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Research Article

Egg White—a Polymer Gel Electrolyte with Exceptionally High Ionic Conductivity

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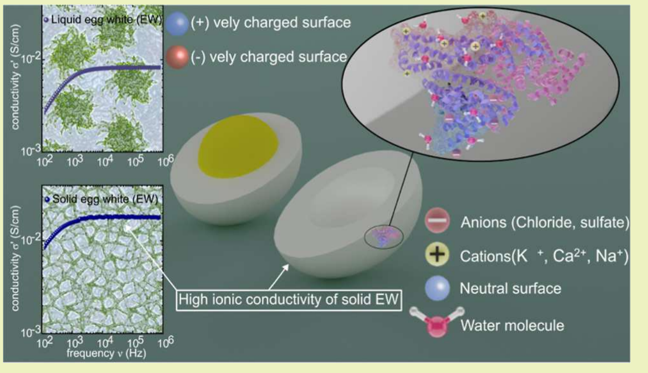
ABSTRACT: A considerable amount of research is currently focusing on mitigating the adverse impact of current energy storing electrolytes on both environment and human health, targeting the development of alternative, biologically friendly materials. In this regard, we propose a mechanically free-standing electrolyte derived simply from hen egg white. The present study reveals that the conductivity of the temperature-induced gel-like, mechanically stable structure of this biological material is truly remarkable, varying between 1 and 10 mS/cm at room temperature. Our conductivity and nuclear magnetic resonance results demonstrate that a solid ionic conductor, with a conductivity significantly higher than those displayed by many inorganic and organic electrolytes, can be attained from a cheap, green, biological source which could serve as a landmark template for future bioinspired electronics.



KEYWORDS: *biodegradable electrolyte, protein matrix, ionogel, ion conductivity, ion diffusion*

■ INTRODUCTION

Owing to their superior stability and safety properties, solid-state ionic conductors have become essential materials in the fast-growing market of electrochemical devices. However, their rapid improvement comes with the price of reducing the effective lifespan of electronics, triggering generation of e-wastes at unprecedented rates.[1] A considerable amount of research is currently focusing on mitigating their adverse impact on both environment and human health,[2] calling for the development of alternative, biologically friendly materials.[3−6] The solid-state conductors currently employed in modern devices largely fall behind these requirements for sustainability and biological compatibility. These include metal−organic frameworks, inorganic oxides, polymeric membranes, and crystalline/ceramic electrolytes which, besides their poor biodegradability issues, often fall short in terms of intrinsic ionic conductivity.

In conventional polymer-based electrolytes, for example, the mobility of ions is usually coupled with the local segmental motions resulting in poor solid-state ionic conductivity. To alleviate reduced energy densities due to low conductivity and use of separators, ionogels were proposed as an emerging class of solid electrolyte materials.[7] These are basically ionic liquids, hence exhibiting high conductivities, confined in inorganic, organic, or hybrid supramolecular frameworks providing their mechanical stability. An overview of the influence of different parameters of polymer matrices on the overall performance of polymer gel electrolytes and their role for electrode protection and thermal stability for electrochemical storage can be found in ref 8. While some recently developed polymer gel electrolytes are able to simultaneously display high conductivity, high cation transport number and large electrochemical window,[9] their high toxicity pose serious concerns toward the use of such ILs-based electrolytes.[10]

On the opposite side of environmental compatibility spectrum, proteins are natural polymers with amino acid units covalently bonded by peptide linkages. Since proteins are rather complex in terms of structure, chemical, and physical properties, they have been recently explored in designing solid-state conductors due to their multifunctional sites. Several works on protein-based ionic conductors, with the proteins serving as matrices for conducting polymers, were recently reported.[11−14] However, the ionic conductivity levels of these hybrid materials have been reported to vary around 10^{-5} S/cm.[15] Consequently, proteins triggered less appreciation despite the fact that they exhibit several advantages such as tunable physical properties, abundance of functional groups for

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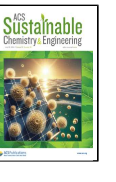

