

Temperature gradient independent thermoelectric conversion based on asymmetric interfacial ion rearrangement

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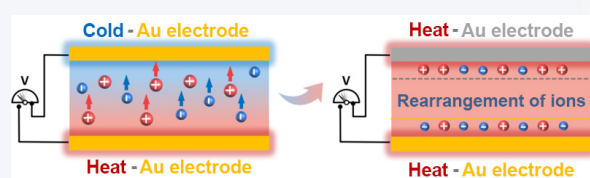
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ABSTRACT: Ionic thermoelectric conversion based on Soret effect, a phenomenon of converting heat into electricity, has been popularized in establishing self-powering systems where thermal exchange is associated. It works in the presence of a temperature gradient, but becomes invalid if there is no significant temperature difference between two electrodes, especially in miniaturized devices and large thermal field. Herein, we break this cognition by discovering thermoelectric conversion in isothermal environments resulting from a heterogeneous system consisting of Metal A/Ionic liquid/Metal B. Asymmetric ion rearrangement is proposed on two ionic liquid/electrode interfaces when temperature varies, accounting for the generation of thermoelectric voltage. This principle can be applied to many electrodes/ionic liquids groups. The advantages of temperature gradient independence lay the ground for the creation of powerless thermometers to alarm large area of fire.

KEYWORDS: ionic thermoelectric, ionic liquid, temperature gradient-independent, thermopower, fire alarming



1 Introduction

Based on the Seebeck effect, which is derived from the directional diffusion of electrons driven by heat flow, thermoelectric materials can generate a voltage across them in the presence of a temperature gradient [1–3]. This discovery of note makes it possible for sensing abnormal temperature or charging Internet of Things (IoT) systems independent on power supplies and cables [4–6]. Thermoelectric phenomenon has also been observed in some ionic materials, represented by ionic liquids [7, 8], ionic gels [9–12], and polyelectrolytes [13–15]. The underlying mechanism is generally attributed to the Soret effect [16, 17]. Unlike the Seebeck effect, the Soret effect refers to the thermal diffusion of ions rather than electrons under a temperature gradient. The differential anion and cation migrations lead to an imbalance in the concentrations of each type of ion, which changes the original electric double layer (EDL) at the electrode interface, thereby causing a potential change between two electrodes [18]. Remarkably, the thermoelectric coefficient of ionic thermoelectric materials based on Soret effect can reach tens of mV/K [19–21], offering great promise for developing self-powering devices with highly sensitive response to heat signal.

Ionic thermoelectric conversion based on the Soret effect does not essentially involve Faraday process at the electrode interface, however it must be dependent on the spatial temperature gradient. Yet, this principle is inapplicable to the isothermal environments, such as high temperature and the miniaturized devices where thermal distribution quickly becomes uniform. Such a limitation has been raised as to whether or not the thermoelectric conversion can happen independent on temperature gradient. Although the Soret effect is the prevailing theory for explaining this ionic thermoelectric conversion, ion rearrangement on electrode surfaces has also been proposed as one important contributor accounting for the generation of thermoelectric voltage [7, 22–24]. Owing to the asynchronous desorption of cations and anions, the potential on the heated electrode is no longer equal to that on the electrode without thermal disturbance. The termed thermoelectric voltage is closely relevant to the difference in cation and anion desorption, as well as the temperature difference between two electrodes. This knowledge has always been focused in the systems with symmetric electrodes.

In this project, we established an ionic thermoelectric conversion system using asymmetric electrodes which mainly highlights the dominant contribution of interfacial ion rearrangement to thermoelectric voltage, and its novelty lies in the independence on temperature gradient (Fig. 1(a) and Fig. S1 in the Electronic Supplementary Material (ESM)). The asymmetric electrodes refer to two different metal electrodes, on which ions exhibit varied degree of desorption during the overall heating. Such a Metal

