



Materials research and development needs to enable efficient and electrified buildings

Shuang Cui, Adewale Odukamaiya,*^{ORCID} and Judith Vidal*

Because of the complexity of modern buildings—with many interconnected materials, components, and systems—fully electrifying buildings will require targeted R&D and efficient coordination across those material, component, and system levels. Because buildings that consume the smallest amount of energy are easier to electrify, energy efficiency is a crucial step toward fully electrified buildings. Materials advances will play an important role in both reducing the energy intensity of buildings and electrifying their remaining energy use. Materials are currently being explored, discovered, synthesized, evaluated, optimized, and implemented across many building components, including solid-state lighting; dynamic windows and opaque envelopes; cold climate heat pumps; thermal energy storage; heating, ventilating, and air conditioning (HVAC); refrigeration; non-vapor compression HVAC; and more. In this article, we review the current state-of-the-art of materials for various buildings end uses and discuss R&D challenges and opportunities for both efficiency and electrification.

Introduction

Modern buildings require energy: to provide occupant comfort, operate appliances and devices, and illuminate interior and exterior spaces. In 2018, residential and commercial buildings in the United States used 20.4 and 18.3 quadrillion Btu (quads), or 20.6% and 18.5% of total US primary energy use, respectively.¹ On a primary energy basis, electricity comprises a majority of building energy use: 27.8 quads or 71.9% of all building energy use. Direct natural gas use in buildings is limited to a few end uses, such as heating, water heating, cooking, and clothes drying, but still represents 8.4 quads or 21.6% of primary energy use. As such, electrifying buildings is essential to reducing energy consumption and improving energy efficiency. In buildings, electrification involves substituting fossil-fuel fired combustion driven technologies with electric technologies, as well as improving efficiency of end uses that are currently powered by electricity. For example, replacing gas-powered furnaces with heat pumps for space heating, and replacing electric resistance water heaters with heat pump water heaters.²⁻⁴

In this article, we review advanced materials that enable efficient and electrified buildings, and discuss the possible benefits and barriers these materials might present to greater

electrification in buildings.⁵ **Figure 1** depicts the electric alternatives that exist for all major energy end uses in buildings, such as building envelopes, heat pumps, and heating, ventilating, and air-conditioning (HVAC) systems. The electrification of buildings with smart controls and thermal energy storage (TES) systems will benefit the future grid by providing greater flexibility and resilience, especially when integrating a large amount of variable renewable energy resources such as wind and solar. It also provides synergistic opportunities for electric vehicles (EVs) and distributed storage. The electrification of buildings for increased efficiency is also an important pathway to decarbonize the energy system.

State of the art and opportunities

Building envelopes

The building envelope affects HVAC energy use; these end uses are among the largest contributors to total energy use in buildings. The thermal performance of building envelopes (both opaque and transparent) represents a key opportunity to increase the energy efficiency of the buildings sector and to reduce greenhouse gas emissions.

Shuang Cui, The University of Texas at Dallas, USA; Shuang.Cui@utdallas.edu
Adewale Odukamaiya, National Renewable Energy Laboratory, USA; Wale.Odukamaiya@nrel.gov
Judith Vidal, National Renewable Energy Laboratory, USA; Judith.Vidal@nrel.gov
*Corresponding author
doi:10.1557/s43577-021-00241-x

Opaque envelope

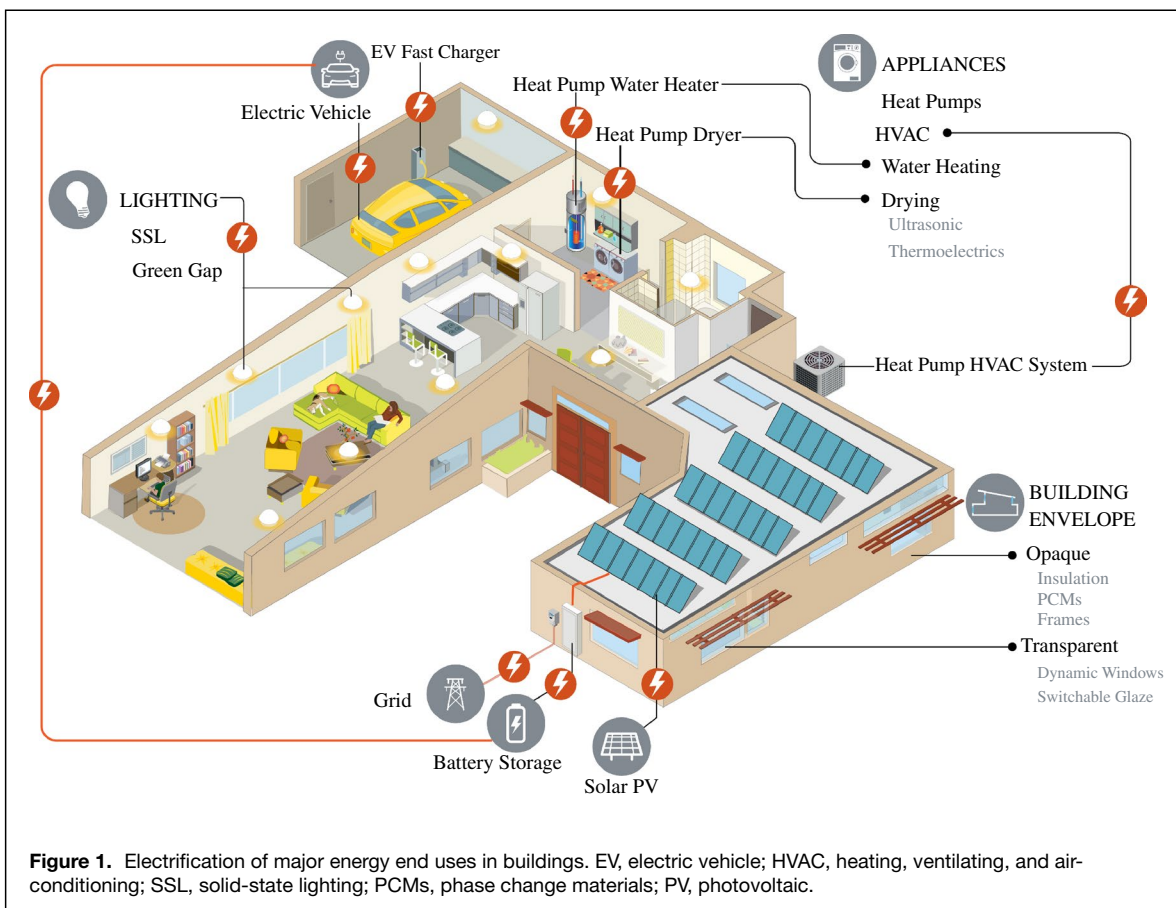
The opaque building envelope—walls, roof, and foundation—affects 25% of building energy use, or 10% of total US primary energy use.⁶ Improving the energy performance of the opaque envelope in US buildings is critical to reducing the total electricity use. The most common ways are reducing air leakage, improving moisture management, and increasing R-value—a measure of how insulative a wall/envelope section is—through passive and/or dynamic solutions. Passive solutions include:

- Insulation materials with ultra-high R-value for reducing the energy and cost of new constructions and retrofits of existing buildings; for example, aerogels, vacuum-insulating panels, and nano-insulation materials
- TES systems (heat and moisture) for shifting the energy demand, improving thermal comfort, and offsetting energy use
- Radiative cooling roofs for reducing energy use

The most common passive materials are thermal insulation materials.⁷ Adding thermal insulation materials in buildings is a simple yet highly energy-efficient method that can be applied to residential, commercial, and industrial buildings.

The principle of insulation materials is to reduce heat loss or heat gain, which leads to the reduction of energy use for space heating/cooling. Insulation materials can be categorized into two main classes per their function in manipulating the heat transfer: mass insulation and reflective insulation.

Mass insulations are those that retard the heat flow by conduction, whereas reflective insulations are ones that reduce the amount of heat transfer by radiation. Various substances, such as fiberglass, polystyrene, polyurethane foam, mineral wool, aerogels, and vacuum-insulating panels, are typically used as mass insulations.^{8,9} These substances have a high thermal resistance, thus retarding the heat flow rate by conduction. When applied to buildings, the effectiveness of mass insulation highly depends on the thickness. For example, the thicker the insulation layer, the better the performance in terms of reducing heat loss or heat gain.⁹ Besides conventional mass insulation materials, hydrogels and phase change materials (PCMs) have attracted attention for the thermal management of buildings over the past two decades.^{10,11} Hydrogels are hydrophilic polymers and can absorb >90 wt% water. They provide evaporative cooling by mimicking the transpiration or perspiration of plants and humans. By applying hydrogel coatings to the roofs of buildings, heat dissipation is enhanced through the evaporation of water inside the hydrogel, enabling surface temperature



reductions. The evaporative cooling capacity depends on the water storage capacity inside hydrogels.

PCMs have also attracted attention over the past few decades. PCMs, like salt hydrates and paraffin wax, can store and release the thermal energy associated with the latent phase change with slight temperature changes upon melting and solidification.¹² Integration of PCMs into building materials (e.g., concretes, plaster, and stuccos)¹³ can effectively reduce and shift energy use. Among all different forms of PCMs, shape-stabilized PCMs—which encapsulate solid-liquid PCMs within porous matrices to prevent the leakage of liquid phase during the melting—are promising due to ease of implementation.^{12,14} The phase-change properties, such as transition temperature, energy density, and enthalpy-temperature curve shapes, influence the energy saving potential depending on the application climates and operating conditions.¹³ Reflective insulation is thermal insulation that reflects radiative heat and is usually used in home attics, roofing, and wall systems.¹⁵ This insulation reduces the solar heat gain and prevents heat transfer due to a reflective (or low emissivity) surface.⁷ Reflective insulation utilizes one or more low-emittance reflective surfaces that enclose air spaces. An ordinary reflective insulation material is white paint, which has high values of solar reflectance (up to 0.9) to reflect most of the solar energy in the visible spectrum (0.4–0.7 μm).¹⁶ Using radiative cooling materials is another effective way to reduce the cooling energy required by buildings.¹⁷ Previously, radiative cooling materials were limited to nighttime because radiators with strong thermal radiation lack high reflectivity in the solar radiation band. With the recent technological advancements in radiators, such as photonic radiators and metamaterials, the advantages of diurnal radiative cooling has been demonstrated¹⁸ by involving the reflection of sunlight (wavelengths ~ 0.3 – 2.5 μm) and emission of long-wave infrared (wavelength ~ 8 – 13 μm) radiation through the respective atmospheric transmission windows into outer space. Other passive solutions include a combination of some materials discussed above, like combining an evaporative hydrogel and radiative cooling material.¹⁹

