ELECTROCALORICS

## It's not about the mass

Electrocaloric cooling devices are traditionally based on sub-millimetre-thick ceramic working bodies. Using instead a flexible polymer that is one order-of-magnitude thinner yields lightweight devices that have now been stacked to pump heat across a relatively wide temperature span.

Xavier Moya and Neil Mathur

There is currently growing interest in cooling devices based on caloric materials, in which phase transitions, and thus thermal changes, are driven near room temperature by magnetic fields, electric fields or mechanical fields[1]. The electric fields are easy to administer as one simply applies a voltage, but the resulting electrocaloric (EC) effects can only be driven hard without breakdown in layers no thicker than several tens of microns. This timeless axiom was responsible for limiting the temperature change of the working bodies that were used in the very first EC prototypes, namely sub-millimetre-thick plates of a monolithic ceramic[2-4] (Fig. 1a). By contrast, sub-millimetre-thick multilayer capacitors (MLCs) that show larger changes of temperature[5] have only just been exploited in prototype EC coolers[6,7] (Fig. 1b,c). Switching to a low-density material, Qibing Pei (UCLA) and colleagues described in 2017 an EC device based on a 0.1 mm-thick polymer bilayer[8] (Fig. 1d). Writing now in *Nature Energy*[9], Pei and colleagues show that four such lightweight devices can be stacked to pump heat across a wider temperature span.

The temperature span between the hot and cold ends of a cooling device need not be limited by the voltage-driven temperature change that can be achieved under adiabatic conditions in an EC material on which it is based. Using passive regeneration, which was originally demonstrated using magnetocaloric gadolinium[10], a large temperature gradient may be established along a column of fluid that is translated with respect to a set of immersed EC working bodies. Fig. 1b shows a single set of working bodies in a single column of fluid[6], while the doubling shown in Fig. 1a permits antiphase operation[2-4]. The working bodies dump heat at one end of the fluid column due to field application, absorb heat at the other end due to field removal, and passively exchange heat with the fluid while the translation is in progress. Alternatively, a temperature gradient can be developed along a line of EC working bodies that are translated to make and break contact with a second such line[7] (Fig. 1c). Both lines undergo suitably timed EC cycles, and the two lines represent a discontinuous active regenerator, such that one could aspire to optimize each working body for the temperature at which it operates.

Conceptually, it is perhaps simpler to repeatedly translate a single EC working body between a hot end where it dumps heat, and a cold end where it absorbs heat[11]. In their 2017 report[8], Pei and colleagues employed this strategy in an elegant way by electrostatically actuating the flexible EC bilayer itself (Fig. 1d), but device temperature span was limited to 2.8 K. By stacking four such lightweight devices, they have now achieved a temperature span of 8.7 K at zero cooling power, or a cooling power of 906 mW at zero temperature span[9]. These figures are similar to the figures of 13 K and 1.22 W for the prototype with ceramic MLCs and passive regeneration[6] (Fig. 1b). This equivalence would appear to challenge the conventional wisdom that it is desirable to employ the largest possible net mass of EC material, which should be distributed to achieve a large surface-area-to-volume ratio, given the low thermal conductivity of EC materials.

The ability to stack four devices was facilitated by the simplicity and compactness that arose from electrostatically actuating the EC bilayers. By thus avoiding the pumps and actuators that have been employed elsewhere (Fig. 1a-c), it was possible to reduce thermal deadweight and unwanted thermal pathways. Moreover, two positive outcomes arose from the fact that each bilayer device in the stack was cycled in antiphase with respect to a neighbour. First, the instantaneous flow of heat was relatively smooth because one bilayer could absorb heat from its cold end while its counterpart dumped heat at its hot end, cf. the antiphase operation to which we referred in the context of Fig. 1a. Second, energy efficiency was improved because each discharging bilayer could help charge its neighbour, as originally demonstrated using two MLCs[11].

In the future, it may be challenging to improve the cooling power of EC devices based on flexible bilayers, as the active thermal mass is small. However, the aforementioned similarity of performance with respect to an MLC-based prototype[6] (Fig. 1b) gives cause for optimism. Therefore it remains to be seen whether the push for applications will be best served by using flexible polymer bilayers with a relatively low active mass that is accompanied by a relatively low inactive mass, or inflexible ceramic MLCs with a relatively high active mass that is accompanied by a relatively high inactive mass. But if the circumstantial correlation between active and inactive mass were broken then it might be all about the mass, after all.

