

Energy saving thermal adaptive liquid gating system

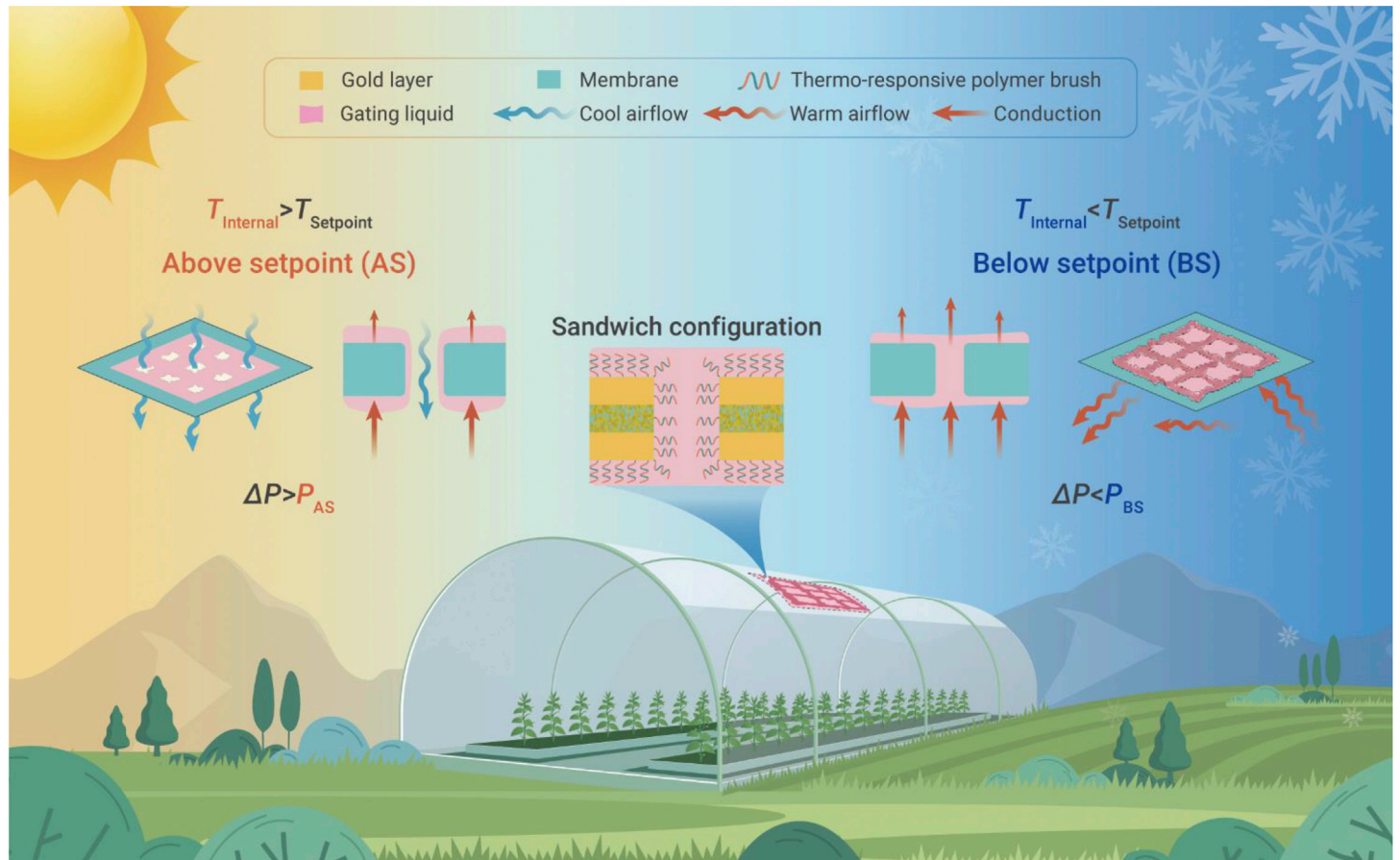
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- An energy saving thermal adaptive liquid gating system is constructed
- The system uses functional liquid to exhibit high metastability, providing durability
- The system is used as an energy saving patch to greenhouse by sandwich configuration
- The system shows energy consumption reduction of $\sim 11.6\%$ than traditional greenhouse



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Thermal transfer systems involving temperature control through heating, ventilation, and air conditioning applications have emerged as one of the largest energy issues in buildings. Traditional approaches mainly comprise closed and open systems, both of which have certain advantages and disadvantages in a single heating or cooling process. Here we report a thermal adaptive system with beneficial energy-saving properties, which uses functional liquid to exhibit high metastability, providing durability in a temperature-responsive liquid gating system. With an efficient use of energy, this system achieves smart “breathing” during both heating and cooling processes to dynamically tune the indoor temperature. Theoretical modeling and experiments demonstrate that the adaptive, sandwich-structured, membrane-based system can achieve temperature control, producing obvious advantages of energy saving compared with both closed and open systems through the bistable interfacial design of the liquid gating membrane. Further energy saving evaluation of the system on the basis of simulation with current global greenhouse plantation data shows a reduction of energy consumption of 7.9×10^{13} kJ/year, a percentage change of $\sim 11.6\%$. Because the adaptive system can be applied to a variety of thermal transfer processes, we expect it to prove useful in a wide range of real-world applications.

