# Stumbling-to-Fetters mechanism and Virginia Creeper model in hydrogel for designing bionic cardiovascular system

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## Abstract

Manufacturing hydrogels with identical electrochemical properties are typically riddled with unresolved inquiries and challenges. Here, we utilized ultra-light graphene flakes to trace the influence of convection phenomena during reactions on hydrogels' formation and structural non-uniformity, elucidating its mechanisms. Furthermore, we confirmed that an external electric field induced the orientation of functional groups of hydrogels along the direction of this field, revealing the mechanism of its influence on the structural non-uniformity and electrochemical properties of hydrogels. Additionally, we discovered that ion diffusion was Stumbling-to-Fetters by the functional groups on the polymer chains within the hydrogel, unveiling this mechanism and developing the Virginia Creeper (VC) model for hydrogels. We demonstrated the scalability and application of the VC model. Furthermore, we proposed a molecular-ion diffusion and current decay equation to describe the electrochemical properties of hydrogels. As an application of the VC model, we developed a bionic cardiovascular system and proved its potential to seamlessly interface with living organisms and generate bio-like bioelectricity. Our findings provide novel insights into triboelectricity and guidance for producing hydrogels with identical electrochemical properties, and offer a new pathway for bioelectric generation and the design of new hydrogel devices.

## Full Text

## Stumbling-to-Fetters Mechanism and Virginia Creeper Model in Hydrogel for Designing Bionic Cardiovascular Systems

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**Abstract**

Manufacturing hydrogels with reproducible electrochemical properties remains fraught with unresolved challenges. Here, we employed ultra-light graphene flakes to trace convection phenomena during hydrogel formation, elucidating their influence on structural non-uniformity. We further demonstrated that external electric fields induce orientation of hydrogel functional groups along the field direction, revealing their impact on structural heterogeneity and electrochemical properties. Additionally, we discovered that ion diffusion is "Stumbling-to-Fetters"by functional groups on polymer chains within the hydrogel, unveiling this mechanism and developing the Virginia Creeper (VC) model for hydrogels. We demonstrated the scalability and application of the VC model and proposed a molecular-ion diffusion and current decay equation to describe hydrogel electrochemical properties. As a proof-of-concept application, we developed a bionic cardiovascular system and validated its potential for seamless interfacing with living organisms and generation of bio-like bioelectricity. Our findings provide novel insights into triboelectricity, guidance for producing hydrogels with reproducible electrochemical properties, and a new pathway for bioelectric generation and hydrogel device design.

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**Introduction**

Hydrogels have emerged as quintessential mimics of living tissues, attributed to their remarkable water content and retention, unparalleled flexibility, eminent biocompatibility, and microstructures that echo the extracellular matrix [**?**, **?**]. Nonetheless, widespread hydrogel applications remain hampered by challenges encompassing electrochemical stability, electrical conductivity, biocompatibility, controllable degradation, interface adhesion, and scalability of production and integration [**?**]. Among these, electrochemical stability is pivotal for facile

module replacement in prolonged applications spanning wearable devices, flexible electronics, biomedical implants, prostheses, drug delivery systems, and medical dressings [**?**, **?**].

While producing hydrogels with identical mechanical properties has been achieved [**?**], reproducing electrochemical performance is generally considered unattainable and remains an evasive topic [**?**]. Hitherto, the intricacies of mechanisms underpinning disparities in hydrogel electrochemical behavior remain enigmatic [**?**]. Unraveling this enigma is imperative for surmounting core technical barriers such as interface compatibility, stability and reliability of synergistic outputs, and replaceability of integrated assemblies. This endeavor is not only pivotal for cost-effective manufacture of hydrogels with reproducible properties but also crucial for their seamless amalgamation with electronic devices or biological matrices.

Free radical polymerization is ubiquitously utilized to synthesize hydrogels [**?**]. However, hydrogels fabricated through this technique frequently exhibit electrochemical properties that are challenging to reproduce, a phenomenon that is largely inevitable. We speculate this arises primarily from internal structural inhomogeneity, which may impact ion and water molecule diffusion within the hydrogel, thereby affecting electrochemical performance consistency.

To explore structural non-uniformity, we employed the polyacrylamide hydrogel system as a representative example. Through free radical polymerization, we synthesized various types of ionic polyacrylamide hydrogels (e.g., PAM(H-NaCl), PAM(L-NaCl), PAM(L-KCl)), cationic polyacrylamide hydrogels (e.g., PAM-ATCS, PAM-ATCS(H-KCl), PAM-ATCS(H-NaCl)), anionic polyacrylamide hydrogels (e.g., PAM-SHES, PAM-SHES(H-KCl), PAM-SHES(H-NaCl)), and their composites with reduced graphene oxide (e.g., PAM@rGO(L-NaCl), PAM@rGO(L-KCl), PAM-ATCS@rGO(H-NaCl), PAM-ATCS@rGO(H-KCl), PAM-SHES@rGO(H-NaCl), PAM-SHES@rGO(H-KCl)) under standardized conditions. We designated the hydrogel surface contacting the mold bottom as the "Bottom" surface and the opposite side as the "Top" surface. If hydrogels from the same batch exhibit similar trends but significant differences in electrochemical performance when integrated into testing circuits in "Top→Bottom" versus "Bottom→Top" sequences, this indicates structural non-uniformity in hydrogels prepared via free radical polymerization.