In addition to the passive materials mentioned before, dynamic and active building envelopes also require materials that can change thermal capacitance or heat transfer at critical times in response to electric grid needs and interior/exterior conditions. The requests for flexible operation could be only a few days per year (e.g., reliability-based demand response), or on a daily, hourly, or even continuous basis, which depends on the market and grid condition at that given time. Dynamic and active building envelopes have co-benefits for both energy efficiency and grid flexibility. To achieve this, non-linear thermal transport materials or devices with variable thermal conductance are needed.²⁰ The most common materials for dynamic building envelopes are dynamic insulations, thermal diodes, and thermal switches.^{21,22} These materials allow building walls to be insulated on a hot day to prevent heat from leaking in from the outside by switching the envelopes to a low thermal conductivity mode. But if the

temperature drops at night, the walls could detect the change and switch to conducting mode to allow heat from the building to escape, thereby enabling free cooling. The efficiency of the dynamic materials highly depends on the switch on/off ratio (conducting versus insulating). Some static materials can also be used to achieve dynamic building envelopes. For example, a thermal diode composed of thermo-responsive hydrogels and PCMs is a non-linear structure featured by a preferential directional transport of heat, analogous to the electrical diode. It can achieve a rectification ratio (a ratio of the larger heat flux or thermal conductivity to the smaller heat flux or thermal conductivity) > 2 based on the mechanism of the temperature dependent thermal conductivity by increasing or reducing the effective thermal conductivity at the forward or reverse mode.^{22,23} Dynamic PCM research is also increasing to achieve tunable TES systems. By fine-tuning the transition temperature of PCMs, TES can be active not just in winter or summer, but throughout the whole year.

Transparent envelope

The transparent envelope—windows—is responsible for $\sim 10\%$ of energy use in buildings. Modern windows provide improved thermal performance, including reduced air leakage but also offer enhanced amenities such as daylighting and views to the outdoors. Presently two approaches are used in glazing systems to control solar heat gain and minimize heat loss/gain. Passive solutions include:

- Highly insulated glass units with multiple panes or transparent thermally resistive inserts in between panes, such as aerogels, inert gas, or vacuum
- High-performance frames (polymer-fiber composites and unplasticized polyvinyl chloride frames with multiple frame cavities filled with insulating materials; air infiltration/exfiltration through interfaces between operable window elements)
- Daylight redirection for redirecting sunlight deeper into spaces
- Static photovoltaic (PV) glazing
- Low-emissivity coatings

Most passive mass insulations used in opaque envelopes cannot be directly applied to transparent envelopes due to their opacity. Instead, low-emissivity coatings can be applied to glazing to reduce solar heat gain. A low-emissivity coating for glazing should show spectral selectivity; this means high transmittance in the visible range, but high reflectance in the near-infrared range.²⁴ This technology transmits energy from the visible portion of the solar spectrum into buildings, while minimizing the amount of ultraviolet and infrared light or heat energy that passes through. The most common low-emissivity materials that work for windows are metal oxides, such as zinc, tin, bismuth, and titanium oxides. PV glazing is another popular research topic.²⁵ Organic- or inorganic-type PV cells are placed between the two glass panes. They reduce

glazing transparency and absorb less solar heat, thus reducing the cooling load. Another advantage of this type of glazing is that the PV provides small-scale electricity generation. It can also be a part of building-integrated PV, widely used on facades or roofs.

More attention recently has been given to switchable/smart/dynamic/adaptative glazing applications, particularly electrically activated switchable devices and systems.²² Dynamic facades and glazing with variable solar heat gain control characteristics act in response to diurnal and seasonal changes in heating and cooling demand, occupancy, and available daylight. Current state-of-the-art dynamic glazing technologies—using thermochromics and electrochromic materials—attenuate both visible and near-infrared wavelengths. Decoupling switching in the visible and near-infrared ranges would enable independent control of glare (tinting) while admitting some solar heat gains in the winter, or restricting some solar heat gains and allowing in daylight in the summer, particularly for buildings that are occupied during daylight hours.^{26,27} The most used dynamic glazing materials are:

- Thermochromics and thermotropic materials for transmitting/blocking the sunlight for energy efficiency
- Electrically actuated switchable materials like electrochromic materials
- Electrically self-powering materials (e.g., using PV cells) for automated attachments^{10,25}

Assuming the same payback period target of 5 years as with static high-performance windows, dynamic glazing will require estimated window cost of \$2.9/ft² for residential buildings and \$14.6/ft² for commercial buildings by 2030.²⁶ These values correspond to an operating range between 0.05 and 0.65 solar heat gain coefficients, which represents a climate-zone-specific compromise between reductions in heating energy use and increases in cooling energy use.