INTRODUCTION

Temperature control involves processes to detect changes in the temperature of a space and regulate the passage of energy into and out of this space to achieve a desirable temperature.^{1–3} Although temperature controls using HVAC (heating, ventilation, and air conditioning) facilities have been commonly applied to actively increase or decrease the temperature in order to meet a user-defined setpoint, the use of façade materials to envelop constructions that react by blocking or accelerating energy exchange in response to temperature or environmental variations is becoming more attractive given increasing energy concerns.^{4–8}

Thermal exchange between an enclosed space and its ambient environment can take place in the forms of conduction, convection, and radiation.^{2,9} Although conduction and radiation prevail, convection usually plays a dominant role in the heat transfer of air in space through the movement of fluids to spread heat.^{10–12}

As a result, a closed space can be effectively heated by blocking the fluids in motion,^{13–15} while an open system offers efficiency in cooling processes by encouraging airflow.^{11,12,16} Therefore, systems with adjustable openings for thermal convection passages, allowing a shift between closed and open systems, are highly desirable for realizing temperature control with energy efficiency.

Stomatal transpiration in plants is a phenomenal example of living organisms' making use of adjustable openings of fluid flow to regulate heat transfer.^{17,18} The stomatal apertures open and close as a result of turgidity changes in the guarded cells that arrange peculiarly inside the stoma, and through size control of the stoma, transpiration rates can be regulated.^{19,20} Although considerable effort has been devoted to mimicking the operation of stomatal apertures,^{21–23} a single system capable of dynamically altering thermal transfer passages in response to the difference between the actual temperature and a desired temperature remains a distant prospect.

Here we describe an energy-saving thermal adaptive (TA) system using a bistable liquid gating, sandwich-structured membrane with different opening profiles for different temperature ranges around a set temperature point. The idea of us-

ing a liquid as a structural and functional material to build responsive gates sounds counterintuitive, even verging on science fiction.²⁴ However, this idea has already become a reality; liquid gating technology is an emerging technology using functional liquids as structural materials to build reconfigurable gates with intrinsically different response profiles for providing a special combination of dynamic and interface physicochemical behaviors.^{25–27} Theoretical modeling and experiments have demonstrated that the adaptive system, which is achieved through a bistable interfacial design of the liquid gating membrane, can provide temperature control of internal space and hence help optimize energy performance. Meanwhile, by using sandwiching, porous membranes with affinitive gating liquids, thermal adaptive systems can produce smart “breathing” to regulate indoor temperature. On the basis of the proposed system, a greenhouse membrane prototype was constructed to demonstrate the energy-saving performance of a thermal adaptive system with practical application. Also, energy saving is further evaluated with simulations based on global greenhouse plantation data from 2020,^{28,29} showing a considerable reduction in energy consumption. The thermal adaptive system design incorporating dynamic properties from liquids can be seen as an example of liquid-based intelligent construction, which would enhance the energy and functional performance of membrane materials for the development of “smart” systems for real-world applications.

RESULTS AND DISCUSSION

Thermal transfers in open, closed, and thermal adaptive systems

Figure 1A compares thermal transfer passages in the closed, open, and TA systems with adaptive openings. Generally, with closed and open systems, the passages for thermal transfer remain consistent regardless of temperature variations, as the convective thermal transfers are predominantly prevented by blocking fluid motions in a closed space, whereas air can freely flow continually in an open system. By contrast, with adaptive opening produced by a liquid forming gate, when the internal temperature is above the desired temperature as a user-defined setpoint, the porous matrix stabilizing the liquid through wetting is in a flaccid state, rendering a moderate affinity with the liquid filler. A mild pressure gradient led by the temperature difference between the internal and the external space is provided adequately to force the air to open the liquid-sealed pores. In contrast, when the internal temperature is lower than the desired temperature, the matrix becomes turgid, with stronger affinity to the liquid filler, and the openings are immediately sealed and require comparatively strong pressure to open in the lower range below the setpoint temperature. In this way, a single system capable of switching the passage of thermal transfer in response to temperature variation to a desired temperature can be achieved (Figures S1 and S2).

We quantitatively investigate temperature changes in the three systems under heating and cooling processes by controlling variables of energy input and start temperature (Figure 1B; Table S1). It is shown that during the heating process, with the same amount of thermal energy input, the temperature increase in the adaptive system is significantly lower than that in a closed system and approximate to that in an open system. During the cooling process, starting from the same temperature, the temperature in the adaptive system diminishes more slowly than that in an open system and tends to parallel that in a closed system. As the liquid openings in the adaptive system display different opening behaviors, when the temperature increases above the set temperature, the system autonomously operates the openings to allow convective fluid movement,