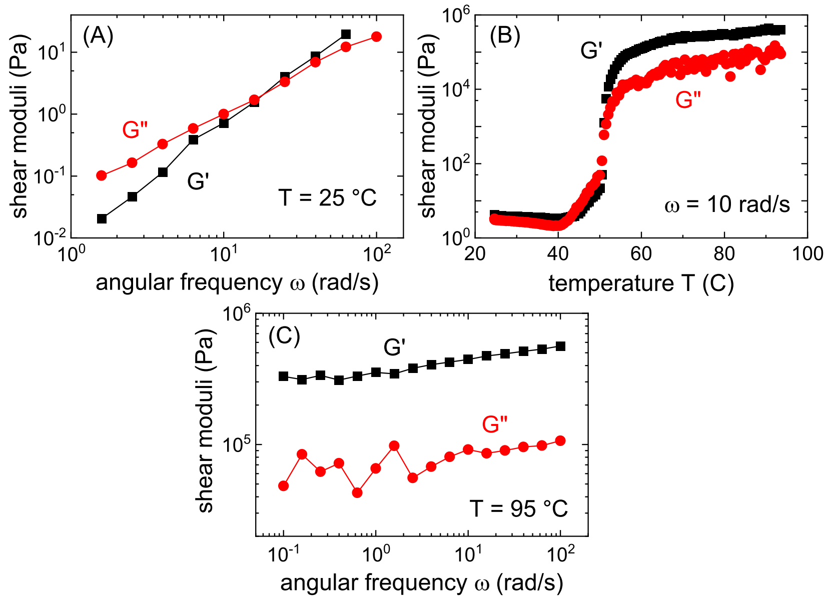

Figure 1. (A) Viscoelastic behavior of liquid EW at room temperature. (B) Shear rheological measurements (temperature sweep experiment, ω = 10 rad/s) of EW showing its solidification beyond 55 °C, where the relatively large magnitude of storage modulus value indicates a solid-like behavior. (C) The storage and loss moduli are shown to be essentially independent of frequencies at T = 95 °C, corresponding to the coagulation into a solid EW structure.

structural modification, and intrinsic biocompatibility, and their potential as ion matrices has been so far incoherently investigated. We note that besides their use in green electrochemical applications, they can alternatively offer exciting solutions in bioelectronics, signal transductions, etc.[16] For example, Gorodetsky et al. reported on a transistor consisting of reflectin (an intrinsically disordered protein)-based conductor with ionic conductivity of $\sim 10^{-4}$ S/cm at room temperature.[17] A similar approach with bovine serum albumin-based protein mat resulted in the solid-state protonic conductivity of 0.05 S/cm.[18]

In view of inherent biocompatibility of protein-based electrolytes, and complementing other recent developments of natural polymer matrices for electrochemical applications,[19] we herein propose a free-standing electrolyte derived simply from hen egg white (EW). The poultry (*Gallus domesticus*) egg is a rich and well-balanced source of essential nutrients for human diet composed of fatty acids, iron, phosphorus, trace elements, vitamins, and proteins of high biological importance. The present study revealed that the solid-state conductivity of "cooked" (a state initiated by denaturation at temperatures >60 °C) EW in its gel-like structure is rather remarkable, varying between ∼1 and 10 mS/cm at room temperature. The heat-treated specimens are mechanically solids (although relatively soft), show no sign of deterioration up to 110 °C, and their conductivity tests were highly reproducible. This study demonstrates that a solid ionic conductor, with a conductivity higher than those typically displayed by its inorganic and organic counterparts can be attained from a cheap, green, biological source which could serve as a landmark template for future bioinspired electronics.

■ EXPERIMENTAL SECTION

Fresh hen eggs were procured from a local supermarket in Lyon. The eggs were broken manually, and the EW suspension was separated from the yolk using a metallic mesh net, followed by the careful removal of the chalazae.