A thermochromic material changes its color by changing its crystal structure above a particular environmental temperature. This type of material can be inorganic and polymer based, including iron oxide, iron silicide, niobium dioxide, nickel silicide, and titanium oxide.²⁸ Vanadium oxide thermochromic glazing is the most popular. It shows a colored phase at 20°C and a colorless phase at 30°C. Other thermochromic materials are nanocrystalline transparent conducting oxide films, particularly tin-doped indium oxide and aluminum-doped zinc oxide, which have been demonstrated to have effective electrochromic properties with near-infrared-only switching.²⁹ Those materials show fast response times, and aluminum-doped zinc oxide has also shown good cycling durability. The combination of thermochromic materials and low-emissivity materials can also enhance performance by showing higher visible and infrared reflectance in the colorless phase than in the colored phase.³⁰ In addition, PCMs and hydrogels can be applied to the transparent envelope as well and are considered thermotropic

materials. These materials can switch from transparent to opaque to avoid excessive solar heat gain and to control glare. Recent studies show that double-glazing windows with PCM fillings can help to reduce solar heat gains in buildings. PCM can also store considerable amounts of energy and release it back to the building for heating later.²⁵ Hydrogels contain more than 90 wt% water and are highly transparent. They can be either sealed between two glass panes or coated onto a single pane of glass depending on the application conditions and chemical compositions. For example, double network hydrogels have been demonstrated to provide effective cooling when coated onto a glass. The temperature of the glass can be reduced by 10°C compared to glass without hydrogel coating due to evaporation cooling.^{10,28}

The main types of electrically controllable active materials include electrochromic glass, suspended particles, liquid crystals, metal hydrides, and plasmonic effects in wide-bandgap electrically conducting nanoparticles.³¹ Among these materials, electrochromic is the most suitable for a building's transparent envelopes considering performance and cost. Transition metal oxides and organic materials can change color due to oxidation or reduction reactions autonomously and reversibly as a response to an external electrical stimulus. Oxide-based electrochromic materials (e.g., tungsten oxides and iridium oxides) are the most common type in buildings. Switching between different control states requires a minimal amount of electricity (2.5 W/m²), and even less is needed to maintain a desired tinted state (less than 0.4 W/m²). Nanocrystal in-glass composite is the most promising electrochromic emerging technology to improve overall performance. It allows regulating transparency separately and independently toward visible light or near-infrared light from the clear to the tinted state.³²

Due to the low energy required, many electrochromic windows can be self-powered by a PV/battery system (1) to avoid increasing the amount of grid supplied electricity consumed and (2) to give a readily installed autonomous device.³³ Depending on the types of solar cells and electrochromic materials, PV-powered electrochromic devices can be classified as Dye-sensitized solar cell based PV-electrochromic, silicon-based PV-electrochromic, semi-transparent perovskite PV, or solid-state electrochromic cells enabling solid-state photovoltachromic devices. Other dynamic alternatives include elastomer-deformation tunable windows and liquid infill tunable windows, which rely on mechanical control.³⁴

Heating and cooling

In the United States and other developed countries, heating and cooling account for roughly 40%³⁵ of total building energy use, with electricity and natural gas being the major contributing fuel types. Meeting building energy efficiency goals is not possible without addressing heating and cooling loads. Additionally, a major barrier to the electrification of buildings is heating in cold climates, where fossil fuels such as natural gas currently account for a significant share of heating supply. Materials advancements

will need to play an important role in reducing the carbon footprint of buildings through energy efficiency and electrification.

Currently, most non-fossil-fuel-based heat pumps (heating and cooling) systems are vapor compression based. Next-generation non-vapor compression heat pump technologies such as vapor sorption, solid-state, and chemical heat pumps have been under development, but have not achieved significant commercial deployment. Materials advancements can play a significant role in improving the performance of vapor-compression heat pumps in the near- to medium-term, while next generation technologies are developed in the long term.

Vapor compression heating and cooling

Vapor compression refrigeration and heat pump systems have significant negative environmental impacts owing to refrigerant charge leaks and their high global warming potential (GWP).³⁶ Consequently, there is a need to develop alternative refrigerants that have zero ozone depletion potential (ODP) and low GWP that can be dropped into existing vapor compression systems. One study estimates that shifting from baseline refrigerants such as R-404A and R-410A to low-GWP refrigerants in commercial refrigeration and residential HVAC could lead to a ~30% drop in kg of CO_{2eq} emitted.³⁶ Desirable characteristics of the next generation of refrigerants include: zero ODP, low GWP, short lifetime in the atmosphere, and high efficiency refrigerants that are also non-toxic, non-flammable, and environmentally friendly.³⁷ Currently, hydrofluorocarbons (HFCs) are the refrigerant of choice in the United States. Many of the HFCs in use today have no ODP, are non-flammable, recyclable, and energy efficient, but they have high GWP³⁸ because they are potent greenhouse gases that survive in the atmosphere for many years. Examples of materials R&D to support phasing out HFCs includes computational screening of alternative refrigerants/small molecules,³⁹ complete experimental characterization of new candidate refrigerants, including determination of thermodynamic properties,⁴⁰ heat transfer and pressure drop measurements,⁴¹ and evaluation of cycle performance.^{42,43} The US Department of Energy Building Technologies Office has identified high-priority initiatives that would have either a direct effect on maintaining or improving energy efficiency while switching to next-generation low-GWP refrigerants.³⁸

Heat pumps use electricity to harness heat from surrounding air, water, or the ground and then pump that heat indoors, essentially acting as an air conditioner operating in reverse.⁴⁴ Heat pumps can be used to heat air for space heating, for water heating for domestic hot water, and for hydronic heating systems. Unlike resistance or fuel-fired heaters that simply convert electricity or fuel to heat, heat pumps pump heat from a lower temperature (exterior) to a higher temperature (interior), and therefore, can achieve efficiencies 3–6× higher than conventional heating technologies.⁴⁵ Due to this higher efficiency and the opportunity to run on electricity from clean/renewable sources, heat pumps have gained popularity with electric utilities as an efficient, low-carbon heating technology.