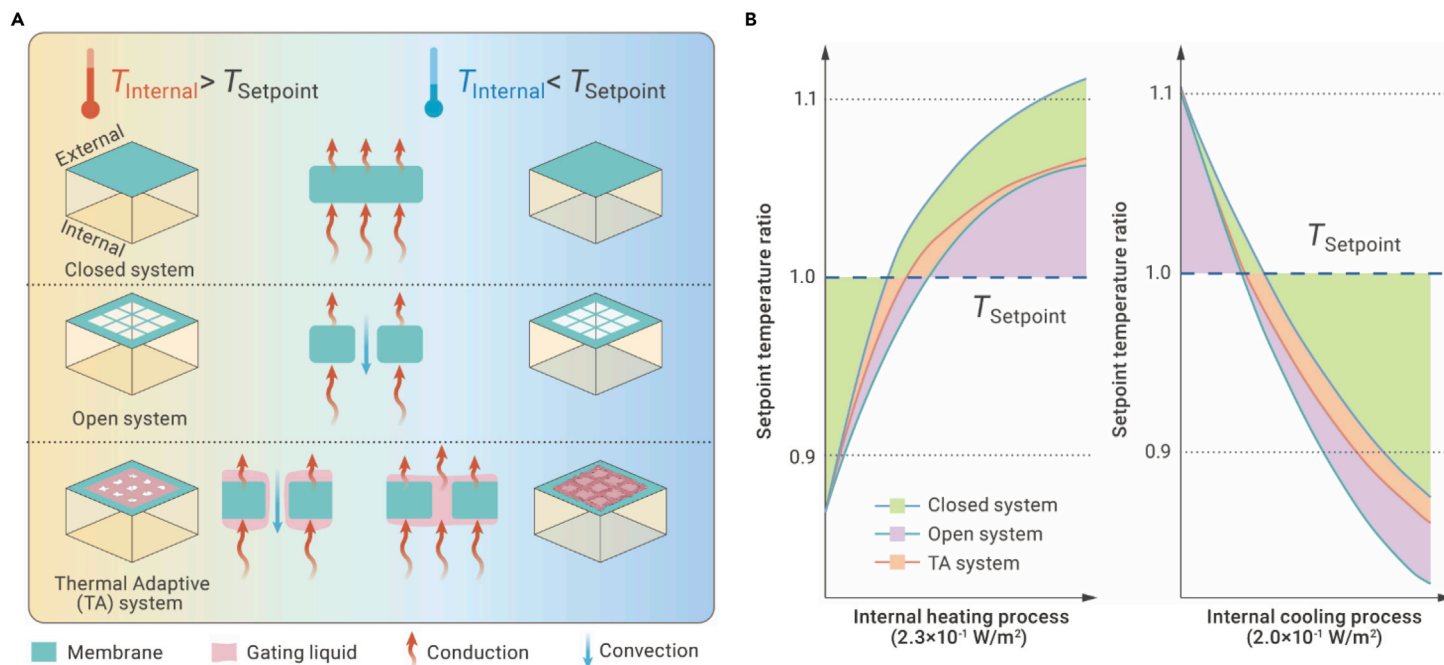


Figure 1. Thermal transfer and energy efficiency analysis of closed, open, and thermal adaptive (TA) systems (A) Thermal transfer of closed, open, and TA systems. The closed and open systems show consistent thermal transfer passages regardless of the temperature change, while the TA system displays thermal adaptive openings enabled by liquid gating. (B) Quantitative comparison of energy consumption for the three systems in heating and cooling processes. Compared with classic thermodynamic systems, the TA system shows lower energy consumption due to its passive temperature control. The shaded area shows the amount of energy consumption.

slowing the temperature increase; during cooling, when the temperature decreases below the setpoint, the system operates to seal the internal space completely. Thus, this design presents a method for adaptive temperature control by minimizing the variations between the actual temperature and the user-defined setpoint, and the autonomous opening and closing functionalities can maintain thermal comfort in buildings,^{30,31} optimal temperatures in greenhouses,^{32,33} and other applications, while alleviating the energy consumption used by active temperature control facilities. In Figure 1B, the shaded areas represent the sums of energy demanded for each temperature point along the curves to achieve the setpoint in the three systems to maintain the set temperature, which equals the definite integral of the differences between the setpoint and transient temperatures obtained by simulation. By the estimation of the convective heat transfer coefficient, convection, including heat and mass convective transfer, is deduced to be the major player in transferring energy in all three systems (Notes S1 and S2). With 6.62% energy transfer via mass convection, the TA system shows the lowest energy demand, using thermal adaptive behaviors of openings for airflow to regulate heat transfer; the ratios are 0% and 28.6%, respectively, for the closed and open systems (Figure S3). Meanwhile, proportional control of temperature proves to be more energy efficient, as it can vary the amount of heating or cooling energy depending on the difference between the actual temperature and the desired temperature. Therefore, autonomously varying the passages of heat transfer to minimize the difference between the actual temperature and the desired temperature can produce significant energy saving.

Design and evaluation of the TA system

Our hypothesis that a liquid-filled thermo-responsive porous membrane could provide a unified temperature control strategy derives from the idea of nature's widespread use of fluids as reconfigurable gates.^{34–40} By incorporating the dynamic nature of liquids with the thermo-responsive properties of membrane materials, we have created a thermal adaptive system that uses liquid controlling thermal transfer passages according to the difference between the actual temperature and a set temperature, with time effectiveness and a full range of operation enabled by liquid sealing performance.^{41,42} The system is composed of a thermo-responsive microporous membrane whose surface wettability is subject to temperature; while wettability varies with temperature change, the affinity between the gating liquid and the membrane is modulated. With differential affinities between the gating liquid and the membrane, the system can render differ-

ential gating profiles, and with dynamic interfacial properties of liquid, the system displays reversible opening behaviors.

