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# Bio-inspired effective and regenerable building cooling using tough hydrogels

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#### HIGHLIGHTS

- Application of tough hydrogel as regenerable 'sweating skin' for building cooling.
- Tough gel exhibits effective evaporative cooling and extraordinary cyclability.
- Charging and discharging capability of the tough gel was retained after 50 cycles.
- Tough gel cooling could lead to substantial energy saving in buildings.

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#### ABSTRACT

# Innovative thermal regulation technologies could provide great potential for reducing energy consumption in buildings. In this work, we report, for the first time, the application of highly stretchable and tough double network hydrogels (DN-Gels) as durable and reusable 'sweating skins' for cooling buildings. These DN-Gels demonstrate outstanding cooling performance, reducing the top roof surface temperature of wooden house models by 25-30 °C for up to 7 h after only a single water hydration charge. More importantly, compared with single network hydrogels (SN-Gels) previously studied for cooling applications, these DN-Gels exhibit extraordinary toughness and cyclability due to their interpenetrated ionically and covalently cross-linked networks, as demonstrated by constant cooling performance over more than 50 cycles. This excellent cyclability is further demonstrated by the unaltered mechanical properties and charging capability of the hydrogels after many cycles, compared to fresh ones. By coating a 100 m<sup>2</sup> roof of a single house with tough DN-Gels, it is estimated that the annual electricity consumption needed for air conditioning can be reduced by ~290 kW h with associated CO<sub>2</sub> emission reductions of 160 kg. Our results suggest that bio-inspired sweat cooling, specifically using tough DN-Gel coatings, represents a promising energy-efficient technology for cooling buildings as well as other devices and systems.

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#### 1. Introduction

In most countries, residential and commercial buildings are one of the highest energy consumption sectors. In the United States in particular, over 40% of energy consumption and greenhouse gas emissions are related to building temperature regulation [1]. Notably, building energy consumption is still increasing at a rate of 0.5–5% annually in developed countries [2] and is expected to increase even more rapidly in developing countries. Therefore, alternative building thermal regulation technologies, based on passive systems, have been extensively studied over past decade [3–5], such

as night-time ventilation in moderate or cold climate [6,7], high infrared (IR) reflective coatings for reducing energy uptake [8,9], and phase change materials (PCMs) for thermal energy storage [10–12]. Nevertheless, no current passive cooling technologies used in buildings possess ideal characteristics such as high cooling efficiency under a variety of weather conditions, high durability including resistance to thermal cycling and UV irradiation, and low cost. For example, PCMs are less effective under high solar intensity fluctuations due to their low latent heat (~hundreds of kJ kg<sup>-1</sup>). More efficient and durable cooling materials and systems are needed for sustainable building cooling.

In nature, plants and animals are autonomously adaptive to increases in environmental temperature through transpiration and perspiration of water, which has one of the highest latent heats among fluids. Inspired by such passive biological cooling processes,





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several self-adaptive technologies involving bio-inspired artificial skins have been reported [13–17]. One of the more promising materials is based on superabsorbent polymers, or hydrogels, which can contain more than ~90 wt% water in their fully swollen state [18,19]. These swollen hydrogels can be applied to the roofs of buildings [20], acting as artificial 'skins' to provide cooling. By applying hydrogel coatings, heat dissipation is enhanced through the evaporation of water inside the hydrogel, enabling surface temperature reductions of 10–30 °C in various objects, such as skin [21,22], handheld electronics [23–25], Li-ion battery packages [26,27], and buildings. This remarkable autonomous cooling capability makes hydrogels an attractive candidate for energy-efficient building cooling.

Importantly, hydrogels previously investigated for cooling applications have not vet demonstrated one key feature required to truly mimic biological skins: durability and reusability for repeatable cooling. Only limited regeneration capability tests have been performed on hydrogels for cooling applications [20,26,27]. The maximum cycling number demonstrated so far is about six, which is far below the amount needed for practical cooling applications. Hydrogels utilized for bio-inspired cooling so far are often brittle, as measured by low fracture energies ( $\sim 10 \mid m^{-2}$ ) [28], which is orders of magnitude lower than that of human skin  $(\sim 1800 \text{ Jm}^{-2})$  [29]. These poor mechanical properties (low stretchability and toughness) severely limit the scope of cooling applications for hydrogels, where reusable and regenerable cooling is important for both long-term performance and costeffectiveness. For example, embedding hydrogels inside roofing [20] makes it unfeasible to replace the gels after only a few cycles. Furthermore, degradation and aging of hydrogels via UV radiation hinders outdoor applications [30-32], such as for coating windows or building facades.