Despite the obvious efficiency and carbon benefits, heat pumps have struggled to operate efficiently in cold climates, and have been limited to more moderate climates, leaving natural-gas-fired heating as the technology of choice to heat homes in cold climates.⁴⁶ Electrifying and consequently decarbonizing buildings will largely depend on developing heat pump technologies that can perform well in cold climates, eliminating the need for fossil-fuel-based heating. Technologists are currently working on this—to further advance cold climate heat pumps, recent work has focused on multi-stage, variable-speed, or booster compressors; advanced refrigerant management; improved defrost control; and alternative refrigerants.⁴⁷

Material development will play a strong role in the latter three improvements. For example, air-source heat pump evaporators are prone to frosting in cold climates, which can severely limit their performance because the insulating ice layer restricts heat transfer, increases the required fan power, and requires an energy-intensive defrost cycle.⁴⁸ To mitigate these issues, researchers have developed frost-resistant surfaces and coatings to limit frost accumulation to a small area (hygroscopic or biphilic surfaces)^{49,50} or to encourage droplet removal to prevent or delay frost growth.^{51,52} Although historically these engineered surfaces have been limited to small samples, inexpensive batch coating processes have been developed in recent years to treat a fully assembled heat exchanger.^{48,53,54} These coated evaporators have been shown to delay frost formation by 3×, reduce defrost energy consumption by 50%,⁴⁸ and boost average system efficiency in cold climates.⁵⁴ Other opportunities for improving the performance of vapor compression heat pumps exist, such as better heat exchanger materials and working fluids with better heat transfer performance, without sacrificing ODP or GWP.⁴⁷

Non-vapor compression heating and cooling

In the medium to long term, eliminating vapor compression heat pumps altogether in favor of advanced non-vapor compression heat pumps such as vapor sorption, solid-state, and chemical heat pumps could drastically improve heating/cooling efficiency and provide further opportunities for deeper electrification. Absorption systems are one type of vapor sorption cycle that utilize thermal energy to drive a heat pump where a refrigerant is absorbed and desorbed from a secondary fluid in cycles.^{55,56} Absorption heat pumps can be configured to provide heating, cooling, or both through a reversible cycle. Although cooling efficiencies are typically less than those for vapor-compression systems, absorption heat pumps offer large potential energy and cost savings, especially for heating-dominated climates. Materials advances that would advance the state-of-the-art of absorption heat pumps include: developing benign refrigerant pairs or introducing a third working fluid that inhibits the crystallization process (toxicity and crystallization are main challenges),⁵⁷ using microchannel⁵⁸ or membrane absorbers and desorbers,^{59,60} and utilizing a cascade reverse-osmosis system to separate the refrigerant-absorbent pair in place of a thermal generator.⁶¹

Adsorption heat pumps are another type of vapor sorption heat pump. In its most basic embodiment, an adsorption heat pump comprises an adsorbent material that is packed or coated on an adsorbent bed, an evaporator, a condenser, an expansion valve, and a working fluid (adsorbate) to move heat in and out of the adsorbent bed.⁶² A critical determinant of the performance of adsorption heat pumps is the choice of adsorbent-adsorbate working pair. Examples of common adsorbent-adsorbate pairs that have been studied extensively in the literature include zeolite-water,⁶³ carbon-methanol,⁶⁴ and silica gel-water.⁶⁵ While many working pairs have been proposed, they are often expensive or unavailable in large enough quantities. There is a need for adsorbent-adsorbate working pairs that are abundant, low cost, environmentally friendly, and can be regenerated with low energy input. There is also an opportunity to improve adsorbent material properties, in particular, increasing the thermal conductivity and permeability,⁶⁶ which is challenging because enhancing one typically diminishes the other. Recent efforts have investigated coating the adsorbent in a thin layer onto the heat exchanger surfaces to improve heating/cooling power density by reducing heat transfer resistance.⁶⁷ Developing novel adsorbent formulations and processing techniques optimized for coating heat exchangers could lead to important improvements in adsorbent heat pump technology.

Absorption and adsorption systems are thermally driven cycles, so currently, they are typically gas driven. However, there is an opportunity to drive these systems using waste heat sources, reducing energy use, and improving space conditioning efficiency. As previously mentioned, the thermal generator can also be replaced by a reverse-osmosis system to enable the use of electricity to drive these systems.