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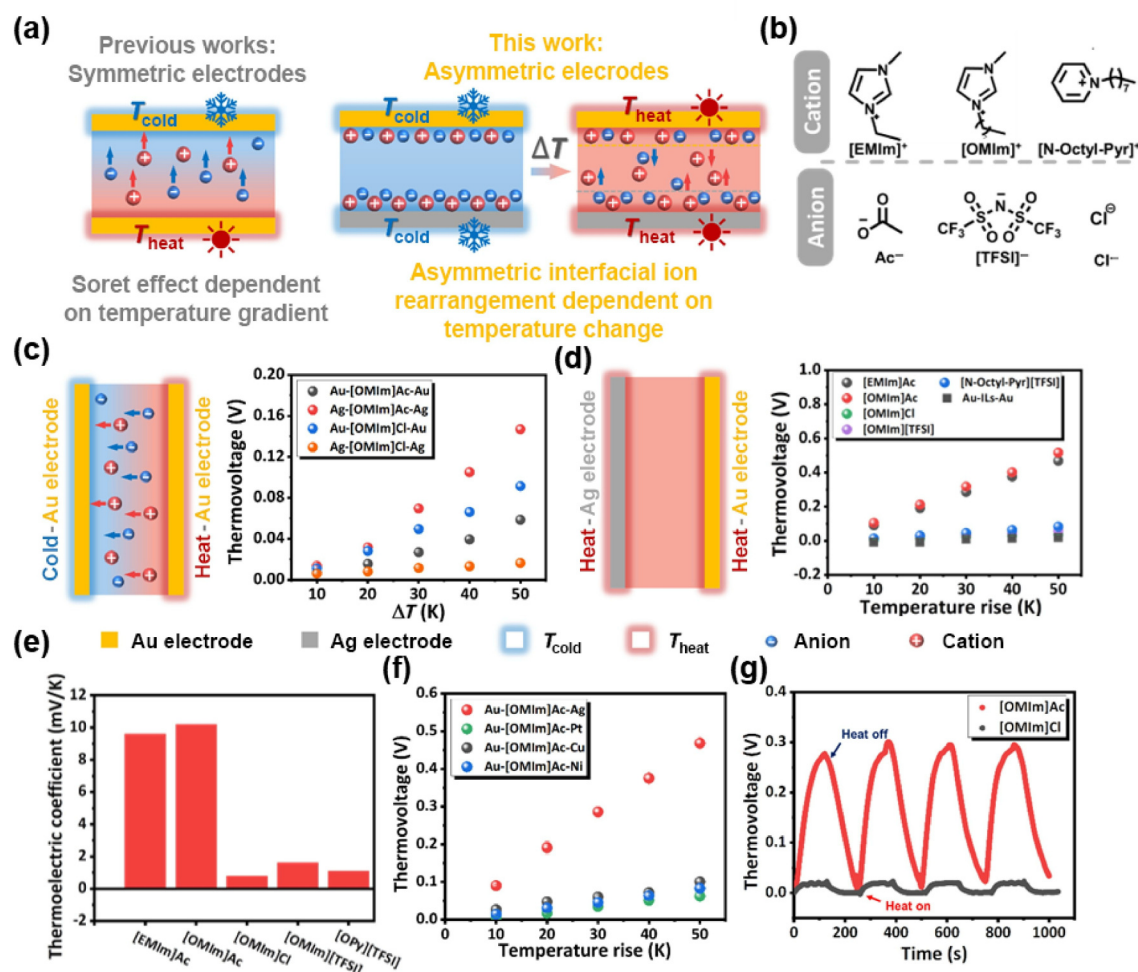


Figure 1 Principle and characterizations of the ionic thermoelectric conversion that is independent on temperature gradient. (a) Schematic illustration of ionic thermoelectric converters made of symmetric (Metal A/ionic liquid/Metal A) or asymmetric electrodes (Metal A/ionic liquid/Metal B). (b) Some representative ionic liquids used in thermoelectric experiments. (c) Thermoelectric voltages of ionic thermoelectric converters based on symmetric electrodes under temperature gradient. (d) Thermoelectric voltages (termed as $\phi_{Ag} - \phi_{Au}$) of ionic thermoelectric converters based on asymmetric electrodes during overall heating. (e) Thermoelectric coefficients of ionic thermoelectric converters corresponding to (d). (f) Thermoelectric voltages of the ionic thermoelectric converters by combining gold and several other metals as asymmetric electrodes. (g) On-off cycles of the thermoelectric response of ionic thermoelectric converters with [OMIm]Ac and [OMIm]Cl as ionic components, and Au-Ag as asymmetric electrodes. The applied temperature increased from 25 to 55 °C.

