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Upcycling Chips-Bags for Passive Daytime Cooling

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Plastic pollution has caused numerous environmental issues in recent decades. As one of the most commonly used packaging materials, aluminum-plastic laminates (APL) are particularly challenging for recycling purposes due to their sophisticated materials components. This work reveals a new strategy to upcycle such post-consumer APL packaging waste, e.g., chips-bags, for passive daytime cooling applications. This opens an attractive route to reuse APLs while at the same time reducing global energy consumption and carbon emissions. The mirror-like appearance of the APLs possesses a strong solar reflection, up to 86%. By coating, this reflective layer of the APL waste with a high emissive polydimethylsiloxane layer, a simple but effective passive daytime cooling foil is constructed, which shows promising passive cooling performance theoretically and practically. More importantly, the passive cooling foil based on APL waste is flexible and can be applied to any target object, protecting it from harsh sunlight. The low-cost APL waste-based passive cooling foil proposed in this work will significantly contribute to both energy and environmental issues that humans face today.

1. Introduction

Aluminum-plastic laminates (APL) have been massively used as packaging materials to extend the shelf-life of products like chips, roasted, and powdered coffee, as well as milk and juices.[1] Besides, APL has extended its applications dramatically in the pharmaceutical packaging of FFP2 masks and rapid tests over the last few years because of the COVID-19 pandemic. With aluminum as a barrier layer, the APL provides satisfactory product protection from various deteriorating factors, including oxygen, moisture, and light.[2] However, the complex construction, consisting of aluminum and multiple layers of polymer, makes this sort of packaging foil exceedingly challenging to recycle.[1,3] Consequently, most APL waste is treated by incineration or discarded in landfills, causing severe environmental sustainability issues, including the release of greenhouse gas emission, air pollution, and soil infertility.[4]

The industrial recycling of APL faces several challenges, such as the segregation of the different layers as well as cleaning and sorting issues.[3b,c] One technique for recovery of aluminum and individual polymer components is chemical delamination, where the individual polymer layer is dissolved with the corresponding solvent.[5] Besides, thermal and catalytic pyrolysis have also been developed to extract aluminum and energy from APLs.[6] More recently, various innovative strategies for APL recycling have been reported. For instance, thermal delamination[7] and enzymatic bioleaching recycling methods.[8] Despite the progress in material and energy extraction from APL, it is still considered unrecyclable due to its low recycling efficiency. The European plastic strategy aims for all used packaging to be recyclable or reusable by 2030.[9] For this to be successful and to reduce the impact of APL waste on the environment, advanced strategies for cost-effective recycling or repurposing of APL are urgently desired.

Another rising concern is global warming, caused by the overuse of fossil fuels and resulting greenhouse gas emissions.[10] As one of the consequences, the frequency of intense heat waves has increased worldwide, threatening human and ecological health.[11] Passive daytime radiative cooling (PDRC) is regarded as a promising strategy to combat global warming by reducing the energy demand for cooling.[12] By emitting thermal irradiation to cold outer space (3 K) through the atmospheric transparency window (8–13 μm), terrestrial materials with engineered optical properties autonomously cool down