Unlike refrigerant or fluid-based systems that compress or pump a working fluid around in a cycle, solid-state heat pumps have few or no moving parts and can be configured to not require a working fluid.^{68,69} This provides several potential advantages over conventional systems. No moving parts means silent operation, higher reliability and system lifetime, and no working fluid means no potential for refrigerants with GWP or ODP to leak out into the atmosphere. Solid-state heat pumps include cycles based on the magnetocaloric, electrocaloric, elastocaloric, and barocaloric effects.⁶⁸

The general principle of caloric heat pumps is the application of an external field or stimuli causing a phenomenon with an associated temperature change in a caloric material, which can be exploited to provide heating or cooling.⁶⁸ In magnetocalorics, the external stimulus is a magnetic field, and the resulting phenomenon is magnetization. In electrocalorics, it is an electric field, and polarization. In elastocalorics, it is applied mechanical stress, and resulting strain. In barocalorics, it is an applied pressure field, resulting in a volume change. Magnetocaloric materials (e.g., Gd LaFeSi or Fe₂P) have been investigated for several decades, while their electro-, elasto-, and baro- counterparts have only been investigated extensively for a little more than a decade.⁷⁰ Future

improvements in magnetocaloric heat pumps could arise from materials improvements such as new magnetocaloric material combinations (e.g., ones with first- and second-order phase transitions) to achieve high specific power and adiabatic temperature change,^{71,72} and novel particle and bed geometries.⁷¹ The field of barocalorics is still very nascent. Recently, barocaloric effects have been demonstrated in various materials, including magnetic alloys, ceramic ferroelectrics, ionic salts, fluorides, organic-inorganic perovskites, and even rubber.⁷⁰ Material discovery to identify barocaloric materials with large entropy change is ongoing. Material discovery can also play an important role in the field of elastocalorics, where materials with large elastocaloric effects are needed.⁷³ Due to the cyclic operation of caloric heat pumps, enabling material technologies to manage heat flow such as thermal switches, diodes, and heat pipes are also needed.⁷⁴

Separate sensible and latent cooling for humid climates

Traditional air conditioners serve dual functions in humid climates: sensible cooling (i.e., reducing air temperature to comfortable levels), and latent cooling (i.e., reducing indoor humidity to comfortable levels). This typically requires cooling the air temperature beyond what is necessary in order to condense enough water vapor out of the room air as it is passed over the air-conditioner's indoor heat exchanger.⁷⁵ This is detrimental to the efficiency of vapor-compression air conditioners. To mitigate this, approaches to provide independent control (separate) over the sensible and latent portions of cooling have been under development for some time.⁷⁶ It has been estimated that separate sensible and latent cooling (SSLC) has the potential to provide energy savings of 30% and greater.⁷⁷ Several approaches have been proposed for SSLC, which can be categorized as systems approaches (e.g., using a variable speed compressor or cascading vapor compression cycles), and materials approaches.^{75,78} In materials approaches, liquid desiccants, solid desiccants, or membranes that trap or manage moisture in the form of water vapor with little-to-no change in air temperature are integrated into the air-conditioning system.⁷⁹⁻⁸² The biggest opportunity for materials advances to support desiccant-based SSLC is in developing and evaluating solid and liquid desiccants with high moisture ad/absorption capacity (grams of water vapor per gram of dry desiccant) with low regeneration energy requirements.⁸³ It is also important to characterize steady state and transient properties of new high-potential desiccants to support modeling of SSLC concepts. For membrane-based SSLC, higher permeability, higher strength, lower-cost membranes that are durable and resistant to fouling are needed, in addition to membranes that have higher selectivity of water to air (or H₂O/N₂).⁷⁹

Equipment/appliance integrated thermal energy storage

Achieving high levels of renewables penetration will require large amounts of cost-effective energy storage due to the

inherent intermittency of most renewable sources. Electricity consumption load profiles in buildings are dominated by thermal end uses (e.g., space heating, space cooling, water heating, refrigeration, clothes drying), especially during peak electricity consumption periods,⁸⁴ which also often do not align with renewables generation.⁸⁵ This creates a significant need for behind-the-meter energy storage in buildings. TES can be deployed at lower cost than electrochemical energy storage to meet this need.²⁰ There is a huge opportunity to develop packaged heating/cooling equipment and appliances with onboard TES to shift loads of these individual end uses. TES can be in various forms, including sensible, latent, and chemical. Early examples of building appliance/equipment-integrated TES have been reported in the literature.^{86–88} To support this, TES materials that are energy and power dense, low cost, and have long lifetimes are needed. High thermal conductivity is important to achieve high power density, but traditional approaches increase thermal conductivity at the expense of energy density because they displace PCM volume.⁸⁹ Furthermore, TES materials that can dynamically tune their phase change temperature or other properties to operate optimally in heating and cooling seasons are needed to increase the utilization factor of TES, lowering the leveled cost.⁸⁵ As demonstrated by Woods et al.⁸⁹ defining material property targets should be driven by component- and system-level performance requirements.

Water heating and clothes drying

In 2014 in the United States, water heating and clothes drying accounted for 3.54, and 0.68 quads of energy, representing 9% and 2%, respectively, of total primary energy use in buildings.⁹⁰