The conductivity measurements were performed using an Alpha-A High Performance Modular Measurement System from Novocontrol. The frequency was varied from 0.1 Hz to 10 MHz. The denatured hen EW specimens with 10 × 10 × 3 mm dimension were placed between two polished brass electrodes (thickness 2 mm, diameter 20 mm). For the EW suspensions, the sample cell was constructed from polypropylene cylinder of 14 mm length and 20 mm diameter, sealed by polished brass electrodes at both ends. The applied testing voltage was 0.1 V_{rms}. For measuring conductivities as a function of temperature the dielectric cell containing the liquid EW was inserted into a cryostat coupled with cooling (liquid N_2) and heating modules. All tests were repeated several times to ensure reproducibility and consistency.

The nuclear magnetic resonance (NMR) measurements were performed with a 400 MHz Bruker spectrometer AVANCE-III HD Nanobay, on sealed glass tubes containing uncooked and cooked EW. For the diffusivity tests we used the diffusion ordered spectroscopy (DOSY) experiment of bipolar gradient pulses with convection compensation.[20]

■ RESULTS AND DISCUSSION

The lack of fluidity of hard-boiled eggs is known to anyone with minimal cooking experience. Ovalbumin is the major protein in the EW, and much work has been conducted on its thermal aggregation and gelation.[21−25] It was found that ovalbumin behaves as a mixture of two proteins, where the amount of S-ovalbumin depends on the storage time and pH of the eggs. Native (N-) ovalbumin can be converted into Stable (S-) ovalbumin through the formation of an Intermediate (I-). All these species are able to aggregate. Ovalbumin contains four free SH groups within its native structure and, upon heating, these groups form disulfide bonds between denatured proteins.[26,27] The thermal denaturation of ovalbumin was shown to follow a first-order kinetics, and previous studies indicated that this process is primally responsible for the heat-induced aggregation (gelation) of EW.[28]

To quantify the mechanical stiffness, we performed shear rheological measurements (linear viscoelasticity) on EW during a heating scan (Figure 1, temperature sweep experi-

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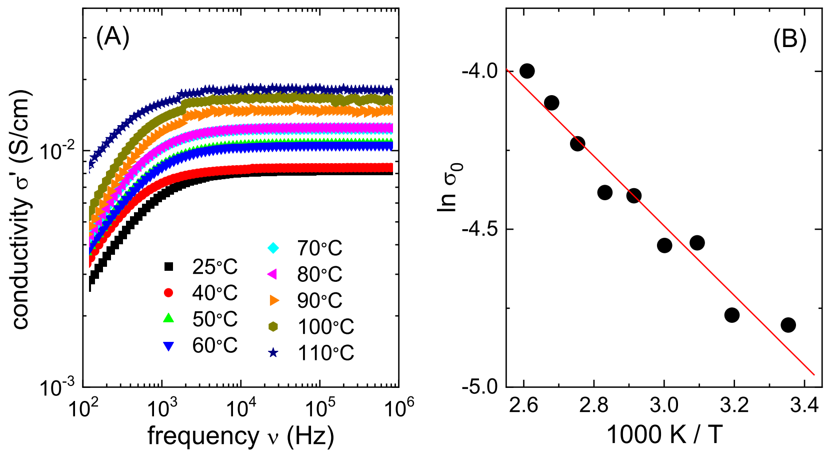

Figure 2. (A) Conductivity of cooked EW vs frequency from 25 to 110 °C. (B) Arrhenius plot of boiled EW conductivity.

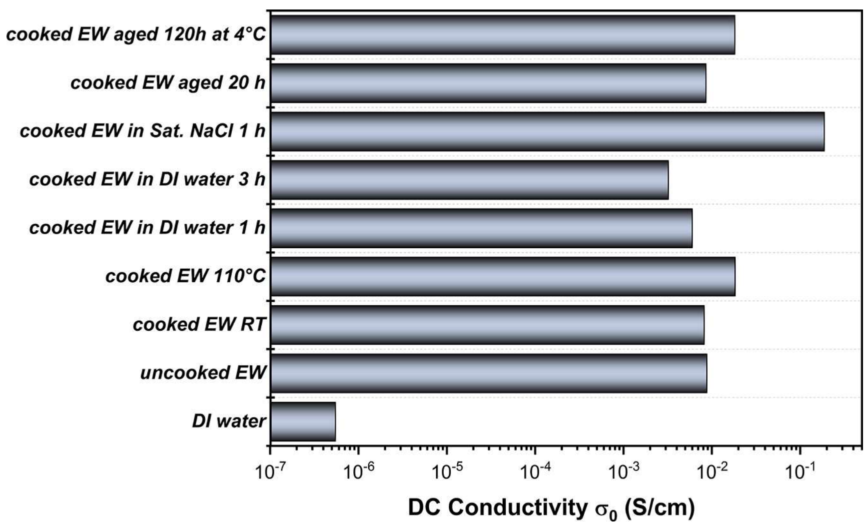

Figure 3. Room-temperature ionic conductivity of different EW samples and of DI water at 1 MHz.