The feasibility of temperature control with a thermal adaptive system relies on the temperature responsiveness of the porous membrane and the stability and thermal adaptive gating performance of the gating liquid. We illustrate temperature-responsive controllability by using the wettability change of the thermo-responsive polymer (poly [*N*-isopropylacrylamide], or PNIPAAm),^{43,44} because its lower critical solution temperature (LCST), the response temperature of the polymer, also functions as the set temperature of the thermal adaptive system and is approximate to the optimal greenhouse temperature ($\sim 30^\circ\text{C}$), which is a practical example of a desired temperature.¹⁹ Hydrophobic or hydrophilic monomers (HBMs or HLMS) are copolymerized with the NIPAAm monomer to adjust LCST (Figure 2A, left). LCST of the copolymer linearly increases with increasing HLM molar ratio and linearly decreases with increasing HBM molar ratio because of the changes of the hydrogen bond donor content in copolymer (Figures 2A, right; Figure S4). However, the change in temperature will lead to a change in the wettability of the membrane, and the change in wettability will inevitably affect the stability of the gating liquid to the membrane, which presented a special design challenge to the system, as the stability of liquid gate cannot be sustained with single structure-based membrane materials. As shown in Figure 2B (left), the gating liquid has good adhesive properties (being close to the optimal straight line) and spreading behavior (being in 65° wetting envelope) on PNIPAAm when the temperature is below its LCST, because of the strong interaction between the aqueous functional liquid and the hydrophilic polymer molecules. But when above the LCST, the gating liquid shows poor wetting performance and a high contact angle, because the molecular polarity of PNIPAAm decreases, and the polar interaction between the liquid and solid phases weakens, which leads to the loss of gating liquid and diminished performance of the liquid gating system. Therefore, we propose dual-complex structure-based materials with a permanent hydrophilic part and a responsive wettability conversion part that can be prepared to achieve stable interfaces regardless of temperature changes (Figures 2B, right; Figure S5; Note S3). This dual-complex structure possesses both temperature- and wettability-responsive properties and low interfacial tension to the gating liquid: the temperature- and wettability-responsive properties cause variation of the adhesion force between the solid substrate and gating liquid below and above the LCST, which produces the change in air transmembrane pressures; the permanent hydrophilic part provides bistability of wettability and adhesion between the