For water heating, fuel-based heaters (mostly natural gas with some propane and fuel oil/kerosene) accounted for about 50% of total water heating energy use, with only ~25% from electric heating.⁹¹ Of that 25%, 24% is from electric resistance heating, and only 1% is from heat pump water heaters (HPWH).⁹² Electric resistance heaters convert each unit of electricity to one unit of heat, whereas heat pumps convert each unit of electricity to greater than one unit of heat (typically 1.8–2.5⁹³) because they pump additional heat from a lower temperature to a higher temperature.^{44,94} Consequently, HPWHs are significantly more energy efficient than electric resistance heaters. One of the major challenges preventing greater adoption of HPWHs is that they cool the space that they are in, because they extract heat from ambient air. This causes an issue in heating-dominated climates because occupants do not want the water heater producing cold air inside the house, but it is too cold to place the heater outside. One strategy for mitigating this problem is to utilize CO₂ as the refrigerant. It can operate in much colder ambient temperatures, so the water heater can be placed outside. This strategy also solves the problem of using high-GWP refrigerants, as state-of-the-art HPWHs typically use R134a or R410a as the refrigerant. HPWHs can also benefit from integrating PCMs

for thermal storage. A major disadvantage of HPWHs is that they heat slower than electric resistance water heaters. The current solutions are to add backup electric resistance elements (which require a 240-V circuit), increase tank size (which is space inefficient), or increase temperature set point (which creates safety concerns). Alternatively, a high energy density PCM can be integrated, allowing the tank to store more thermal energy compactly.^{95,96}

Currently, the clothes drying market is dominated by electric resistance dryers in residential buildings and gas dryers in commercial buildings. There is a need for heat pumps in electric drying; however, similar to HPWHs, heating is slow, leading to excessively long dry times. Cost is also a challenge. Recently, technologists have developed alternative approaches such as thermoelectric dryers⁹⁷ and ultrasonic dryers.⁹⁸ Patel et al. showed that existing BiTe thermoelectric technology can be performance competitive with vapor compression heat pumps for clothes drying,⁹⁷ but lower cost thermoelectric materials are needed. Ultrasonic dryers operate by using piezoelectric transducer-induced high-frequency vibration to expel water out of fabric as a cold mist.⁹⁹ This approach is very promising because mechanical extraction is far more energy efficient than evaporation.¹⁰⁰ Lead zirconate titanate ceramic is the most widely used material for fabrication of ultrasonic transducers. Although lead zirconate titanate is well established and low cost, consumers may not take well to the presence of lead oxide, even though they only pose a potential hazard if the transducer becomes damaged or chipped. Consequently, alternative piezoelectric transducer materials would be welcomed. The piezo assembly in each transducer is subjected to billions of cycles per dry cycle, so robust piezoelectric materials with minimized hysteresis that have fewer defects and are less prone to fatigue failure are needed. Additional desirable properties include high piezoelectric charge constant, higher mechanical quality factor reducing mechanical loss, low dissipation factor ensuring cooler operation, and high dielectric stability. There is also a fabrication challenge limiting scale up. Current commercially available transducers are limited to ~50 mm diameter. Larger diameter transducers would facilitate scale up.

Solid-state lighting

Advances in solid-state lighting (SSL) technology could unlock \$50 billion in annual energy savings, amounting to about a 5% reduction in the total primary energy budget of the United States.¹⁰¹ The efficiency of LED lighting (luminous efficacy) is measured in units of lumens per watt (lm/W), with continued R&D advancing toward the practical limit of 255 lm/W for state-of-the-art approaches, and the ultimate theoretical limit of 325 lm/W for next-generation technology,¹⁰² compared to current state of the art, which is around 100–150 lm/W.¹⁰³ White light is, most effectively, a combination of blue, green, amber, and near-red, which requires emitters in the 440–460 nm, 530–550 nm, 570–590 nm, and 610–620-nm wavelength bands, respectively. State-of-the-art

emitters are III-V semiconductors such as GaN/InGaN LEDs, which do not emit efficiently in the green/amber wavelengths (500–600 nm) due to a lattice mismatch between GaN and the high (>25%) In-composition InGaN layers required to reach these wavelengths.^{104,105} The lack of efficient emitters in this wavelength range is known as the “green gap.” Currently, white light is achieved by down-converting a high-intensity blue GaN/InGaN emitter to green, amber, and near-red wavelengths by coating with phosphors.¹⁰⁶ This approach, known as phosphor-converted LED (pc-LED), is fundamentally inefficient because the excess energy of the blue light is dissipated as heat.

Reaching the ultimate theoretical luminous efficacy limit will require moving from pc-LEDs to direct emitting color-mixed LEDs, which means improving the efficiency of green/amber emitters. Current approaches involve developing non-polar or semi-polar GaN growth and substrates to reduce the impact of polarization fields (a major source of efficiency loss)¹⁰⁷ or developing new emitter materials that are more efficient.¹⁰⁸ The first approach requires advancements in semiconductor growth and fabrication to enable low-cost, large-area GaN substrates grown or sliced in the non-polar orientation.¹⁰⁷ The latter approach requires new semiconductor material systems that emit efficiently in 500–600 nm wavelengths, have the right lattice constant, possess a direct bandgap, and are synthesizable with sufficient crystal quality such as ZnGeN₂ recently demonstrated by researchers at the National Renewable Energy Laboratory and The Ohio State University.^{109,110}

Another major challenge with nitride LEDs is droop due to Auger recombination, where there is an efficiency loss at high current when brighter LEDs are needed.¹¹¹ A deeper fundamental understanding of non-radiative recombination mechanisms is required to enable device designs that minimize current droop. Efficient approaches to heat dissipation, or materials that are more tolerant to high temperature, are also needed to avoid efficiency loss due to heat, known as thermal droop. Finally, current materials used as phosphors in pc-LED are rare-earth-containing materials,¹¹² which suffer from high cost and material supply chain problems. There is therefore a need for phosphors that are free of rare-earth elements.