ment). First, it can be noted that at room temperature the EW behaves as a viscoelastic liquid (Figure 1A). Up to 40 °C the moduli decreases upon heating, following a trivial temperature dependence. The solidification process starts at $T = 40$ °C with a strong increase of both moduli G' and G'' (Figure 1B). This is essentially a two-step process, the shear moduli initially increasing as a result of increased molecular dimensions due to unfolding of ovalbumin molecule upon heating (Figure 1B). In the second stage, molecular aggregation aided by covalent bonding and hydrophobic interactions, leads toward the formation of continuous aggregates and gelation, resembling a quasi-solid structure as confirmed in Figure 1C ($G' > G''$ over the entire range of frequencies). The gel point is usually defined as the crossover of the storage G' and the loss G'' moduli, and the corresponding temperature for the investigated EW suspension was 52 °C (Figure 1B). The storage and loss moduli appear to be independent of temperature beyond 60 °C, indicating irreversible coagulation and aggregation of the major protein ovalbumin into a quasi-solid (gel) EW structure.

Before presenting the dielectric properties of this material, we note that pristine EW suspensions contain about 90% water and a wide variety of cations (calcium, magnetism, iron, sodium, potassium, copper, and zinc) and anions (chloride, sulfate, and phosphate).[29] These ions coexist with the proteins; hence, one would already expect that this material exhibit relatively large conductivities under ambient conditions. The conductivity spectra obtained for the solid (boiled) EW samples at several temperatures are shown in Figure 2A. Here, one can recognize at high frequencies (above 10^3 Hz) the DC plateaus, with their amplitudes defining the characteristic DC conductivities, while the strong decrease at lower frequencies is a nonintrinsic effect (so-called electrode polarization) due to the blocking of migrating charge carriers at the electrode interfaces. The temperature-dependent DC conductivities (σ_T) obtained at 1 MHz in the temperature range between 25 and 110 °C are presented in Figure 2B. They reveal a roughly linear dependence between log (σ_T) and $1/T$ and indicate that no phase transition occurred over the investigated temperature range. Accordingly, the conductivity follows the Arrhenius equation: $\sigma_T = \sigma_0 \exp(-E_a/k_B T)$, where σ_0 is the pre-exponential factor, and E_a is the activation energy. In the absence of a significant change in the microscopic structure, E_a corresponds to the energy barrier controlling transport of charge carriers through the water−protein network. By fitting the data in Figure 2B, we estimated the activation energy to be 0.094 ± 0.02 eV (9.1 ± 2 kJ/mol), which is rather small as compared with other nonliquid electrolytes.

The room temperature conductivity of several EW specimens prepared under different conditions are summarized in

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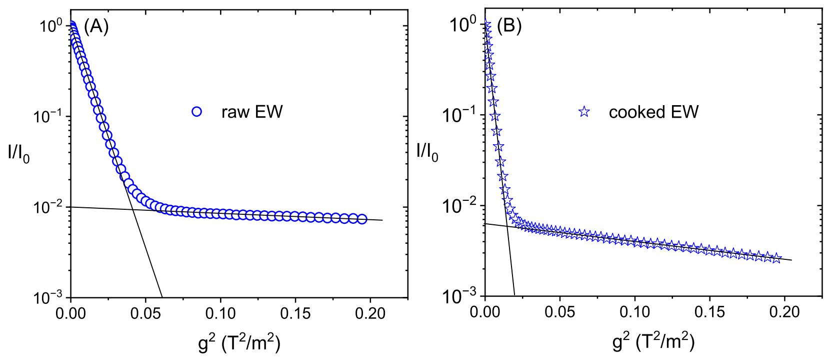

Figure 4. Intensities of ^{1}H NMR lines as a function of the square of gradient strength for (A) raw and (B) cooked EW.