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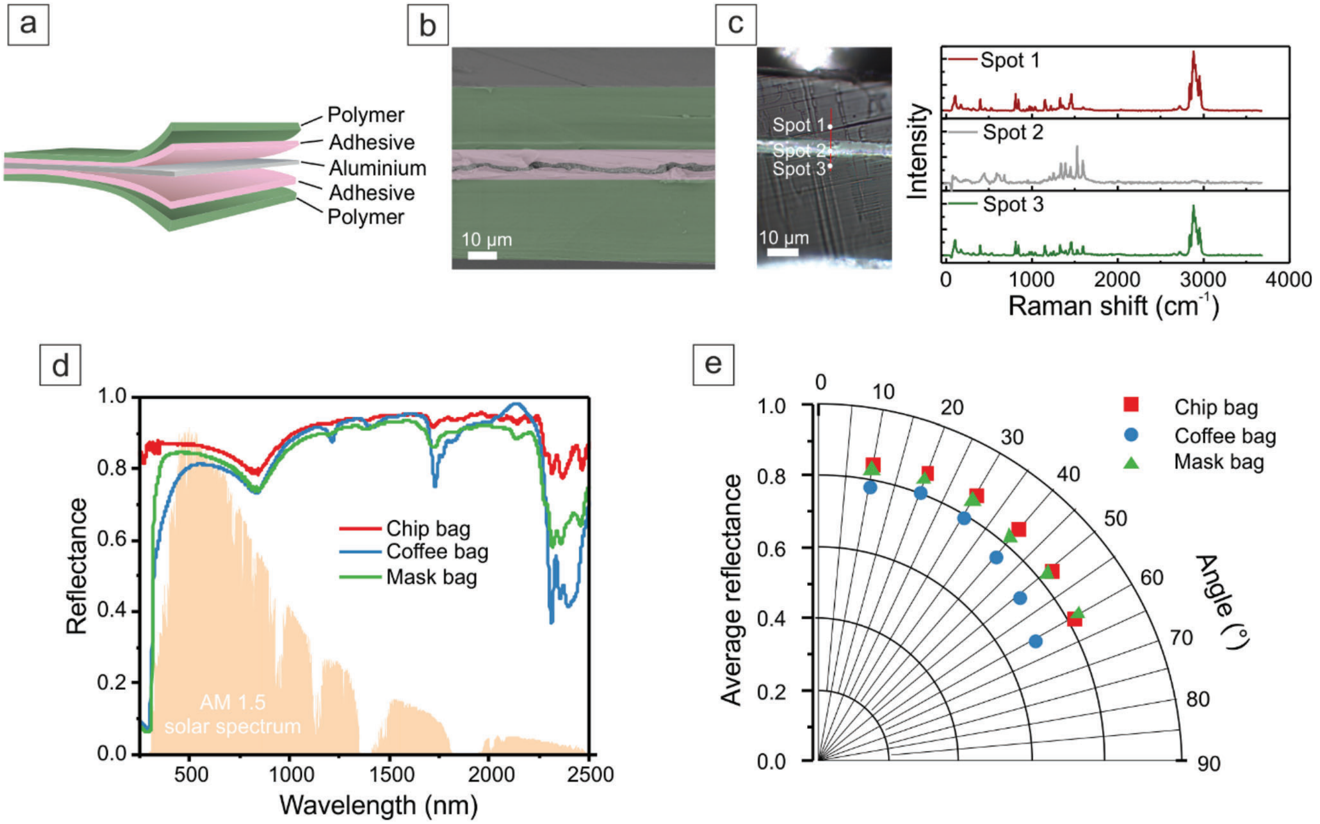

Figure 1. a) Schematic of the multilayer structure of an APL used for packaging. b) SEM image of the cross-section of an APL from a chip bag. c) Raman spectra of the cross-section of the APL. d) Solar reflectance of the goods contacting side of the APLs collected from different packaging bags: chip bag, coffee bag, and FFP2 mask bag, with an incidence angle of 8°. The AM 1.5 solar spectrum is plotted as the background.[18] e) Average solar reflectance of the goods contacting side of the chip bag, coffee bag, and FFP2 mask bag at different incidence angles.

to subambient temperatures in the nighttime, without external energy input. By minimizing the absorption of a material in the solar regime (0.3–2.5 μm), the passive cooling properties can be preserved during the daytime, even under direct sunlight illumination. A passive cooling device can be readily constructed by combining a highly solar transparent and emissive layer, like polydimethylsiloxane (PDMS), with a back reflector, e.g., silver (Ag) or aluminum (Al).[13] To circumvent the usage of the metallic back reflector, in recent years, advanced structured materials have been fabricated to strongly scatter sunlight,[14] including micro-/nanoporous materials,[15] and nanoparticles-based composites.[16] Though an increasing number of passive cooling devices with remarkable daytime cooling performance have been reported, the transition from fundamental scientific studies to a widely distributed cooling technology is still missing. Scalability, practicality, durability, and manufacturing costs are insurmountable problems restricting this technique from practically transitioning to real-life applications. Considering that the amount of energy saved with PDRC is proportional to the applied area, it is imperative to design a cost-effective, scalable, and easily applicable PDRC foil.