To validate this hypothesis, we integrated hydrogels into testing circuits in both configurations and characterized their current-time relationships (Fig. S1). Excitingly, results clearly demonstrated that currents generated in the "Top→Bottom" sequence consistently exceeded those in the "Bottom→Top" sequence, directly reflecting internal structural heterogeneity post-formation (Fig. S1A-C). Higher ion concentrations facilitated diffusion rates of ions and water molecules within the hydrogel, generating larger current values. Additionally, higher ion concentrations helped mitigate negative impacts of structural non-uniformity on electrochemical performance (Fig. S1A-C, E, F, H, I). Reduced graphene oxide (rGO) addition enhanced electronic conduc-

tivity of polyacrylamide hydrogels (Fig. S1B and C, S1J and M). However, rGO exacerbated structural non-uniformity in cationic and anionic polyacrylamide hydrogel composites, leading to further electrochemical performance discrepancies (Fig. S1E and K, S1F and N, S1H and L, S1I and O).

Notably, precursor solutions for these hydrogels were thoroughly stirred and evenly mixed. Without interfering factors, hydrogels formed from uniformly mixed precursors should possess homogeneous internal structures. However, results indicated that hydrogels prepared using different raw materials all exhibited structural non-uniformity, leading to strikingly similar trends in electrochemical performance variations (Fig. S1), suggesting additional factors influence internal non-uniformity.

Since chemical reactions invariably involve bond breaking and formation, internal temperature changes during hydrogel preparation inevitably induce molecular convection within the reaction solution. Structural and morphological non-uniformities caused by molecular convection are prevalent in material synthesis, particularly during polymer synthesis [**?**]. We therefore hypothesized that convection during chemical reaction is a key factor contributing to hydrogel structural non-uniformity, as convection may affect molecular distribution within the solution.

To better observe convection phenomena, we utilized ultra-light graphene flakes as tracers and reduced initiator quantity. Surprisingly, experimental videos clearly show that upon initiator addition, noticeable convection movements of graphene flakes occur even with slight temperature fluctuations at room temperature ($24\$\pm0.4°C$), $resultinginbriefrapidconvectionperiods(SupplementaryVideo1).Asthereactionprogresse$ °C) in hydrogel precursor solutions, affecting formation processes and internal structural uniformity.

To elucidate how molecular convection generated by local temperature gradients influences internal material structure, we maintained constant ambient temperature while systematically varying reaction solution temperatures (40, 60, 80, and 100 °C) in multi-physics simulations. Reported results indicate that in free radical polymerization, the reaction solution leads local reaction and releases heat under initiator action [**?**]. Our simulations demonstrate that even slight temperature variations from localized heating can create non-uniform local densities, consequently generating buoyancy-driven flow within the reaction vessel (Fig. S2A). These buoyancy-driven flows create convective zones clearly visible in velocity field streamlines (Fig. S2). These findings demonstrate that recirculation zones during hydrogel preparation are driven by temperature differentials between reaction solution and environment (Fig. S2), with more significant temperature differences producing denser convective zones (Fig. S2B-E).

Furthermore, as reaction solution viscosity increases during free radical polymerization, thermal convection presumably induces counter-directional orientation of functional groups on elongated hydrogel molecular chains. This orientation can alter uniform functional group distribution along hydrogel molecular chains,

leading to structural non-uniformity that results in higher currents when hydrogels are integrated in "Top→Bottom" versus "Bottom→Top" sequences (Fig. S1). Together, these findings suggest that appropriately increasing temperature differentials between reaction solution and ambient environment during hydrogel preparation can expedite reaction time and achieve better internal structural uniformity.

Considering that hydrogels are prepared within containers or molds, we hypothesized that external electric field forces may also influence non-uniform internal structure during preparation, such as from container or mold surface charges. To clarify this impact mechanism, we constructed molecular dynamics models of hydrogels subjected to external electric fields along the Z-axis (field strengths of 2$×10^{-21}, 2×10^{-12}, 2×10^{-8}, and 2×10^{-4}$ Å/mV) and systematically investigated diffusion behavior of free ions and water molecules within the hydrogel (Fig. S3A-D). Results indicate that both ions and water molecules exhibited enhanced diffusion along the Z-axis under external electric fields (Fig. S3A-D), suggesting that electric field influence leads to concentrated rather than uniform or random distributions, thereby contributing to structural non-uniformity.

More specifically, under external electric fields, sodium ions concentrated their diffusion along the Z-axis (Fig. S3E-H) with diffusion coefficients of 2.71 (2$×10^{-21}$ Å/mV), 1.80 (2$×10^{-12}$ Å/mV), 1.67 (2$×10^{-8}$ Å/mV), and 1.84 (2$×10^{-4}$ Å/mV) $cm^2\ s^{-1}$ (Fig. S3A-D). Chloride ions similarly showed concentrated diffusion along the Z-axis (Fig. S3I-L) with diffusion coefficients of 2.44 (2$×10^{-21}$ Å/mV), 1.11 (2$×10^{-12}$ Å/mV), 1.93 (2$×10^{-8}$ Å/mV), and 1.68 (2$×10^{-4}$ Å/mV) $cm^2\ s^{-1}$ (Fig. S3A-D). Water molecules primarily diffused along the Z-axis under external electric fields (Fig. S3M-P) with diffusion coefficients of 3.64$×10^{-4}$ (2$×10^{-21}$ Å/mV), 3.49$×10^{-4}$ (2$×10^{-12}$ Å/mV), 4.81$×10^{-4}$ (2$×10^{-8}$ Å/mV), and 2.91$×10^{-4}$ (2$×10^{-4}$ Å/mV) $cm^2\ s^{-1}$ (Fig. S3A-D). These results show that ion diffusion rates in hydrogels under external electric fields are significantly higher than those of water molecules.

Furthermore, Fig. S3M-P indicates that water molecules demonstrate noticeable diffusion along X/Y axes in hydrogels subject to varying intensities of external electric fields applied along the Z-axis. Additional findings suggest that while external electric fields cannot wholly control concentrated diffusion of water molecules along the field direction, they can create an environment influencing diffusion direction and speed, constraining water molecules within specific spatial trajectories (Fig. S3Q-T). As external electric field strength increases, water molecule distribution in hydrogels transitions from continuous to multi-layered, concentrating more specifically in certain spatial zones (Fig. S3Q-T). These results suggest that water molecule diffusion within the hydrogel is "Stumbling-to-Fetters" by groups on the molecular chain.