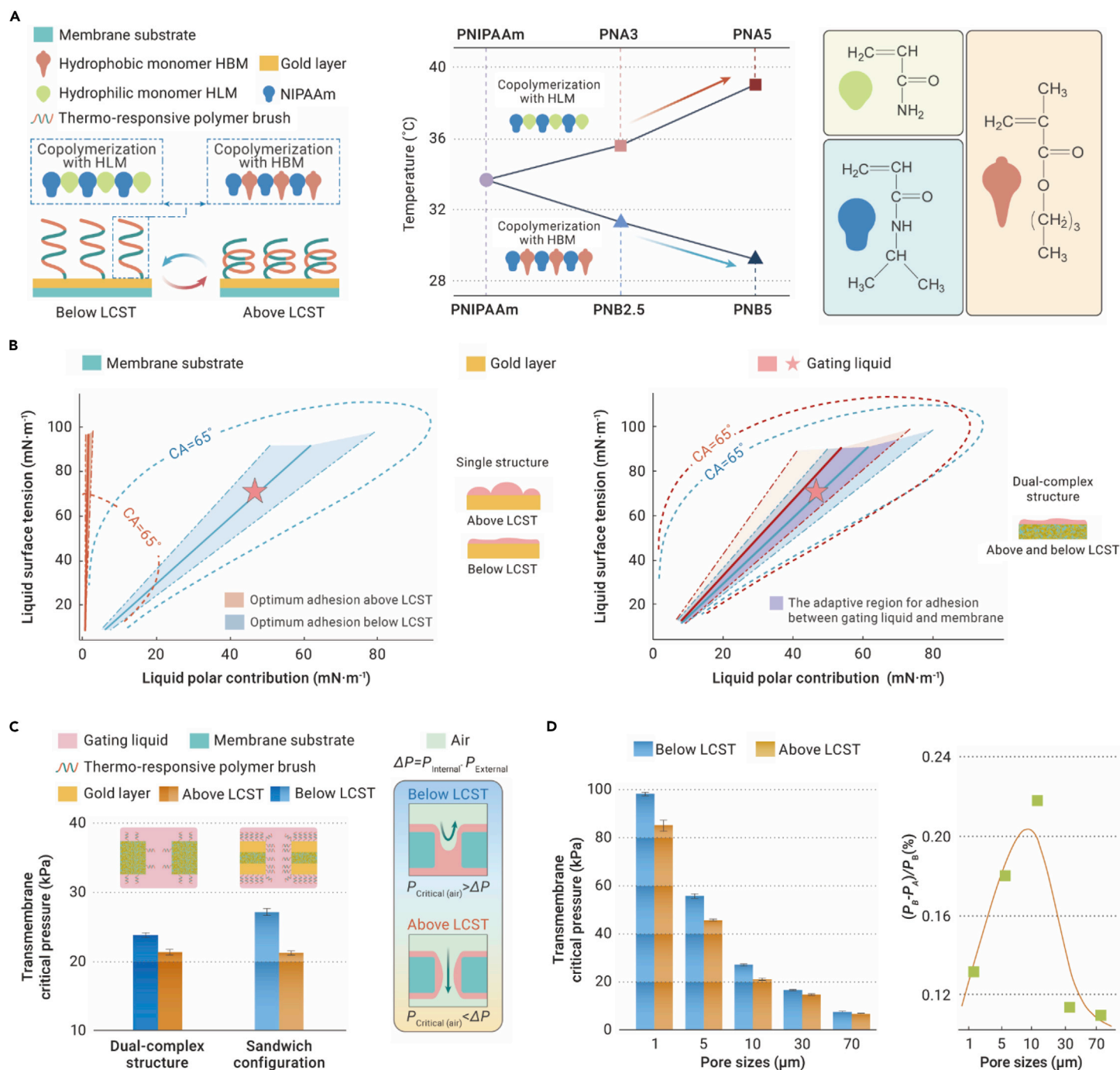


Figure 2. Design, evaluation, and working principles of the TA system (A) Design and conformation transition of the thermo-responsive polymer responding to the temperature change (left). Controllable LCST of the thermo-responsive copolymer with different molar ratio of HLM and HBM incorporation (right). (B) Interfacial design of the TA system with bistability. Adhesion work analysis with the wetting envelope and the corridor for optimal adhesion, as well as the value for gating liquid on single (left) and dual-complex (right) structures. Inset diagrams depict the wetting behavior of the gating liquid on single and dual-complex structures. (C) Transmembrane critical pressures required for the air to force through TA systems with original dual-complex structure and sandwich configuration, below and above the LCST, respectively (left); insets show the geometry and chemical modification hypothesis of porous membrane cross-section. Schematic illustration of gating liquid reconfiguration for realizing intelligent control of air movement below and above the LCST (right). (D) Transmembrane critical pressures for air through TA systems with different pore sizes. Inset shows the ratio of pressure difference versus different membrane sizes. All error bars indicate SD. LCST, lower critical solution temperature of thermo-responsive polymer; HLM, hydrophilic monomer; HBM, hydrophobic monomer.

porous membrane and gating liquid regardless of the variation of temperature, which ensures the working stability of the liquid gating system. This design allows a unique strategy to solve problems concerning wetting and adhesion of liquids on solids in a quick and cost-effective way.

It is worth mentioning that with the dynamic nature of liquid, thermal adaptive gating performance is variably affected by porous geometries, surface chemistries, pore sizes, and gating liquids. In order to optimize gating performance, we use a sandwich configuration design (Figure S6). Compared with the original dual-complex structure, which achieved both stable interfacial adhesion and thermal adaptive wettability to the gating liquid, the sandwich configuration with spe-

cific geometries and surface chemistries (Figure S7) further improves the functionality and stability of the TA system through continuous Au sputtering on the dual-complex structure. That is, the sandwich configuration with specific geometries and surface chemistries increases the active modification sites of thermo-responsive PNIPAAm, thus producing more sensitive responses while ensuring the stability of the gating liquid inside the porous membrane, thereby enlarging the threshold difference of the liquid gating substantial air transmembrane critical pressures ($P_{\text{Critical (air)}}$), the forces required to overcome the capillary pressure at the liquid-air interface under different temperatures (Figure 2C, left). As shown in Figure 2C (right), the air must deform the pore-filling gating liquid