In our contribution, we outline a comprehensive upcycling strategy to repurpose APL waste and to access PDRC foils in a cost-effective manner. This combination has a twofold beneficial consequence for the environment due to the reduction of APL waste, and the fabrication of PDRC foils with already available materials. Optical property characterization elucidates that the plain APL possesses a substantial solar light reflection (up to 86%), owing to the presence of the Al layer, which makes APL a promising candidate for the back reflector of the PDRC material. By adding an emissive layer, e.g., PDMS, to enhance the radiative heat release, the cooling capacity of APL waste-based foils is theoretically and experimentally demonstrated. Moreover, the cooling foil is scalable, flexible, and can be readily applied to various surfaces, protecting the target object from harsh sunlight and heat. Our approach is the first step toward a widespread and low-cost utilization of optically engineered passive cooling foils.

2. Results and Discussion

APL is generally constituted by several polymer layers and aluminum laminated with adhesives, as illustrated in **Figure 1**a. A scanning electron microscope (SEM) image of a cross-section of APL used for a chip bag shows its multilayer structure. A thickness of about 18.9, 2.7, 1.7, 3.2, and 31.2 μm is observed for respective layers (from the top, goods contacting side, to bottom, ink side). By using confocal Raman spectroscopy, we determined the chemical composition of each layer. The Raman spectra show that both polymer layers are polypropylene (PP) for the tested APL.[17] APL shows a mirror-like appearance due to the presence of Al laminated at the core, demonstrating its potential as a back reflector. To reveal the capability of APL as a back reflector for PDRC, we examined the solar reflectance of APL from various packaging bags, as shown in Figure 1d; and Figure S1 (Supporting Information). As representative samples, APLs from bags originally used for potato chips, coffee, and face masks are mainly focused on in this work due to their widespread usage in the market. An impressive solar reflectance is observed for most of the APLs. As shown in Figure 1d, an average solar reflectance of 0.86, 0.81, and 0.83 was obtained from the chip, coffee, and mask bags, respectively. The decent solar reflectance verifies that APL holds considerable potential as the back reflector for passive daytime cooling materials. Moreover, APLs, especially the chip bag and the mask bag, exhibit a constant reflectance at a large incident angular range (Figure 1e). This stable and high solar reflectance allows them to face and reflect the sun at various angles. Even when substantially wrinkled, as expected from waste stream collection, the APL foils retain their high solar reflectance (Figure S2, Supporting Information).

For PDRC materials, absorption in the solar range determines the energy uptake from the sun in the daytime, while mid-infrared (MIR) emissivity controls the radiative heat release. Therefore, both aspects of the material need to be optimized to achieve a net passive cooling performance in the daytime. In the market, various polymers have been utilized to compose the APL, including PP, low-density polyethylene (PE), and polyethylene terephthalate (PET).[4b] APL varies in composition and layer thickness depending on the end-use product, resulting in different MIR emissivities. To reveal the MIR emissivity of the tested plain packaging bags, we measured their absorptance (1 - reflection) spectra at a wavelength range of 2–18 µm by using Fourier-transform infrared spectroscopy (FTIR) (**Figure 2**a). Average MIR absorptances of 0.23, 0.51, and 0.70 were obtained for the plain chip bag, coffee bag, and mask bag, respectively, which are related to the layer composition (Figures S3 and S4, Supporting Information) and thickness. The impact of the layer thickness on material optical properties in both the solar and MIR ranges has been investigated in a previous study.[19]

To experimentally demonstrate the capability of the plain APL for PDRC, the passive cooling performance of the APLs, i.e., chip bag, coffee bag, and mask bag, was determined with a home-made indoor setup. Figure 2b shows the schematic of the indoor setup, and detailed information on the design is presented in our previous work.[20] Compared to conventional field testing under uncontrollable weather conditions, indoor measurements ensure reproducible measurements under controlled and stable conditions, providing better cooling performance evaluation, especially for comparing different samples. In addition, various parameters, such as solar intensity and ambient temperature, are independent and can be tuned individually with the designed indoor setup. By contrast, the study of the influence of a single parameter on material cooling performance is not possible for conventional field testing due to the time-varying weather conditions.