A/ionic liquid/Metal B structure can generate a potential difference between two electrodes even they both are in an isothermal environment.

2 Results and discussion

2.1 Thermoelectric effect independent on temperature gradient

As presented in Fig. 1(b), cations and anions with large ion radius can be combined to form ionic liquids with low melting point. Most of them have been confirmed to possess the thermoelectric conversion properties over temperature gradients [25, 26]. As explained by Soret effect, there is a difference in the migration of anions and cations from the hot electrode to the cold electrode, forming the asymmetric distribution of ions at the surface of both electrodes along with the asymmetric electrode potential, which is responsible for the generation of thermoelectric voltage [27]. If the resulted thermoelectric voltage is fully attributed to the differential migration of anions and cations, then changing the type of

electrode should not affect the magnitude of the output voltage. However, as shown in Fig. 1(c), for the same ionic liquid, the type of electrode has a significant effect on the thermoelectric voltage (Figs. S2–S5 in the ESM). It can be rationally inferred that there is an ion-arrangement process at the electrode interface related to the nature of electrode itself, which likewise contributes to the generated thermoelectric voltage. Differential ion adsorption states, closely related to the nature of the electrodes, are bound to exist at the interface of the two electrodes with different temperatures. Therefore, combined with the experimental phenomena in Fig. 1(c), a hypothesis is proposed that the differential ion adsorption is the fundamental reason why the electrode species change the thermoelectric voltage. Based on this hypothesis, subjecting the two types of electrodes to the same varying temperature field can also create different ion adsorption and desorption processes, which are reflected as a change in the potential difference between the electrodes. Intriguingly, as shown in Fig. 1(d) and Fig. S6 in the ESM, when the device employing the gold-silver electrode pair (Au-Ag) was heated or cooled as a whole, a considerable voltage signal was output between the two

electrodes, and this experimental observation persuasively corroborated the aforementioned hypothesis. Compared with the traditional thermoelectric conversion system based on the Soret effect, the ionic thermoelectric conversion constructed with asymmetric heterogeneous electrodes gets rid of the dependence on the temperature gradient, and the thermoelectric voltage can be generated during the overall heating or cooling process. In the case of Au-Ag asymmetric electrodes, the thermoelectric voltage is defined as the potential difference between Ag and Au electrodes ($\phi_{\text{Ag}} - \phi_{\text{Au}}$), and the converters containing imidazole or pyridine-based ionic liquids have positive thermoelectric coefficients (Fig. 1(e)), suggesting that the potential rise of the Ag electrode is greater compared to that of the Au electrode as the temperature increases. The phenomenon of ionic thermoelectric conversion based on asymmetric ion adsorption and desorption processes has been demonstrated for many electrode pairs, such as Au-Pt, Au-Cu, and Au-Ni (Fig. 1(f) and Figs. S7–S9 in the ESM). It is worth noting that the proposed theory is inherently different from the mechanisms of thermogalvanic cells and thermally regenerative electrochemical cycle, because the ionic liquids involved do not contain redox pairs that can undergo Faraday processes, and the electrodes employed are relatively inert. In addition, as shown in Fig. S10 in the ESM, in the thermoelectric cycle, the loop current is in the order of 10^{-8} A, suggesting an almost negligible

electrochemical reaction of the system. Cyclic heating and cooling tests confirmed the stability and reliability of these asymmetric electrodes-based thermoelectric conversion systems (Fig. 1(g)).

