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VIEWPOINT

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Pumping into a cool future: electrocaloric materials for zero-carbon refrigeration

Existing commercial heat pumps, such as air conditioners (A/C) and refrigerators, possess a coefficient of performance (COP) up to and in some cases even higher than 6. However, most existing space heating techniques rely heavily on fossil fuels as their direct (burning) or secondary (electric heating) energy supply. Space heating is responsible for 45% of building emissions. In particular, approximately 4.3 Gt of CO_2 was released into the atmosphere in 2019 for heating building spaces, representing approximately 12% of global energy and process-related CO_2 emissions, according to the most recent International Energy Agency (IEA) estimation [1]. With its inherited advantages in energy efficiency, heat pump technology has considerable potential in the building sector, especially for space heating. Currently, most of the purchased heat pump units are applied for space cooling. In 2019, space cooling consumed 15% of the energy used for space heating, which is close to 1 Gt CO_2 as an indirect input in global carbon footprint [1]. Meanwhile, the direct equivalent CO_2 emissions cannot be ignored, as most heat pumps in the market still utilize hydrochlorofluorocarbons (HCFCs) that possess a high global warming potential (GWP) [2].

The carbon emission for space cooling is far from reaching its peak. Recently, there has been a surge in electricity use in several major cities that are experiencing heatwaves more frequently than in previous decades, indicating a continuous climate shift, hence causing more A/C-induced CO_2 emissions in the coming years. In addition, the IEA assessed that approximately 35% of the global population lives in areas with a high number of cooling degree-days (a metric that depicts the cooling needs), only 15% own an air conditioner (see Fig. 1).

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By 2050, considering the improvement in living standards in developing countries and the climate shift, this share is projected to be quadrupled, i.e., 60%. With the stated policies scenario (STEPS), the IEA expected that the electricity consumption for cooling to be more than doubled by 2060. Therefore, the technical advances in refrigeration have been ranked first in the drawdown of CO_2 emissions [3].

A major factor affecting the building energy cost is the operating temperature range or the "neutral-band", which is the temperature range in which the centralized HVAC systems require no heating or cooling. In a building with central A/C system, the neutral band is usually set between 21 °C and 24 °C, which is tighter than the ANSI/ASHRAE standards. To meet the sustainable development scenario, the society calls for disruptive innovations in heat pump technologies, which should be scalable, customizable, exhibit no direct and indirect CO_2 emissions, capable of being operated for a considerable time, low noise, and economical. However, currently commercialized technologies were not equipped with above advantages to answer the urgent call.

In several alternative heat pump technologies currently under development, electrocaloric (EC) refrigeration may be integrated in as one of the solutions to ease the urge. Electrocaloric effect (ECE) links two dipolar entropy states by applying and removing electric fields to condensed matter (see Fig. 2). The phenomenon of the ECE was first discovered in 1930 [4]. However, the cooling effect discovered then was too weak. Hence, it was considered impractical compared to the conversed physical effect of pyroelectrics. In this century, the prediction and direct observation of the giant electrocaloric effect (GECE) has revived this research field [5,6]. The 5–10 K of temperature change induced by electrical charging/discharging of condensed matter suddenly made ECE utilizable in several potential applications.