The indoor measurement was first carried out without (w/o) solar light. A steady-state temperature of 18.4, 16.4, and 13.8 °C was observed for the chip bag, coffee bag, and mask bag, respectively (Figure 2c–e). Compared to the “ambient temperature” (20.7 °C), obtained with an Ag mirror under identical measurement conditions without solar light, all the APLs show varying degrees of passive cooling performance. The mask bag provides the best subambient cooling, 6.9 K, followed by the chip bag (2.3 K) and coffee bag (4.3 K), owing to the higher emittance in the MIR regime. Subsequently, AM 1.5 solar light was applied to the samples by a solar simulator to imitate the cooling performance characterization in the daytime. To reveal the impact of the solar radiation intensity on the passive cooling performance, the solar intensity was increased stepwise from 0% to 25%, 50%, 75%, and 100% of one sun power (1000 W m^{-2}), after the sample's temperature reaches a steady state. Note that our previous study showed that the intensity of the light reaching the sample is about 75% of the initial power due to the absorption, scattering, and reflection of the convection shield in the indoor setup.[20] As shown in Figure 2c–e, the solar illumination increases the temperature of all APL samples until a steady state is reached. Moreover, the solar energy uptake exceeds the radiative heat loss at a certain level of solar intensity for the chip bag and coffee bag. As a result, the steady-state temperature is higher than the ambient temperature, for instance, 50% solar intensity for the chip bag and 75% solar intensity for the coffee bag, which follows the degree of emittance of those sample in the sky-window range. This observation implies that a net heat loss can only be achieved under a certain solar intensity level for the chip bag and the coffee bag due to their inherently low MIR emittance. By contrast, the mask bag allows net passive cooling even under a sunlight power of 100%.

To enhance the radiative heat release by increasing the MIR emittance, we introduced PDMS onto the APLs. PDMS, a widespread and easily processable polymer, has been widely used as standard material for fabricating passive daytime cooling devices, ensuring a direct comparison between the APL waste-based cooling foil and PDMS-based cooling devices reported in the literature. Furthermore, its optical transparency is ideal for retaining the reflective properties of the APL support structure and its flexibility ensure a good adhesion to the APL foil. In particular, a PDMS layer (≈200 µm) was coated onto the APLs via the doctor blading technique. The IR absorption spectra illustrate the significant enhancement of the APLs in the MIR region (Figure 2f). An average emittance of up to 0.94 was obtained for all PDMS-coated APLs. The cooling performance of plain APLs and PDMS-coated APLs, determined by the indoor setup, is summarized in Figure 2g. Compared to plain APLs, a promising PDCR performance is obtained with all PDMS-coated APLs. A subambient cooling was observed with all PDMS-coated APLs for both dark and light conditions. Furthermore, no pronounced difference was observed between the PDMS-coated chip bag, coffee bag, and mask bag for measurements in the dark. However, with the presence of 100% solar light, the PDMS-coated chip bag shows a slightly lower temperature, owing to its high solar reflectance (Figure 1d) compared to the coffee and mask bag.

Apart from the steady state temperature reduction measurements, the indoor setup also allows for determining the net cooling power. To measure the net cooling power of the plain and PDMS-coated APLs, the samples were heated to the “ambient” temperature. The required heating power at this temperature represents the net cooling power of the emitter. An observation similar to the steady-state temperature measurements was obtained for the cooling power measurements (Figures S6 and S8, Supporting Information). Solar illumination decreases the net