Figure 3. The results demonstrate that the conductivity level of EW is significantly larger than that of deionized (DI) water and does not change significantly upon heat denaturation, although the latter significantly impacts the mechanical properties of this material (Figure 1).

Having demonstrated that EW is an excellent electrolyte, the question arises what microscopic mechanism gives rise to its high conductivity. Is the latter related to protonic or ionic transport? For proton conductivity, two types of mechanism have been proposed, the proton transfer (in its collective form known as the Grotthuss mechanism) and the vehicular transport.[30] Grotthuss mechanism is associated with proton hopping from one proton carrier to a neighboring one along the hydrogen bonded network. This type of conduction involves a significant amount of water. In the vehicular mechanism, the protons might diffuse along with water molecules via H_3O^+ ions. In the case of ionic transport, the latter is expected to be strongly dependent on ion concentration and solution viscosity.

The DC conductivity of any conductor is proportional to both the effective number density of charge carriers n_{eff} (which may be affected by cross-correlation effects in highly concentrated electrolytes)[31] and their diffusivity D. According to the Nernst−Einstein relation

$$\sigma = \sum_{i} \frac{n_{\mathrm{eff},i}(z_i q)^2}{k_B T} D_i \qquad (1)$$

where the index "i" corresponds to different charge carriers, z is their charge number, and q the elementary charge. To gain information on the composition and diffusivities of different components present in both uncooked and cooked EWs we performed NMR investigations covering a temperature range comparable to that in which their DC conductivities have been determined. The NMR investigations revealed the presence of significant amounts of elements containing ^{31}P, ^{23}Na, and ^{35}Cl nuclei (hence salt), and protein and sugars in addition to water as the dominant compound. The latter forms the mesoscopic matrix in which the charges are being transported, whereas denatured proteins build the (mechanically stable) embedding network at a macroscopic level (see Figure S1 in the Supporting Information).

To understand the mechanism of conductivity in EWs it is crucial to determine the proton diffusivities in these materials. This has been done in this work using NMR based DOSY. The obtained intensities of the ^{1}H lines are shown in Figure 4 as a function of the gradient strength at a representative temperature of 25 °C.

These data reveal two populations of protons with strikingly different mobilities, as for both uncooked and cooked materials the normalized NMR intensities decay in a two-step manner. Their slopes define the corresponding diffusion coefficients (multiplied with some experimental factor), whereas their intercept with the ordinate provide their corresponding populations. For the uncooked EW, we found that at room temperature the fast species are characterized by a diffusivity of 1.85×10^{-9} m^2/s and they represent about 99% of the total fraction of ^{1}H translating carriers, while for the cooked material, the corresponding values are 1.74×10^{-9} m^2/s and 99.3%. The remaining (small) amounts of protons belong to the less mobile species (which might be attributed to bonded water and proteins), with $D \sim 2.2 \times 10^{-11}$ m^2/s for both uncooked and cooked samples. Since the diffusivities of the fast species are very similar to that of neat water at room temperature, one may conclude (i) that essentially free water exists in a very large amount in the protein matrix of the cooked EW. Since in most hydrogels the water content varies between 60 and 90%,[32] cooked EW can be considered as a close to perfect hydrogel and (ii) that the high conductivity values of egg whites cannot be explained by an accelerated (increased diffusivity with respect to water) proton transfer. As we will discuss later, pH analysis suggests a decrease in free protons in EW relative to DI water, further excluding the role of proton transport.

