILAE," Clinical Neurophysiology, vol. 147, pp. 108–120, 2023, doi: 10.1016/j.clinph.2023.01.002.

[18] M. M. Rahman et al., "Sources, extractions, and applications of alginate: a review," Discover Applied Sciences, vol. 6, art. 443, 2024, doi: 10.1007/s42452-024-06151-2.

[19] Y. Ren et al., "Alginate-based hydrogels mediated biomedical applications: a review," International Journal of Biological Macromolecules, vol. 279, 135019, 2024, doi: 10.1016/j.ijbiomac.2024.135019.

[20] N. Rossi, C. Grosso, and C. Delerue-Matos, "Shrimp waste upcycling: Unveiling the potential of polysaccharides, proteins, carotenoids, and fatty acids with emphasis on extraction techniques and bioactive properties," Marine Drugs, vol. 22, no. 4, art. 153, 2024, doi: 10.3390/md22040153.

[21] S. Shetranjiwalla and A. Ononiwu, "Identifying barriers to scaled-up production and commercialization of chitin and chitosan using green technologies," International Journal of Biological Macromolecules, vol. 305, 141062, 2025, doi: 10.1016/j.ijbiomac.2025.141062.

[22] X. Su and B. Chen, "Transparent, UV-proof and mechanically strong montmorillonite/alginate/Ca2+ nanocomposite hydrogel films," Applied Clay Science, vol. 165, pp. 223–233, 2018, doi: 10.1016/j.clay.2018.07.044.

[23] N. V. Thakor and J. G. Webster, "Ground electrode motion artifact in biopotential recordings," Journal of Biomedical Engineering, vol. 2, no. 4, pp. 291–294, 1980.

[24] U.S. Food and Drug Administration, "Cutaneous electrodes for recording purposes—Performance criteria for safety and performance based pathway," Guidance for Industry and Food and Drug Administration Staff, Aug. 2020. [Online]. Available: FDA.

[25] U.S. Food and Drug Administration, "Use of International Standard ISO 10993-1, Biological evaluation of medical devices—Part 1," Guidance for Industry and FDA Staff, 2023. [Online]. Available: https://www.fda.gov/regulatory-information/search-fda-guidance-documents

[26] K. Vinchurkar et al., "Advances in sustainable biomaterials: characterizations, and applications in medicine," Discover Polymers, vol. 2, art. 2, 2025, doi: 10.1007/s44347-02500014-8.

[27] L. Xiong, N. Li, Y. Luo, and L. Chen, "Sustainable development of electroencephalography materials and technology," SusMat, vol. 4, no. 2, e195, 2024, doi: 10.1002/sus2.195.

[28] J. Xu, Y.-L. Tsai, and S.-H. Hsu, "Design strategies of conductive hydrogel for biomedical applications," Molecules, vol. 25, no. 22, 5296, 2020, doi: 10.3390/molecules25225296.

[29] H. Xue et al., "Hydrogel electrodes with conductive and substrate-adhesive layers for noninvasive long-term EEG acquisition," Microsystems & Nanoengineering, vol. 9, art. 79, 2023, doi: 10.1038/s41378-023-00524-0.

[30] R. Ye et al., "Synthesis, characterization, properties, and biomedical application of chitosan-based hydrogels," Polymers, vol. 15, no. 11, 2482, 2023, doi: 10.3390/polym15112482.