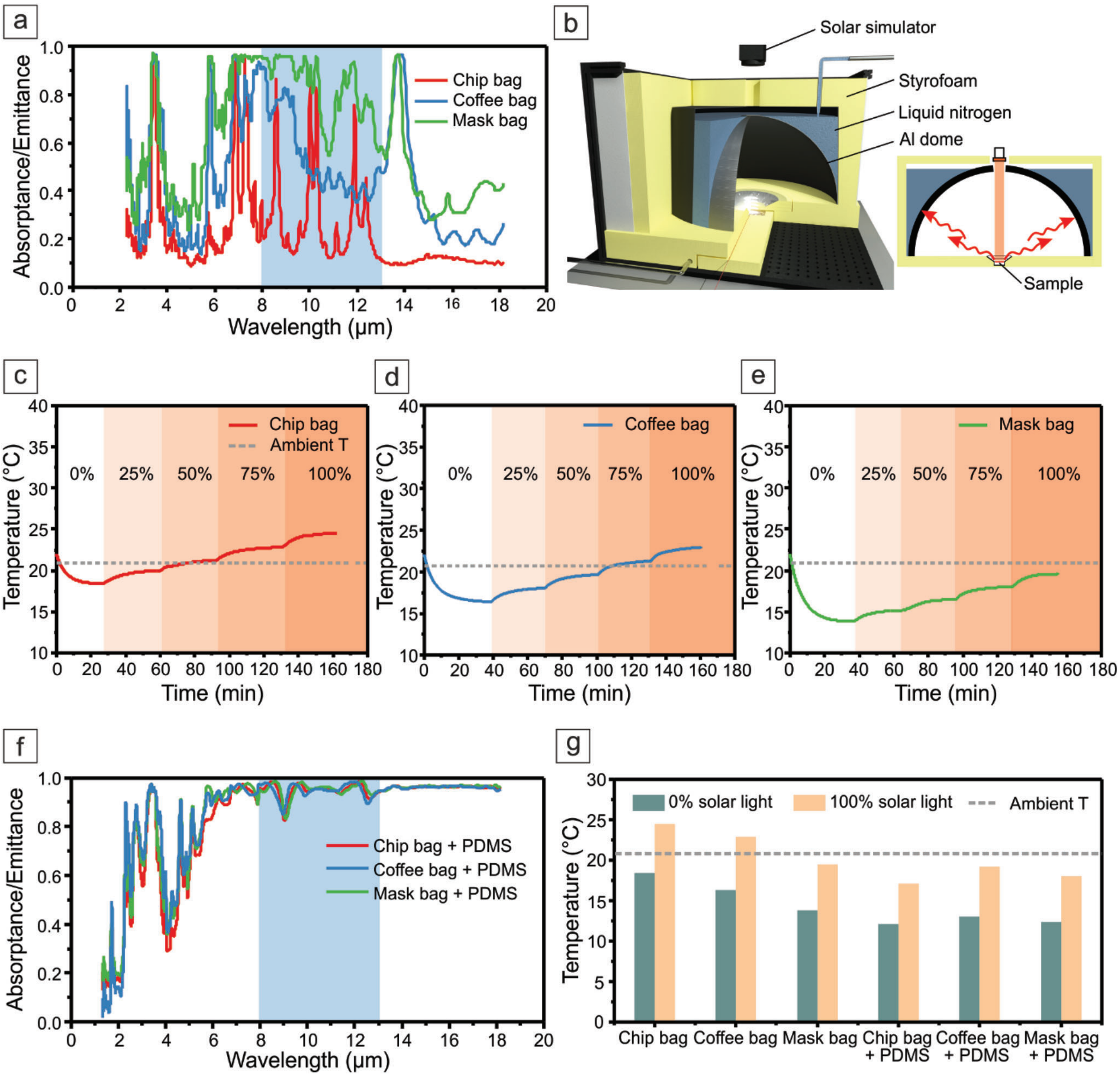

Figure 2. Indoor characterization of plain and PDMS modified APL bags: chip bag, coffee bag, and FFP2 mask bag. a) Optical properties of plain APL bags in the MIR range. The blue shaded area indicates the atmosphere window (8–13 µm). b) Schematic of the setup for indoor measurements. Continuous temperature measurements of the APL bags with stepwise increase of the solar intensity from 0% to 100% of one sun for a c) chip bag, d) coffee bag, and e) FFP2 mask bag. f) Optical properties of PDMS (thickness ≈200 µm) coated APL bags. g) Steady-state temperature of plain and PDMS-coated laminated Al packaging bags with and without 100% solar light. The dotted line indicates the ambient temperature.

cooling power of all tested samples. The plain chip bag and coffee bag only possess net cooling power below a certain level of solar intensity. In contrast, the plain mask bag shows net cooling power even under illumination with full power. The solar light intensity follows a linear dependency for the steady-state temperature (Figure S5, Supporting Information), and the net cooling power (Figure S7, Supporting Information), indicating the stable and constant measurement conditions. In addition, the PDMS coating dramatically enhances the net cooling power for all APLs.