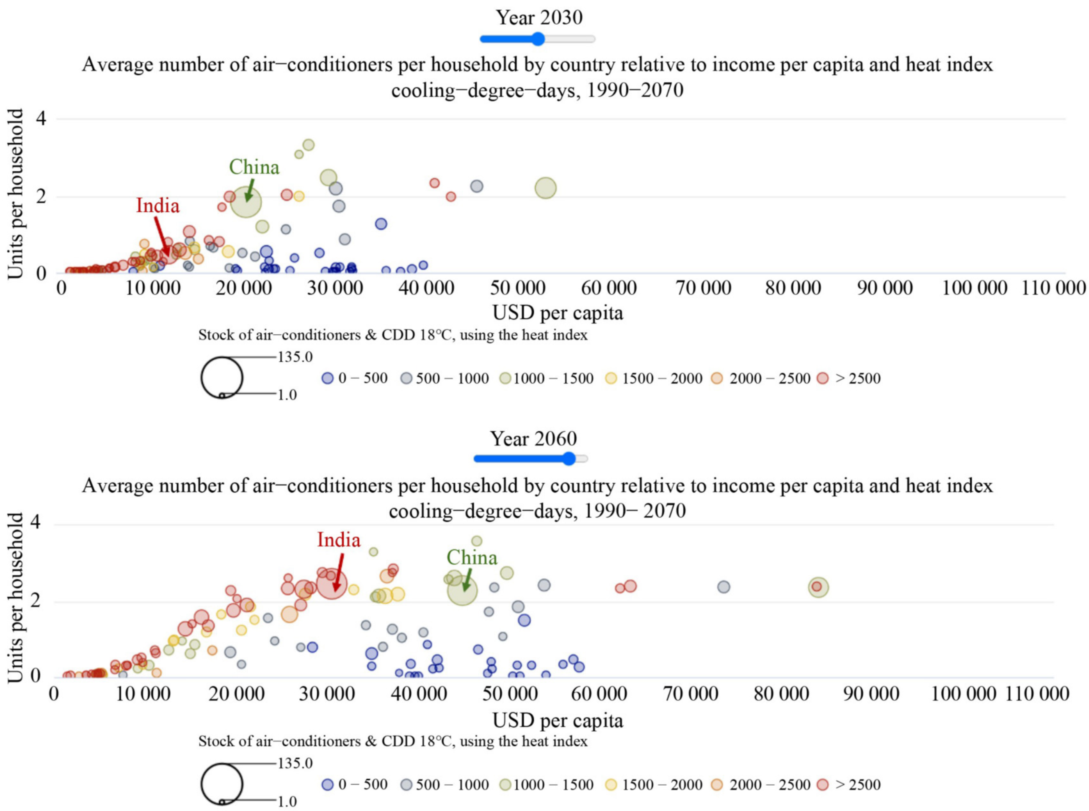

Fig. 1 Estimated average number of A/C units per household by country according to income per capita and heat index cooling degree days, in 2030 and 2060 by IEA (adapted from Ref. [1]).

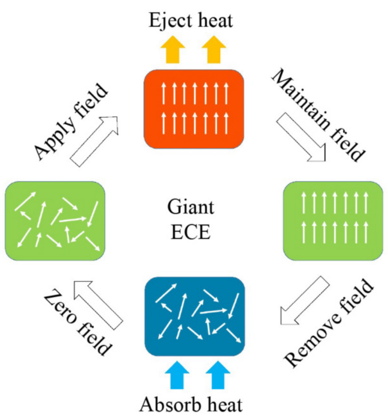

Fig. 2 Schematics of an EC refrigeration cycle.

Despite the EC fluid [7], which has been predicted in fluidic liquid crystals, the EC materials discovered are generally solids [8]. The cooling effect has been demonstrated in organic and inorganic ferroelectrics, such as lead-based [9] and lead-free ceramics [10], single crystals [11], and in polyvinylidene fluoride (PVDF)-based ferroelectric polymers [12]. Owing to the high demand for energy reversibility, the material should be processed into a state of relaxor ferroelectrics [13]. Moreover, considering the large pool of inorganic ferroelectrics, the inorganic EC materials being studied are perovskites that are $PbTiO_3$-based, $BaTiO_3$-based, $KNbO_3$-based, $SrTiO_3$-based single crystals, ceramics, solid solutions, and several 2-D inorganic materials. A recent breakthrough has been achieved in multilayer ceramic capacitors (MLCCs) of $PbSc_{0.5}Ta_{0.5}O_3$, in which a large ECE is directly recorded in a thermally bulk material that can be fabricated by commercial manufacturing processes [14].

The polymeric EC materials may lead to more flexural applications. Polymers, such as rubber bands, are mechanically/dielectrically strong and can withstand large strains without failure. In addition, they can be easily fabricated into thin films, multilayer capacitors, etc., via the roll-to-roll processes, which do not require ultrahigh temperatures. An electrically driven polymeric refrigerant can provide thermal solutions to address the challenges of numerous emerging applications. A plethora of EC polymer-based nanocomposites have also been studied [15,16]. To date, EC polymer-based cooling devices have been demonstrated to have high performance and unique device configurations [17–20].

Similar to magnetocaloric (MC) heat pumps, where magnetic entropy is utilized to form thermodynamic cycles, many EC heat pumps have been developed using various geometries, such as linear, rotary, and cascaded; different heat transfer mechanisms, such as solid-fluid and solid-solid heat transfer; and active regeneration [21,22]. The EC working body is essentially a dielectric capacitor exhibiting zero-GWP and is highly efficient during the charging and discharging process; hence, it enables low-carbon heat pumps. In addition, the uniqueness of the EC heat pumps depends on the system size and weight. By utilizing electricity directly and efficiently, the EC heat pumps can be designed to be compact, and if necessary, even miniaturized. This is because their power source, control board, and other axillary parts can be integrated into daily electronics. The EC devices have demonstrated the highest device-specific cooling power among the major alternative refrigeration technologies under development [23].