Notably, water molecule diffusion coefficients within the hydrogel were greater than in other groups at an electric field strength of 2$×10^{-8}$ Å/mV. When external electric field strength was below or above 2$×10^{-8}$ Å/mV, sodium

ion diffusion coefficients exceeded those of chloride ions. Moreover, at 2$×10^{-21}$ Å/mV, ion diffusion coefficients were greater than in other groups. These results thoroughly demonstrate that concentrated distribution of free ions and water molecules under external electric forces leads to structural non-uniformity, while also suggesting that free ion and water molecule distribution can be controlled by applying external forces (not limited to electric fields) to regulate hydrogel electrical performance during synthesis. Most importantly, our simulations confirm that external electric fields induced orientation of functional groups along hydrogel molecular chains in the field direction (Fig. S4), which undoubtedly enhanced the "Stumbling-to-Fetters" effect of molecular chain groups on ion and water molecule diffusion. Consequently, greater current generation in "Top→Bottom" versus "Bottom→Top" sequences can be attributed to this "Stumbling-to-Fetters" phenomenon (Fig. S1).

To further test whether the "Stumbling-to-Fetters" effect exists without electric field forces, we discovered anisotropic diffusion of free ions and water molecules within hydrogels in the absence of electric fields, suggesting their diffusion is not entirely unrestricted (Fig. 1A [Figure 1: see original paper]). Without electric field influence, water molecules primarily diffused along X/Y/Z axes and the XY plane within the hydrogel (Fig. 1B-D) with diffusion coefficients of $1.8$\times10^{-6}, 2.6\times10^{-6}, 1.7\times10^{-6}, and 1.2\times10^{-6}$ cm$^2$ s$^{-1}$, respectively. Sodium ions mainly diffused along X/Y/Z axes (Fig. 1B-D) with diffusion coefficients of $2.3$\times10^{-7}, 1.8\times10^{-7}, 1.6\times10^{-7}, and 3.6\times10^{-7}$ cm$^2$ s$^{-1}$, respectively. Chloride ions primarily diffused along X/Y/Z axes and the YZ plane (Fig. 1B-D) with diffusion coefficients of $5.1$\times10^{-7}, 7.6\times10^{-7}, 5.3\times10^{-7}, 2.4\times10^{-7}, and 2.2\times10^{-7}$ cm$^2$ s$^{-1}$, respectively. These results demonstrate anisotropic diffusion of free ions and water molecules, with ion diffusion rates significantly lower than water molecule rates, suggesting ion diffusion is "Stumbling-to-Fetters" by entanglement with functional groups on polymer chains even without electric field influence.

To reveal the "Stumbling-to-Fetters" mechanism, we investigated radial distribution functions (RDF) of sodium ions, chloride ions, and water molecules (Fig. 1E-H). We found strong hydrogen bonding interactions between sodium ions and water molecules, and between sodium and chloride ions (Fig. 1E). Chloride ions exhibit strong hydrogen bonds with hydrogen atoms in -$NH_2$ groups on polymer chains (Fig. 1F). Moreover, chloride ions interact with water molecules through both hydrogen bonding and van der Waals forces (Fig. 1E). These interactions suggest that water molecule and chloride ion diffusion directly influences sodium ion diffusion. We further found that water molecules and sodium ions concentrate around double-bonded oxygen atoms (-X=O) on polymer chains (Fig. 1G), where they engage in significant hydrogen bonding interactions (Fig. 1F-H). This aligns with water molecule diffusion trajectories concentrating in specific regions (Fig. 1I-L). These results indicate that diffusion of water molecules, chloride ions, and double-bonded oxygen atoms (-X=O) on polymer chains affects sodium ion diffusion. Summarizing, these findings demonstrate that functional groups on polymer chains in hydrogels form strong

hydrogen bonding interactions with free ions and water molecules, thereby exerting a "Stumbling-to-Fetters" effect on their distribution and contributing to structural non-uniformity.

To investigate whether the "Stumbling-to-Fetters" mechanism persists in different-density hydrogels, we utilized molecular dynamics simulations to calculate diffusion coefficients of sodium ions, chloride ions, and water molecules in hydrogels with varying polymer chain numbers (Fig. 1M). Results show that sodium ion and water molecule diffusion coefficients generally follow similar trends with hydrogel density changes, likely due to hydrogen bonding between sodium ions and water molecules. However, chloride ion diffusion coefficients display notably different trends (particularly from 6PAM to 8PAM and from 14PAM to 16PAM), possibly due to hydrogen bonding between chloride ions and both sodium ions/water molecules as well as hydrogen atoms in -$NH_2$ groups on polymer chains. We also studied diffusion coefficients in various directions within different-density hydrogels (Fig. S5). As hydrogel density increased, chloride ions primarily diffused along X/Y/Z axes and the XZ plane, while water molecules diffused mainly along X/Y/Z axes. Sodium ion diffusion in various directions showed no clear consistent trend with increasing density, potentially due to influences from water molecules, chloride ions, and double-bonded oxygen atoms (-X=O) on polymer chains (Fig. 1E-H). Irregular diffusion radar charts for water molecules, chloride ions, and sodium ions illustrate that their diffusion in specific directions is significantly influenced by hydrogel density (Fig. S5). These findings suggest the "Stumbling-to-Fetters" mechanism by which functional groups impede free ion and water molecule diffusion remains present across densities.