Due to the absence of the atmospheric window in the indoor setup, the net cooling power measurements only allow an indirect comparison to the outdoor field tests. To illustrate the cooling performance of PDMS-coated APLs more clearly, we normalized the cooling power of the plain and PDMS-coated APLs by the cooling power measured with a standard passive daytime cooling sample namely a PDMS-coated Ag mirror (Figure S9, Supporting Information). Compared to this standard passive cooling sample, a comparable cooling power can be observed for all PDMS-coated

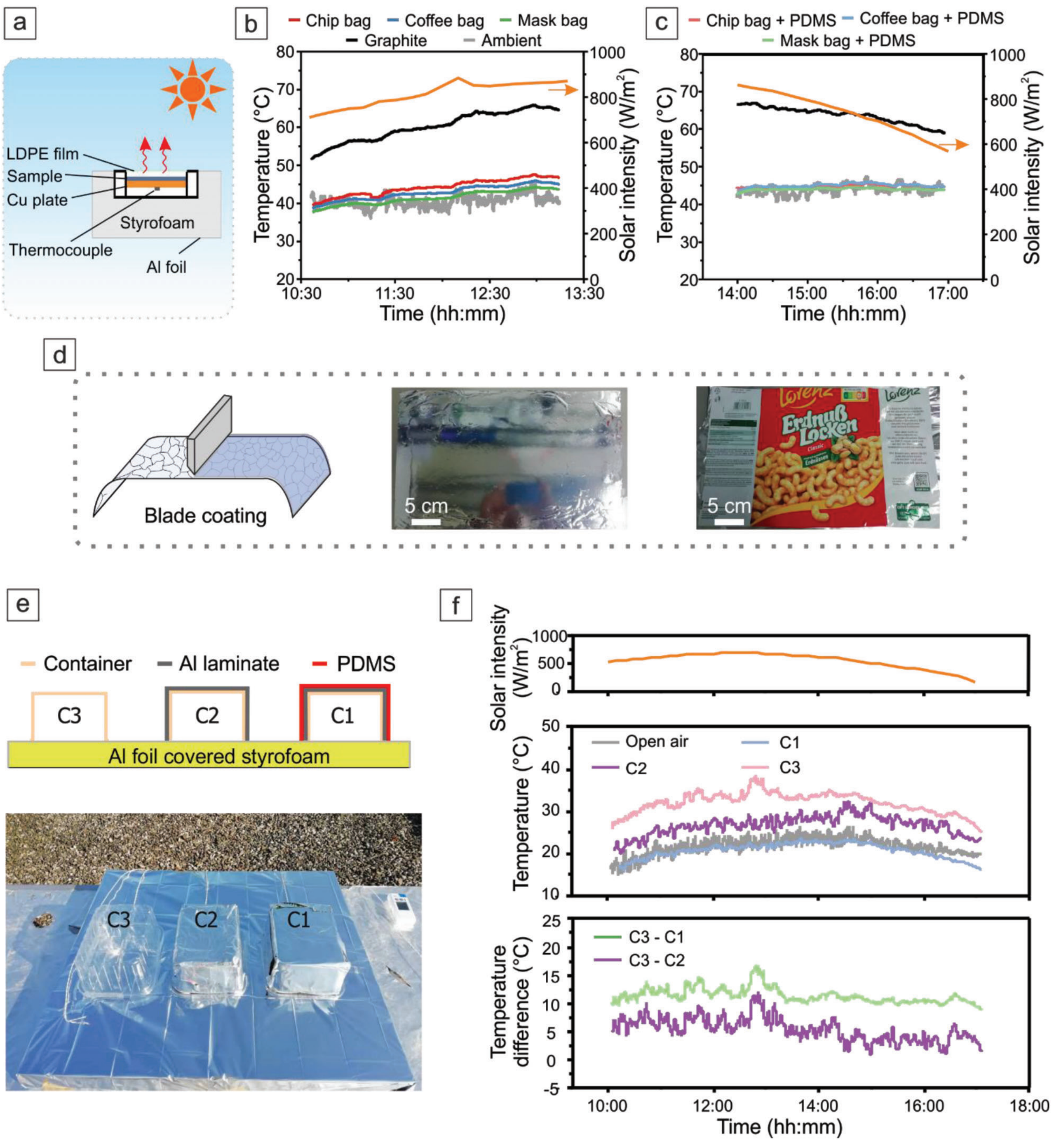

Figure 3. a) Schematic of the setup used for outdoor measurements. Outdoor measurements for b) plain and c) PDMS-coated APL bags. The measurement was carried out under a clear sky on 18.06.2022 in Bayreuth, Germany. d) Schematic of the blade coating technique and photographs of the entire piece of the PDMS-coated chip bag, goods contacting side (left), and ink side (right). Application of the APL waste-based cooling foil on a target surface. e) Schematic and photograph of temperature measurements for different samples. C1, transparent PET box covered with PDMS-coated chip bag; C2, transparent PET box covered with plain chip bag; C3, transparent PET box. f) Temperature tracking of different samples and temperature differences shown in (e). The measurement was carried out under a clear sky on 24.03.2022 in Bayreuth, Germany.

APLs in the dark. With the full power of solar light, the PDMS-coated APLs could still provide up to 25% of the net cooling power of the reference material.

To further verify the potential of APL waste as PDRC material for energy saving, we determined the cooling performance of plain and PDMS-coated APLs with field testing under a clear sky in Bayreuth, Germany. **Figure 3**a illustrates the schematic of the homemade setup, and a photograph of the setup is shown in Figure S10 (Supporting Information). During the measurement, the samples were placed into identical sample holders surrounded by styrofoam to block undesired thermal conduction. A highly transparent, low-density polyethylene (LDPE) foil suppresses convective heat losses. Besides the tested sample APLs, the temperature of the ambient and a graphite sample were also collected for comparison. Figure 3b,c shows the real-time temperature of the tested plain and PDMS-coated APLs under direct sunlight. Due to substantial solar reflectance, the plain APLs exhibit a much lower temperature than graphite. The temperature difference was 18.1, 19.7, and 21.2 K between graphite and the chip bag, coffee bag, and mask bag, respectively, under average solar irradiation of 866.2 W m^{-2}. The significant temperature difference confirms the potential of APLs as a back reflector for PDRC. However, due to inadequate radiative heat release, the temperature of all tested packaging bags is slightly higher than the ambient. With the introduction of the PDMS coating, thermal radiation is greatly facilitated for all APLs. This leads