2.2 Mechanism of thermoelectric effect independent on temperature gradient

As stated above, the ionic re-arrangement on two different electrodes accounts for this temperature gradient-independent ionic thermoelectric conversion. Taking the Ag-[OMIm]Ac-Au system with high thermoelectric coefficient as an illustration, when the overall converter is at a lower temperature, attributed to the interaction between ions and electrodes as well as the difference in the radius of the metal atoms, the ionic liquids have inconsistent adsorption states at the interface of the two electrodes. As the temperature rises, the anions and cations are respectively desorbed to reach a new equilibrium. Consequently, the adsorption state of ions at the interface of the two electrodes is still different. It is reflected in the existence of a certain difference between the induced potentials of the two electrodes, known as the thermoelectric voltage (Fig. 2(a)).

To support the proposed theory, molecular dynamics simulations are conducted to provide insight into the adsorption state of the [OMIm]Ac on the interface of Au and Ag electrodes, respectively (Fig. 2(b) and Figs. S11–S22 in the ESM). As shown in

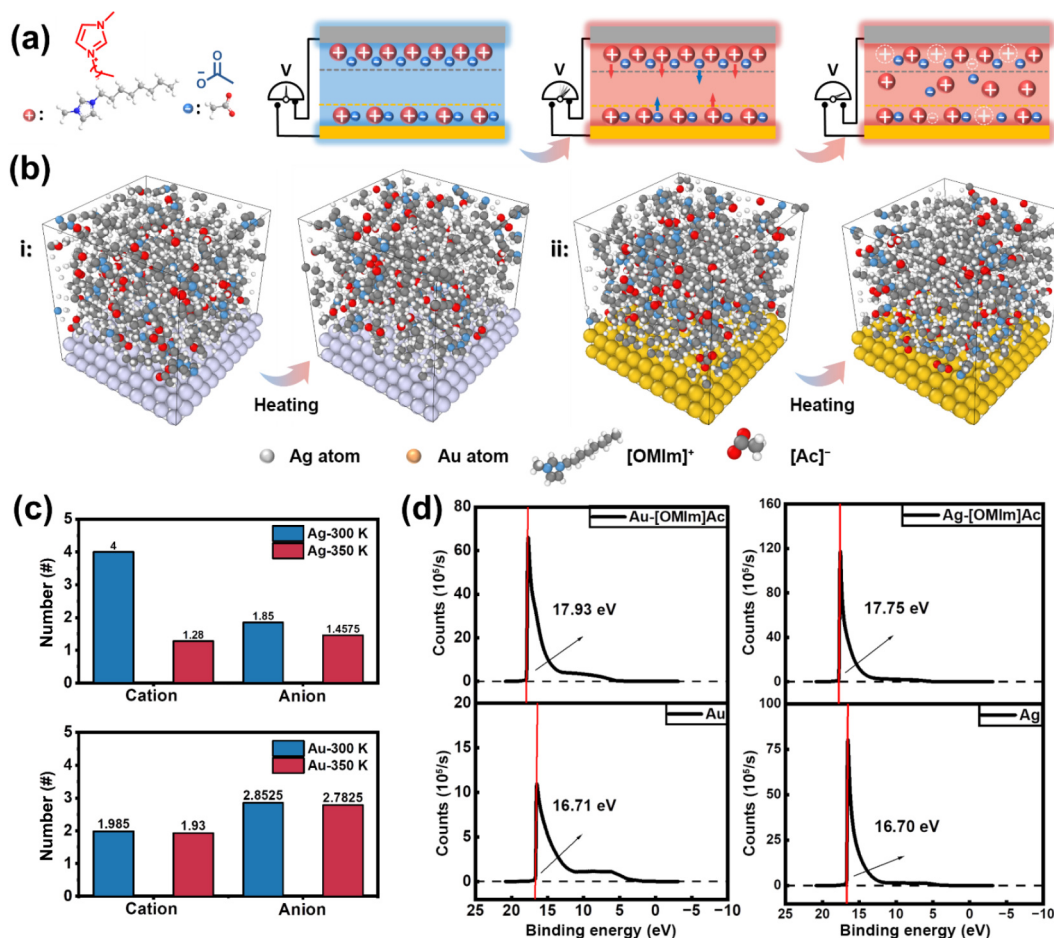


Figure 2 Mechanism of the temperature-gradient-dependent device. (a) Schematic diagram of the thermoelectric conversion mechanism of Ag-[OMIm]Ac-Au system. (b) Molecular dynamics simulation of temperature-dependent desorption processes of [OMIm]Ac at Au and Ag surfaces. The temperature increases from 300 to 350 K. (c) The number of anions and cations adsorbed at the surface of Au and Ag electrodes at 300 and 350 K. The electrode area used for the calculation is $28.89 \text{ \AA} \times 28.89 \text{ \AA}$. (d) Work function of Au and Ag electrodes before and after coating with [OMIm]Ac.