A bold conjecture emerged: hydrogels composed only of polymer and water molecules, without free ions, still exhibit current signals (Fig. S1D, G). We questioned whether this current arises from the "Stumbling-to-Fetters" mechanism. To explore this, we employed molecular dynamics simulations to study water molecule diffusion coefficients in different-density hydrogels composed solely of polymer and water molecules (Fig. S6) and traced water molecule diffusion trajectories (Fig. 1N and Supplementary Video 3). Results indicate that as water molecules shuttle between polymer chains, they interact with and cause vibrations in charged groups on polymer chains (Fig. 1O). These charged group vibrations facilitate current signal generation in hydrogels composed solely of polymer and water molecules, aligning with established conditions for current generation reported in literature [**?**].

Additionally, our results illustrate that overall water molecule diffusion coefficient changes with hydrogel density, decreasing as density increases (Fig. S6A-I). This suggests that increasing hydrogel density augments the number of functional groups (-X=O) capable of establishing hydrogen bonds with water (Fig. S7A). Consequently, this reduces water molecule diffusion coefficients and decreases entanglement interaction likelihood between functional groups and water molecules, ultimately diminishing current. Since PAM-ATCS hydrogels have

fewer -X=O groups capable of forming hydrogen bonds with water compared to PAM-SHES hydrogels (Fig. S7B), the probability of "Stumbling-to-Fetters" interactions is higher in PAM-ATCS, resulting in higher currents generated during testing in both "Top→Bottom" and "Bottom→Top" sequences compared to PAM-SHES hydrogels. These results demonstrate that hydrogels containing only polymer molecules and water can indeed produce current signals through the "Stumbling-to-Fetters" mechanism.

Based on these results, we developed the Virginia Creeper (VC) model for hydrogels with electrochemical properties (Fig. 1P) and derived a molecular-ion diffusion and current decay equation to describe hydrogel electrochemical properties. By fitting data from Fig. S1, we found that current-time relationships for various hydrogel types consistently adhere to the following expression (Eq. 1):

$$y = A_1 \exp(-\frac{x}{t_1}) + A_2 \exp(-\frac{x}{t_2}) + A_3 \exp(-\frac{x}{t_3}) + y_0 \quad (1)$$

Combining our proven conclusions with the Boltzmann distribution equation, we derived that the current-time relationship of hydrogels conforms to the following molecular-ion diffusion and current decay equation (Eq. 2):

$$I(T,t) = n_{\text{cation}} Q_{\text{cation}} S v_{\text{cation}}^2 \exp\left(-\frac{\beta_{\text{cation}} \pi D_{\text{cation}}}{kTt}\right) + n_{\text{anion}} Q_{\text{anion}} S v_{\text{anion}}^2 \exp\left(-\frac{\beta_{\text{anion}} \pi D_{\text{anion}}}{kTt}\right) + n_{\text{hydrone}} Q$$

$$\beta = \alpha - 1 \quad (3)$$

$$\gamma = \alpha + 1 \quad (4)$$

Where $I(T,t)$ represents the total current density from cations, anions, and functional groups. Additionally, $k$, $\pi$, $T$, $t$, and $m$ are the Boltzmann constant, circular constant, temperature, time, and relative molecular mass, respectively. $I_0$ is the initial charge density and $v$ expresses the average rate. Moreover, $Q_{\text{cation}}$, $Q_{\text{anion}}$, and $Q_{\text{hydrone}}$ express the valence of cations, anions, and functional groups, respectively. $\alpha$, $\beta$, and $\gamma$ are constants. $D_{\text{cation}}$ and $D_{\text{anion}}$ show diffusion coefficients of cations and anions, respectively, and $D_{\text{hydrone}}$ shows the oscillating coefficient of surrounding functional groups. Then, $n_{\text{cation}}$ displays the number of cations per unit volume, $n_{\text{anion}}$ displays the number of anions per unit volume, and $n_{\text{hydrone}}$ displays the number of oscillating surrounding functional groups per unit volume during "Stumbling-to-Fetters" interactions. Additionally, $v_{\text{cation}}$, $v_{\text{anion}}$, and $v_{\text{hydrone}}$ express the average rates of cations, anions, and functional groups, respectively. $S$ represents the cross-sectional area of the hydrogel.

Summarizing, we outline the main contents of the VC model as follows: (I) Generally, hydrogel structures synthesized through free radical polymerization are shaped by external electric fields and chemical reaction thermodynamics, reflecting the entangled structure typical of the VC model. (II) Electrical signals generated by hydrogels result from combined effects of free ion motion and "Stumbling-to-Fetters" interactions between functional groups on polymer chains and water molecules. (III) Electrical signals from hydrogels follow a molecular-ion diffusion and current decay equation. These findings herald a new turning point for applications requiring precise control and predictable behavior, such as precise sensing, biomedical applications, customized tissue engineering, scientific experimentation, and reproducibility of hydrogel products.

**Scalability and Application Potential of the VC Model**

To demonstrate scalability and application potential, we utilized chicken intestines and tracheae as test subjects (Fig. 2A [Figure 2: see original paper], B). Since chicken trachea inner surfaces are lined with profuse cilia and chicken intestine mucosa is covered with numerous microvilli, fluid flow can elicit motion in these structures. If the VC model can be extended, flowing pure or saline water should cause cilia and microvilli oscillation to generate current and voltage signals.

Results show that flowing pure water disturbs intestinal cilia, producing weak current signals and high voltages over 0.9 V, with signals reaching up to 1.4 V (Fig. 2C). Flowing saline water similarly disturbs cilia (Fig. 2D, Supplementary Video 4). Remarkably, flowing pure water disturbs tracheal microvilli, generating voltages above 1.4 V, with signals reaching 2.1 V (Fig. 2E). Moreover, flowing saline water disturbed tracheal microvilli, reaching over 0.21 A after 8.9 seconds (up to 0.37 A) (Fig. 2F), significantly surpassing other experimental groups (Fig. 2C-E). Recorded voltage exceeded 0.64 V (Fig. 2F, Supplementary Video 5). The chicken trachea is a spring-like structure composed mainly of cartilage rings, tapering from proximal to distal ends (Fig. 2B), with smaller inner diameter compared to intestines (Fig. 2A, B). With equivalent pump input, fluid flow rate in the trachea is larger, which may explain this phenomenon (Fig. 2C-E). Additionally, currents generated by cilia or microvilli disturbance from flowing saline water exceeded those from pure water.