EC heat pumps can be utilized as localized thermal management systems (LTMSs), offering solutions for reducing building heating and cooling energy requirements, as shown in Fig. 3. The LTMSs modify the local thermal environment surrounding the human body rather than the entire building, enabling wider neutral band temperature setpoints without the loss of human comfort. Hoyt et al. showed that annual energy savings for four different cities when setpoints were expanded from a typical baseline of 21–24 °C [24]. Setpoint expansions of only 2 °C in each direction yielded more than 15% energy savings. Thus, the LTMS solutions that reduce the need for tightly controlled building environments lay the foundation for new sustainable architectures in next-generation building designs. For many emerging application scenarios such as localized and wearable cooling, lightweight, compact size, and even pliable heat pumps are highly desirable in addition to energy and environmental concerns, whereas conventional VCC-based heat pumps may not meet these requirements.

Current challenges in the further implementation of EC technology remain unaddressed in many aspects (TRL3-5). It is essential to prioritize the material properties in material engineering to allow large-scale manufacturing and integration and maintain the high quality of the working substance. Li et al. proposed the design of an EC device with kilowatt cooling power using state-of-the-art EC materials [25,26]. The EC material for a practical device should integrate into a working body with considerable refrigeration capacity to handle real-world tasks [14]. Research on thin films [27] (nanometer scale) should translate into material systems with a large refrigeration capacity comparable to the external thermal load. Considering the EC cooling body characteristics, reliability and long operation lifetimes are as important as significant cooling effects [28]. Therefore, it is essential to operate them at low fields [29], which should be 20%–30% of the breakdown field of the integrated dielectrics. Considering the potential application in wearable cooling devices, voltage application is also a key issue in addition to electric fields. It is also crucial to reduce the specific heat of the working body, as the final goal is to pump heat in the ambient environment rather than to change the temperature of the working body itself. To achieve zero-carbon refrigeration, the EC heat pumps should be studied regarding how to integrate them into renewable energy systems, such as on-site solar photovoltaic (PV), storage (battery systems), and micro-grids. To meet the demands for the dual-carbon goal and zero-carbon thermal management solutions, the society needs quick implementation of advanced heat pump technologies, which may be personalized, localized, and wearable, so that they can be gradually merged into current technical strategies, serving as supplementary technologies in applications for which the existing

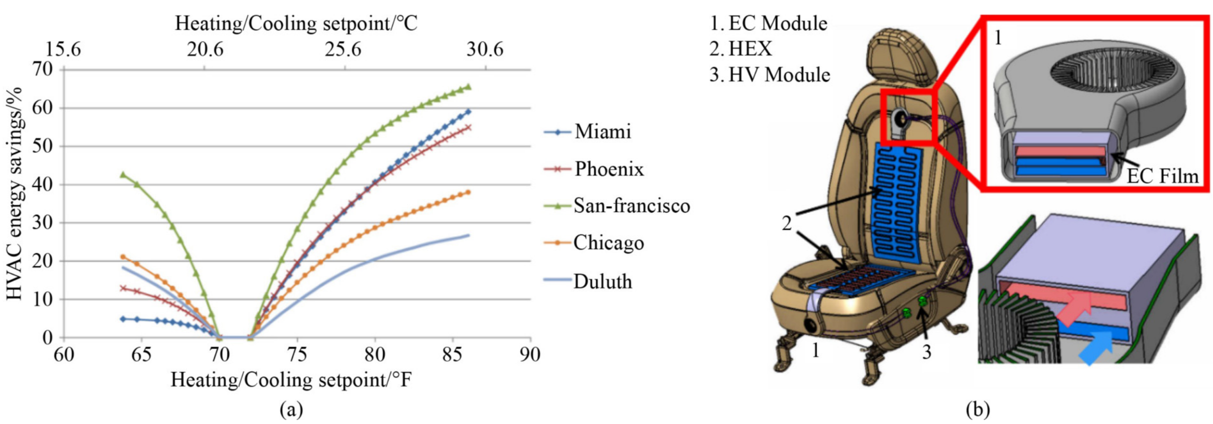

Fig. 3 Localized thermal management could draw down the energy consumption in building sector.
(a) Summary of average HVAC energy savings over five model types (excluding High-Existing-VAV Fixed), compared to baseline (adapted with permission from Ref. [24]); (b) localized cooling seat designed for electric vehicles, provided by X. Qian group at Shanghai Jiao Tong University. Copyright 2015 and 2019 Elsevier, respectively.

commercial technologies are not designed.

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