Figure 5 presents the obtained temperature variation of diffusivities of fast proton containing species in the uncooked

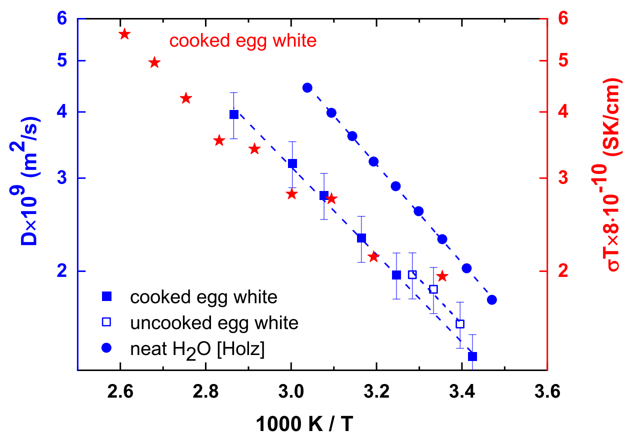

Figure 5. Temperature dependence of the fast component of proton diffusivity of pristine (open squares) and cooked (filled squares) EW compared with that of conductivity of cooked EW. The proton diffusivity of water (dots) are included for comparison.

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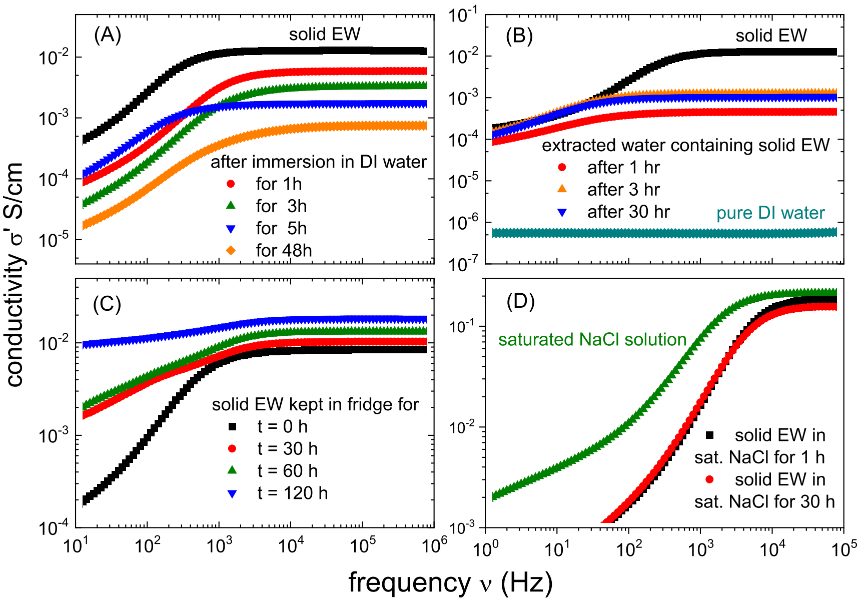

Figure 6. (A) Conductivity of a slice of cooked egg after keeping it in DI water for certain times; (B) conductivity of water solution (extracted water) containing white solid egg with a specified dimension for certain times. The conductivities of pure water and solid EW are also shown; (C) conductivity of cooked EW measured at different times after keeping the egg in the fridge; and (D) conductivity of cooked EW after keeping it in saturated NaCl solution for different times.

and the cooked materials. Fits with Arrhenius laws of the fast component diffusivity data (indicated by the dashed lines in Figure 5) estimate the activation energies ∼0.16 eV for the cooked EW, ∼0.17 eV for uncooked EW, and ∼0.18 eV for neat water.

Assuming that the diffusivities of the intrinsic ionic species are of the same order as the diffusivity of water molecules in this natural hydrogel, i.e., $D_i \sim D$, eq 1 can be rewritten as

$$D = \sigma T\left(\frac{k_B}{q^2}\frac{1}{\sum_i n_{\text{eff},i} z_i^2}\right) \tag{2}$$

In this way, one may attempt to gain some information regarding the static term contained between the brackets of eq 2. According to the normalization factor of the right y-axis of the Figure 5, this term is about 8×10^{-12} $m^3/(s \cdot S \cdot K)$, corresponding to $\sum_i n_{\text{eff},i} z_i^2 \sim 6 \times 10^{25}$ m^{-3}. Further assuming $z_i = 1$ and using the number density of molecules in neat water 3.3×10^{28} m^{-3}, the effective number density of charge carriers in EW is of the order of 10^{-3} with respect to that of water molecules. This amount agrees with the content of various ions in this material (see Table 1 of ref 33). Thus, relatively high ion concentration combined with their large mobility mediated by the fast rearrangements of the water molecules are responsible for the large conductivity exhibited by these materials. As shown in Figure S2 of the Supporting Information, this fast charge transport remains preserved (independent of thermal history) up to 100 °C.