Xavier Moya and Neil Mathur
Department of Materials Science, University of Cambridge, Cambridge, CB3 0FS, UK
e-mail xm212@cam.ac.uk, ndm12@cam.ac.uk

**References**

1. X. Moya and N. D. Mathur *Science* DOI:10.1126/science.abb0973 (2020).
2. Y. V. Sinyavsky et al. *Ferroelectrics* **90,** 213-217 (1989).
3. Y. V. Sinyavskii & V. M. Brodyansky *Ferroelectrics* **131,** 321-325 (1992).
4. Y. V. Sinyavskii, *Chem. Petrol. Eng.* **31,** 295-306 (1995).
5. B. Nair et al. *Nature* **575,** 468-472 (2019).
6. A. Torelló et al. *Science* **370,** 125-129 (2020).
7. Y. Wang et al. *Science* **370,** 129-133 (2020).
8. R. Ma et al. *Science* **357,** 1130-1134 (2017).
9. Y. Meng et al. *Nature Energy* **XXX,** XXX-XXX (2020).
10. G. V. Brown, *J. Appl. Phys.* **47,** 3673-3680 (1976).
11. E. Defay et al. *Nat. Commun.* **9,** 1827 (2018).

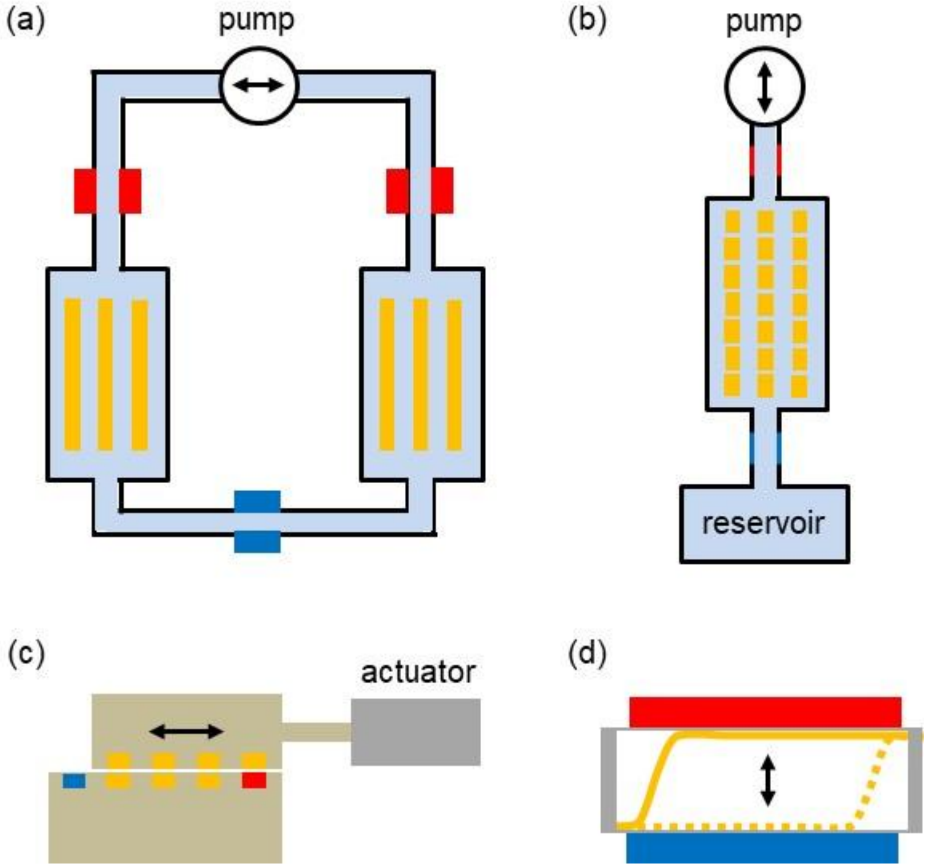


**Fig. 1. Electrocaloric prototypes through the ages.** Simplified schematics, not to scale, show devices that pump heat from cold ends (blue) to hot ends (red) by cyclically driving electrocaloric (EC) working bodies (yellow) with a voltage while cyclically translating either (a,b) a fluid or (c,d) the EC working bodies themselves. The working bodies comprise (a) sub-millimetre-thick plates of PST ($PbSc_{0.5}Ta_{0.5}O_3$, ), (b,c) sub-millimetre-thick MLCs of PST, and (d) a 0.1 mm-thick polymer bilayer of P(VDF-TrFE-CFE), that is, poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene). Images are based on references published in (a) 1989-1992[2-4], (b) 2020[6], (c) 2020[7] and (d) 2017[8]. The new work by Meng *et al*.[9] stacks four of the devices shown in (d).