Fig. 2(c), there is a significant difference in the number of anions and cations occupying the surface of the Au and Ag electrodes, regardless of whether they are at 300 K or at 350 K. Moreover, when the temperature increases from 300 to 350 K, both cation and anion at the interface of Ag electrode show obvious desorption process while that at the interface of Au electrode change little. The difference in ion distribution between Au and Ag surfaces at room temperature was further verified by testing the work function, which refers to the minimum thermodynamic work needed to remove an electron from a solid to a point in the vacuum immediately outside the solid surface. Ultraviolet photoelectron spectroscopy results indicate that after spin-coating a relatively thin layer of [OMIm]Ac, the work function of Au film electrodes decreases from 4.51 to 3.29 eV, while that of Ag film electrodes decreases from 4.52 to 3.47 eV (Fig. 2(d)). The relatively small change in the work function of Ag is due to the greater number of surface-occupying cations, which attract electrons and prevent their escape. This result contributes to our understanding of the interaction between ionic liquids and different electrodes.

In addition to investigating the difference in the adsorption of ionic liquids on asymmetric heterogeneous electrode pairs, constructing a quantitative relationship between ion adsorption at the electrode interface and the thermoelectric voltage can deepen the understanding of the thermoelectric conversion mechanism. Again taking the Ag-[OMIm]Ac-Au system as an example (Fig. S23 in the ESM), the potential difference between the two electrodes at a given temperature can be derived from Eq. (1)

$$U_{(Ag/Au),T} = \vec{U}_{s/Ag} - \vec{U}_{s/Au} = \frac{Q_{s/Ag}}{C_{s/Ag}} - \frac{Q_{s/Au}}{C_{s/Au}} \quad (1)$$

where $U_{S/Metal}$, Q_s , and C_s are the interfacial potential difference between electrode and ionic liquid, the net charge adsorbed on the electrode surface and the interfacial capacitance, respectively. $U_{S/Metal}$ is a vector whose direction is determined by the kind of net charge

adsorbed at the electrode interface. Theoretically, the thermoelectric voltage is defined as Eq. (2)

$$U_{(Ag/Au),T} = U_{(Ag/Au),T} - U_{(Ag/Au),T_0} \quad (2)$$

Here T_0 and T refer to the initial and thermal treatment temperature of the thermoelectric converter as a whole, respectively. The specific value of Q_s can be confirmed according to the results of molecular dynamics simulation, and the specific value is shown in Table S1 in the ESM. To obtain the interfacial capacitance, electrochemical impedance spectroscopy tests were implemented. An equivalent circuit is shown in Fig. 3(a), where CPE, R_E , and R_S represent the constant phase element, bulk phase resistance, and interfacial electron transfer resistance, respectively. In principle, the R_S in the equivalent circuit originates from the interfacial electron transfer due to the Faraday process (Figs. 3(b) and 3(c)). However, according to the Nyquist plots and the fitted electronic component parameters, the R_S at both the Au and Ag interfaces are in the order of $10^5 \Omega$, which further confirms that it is more difficult for the interfaces to exhibit electrical signals similar to those of the thermoelectrochemistry (Table S1 in the ESM). Interestingly, the change of interfacial capacitance of Ag system is far larger than that of Au system (Fig. 3(d)), which is consistent with the simulated ionic movement shown in Fig. 2(c). Based on Eqs. (1) and (2) and Fig. 3(d), the thermoelectric voltage value generated by the proposed thermoelectric conversion model under the condition of temperature rising from 300 to 350 K is a positive response of 381 mV, which is close to the experimentally tested value.