These results demonstrate both scalability and application potential of the VC model, while providing new understanding of triboelectrification that can guide development of new power generation equipment (such as polymer evaporation power generation) [**?**, **?**], self-powered devices [**?**, **?**], micro-power sensors [**?**, **?**], disease prevention and diagnosis, artificial blood vessels, and artificial lungs. Most significantly, these findings suggest a novel avenue for bioelectricity generation, whereby biological systems such as esophagus, intestine, and trachea could generate bioelectricity during eating, drinking, or even sneezing.

### Bionic Cardiovascular System Application

As proof-of-concept application, we mimicked the human cardiovascular system (Fig. 3A [Figure 3: see original paper]) and developed a bionic cardiovascular system. Based on circulatory principles, we used PAM-ATCS and PAM-SHES hydrogels to construct venous and arterial vessels (Fig. 3B), respectively. High-concentration NaCl solution mimicked blood, while low-concentration NaCl solution simulated interstitial fluid. A mechanical pump replicated heart function, creating a bionic cardiovascular system (Fig. 3B). The system operated with 12 V driving voltage and 5 W power, circulating fluid at 60 mL/min, with hydrogel cardiovascular tubes having 2.5 mm inner radius. PAM-ATCS hydrogel primarily allowed chloride ion passage while inhibiting sodium ions, whereas PAM-SHES hydrogel mainly facilitated sodium ion passage while restricting chloride ions.

Superbly, results demonstrate that the bionic cardiovascular system can output currents above 0.85 A and voltages over 0.22 V (Fig. 3C, D and Supplementary Video 6), comparable to magnitudes of natural bioelectricity [**?**]. Our simulations reveal that ions in high-concentration NaCl solution can permeate through PAM-ATCS and PAM-SHES hydrogel tubes, facilitating ion exchange with low-concentration NaCl solution (Fig. 3E). Simultaneously, water molecules from low-concentration solution can pass through hydrogel tubes, diluting the high-concentration solution. Given the maintained concentration gradient introduced by the mechanical pump, the bionic cardiovascular system exhibits potential for sustained, relatively stable current and voltage output (Fig. 3C, D). These findings suggest that bionic organisms can potentially rely on their bionic cardiovascular systems to continuously generate stable bio-like bioelectricity, resembling natural biological systems.

To validate application potential and prevent rabbit mortality, we adopted an open-source approach, preventing blood from re-entering the rabbit' s body. Concurrently, rabbits were injected with saline solution to dilute blood concentration, avoiding death from excessive blood loss. Furthermore, originally separate PAM-ATCS and PAM-SHES hydrogel tubes were combined into a single hydrogel tube (Fig. 3F). The system operated with 12 V driving voltage and 5 W power, circulating fluid at approximately 6 mL/min, with hydrogel cardiovascular tube inner radius of 0.7 mm. Rabbits weighed 3 kg (whole blood volume: 55-70 mL $kg^{-1}$). As shown in Fig. 3G, when rabbit blood flowed through the bionic cardiovascular system, notable current output signals were recorded, reaching up to 5 A. This observation underscores the potential of the bionic cardiovascular system to seamlessly interface with living organisms and generate bio-like bioelectricity.

### Conclusion

This study aimed to resolve the riddle of how hydrogel structure impacts reproducible electrochemical performance. We discovered that convection phe-

nomena during chemical reactions and external electric fields influence internal structural homogeneity, leading to uneven electrochemical properties. We elucidated the "Stumbling-to-Fetters" mechanism of hydrogel functional groups on water molecule and ion diffusion, established the VC model, and proposed a molecular-ion diffusion and current decay equation to describe hydrogel electrochemical properties. Additionally, we systematically demonstrated scalability and application potential of the VC model. Our findings will provide novel insights for designing innovative hydrogel or polymer devices and offer a new avenue for developing flexible electronics, bioelectronics, soft robots, and related applications.

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**Competing interests:** Authors declare that they have no competing interests.

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**Supplementary Materials**

**Materials and Methods**
**Figs. S1-S7**
**Tables S1-S5**
**Supplementary Videos S1-S6**

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## Materials and Methods Section 1. Animal Use

Ethics statement: All protocols and surgical procedures were designed to prevent animal discomfort and suffering at any moment. These were approved by Shanghai Shengchang Biotechnology Co., LTD Experimental Animal Ethics Committee (Lunsheng Lot No. 2022-10-KQ-WXL-021), and follow guidelines provided by Fudan University Institutional Animal Care and Use Committees. Animals were housed in a pathogen-free facility, with two animals per ventilated cage, in a room maintained at 25 $\pm$ 1°C with 35-45% humidity and a 12/12-h day/night cycle. Animals had free access to food and water. At study termination, euthanasia was performed by decapitation during deep isoflurane anesthesia.

## Section 2. Synthesis of Materials

Raw materials comprised acrylamide (Am), ammonium persulfate (APS, Aladdin), N,N' -Methylenebisacrylamide (Bis, Aladdin), N,N,N' ,N' -Tetraethylethylenediamine (TEMED, Aladdin), (3-Acrylamidopropyl)trimethylammonium chloride solution (75 wt.% in $H_2O$, ATCS, Sigma-Aldrich), sodium isethionate (SHES, Aladdin), sodium chloride (Aladdin), potassium chloride (Aladdin), reduced graphene oxide (rGO, Aladdin), and deionized water.