In this work, we report, for the first time, the application of a highly stretchable and tough double network hydrogel (DN-Gel) [33,34] as a regenerable 'sweating skin' for cooling buildings. Compared with single network hydrogels (SN-Gels) used in previous cooling studies [20–27], DN-Gels have significantly higher fracture energy (~9000 J m<sup>-2</sup>) [35], comparable to that of animal skin [29,36]. While the toughness of DN-Gels is well established, their suitability for cyclic cooling or heating has not been studied. During the cooling process, the internal morphology (e.g., porosity) could change upon swelling/deswelling cycles, or the material may degrade after exposure to heat and UV radiation. Herein, we show that DN-Gels exhibit excellent evaporative cooling performance as well as extraordinary toughness and cyclability, as demonstrated by continual cooling performance over more than 50 cycles.

#### 2. Materials and methods

#### 2.1. Reagents and materials

The DN-Gel monomers, alginate (AG) and acrylamide (AAm) (99+% electrophoresis grade) were purchased from FMC BioPolymer and Alfa Aesar, respectively. A crosslinker, N,N'-methylenebis (acrylamide) (MBAA) (>98.0%) and calcium sulfate dehydrate (>99%), crosslinking accelerator, N,N,N',N'-tetramethylethylenedia

mine (TEMED) (99%), and photoinitiator, ammonium persulfate (AP) (>98.0%), were purchased from Sigma–Aldrich. The SN-Gel monomers, acrylic acid (AAc) (99.5%), and photoinitiator, 2,2-dime thoxy-2-phenylacetophenone (DMPA) (99%), were purchased from Alfa Aesar and Acros, respectively. All chemicals were used as received without any purification (see Table 1).

#### 2.2. Hydrogel preparation

To prepare DN-Gels, AG and AAm were dissolved in DI water with weight ratios of 2%, 12%, and 86% [35]. We then added 0.06 wt% MBAA, as a cross-linker to AAm, and 0.17 wt% AP, as a photoinitiator for AAm, to the solution. After degassing the solution in a vacuum chamber, we added 0.25 wt% TEMED, as a cross-linking accelerator to AAm, and 13 wt% calcium sulfate slurry, as an ionic cross-linker to AG, for homogeneous mixing using a syringe technique. The solution was poured into a plastic petri dish, cured with UV light ( $\lambda = 254$  nm) for 1 h at 50 °C, and then left in a humid box for 24 h to stabilize the reaction.

For SN-Gels, a photoinitiator solution was prepared from 0.1923 g of DMPA in 10.0 mL of DMSO [37]. The solution was sonicated until all the DMPA was dissolved, then covered and kept in the dark. A pre-polymer solution contained the monomers and cross-linking agent in a pH buffer solution with a molar ratio 7:3 of AAc to AAm and a 0.128 mol% crosslink density to the total monomer. The photoinitiator solution and the pre-polymer solution were homogeneously mixed *via* sonication for 1 h. The solution was then poured into a petri dish and polymerization was initiated by exposure to a UV lamp ( $\lambda$  = 365 nm) at room temperature for 10 min.

#### 2.3. Cooling performance experiments

To investigate the cooling performance and cyclability of tough DN-Gels, we conducted cooling experiments on miniaturized model houses under simulated solar irradiation (QL 1500 Series lamp) with a maximum power density of 1000 W m<sup>-2</sup>. Two identical model houses made of oak wood (with thermal conductivity of 0.17 W m<sup>-1</sup> K<sup>-1</sup>, density of 740 kg m<sup>-3</sup>, and thickness of 0.025 m), each with a roof surface area of 64 cm<sup>2</sup> (Fig. 1a), were built to compare the cooling effectiveness of the DN-Gels and SN-Gels. Thermocouples were attached to the top and bottom surfaces of the roof panel on both model houses to monitor the temperature rise induced by simulated solar irradiation. To directly compare cooling performance, both types of hydrogel layers were soaked in deionized (DI) water for  $\sim$ 8 h prior to attaching them to the model house roofs. An acrylic adhesive was used to ensure good thermal contact between the hydrogel layer and roof. A solar simulator was used to apply a normal incident irradiance of  $\sim 800 \text{ W} \text{ m}^{-2}$  to both roofs. Similar experiments were conducted to test the cyclability of the DN-Gels and SN-Gels after reducing soaking and drying time to  $\sim$ 5 h, and varying incident irradiance power (700 W m<sup>-2</sup> and 800 W m<sup>-2</sup>). The thickness of the tough DN-Gel mats synthesized here is initially 1 cm but can reversibly expand up to 2 cm in the swollen state after storing up to 90 wt% water [34,35]. We also synthesized a 0.8 cm thick SN-Gel (Poly (AAm-AAc)) according to a reported recipe [37].

Table 1Reagents for DN-Gel and SN-Gel synthesis.

Reagent	DN-Gel	SN-Gel
Monomer	Alginate (AG)/acrylamide (AAm)	Acrylic acid (AAc)/acrylamide (AAm)
Crosslinker	N,N'-methylenebis (acrylamide) (MBAA)/calcium sulfate	N,N'-methylenebis (acrylamide) (MBAA)
Crosslinkingaccelerator	N,N,N',N'-tetramethylethylenediamine (TEMED)	-
Photoinitiator	Ammonium persulfate (AP)	2,2-Dimethoxy-2-phenylacetophenone (DMPA)

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