to coinciding ambient and sample temperatures under average solar irradiation of 847.8 W m^{-2}. In addition, a temperature difference of about 22.5 K between graphite and the PDMS-coated APLs is obtained. Despite the deviation of the absolute value between the indoor measurement and the field testing, the trend agrees well. The deviation is mainly attributed to the absence of the atmosphere for the indoor setup and the inconsistent conditions between indoor and outdoor measurements, such as solar intensity and humidity.

The cooling foils based on APL waste are flexible, possess excellent mechanical strength, and can be scaled up. In the first step to accessing large area foils, stitching multiple packaging bags via plastic joining techniques, including welding and adhesive bonding,[21] are expected to be suitable processes that need to be adapted to APL waste. Concomitant with the areal scale-up, a consistent high solar reflectance of the patches also needs to be realized. These engineering challenges are beyond the scope of this work. In the second step, an emitter layer, such as PDMS, can be coated with well-established processing techniques, like blade coating and roll-to-roll coating.[22] Figure 3d shows a whole piece of a chip bag coated with PDMS via blade coating. Such low-cost, flexible, and free-standing cooling foils can be practically applied to any target surface. To emphasize this point, we applied the PDMS-coated chip bag to a standard transparent container using double-sided adhesive tape. Subsequently, we monitored the temperature inside the cooling foil-covered container (C1) under direct sunlight. The inside temperature of an identical container (C3) and a plain chip bag-covered box (C2) were also recorded for comparison. Figure 3e shows the schematic and a photograph of the measurement. All samples were placed on an Al foil-covered styrofoam plate to avoid undesired thermal conduction. In Figure 3f, we plotted the continuous inner temperature of different containers and the temperature difference relative to C3, along with the solar irradiation during the measurement. With reflecting most of the solar light, the plain chip bag-coated container (C2) shows a much lower temperature than the bare container (C3), which exhibits a substantial amount of greenhouse warming. The average temperature difference is 7.2 K from 11:30 to 13:30, under an average solar irradiation of 668 W m^{-2}. Furthermore, the cooling foil-covered container (C1) maintained its temperature close to the open air temperature and is much lower than C2 and C3 due to the high solar reflection and superior heat release via MIR radiation. The average temperature difference between C1 and C3 is 12.6 K. The much lower temperature achieved with APL-based cooling foil proves that it can effectively protect the target object from overheating under intensive solar light. In addition, the protection will be more pronounced when applying the cooling foil on surfaces of solar light-absorbing materials.