**PAM(H-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 2.5 M sodium chloride.

**PAM(L-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.015 M sodium chloride.
**PAM(L-KCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.015 M potassium chloride.
**PAM-ATCS:** 2.8 M Am, 0.0065 M Bis, 0.088 M APS, 0.0094 M TEMED, 1.2 M ATCS.
**PAM-ATCS(H-KCl):** 2.8 M Am, 0.0065 M Bis, 0.088 M APS, 0.0094 M TEMED, 1.2 M ATCS, 2.5 M potassium chloride.
**PAM-ATCS(H-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.088 M APS, 0.0094 M TEMED, 1.2 M ATCS, 2.5 M sodium chloride.
**PAM-SHES:** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 1.4 M SHES, 2.5 M potassium chloride.
**PAM-SHES(H-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 1.4 M SHES, 2.5 M sodium chloride.
**PAM@rGO(L-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.03 mg rGO, 0.015 M sodium chloride.
**PAM@rGO(L-KCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.03 mg rGO, 0.015 M potassium chloride.
**PAM-ATCS@rGO(H-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.03 mg rGO, 1.2 M ATCS, 2.5 M sodium chloride.
**PAM-ATCS@rGO(H-KCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.03 mg rGO, 1.2 M ATCS, 2.5 M potassium chloride.
**PAM-SHES@rGO(H-NaCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.03 mg rGO, 1.4 M SHES, 2.5 M sodium chloride.
**PAM-SHES@rGO(H-KCl):** 2.8 M Am, 0.0065 M Bis, 0.0088 M APS, 0.0094 M TEMED, 0.03 mg rGO, 1.4 M SHES, 2.5 M potassium chloride.

After mixing evenly, we used a mold containing many holes with 1 cm inner diameter and 0.4 cm depth to model hydrogels by free radical polymerization.

### Section 3. Experiment of Chicken Intestines and Tracheae

A healthy chicken was purchased at a poultry market. After slaughter, intestines and trachea were removed, cleaned with deionized water, and connected separately to a circulating pump system. The pumped solution was deionized water and 1.2 M sodium chloride solution. The system operated with 12 V driving voltage and 5 W power, circulating fluid at approximately 6 mL/min.

### Section 4. Preparation of Hollow Hydrogel Tubes

Hydrogel tubes were produced using a larger-radius hard plastic tube and smaller-radius rubber hose nested to create a mold, followed by injection of the mixed hydrogel solution into the mold and subsequent demolding.

### Section 5. Experiment of Bionic Cardiovascular System

First, 100 mL sodium chloride solutions with 2.5 M and 0.015 M concentrations were prepared. Hydrogel tubes were inserted into the testing mold (24 cm), and silicone hoses were connected to both ends. The silicone hoses were then con-

nected to a Y-shaped glass tube, which was finally connected to the circulating pump system. One pump continuously pumped 2.5 M sodium chloride solution into hydrogel tubes, while another pump continuously pumped 0.015 M sodium chloride solution into the testing mold to maintain stable concentration differences inside and outside the hydrogel tube. Keithley 2400 SourceMeter testing wiring is shown in Fig. 3B. The system operated with 12 V driving voltage and 5 W power, circulating fluid at 60 mL/min, with hydrogel cardiovascular tubes having 2.5 mm inner radius.

**Section 6. Experiment of Bionic Cardiovascular System Integrated into a Hydrogel Tube**

Briefly, rabbits were anesthetized with 2% isoflurane/oxygen mixture. The bionic cardiovascular system integrated into a hydrogel tube was connected to the rabbit auricular artery. All current-voltage-time relationships were measured by Keithley 2400 SourceMeter. Note: This experiment is particularly prone to failure, especially circulating blood bursting the hydrogel tube. Blood generally cannot flow back as in living organisms. Therefore, all preparatory work must be completed before the experiment, leaving only a short testing window.

**Section 7. Molecular Dynamics Simulations**

Simulations were conducted using Material Studio software (BIOVIA). Dynamic atomistic simulation was performed as follows:

**Step 1: Building Cubic Cells.** All simulation cubic boxes were constructed using the amorphous cell module. For example, based on PAM(H-NaCl) hydrogel masses, we assumed initial PAM(H-NaCl) hydrogel contains 10 NaCl, 200 $H_2O$, and 5 hydrogel polymers. Additionally, 1PAM contains 10 NaCl, 200 $H_2O$, and 1 hydrogel polymer; 2PAM contains 10 NaCl, 200 $H_2O$, and 2 hydrogel polymers; 4PAM contains 10 NaCl, 200 $H_2O$, and 4 hydrogel polymers; and so on up to 16PAM.

**Step 2: Molecular Dynamics Simulation.** Dynamics simulations were performed at 298 K. Cells were subjected to 1,000,000 dynamic steps of 1 fs each at constant mole number, pressure, and temperature (NPT ensemble) to determine density. This was followed by a constant mole number, volume, and temperature (NVT ensemble) refinement stage of 1,000,000 dynamic steps. All molecular dynamics simulations were conducted using the Forcite module with COMPASS III force field. The electrostatic term was considered using Ewald and the van der Waals term using atom-based summation methods with 5$×10^{-19}$ kcal/mol accuracy. The repulsive cutoff for the electrostatic term was 15.5 Å. For NPT molecular dynamics simulations, Nose thermostat and Berendsen barostat were chosen. The key script example for applying electric field forces along the Z-axis is: "ChangeSettings([ ElectricFieldStrength => 1, ElectricFieldX => 0, ElectricFieldY => 0, ElectricFieldZ => 0.2, CounterElectricField => "No"]);"

**Section 8. Multi-Physics Simulations for Convection of Reaction So-**

**lutions**

Analyzing natural convection of reaction solutions involves modeling fluid flow using a "non-isothermal flow" interface. Initially, both container and reaction solution are at 313.15 K (temperatures explored sequentially: 333.15, 353.15, 373.15, 393.15 K). The surrounding environment is maintained at constant 293.15 K. Container walls have limited thickness and specific thermal conductivity. Due to rotational symmetry, we employ axisymmetric geometry to model the entire system in two dimensions. Global mass and momentum balances of non-isothermal flow are coupled with energy balance through convective and conductive heat transfer.