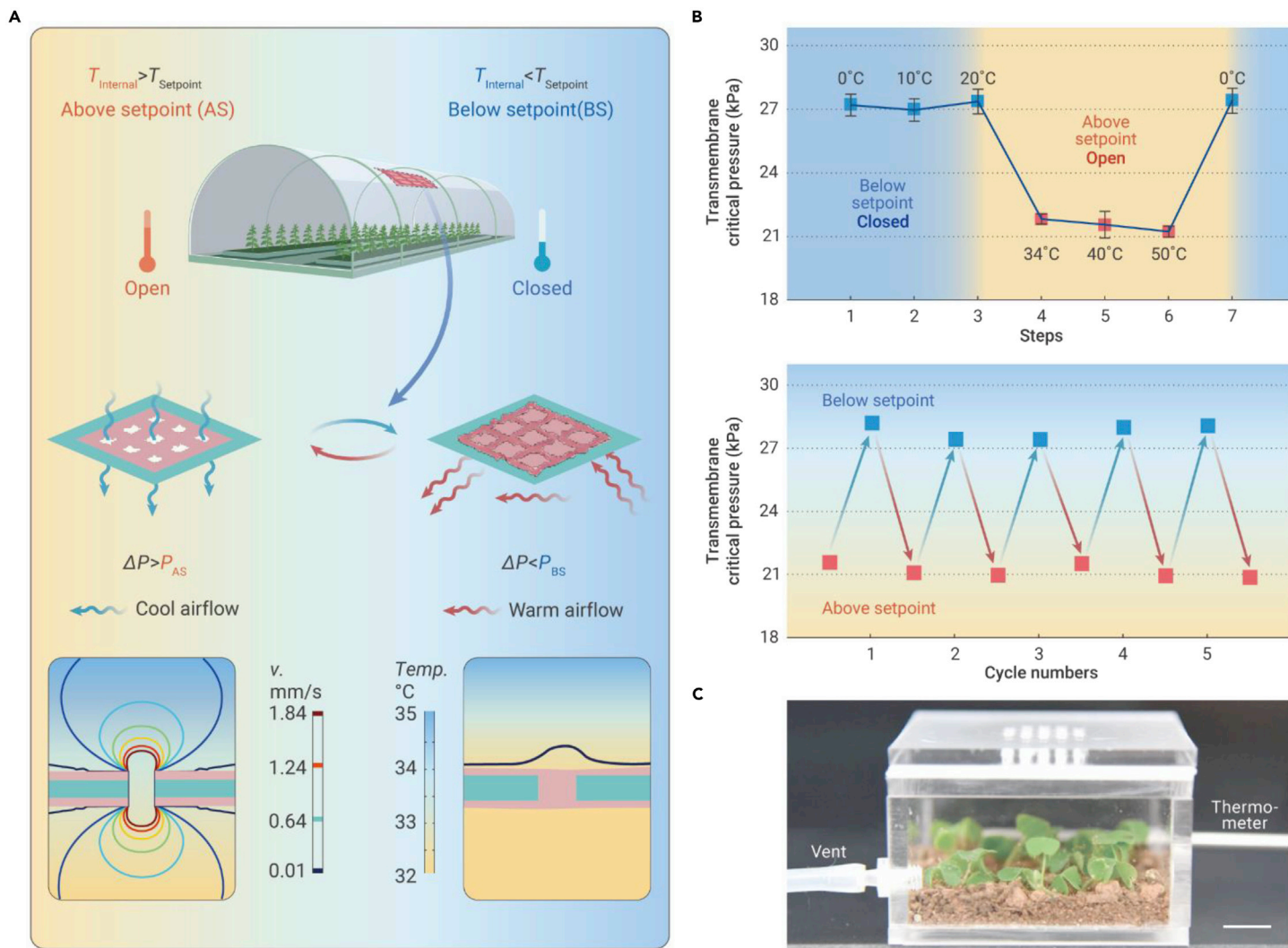


Figure 3. Schematic of a TA system with temperature control used in a greenhouse application (A) Adaptive ventilation of a TA system-patched greenhouse. If the internal temperature is above the setpoint and the pressure difference (ΔP) between internal and external space is higher than the air transmembrane critical pressure (P_{AS}), the TA system opens, cool air from outside enters the greenhouse, and ventilation starts. When the temperature is below the setpoint, and ΔP is lower than the air transmembrane critical pressure (P_{BS}), the TA system is sealed by gating liquid and thermally insulated. Conceptual simulations using COMSOL Multiphysics also show the air velocity and temperature change at open and closed states. (B) Transmembrane critical pressure required for air movement through the TA system at different temperatures. Error bars indicate SD (top). Cyclability of the TA system for air through temperatures alternating below and above the setpoint (bottom). (C) Optical image of a self-designed experimental greenhouse prototype for internal temperature monitoring. Scale bar: 1 cm. Energy saving of the TA system in a greenhouse application.

interface to enter the pores. Because of the stronger affinity for gating liquid to the thermo-responsive membrane below the LCST, a mild pressure difference (ΔP) between both sides of the membrane cannot slide open the liquid gate, but as the temperature increases, the membrane stabilizing the liquid through wetting tends to be flaccid, so the air will create an open, liquid-lined pathway through the liquid-sealed pores. Because this liquid gating mechanism involves structural reconfiguration, rather than expulsion of the liquid, the pores will be thermodynamically primed to close as soon as the temperature drops below the LCST. In the demonstration, through continuous Au sputtering on the dual-complex structure, the sandwich configuration is achieved, and there is an obvious increase in the difference of the air transmembrane critical pressure from 2.5 to 6.0 kPa. In addition, the pore sizes of the membrane directly determine the air transmembrane critical pressure, which can be regulated accurately to adapt to different pressure environments for the thermal adaptive system (Figure 2D). Moreover, through the composition design of the gating liquid, the influence of volatilization and other factors during its function has been reduced. For example, in an environment in which heat radiation and natural convection prevail, water as a gating liquid cannot be sustained for a longer time, but a dual-component 70% (v/v) glycerol/water solution as a gating liquid can greatly reduce the volatilization loss under natural convection and maintain very stable gating performance even at 0°C, under which condition water freezes (Figures S5 and S8).