Materials challenges for efficient and electrified buildings

This review can largely be boiled down to five materials challenges, which if addressed, would enable significant strides toward efficient, electrified buildings:

1. TES materials
2. Zero GWP refrigerants and materials for non-vapor compression cycles
3. Materials to decouple dehumidification and cooling (separate sensible and latent cooling)
4. Dynamically tunable insulation for building envelopes
5. Materials to increase the efficiency of solid-state lighting

Advances in TES materials, components, and systems have the potential to disrupt building envelopes, heating/cooling equipment, water heating, and clothes drying. TES can provide much needed storage at a low enough cost to enable deployment at large enough scales needed to integrate large amounts of intermittent renewable sources to the grid. Refrigerants, on the other hand, are used in vapor compression cycles, which are important to space heating/cooling, water heating, and drying. As we transition away from fuel-fired heating in the winter, heat pumps will be the main alternative for heating, adding a large number of additional equipment and appliances utilizing refrigerants. Transitioning to refrigerants with zero GWP that are also efficient, non-flammable, and non-toxic will be critical to decarbonizing the buildings sector. Doing away with refrigerants entirely by moving to non-vapor compression cooling/heating cycles would also solve this challenge. In addition, separating sensible and latent cooling can greatly reduce cooling energy requirements in humid climates, leading to more energy-efficient buildings that are easier to electrify. Similarly, having dynamic control over when a building’s envelope is insulative and when it is not would allow buildings to leverage diurnal temperature changes for free heating/cooling from the ambient, lowering the need for mechanical heating/cooling, and increasing the building’s energy efficiency. Last, advances in materials for solid state lighting could increase luminous efficacy of LED lighting by solving the infamous “green gap” and droop problems, which could unlock \$50 billion in annual energy savings, amounting to about a 5% reduction in the total primary energy budget of the United States.

Besides the end-use-specific challenges, there are also five cross-cutting materials challenges: (1) scalability, (2) durability, (3) dynamic functionality, (4) circularity, and (5) equity. Considering the lifetime and scale of buildings, the durability and scalability of the materials, especially new materials, are always concerns for builders and homeowners. Although most of the envelope materials, such as PCMs and hydrogels, have been demonstrated with repeatable performance without degradation for hundreds of cycles, it is still below the minimum thousands of cycles required by the buildings industry. The scalability of those materials—without compromising properties, such as mechanical properties and energy-storage capacity—is also very challenging. Adding dynamic functionality to the materials and enabling effective active control of the materials is even more difficult when considering the interaction with the grid for efficiency. Two other challenges that are not discussed in detail in this review are circularity and equity. The construction industry is one of the world’s largest consumers of energy and raw materials, and the largest source of solid waste, so the circularity of the building materials is critical to decarbonize industrial processes. This will require three main tenets: (1) designing out waste and pollution from materials, (2) extending the useful life of materials, and (3) regenerating or recycling the materials. Equity is seldom discussed in the

buildings industry, even though historically, research, development, and demonstration of clean energy technologies has prioritized first adopters, increasing social inequities. If equity is not centered throughout R&D in order to intentionally identify and address unique challenges, new innovations will not scale and achieve national/global impact.

Acknowledgments

This work was co-authored by the National Renewable Energy Laboratory, operated by Alliance for Sustainable Energy, LLC, for the US Department of Energy (DOE) under Contract No. DE-AC36-08GO28308. The views expressed in the article do not necessarily represent the views of the DOE or the US Government. The US Government retains and the publisher, by accepting the article for publication, acknowledges that the US Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for US Government purposes. The authors would like to thank B. Sparr, A. Mahvi, J. Woods, R. Tenent, L. Wheeler, B. Tellekamp, A. Momen, K. Gluesenkamp, V. Patel, and M. Deru for helpful discussions in drafting this review.

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Shuang Cui is an assistant professor in the Department of Mechanical Engineering at the University of Texas at Dallas (UTD) and a joint faculty in the Building Energy Science Group at the National Renewable Energy Laboratory (NREL). Her research focuses on both fundamental study of nanoscale heat transfer and energy conversion and advanced materials development, spanning intelligent soft materials/devices for moisture control and composite phase change materials for thermal energy storage. She is highlighted by the U.S. Department of Energy's "Women @ Energy: STEM Rising" website. Cui can be reached by email at Shuang.Cui@utdallas.edu.



Adewale Odukamaiya is a research engineer and Director's Fellow in the Building Energy Science Group at the National Renewable Energy Laboratory. His research focuses on innovating materials for heat transfer and energy storage in ways that improve building efficiencies and support low-carbon buildings. This research applies fundamental materials science, heat transfer, and thermodynamics to advanced energy materials and components, with an emphasis on thermal and electromechanical energy storage technologies and their advanced manufacturing. Odukamaiya can be reached by email at Wale.Odukamaiya@nrel.gov.



Judith Vidal is the manager of the Building Energy Science Group, the sub-program lead of Buildings Emerging Technologies, and a distinguished member of research staff at the National Renewable Energy Laboratory, and a joint faculty at the Colorado School of Mines. She has established an international reputation for her cutting-edge work on thermal systems, thermomechanical and chemical optimization of materials, and advanced manufacturing. Vidal has diversified her expertise and capabilities in several technologies including building technologies, and high temperature components, among others. She has several patents and has published many impactful journal articles. Vidal can be reached by email at Judith.Vidal@nrel.gov.