Assuming ideal contact between surrounding environment and container bottom, boundary conditions can be set to 293.15 K. On top and outer surfaces, convective heat flux boundary conditions are used, driven by temperature differences between container and environment. For the flow field, no-slip conditions are applied at internal boundaries (between container and reaction solution), axisymmetric conditions at the rotational axis, and slip conditions at open surfaces.

**Section 9. Multi-Physics Simulations for Bionic Cardiovascular System**

During permeation, specific components preferentially transport through hydrogel tube walls. This diffusion-driven process occurs due to concentration differences between dialysate and permeate sides within hydrogel tube walls. Solute separation is achieved through differences in molecular size and solubility, resulting in varying diffusion rates across hydrogel tube walls. This simulation focuses on saline solution transport within and through hollow hydrogel tube walls.

High-concentration saline solution flows inside the hydrogel tube, while low-concentration saline solution flows in co-current mode outside. Ions transport through hydrogel tube walls to the permeate side via diffusion as the sole mechanism. Mass transfer is modeled using the "Dilute Species Transport" interface. To analyze convective fluxes, the "Laminar Flow" interface is utilized, assuming laminar flow. Concentration discontinuities exist at hydrogel tube wall-liquid interfaces, necessitating boundary conditions on both sides.

At inlets of high- and low-concentration saline solutions, "Danckwerts" inflow conditions are set. At outlets, convection is assumed to contribute significantly more to mass transport than diffusion, modeled by outflow conditions. Symmetry applies to the leftmost boundary of this axisymmetric model geometry, with no-flux conditions at hydrogel tube wall edges and far-right boundary due to absence of material passage.

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**Supplementary Figures and Tables Fig. S1.** The current-time relationships tested from top to bottom (I ) and bottom to top (I ). (A) PAM(H-NaCl). (B) PAM(L-NaCl). (C) PAM(L-KCl). (D) PAM-ATCS. (E) PAM-ATCS(H-NaCl). (F) PAM-ATCS(H-KCl). (G) PAM-SHES. (H) PAM-SHES(H-NaCl). (I) PAM-SHES(H-KCl). (J) PAM@rGO(L-NaCl). (K) PAM-ATCS@rGO(H-NaCl). (L) PAM-SHES@rGO(H-NaCl). (M) PAM@rGO(L-KCl). (N) PAM-ATCS@rGO(H-KCl). (O) PAM-SHES@rGO(H-KCl). All fitting lines consistently adhere to Eq. 1.

**Table S1.** Parameter values of fitting curves for current-time relationships of PAM-SHES, PAM-SHES(H-KCl), and PAM(L-KCl).

| Parameter | PAM-SHES Top→Bottom | PAM-SHES Bottom→Top | PAM-SHES(H-KCl) Top→Bottom | PAM-SHES(H-KCl) Bottom→Top | PAM(L-KCl) Top→Bottom | PAM(L-KCl) Bottom→Top |
|---|---|---|---|---|---|---|
| $A_1$ | -28.87715 ± 3.52048e-4 | 8141.70414 ± 2.22782e-4 | 1.59393e-4 ± 1.00689e-4 | 2.03263e-6 ± 339.62874 | 6.67542e-5 ± 1.35131e-4 | |

**Table S2.** Parameter values of fitting curves for current-time relationships of PAM(H-NaCl), PAM(L-NaCl), and PAM-ATCS.

| Parameter | PAM(H-NaCl) Top→Bottom | PAM(H-NaCl) Bottom→Top | PAM(L-NaCl) Top→Bottom | PAM(L-NaCl) Bottom→Top | PAM-ATCS Top→Bottom | PAM-ATCS Bottom→Top |
|---|---|---|---|---|---|---|
| $A_1$ | 7.49498e-4 ± 4.14216e-4 | 3.0286e-5 ± 11574.84165 | 3.48742e-6 ± 1.63417e-5 | 9.37915e-5 ± 1.35131e-4 | | |

**Table S3.** Parameter values of fitting curves for current-time relationships of PAM-ATCS(H-KCl), PAM-ATCS(H-NaCl), and PAM-SHES(H-NaCl).

| Parameter | PAM-ATCS(H-KCl) Top→Bottom | PAM-ATCS(H-KCl) Bottom→Top | PAM-ATCS(H-NaCl) Top→Bottom | PAM-ATCS(H-NaCl) Bottom→Top | PAM-SHES(H-NaCl) Top→Bottom | PAM-SHES(H-NaCl) Bottom→Top |
|---|---|---|---|---|---|---|
| $A_1$ | -388.46386 ± 186.36764 | -378.48664 ± 1.64693e-5 | 392.41062 ± 1.71761e-4 | 9.92523e-6 ± 7.56761e-5 | -28.21981 ± 227.70433 | |

**Table S4.** Parameter values of fitting curves for current-time relationships of PAM@rGO(L-NaCl), PAM-ATCS@rGO(H-KCl), and PAM-SHES@rGO(H-KCl).

| Parameter | PAM@rGO(L-NaCl) Top→Bottom | PAM@rGO(L-NaCl) Bottom→Top | PAM-ATCS@rGO(H-KCl) Top→Bottom | PAM-ATCS@rGO(H-KCl) Bottom→Top | PAM-SHES@rGO(H-KCl) Top→Bottom | PAM-SHES@rGO(H-KCl) Bottom→Top |
|---|---|---|---|---|---|---|
| $A_1$ | 5.80245e-5 ± 4.87306e-5 | 5.59941e-4 ± 9.30346e-4 | 8.92511e-4 ± 1.35131e-4 | | | |