2.3 Proof-of-concept for fire alarming independent on temperature gradient

Abnormal temperature monitoring is one of promising applications for ionic thermoelectric conversion. Conventional ionic thermoelectric conversion based on the Soret effect strictly relies on

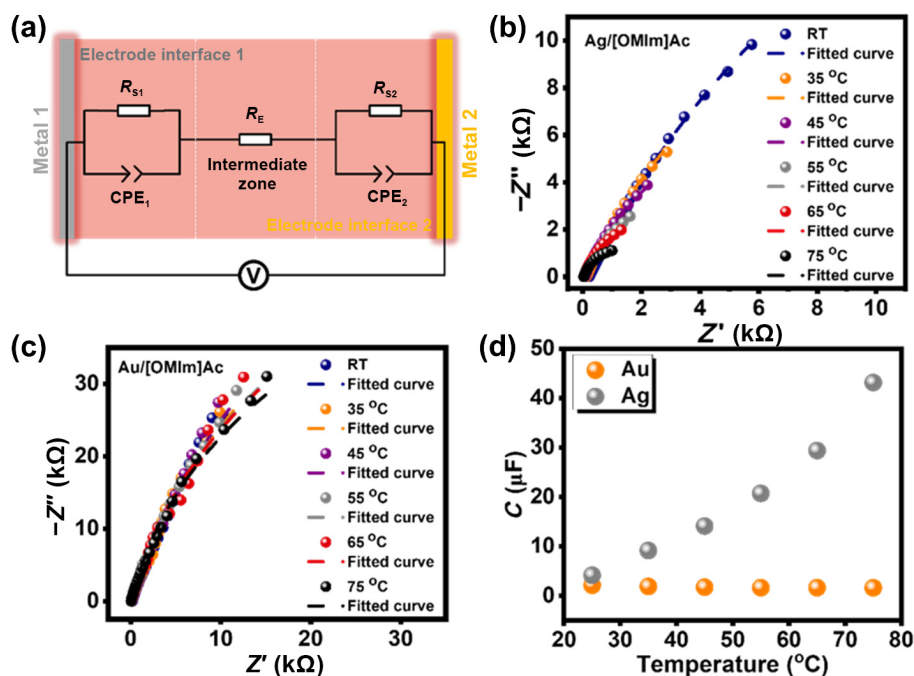


Figure 3 Theoretical derivation of thermoelectric voltage and electrochemical impedance spectroscopy tests. (a) The equivalent circuit of the Metal A/ionic liquid/Metal B system. (b) Nyquist plot of [OMIm]Ac on Ag electrode. (c) Nyquist plot of [OMIm]Ac on Au electrode. (d) Temperature dependence of interfacial capacitance resulting from the combination of Ag and Au electrodes with [OMIm]Ac.

the temperature gradient between two electrodes [28, 29]. However, for large fire areas, the relatively uniform temperature field makes it difficult for the converter to produce the desired signal output. Considering the temperature gradient-independent thermoelectric conversion of the Metal A/ionic liquid/Metal B system, it can be explored as a next-generation breakthrough technology for abnormal temperature monitoring. An amplified thermoelectric signal can also be obtained by connecting the thermoelectric conversion units of Metal A/ionic liquid/Metal B system in series in a head-to-tail fashion (Fig. 4(a)). The electrical response of thermoelectric modules with 1–5 series units was tested, and almost linear amplification can be achieved (Fig. 4(b)). In compliance with this serial approach, a temperature gradient independent thermal sensing module was designed, which consists of a three-layer structure with Au and Ag electrodes alternately distributed on the upper and lower glass substrates (Fig. 4(c) and Fig. S24 in the ESM). Furthermore, the prepared temperature sensing module was combined with a signal transmitter, a wireless signal receiver, and an alarm system for early warning of forest fires (Fig. 4(d)). The feasibility of this system was demonstrated through the real-life scenarios (Fig. 4(e) and Movie S1 in the ESM). When the temperature around shrubs with early warning systems was abnormally high, the operation of the thermoelectric conversion sensing module could be induced to generate a warning thermoelectric signal, which in turn triggered the remote alarm system. When the fire was extinguished, the thermoelectric signal of sensing module dropped dramatically, signifying the end of the alarm process.