Fluid temperature control in greenhouse application

Because greenhouse structures are not open to the atmospheric environment, the temperature inside can be easily built up and maintained.^{45,46} However, ventilation that ensures the movement of air to regulate temperature and CO₂ concentration can be the key to a successful greenhouse, and proper ventilation techniques can save considerable energy by reducing heating and cooling expenses.^{23,31} Thus, a TA system that responds to temperature stimuli to open and close a liquid gate can be provided as an adaptive ventilation system for greenhouse applications, with energy-saving features (Figure 3A). When using a TA system patched on a greenhouse, a constant pressure difference (ΔP) is applied between the internal and external spaces, which is higher than the air transmembrane critical pressure (P_{AS}) above the set temperature and lower than that (P_{BS}) below the set temperature. The cool air from outside would open the TA system patch and penetrate into the greenhouse, providing ventilation when the internal temperature is higher than the setpoint and creating a thermally insulated space by liquid-sealing the TA system patch when the temperature falls. Additionally, the motion of air can be quantitatively predicted by the change of ΔP instantaneously, and the maximum velocity of convective flow can reach a higher rate when the TA system opened (1.64 times than that of closed state), even with a small temperature difference. The modified greenhouse model displays thermal adaptive pressure control and stable performance (Figures 3B and 3C; Figures S9–S11). Thus, the TA system patch with adaptive

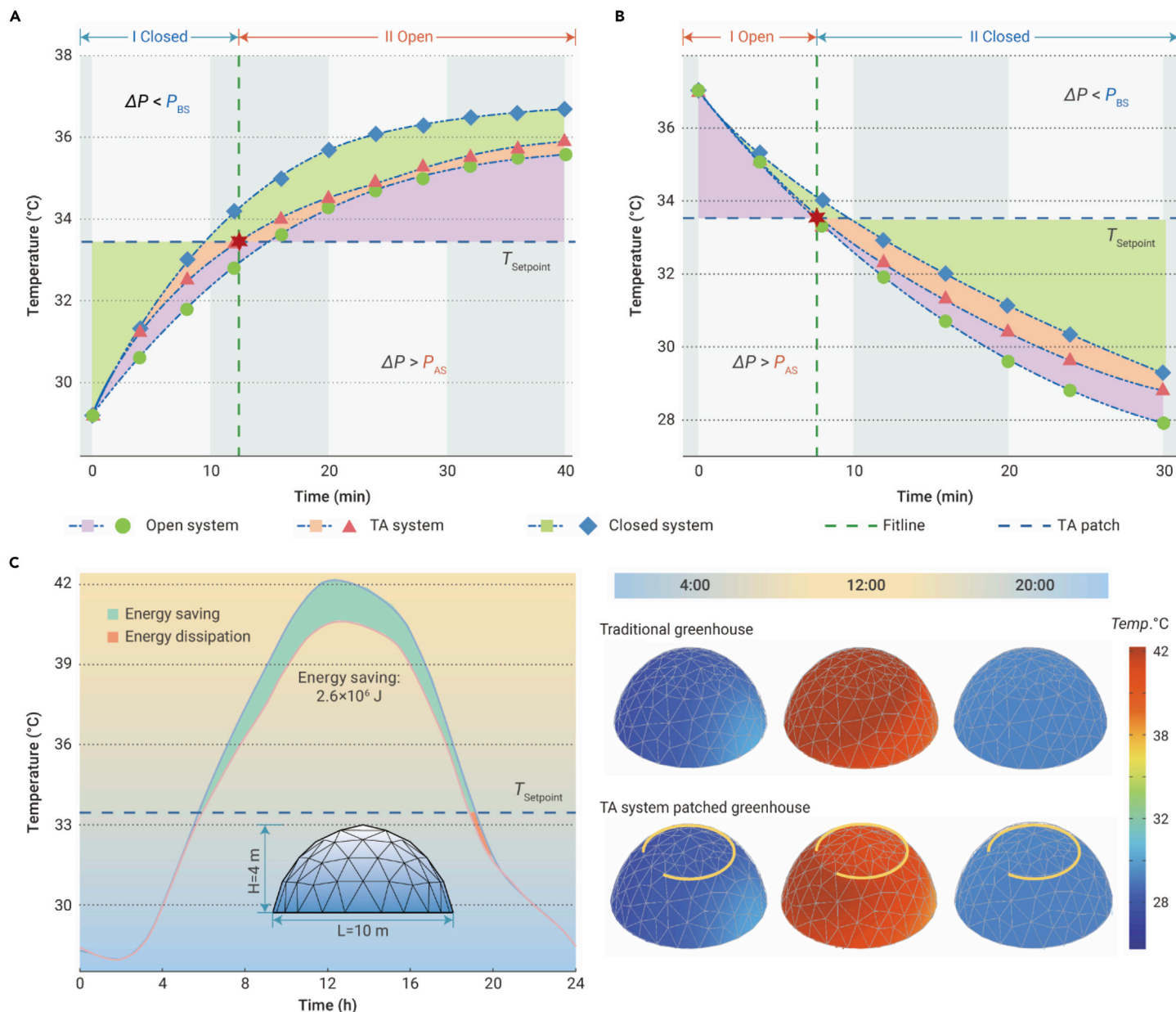


Figure 4. Energy consumption for the closed, open, and TA systems to maintain a set temperature in a greenhouse application and simulation of energy-saving mechanism (A and B) Internal temperature monitoring in real time and comparison of energy consumption for maintaining a set temperature of the closed, open, and TA systems during heating and cooling processes. (C) Thermal transfer simulations of typical dome structural greenhouse for 24 h. The left panel shows the temperature changes in traditional and TA system-patched greenhouses; the dome structure is exhibited as an inset. The right panel shows temperature distributions of the two greenhouses at 4:00, 12:00, and 20:00.

opening produced by a liquid filler can be obtained for the potential application of a smart greenhouse.