**Table S5.** Parameter values of fitting curves for current-time relationships of PAM-ATCS@rGO(H-NaCl), PAM-SHES@rGO(H-NaCl), and PAM@rGO(L-KCl).

| Parameter | PAM-ATCS@rGO(H-NaCl) Top→Bottom | PAM-ATCS@rGO(H-NaCl) Bottom→Top | PAM-SHES@rGO(H-NaCl) Top→Bottom | PAM-SHES@rGO(H-NaCl) Bottom→Top | PAM@rGO(L-KCl) Top→Bottom | PAM@rGO(L-KCl) Bottom→Top |
|---|---|---|---|---|---|---|
| $A_1$ | 1.62861e-4 ± 1.94762e-4 | 2.15665e-4 ± 5.74719e-5 | 5.07896e-5 ± 1.35131e-4 | | | |

**Fig. S2.** Buoyancy-driven flow induces recirculation zones in the reaction container, with velocity field at times 3, 51, and 96 seconds visualized with

streamlines. These recirculation zones are clearly seen in velocity field streamline plots. Initially, container and reaction solution are at 313.15 K (A), with sequential temperatures of 333.15 (B), 353.15 (C), 373.15 (D), and 393.15 K (E). Surrounding environment is maintained at constant 293.15 K.

**Fig. S3.** Diffusion kinetics of free ions and water molecules in hydrogels under external electric field. Diffusion coefficients of water molecules, sodium ions, and chloride ions under field strengths of 2$×10^{-21}$ (A), 2$×10^{-12}$ (B), 2$×10^{-8}$ (C), and 2$×10^{-4}$ (D) Å/mV. Diffusion coefficients of sodium ions (E), chloride ions (I), and water molecules (M) at 2$×10^{-21}$ Å/mV. Diffusion coefficients of sodium ions (F), chloride ions (J), and water molecules (N) at 2$×10^{-12}$ Å/mV. Diffusion coefficients of sodium ions (G), chloride ions (K), and water molecules (O) at 2$×10^{-8}$ Å/mV. Diffusion coefficients of sodium ions (H), chloride ions (L), and water molecules (P) at 2$×10^{-4}$ Å/mV. Three-dimensional motion trajectories of water molecules under field strengths of 2$×10^{-21}$ (Q), 2$×10^{-12}$ (R), 2$×10^{-8}$ (S), and 2$×10^{-4}$ (T) Å/mV.

**Fig. S4.** Orientation of functional groups on hydrogel molecular chains induced by external electric field. Photos of functional group orientation before (A) and after (B) electric field application. Close-up shots of functional group orientation on single hydrogel molecular chain before (C) and after (D) electric field application. Dashed circles randomly mark orientation of some functional groups under external electric field.

**Fig. S5.** Diffusion kinetics of sodium ions, chloride ions, and water molecules in hydrogels with varying polymer chain numbers. Diffusion coefficients for hydrogels with 1 (A), 2 (B), 4 (C), 6 (D), 8 (E), 10 (F), 12 (G), 14 (H), and 16 (I) polymer chains.

**Fig. S6.** Diffusion kinetics of water molecules in different-density hydrogels composed only of polymer and water molecules. Diffusion coefficients for hydrogels with 1 (A), 2 (B), 4 (C), 6 (D), 8 (E), 10 (F), 12 (G), 14 (H), and 16 (I) polymer chains.

**Fig. S7.** Number of functional groups (-X=O) capable of establishing hydrogen bonds with water. (A) Number of functional groups for different-density hydrogels. (B) Number of functional groups for PAM, PAM-ATCS, and PAM-SHES hydrogels.

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**Figure Legends** **Fig. 1.** Kinetic mechanism, "Stumbling-to-Fetters" mechanism, and Virginia Creeper model of hydrogel. (A) Diffusion coefficients of water molecules, sodium ions, and chloride ions for PAM hydrogel. (B-D) Diffusion coefficients along different axes. (E-H) Radial distribution functions of sodium ions, chloride ions, and water molecules. (F) Probability of finding water molecules, sodium ions, and chloride ions around -$NH_2$ groups. (G) Probabil-

ity around double-bonded oxygen atoms (-X=O). (H) Probability around -$CH_3$ groups. (I-L) Three-dimensional motion trajectories and projections. (M) Diffusion coefficients in hydrogels with varying polymer chain numbers. (N) Tracing example of water molecule diffusion trajectory. (O) Model of charged group vibration induced by water molecules shuttling through hydrogel mesh. Diffusion is "Stumbling-to-Fetters" by entanglement with functional groups without electric field influence. (P) Virginia Creeper model for hydrogel. Colored beads represent free ions and water molecules; rings at dendritic polymer chain ends represent functional groups.

**Fig. 2.** Scalability and application potential of the Virginia Creeper model. Experimental photos of chicken intestines (A) and tracheae (B) as test objects. Current-voltage-time relationships from cilia oscillation in chicken intestine caused by flowing pure water (C) and saline water (D). Current-voltage-time relationships from microvilli oscillation in chicken tracheae caused by flowing pure water (E) and saline water (F).

**Fig. 3.** Design, performance, mechanism, and application of bionic cardiovascular system. (A) Cardiovascular system mechanism. Heart contraction and dilation propel oxygenated blood via vascular system, facilitating metabolic waste removal and homeostasis. (B) Experimental photo of bionic cardiovascular system. Yellow solution is high-salinity; green is low-salinity (colored with edible pigments). (C) Circulate voltage-time relationship. (D) Circulate current-time relationship. (E) Multi-physics simulation of ion diffusion from high to low concentration through hydrogel tube wall. Streamlines show total flux with arrows at fixed time intervals. (F) Photo of bionic cardiovascular system integrated into hydrogel tube in living rabbit. (G) Current-time relationship of rabbit blood flowing through integrated system.

*Note: Figure translations are in progress. See original paper for figures.*