3 Conclusions

In summary, an ionic thermoelectric conversion system of Metal A/ionic liquid/Metal B based on asymmetric heterogeneous metal pair was established, which exhibited the unique advantage of being independent on the temperature gradient compared with the traditional ionic thermoelectric conversion based on the Soret effect. The phenomenon can be attributed to the differential adsorption and rearrangement of anions and cations at the interface of asymmetric electrodes. This disruptive thermoelectric conversion concept overcomes the challenge of traditional thermoelectric conversion devices based on Soret effect that fail in a uniform temperature field or isothermal environment, and has great potential for application, especially in miniaturized thermoelectric conversion converter as well as large-area fire warning. In particular, the essence of ionic thermoelectricity has been pointed out to create asymmetric interfaces, which have important implications for the development of ionic thermoelectric materials.

Electronic Supplementary Material: Supplementary material (supporting information and Movie S1) is available in the online version of this article at <https://doi.org/10.26599/NR.2025.94907291>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

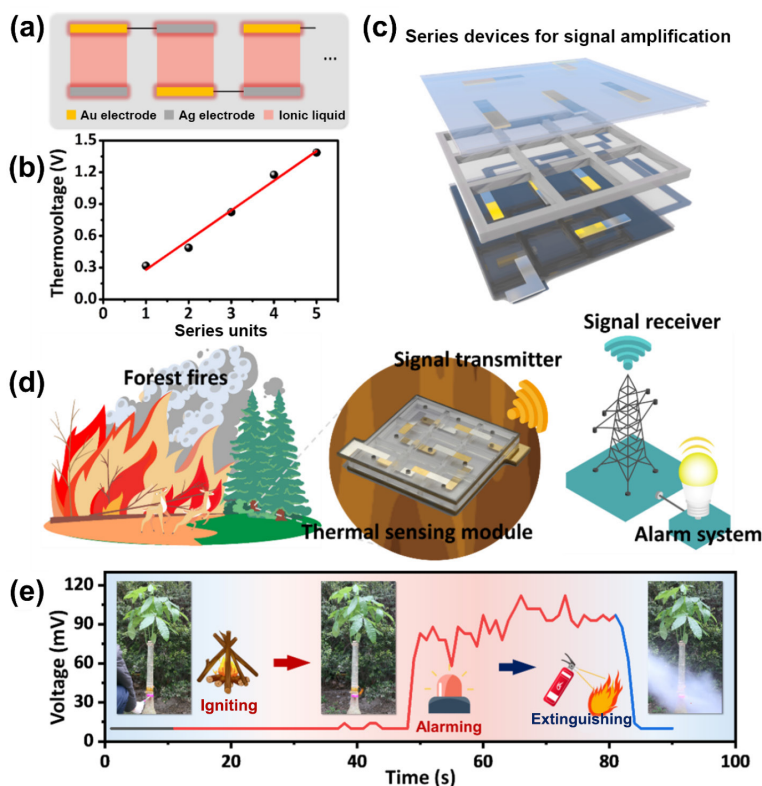


Figure 4 Proof-of-concept for fire alarming independent on temperature gradient. (a) Schematic diagram of series connection of thermoelectric conversion units. (b) Relationship between thermoelectric voltage and number of units in series. (c) Schematic diagram of the series module design for fire alarming. (d) Operational diagram of a wireless fire monitoring system including thermoelectric sensing module, signal transmitting module, signal receiving module, and alarm system. (e) Real-time variation of thermoelectric voltage during fire warning. The inset pictures correspond to the actual scenes before and after the fire as well as the extinguishing of the fire.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

X. W. and Y. L. W. designed the experiment, X. W., Z. W. C., and X. L. L. conducted the experiment, K. L. and Y. L. W. provide molecular dynamics simulation to explain the experiment data. X. W., Z. W. C., and Y. L. W. wrote the manuscript. All authors discussed the results and participated in revising the manuscript.

Use of AI statement

None.

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