The ionic transport hypothesis is further supported by additional experimental results: As indicated in Figure 3, the conductivity of EW decreases significantly after being placed in DI water. Figure 6A demonstrates this decrease develops monotonously as a function of time. Note that after 48 h, the drop in conductivity was found to be almost 2 orders of magnitude as compared to freshly prepared solid EW. Figure 6B complementarily presents the conductivity of the aqueous solution in which EW, revealing that the contribution of charge carriers diffusing out of EW strongly increases the conductivity of the water solute. Clearly, such behavior is not expected if the charge transport in EW would be the same as in water, i.e., dominated by protons.[34]

The stability of the albumen-based solid conductor was additionally studied under fridge storage conditions, at 4 °C. As shown in Figure 6C, the conductivity increases with the annealing time. This can be rationalized by considering that the slow evaporation of water facilitates an increase in number density of charged species responsible for conduction, again, indicating the ionic nature of the latter. Moreover, Figure 6D reveals that at variance for the situation for DI water, immersing the EW material in a saturated NaCl solution increases its conductivity to a value similar to that of the solute itself, with the conduction dominated by the ionic diffusion.

Finally, time-dependent pH values of 10 mL of DI water containing 0.4 cm^3 of cooked EW revealed a significant increase in pH (Figure 7), suggesting a decrease in the number of free protons with adding the solid material. These results additionally indicate that the higher conductivity values of EW compared with DI water is not the result of a larger number

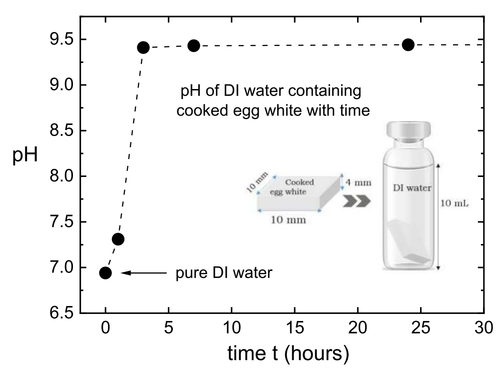

Figure 7. Variation of the pH value of DI water containing EW with time.

density of mobile protons in the former native material, a situation which would have triggered a decrease in the pH of the solute.

■ CONCLUSIONS

In summary, this work introduces a high performance, solid-like, protein-based ionic conductor derived from a biodegradable, abundant green source, i.e., hen EW. Its conductivity was shown to be among the highest ever reported for proteins and protein-derived ionic conductors, i.e., ∼0.01 S/cm. We demonstrate that this conductivity can be even further increased to ∼0.1 S/cm by immersing the cooked hen EW in saturated NaCl solution. These exciting results call for future biophysical studies to check whether this unexpectedly high ionic conductivity of the EW plays any important biological function in providing nutrients and stimulating biochemical reaction during the growth of chicken embryo. Our study also unraveled that the mechanical stability of this natural ionogel is due to a cross-linked protein network embedding a truly remarkable (>90%) water content, which explains the absence of slowing down of water and ion dynamics. Employing easily accessible proteins extracted from abundant natural sources as solid state ionic conducting materials creates exciting new possibilities for future bioelectronic devices.

■ ASSOCIATED CONTENT

Data Availability Statement

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.4c02654.

Schematic illustration of the albumin protein−ionic interactions, optical microscopy images of the EW solid surface, stepwise schematic illustration of thermal denaturation and subsequent aggregation of EW protein, and conductivity of EW in a broad temperature range (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. A.S. started the project and performed preliminary studies to inspire the article. I.A., P.M., W.H., P.A. and P.C. equally contributed to experimental work. I.A., A.S., C.G., and A.P.S. equally contributed on writing the manuscript.

Notes

The authors declare no competing financial interest.

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