Additionally, in our systems, thermal energy relates to temperature T only when the temperature gradient ΔT is small, so we can directly use the microcosmic internal energy equation to quantitatively compare the energy consumption of the three systems to maintain a set temperature (Figures 4A and 4B):

$$U(T, V) = nC_{v,m}T = C_{v,m}TV/V_m \quad (\text{Equation 1})$$

Here, we assume air as a typical diatomic gas, and $C_{v,m}$ can be set at $5/2R$. n denotes the amount of substance of the air. V_m is the molar volume of air, which depends on its temperature and pressure. R is the gas constant. The internal temperature detections of the three systems in real time reveal that the TA system with self-regulated open and closed thermal transfer passages consumes less energy to maintain the set temperature under a heating-cooling cycle than the other two systems (Figure S12).

Considering the compromises between the cost and time duration in maintaining optimal energy performance, we conduct a model from our experimental device to demonstrate that the optimal use-area ratio of the TA membrane to the

area of the whole greenhouse membrane is 22.7% (Figure S13; Note S4). According to the theory of heat transfer, heat transfer efficiency relates to the effective contact area only when the total heat energy is constant,⁴⁷ which indicates that the optimal use-area ratio can be generally suitable for any structural greenhouse. Therefore, to take a commercial dome-structured greenhouse as an example,²⁸ analysis and simulations,⁴⁸ including thermal, laminar fluid, and solar radiation of this greenhouse with the TA system patch or traditional insulation materials for 24 h, are conducted using COMSOL Multiphysics, as shown in Figure 4C (left) (Note S5). Convective mass transfer, heat transfer of conduction, and radiation occur in all three systems. The energy balance leads to⁴⁹

$$q_{input}St + Q_{initial} = q_{conv}tS\phi + q_{cond}th + q_{rad}tS(1 - \phi), \quad (\text{Equation 2})$$

where q_{input} , $q_{conv} = h_{conv}(T - T_{\infty})$, $q_{cond} = \lambda(T - T_{\infty})$, and $q_{rad} = \epsilon(G - e_b[T])$ are the heat flux of input, convection, conduction, and radiation, respectively; h_{conv} and λ are the convective and conductive heat transfer coefficients, which are different among the three systems. T is transient temperature, and T_{∞} is ambient temperature. S is the area of the membrane, ϕ is porosity, t is time, ϵ is surface emissivity, G is irradiation, and $e_b(T)$ is the blackbody hemispherical total emissive

power. The amount of energy saved in the greenhouse with the TA patch for maintaining the set temperature is $\sim 2.6 \times 10^6$ J during 1 day (Note S5). The temperature distribution comparison at 4:00, 12:00, and 20:00 (Figure 4C, right) for the two greenhouses shows the energy-saving mechanism of the TA system-patched greenhouse (Table S1). At 12:00, when solar radiation is intense, the average temperature of the greenhouse with the TA patch is lower compared with the other model, and the temperature difference is especially obvious at the TA membrane, which is caused by its adaptive ventilation and indicates better performance in thermostat applications. The total energy saving could be evaluated by transient heat transfer simulation. As the 2020 estimate statistics show that the global greenhouse plantation area accounts for 6,550 km² (1,618,400 acres),²⁹ and by adopting these data in our simulation, the result shows that the use of the thermal adaptive system could lead to an energy saving of 7.9×10^{13} kJ/year (a percentage change of $\sim 11.6\%$).

CONCLUSIONS

To sum up, we have established an energy-saving thermal adaptive system with a bistable, interfacial, sandwich-structured design, which displays adjustable openings for thermal convection passages, allowing a shift between closed and open systems. Enabled by the sandwich configuration of thermo-responsive porous membranes with affinitive gating liquids, the proposed thermal adaptive system produces different opening behaviors for different temperature ranges around the setpoint and exhibits temperature control by minimizing the variations between the actual and desired temperatures. Additionally, by using the sandwich configuration of porous membranes with affinitive gating liquids, the TA system is demonstrated to be an energy-efficient greenhouse patch, achieving smart “breathing” to provide proper ventilation. We believe that this prototype of an energy-saving thermal adaptive mechanism can open new avenues for further extending the scope of using liquid-based adaptive systems in practice to achieve the goal of carbon neutrality.

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AUTHOR CONTRIBUTIONS

X.H. conceived the idea. X.H. and B.C. designed the research. B.C. performed the experiments. M.Z., Y.H., and H.W. implemented the numerical calculations. X.H., B.C., M.Z., and X.C. analyzed and interpreted the results. X.H., X.C., B.C., M.Z., R.Z., and Y.F. drafted the manuscript. All authors contributed to discussions of the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

SUPPLEMENTAL INFORMATION

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