To theoretically quantify the cooling performance of the plain and PDMS-coated APLs, i.e., chip bag, coffee bag, and mask bag, we calculated their net cooling power for both daytime, and nighttime (Figures S10 and S11, Supporting Information), based on the radiative model,[23] $P_{cool} = P_{mat} - P_{sun} - P_{atm} - P_{nonrad}$. Here, P_{mat} is the emitter thermal irradiation power, P_{sun} is the solar power absorbed by the emitter, P_{atm} is the emitter absorbed power from the atmosphere, and P_{nonrad} is the absorbed power caused by conduction and convection, which is defined as $P_{nonrad} = h_c \cdot (T_{atm} - T_{mat})$, and h_c is the nonradiative heat transfer coefficient. Solar irradiation varies over the daytime. An average solar irradiation of 500 W m^{-2} is thus applied to the daytime calculations (see the Supporting Information for calculation details). For the plain APLs, a net cooling power of −33.7, −12.3, and 38.3 W m^{-2} are expected for the chip, coffee, and mask bags, respectively. Because of the distinct inherent optical properties, only the mask bag exhibits decent PDRC performance due to its high emissivity in the MIR regime. Still, a promising PDRC performance is obtained for all APLs when coated with PDMS. The net cooling power for the PDMS-coated chip, coffee, and mask bag is 71.3, 45.1, and 50.4 W m^{-2}, respectively. Moreover, when the net cooling power is zero, a subambient cooling of 4.9, 3.1, and 3.5 K could be expected for the PDMS-coated chip bag, coffee bag, and mask bag, respectively, with $h_c = 10$ W m^{-2} K^{-1}. In the nighttime, without the uptake energy from solar light, cooling performance, in terms of net cooling power and subambient cooling, is greatly enhanced for all cooling foils (Figures S11 and S12, Supporting Information). The combination of indoor testing with controlled parameters, the outdoor field tests under realistic conditions, and the theoretical description all confirm the adequate optical properties of upcycled APL waste to be reused as (daytime) passive cooling material.

In this work, PDMS is introduced as an exemplary emitter to enhance the thermal radiation of the plain APLs. Various other emitters could be applied to APLs, too, as long as they possess low solar absorption and high mid-infrared emissivity. Generally, PDRC emitters can be classified into two categories: broadband emitters and selective emitters. **Figure 4**a plots the absorptance/emittance spectra of an ideal broadband emitter ($E_{\lambda > 4\,\mu m} = 1$, $E_{\lambda < 4\,\mu m} = 0$) and a selective emitter ($E_{8\,\mu m < \lambda < 13\,\mu m} = 1$, $E_{\lambda < 8\,\mu m \vee \lambda > 13\,\mu m} = 0$) both with zero solar absorption. The ideal broadband emitter has unit emittance over the entire MIR range, while the ideal selective emitter exhibits unit emittance only in the atmosphere window (8–13 µm). To reveal the impact of the emitter selectivity on the PDRC performance of APL waste-based cooling foils, we calculated the net cooling power as a function of average emissivity for both APL-supported broadband emitters and selective emitters. For simplicity, the absorptance of the APL support layer has been taken as constant (14%), and its emissivity in the mid-infrared range was excluded from the calculation. Due to the fact that the solar intensity varies over the daytime, the influence of solar intensity on cooling performance is also considered in the calculation. As shown in Figure 4b, in the

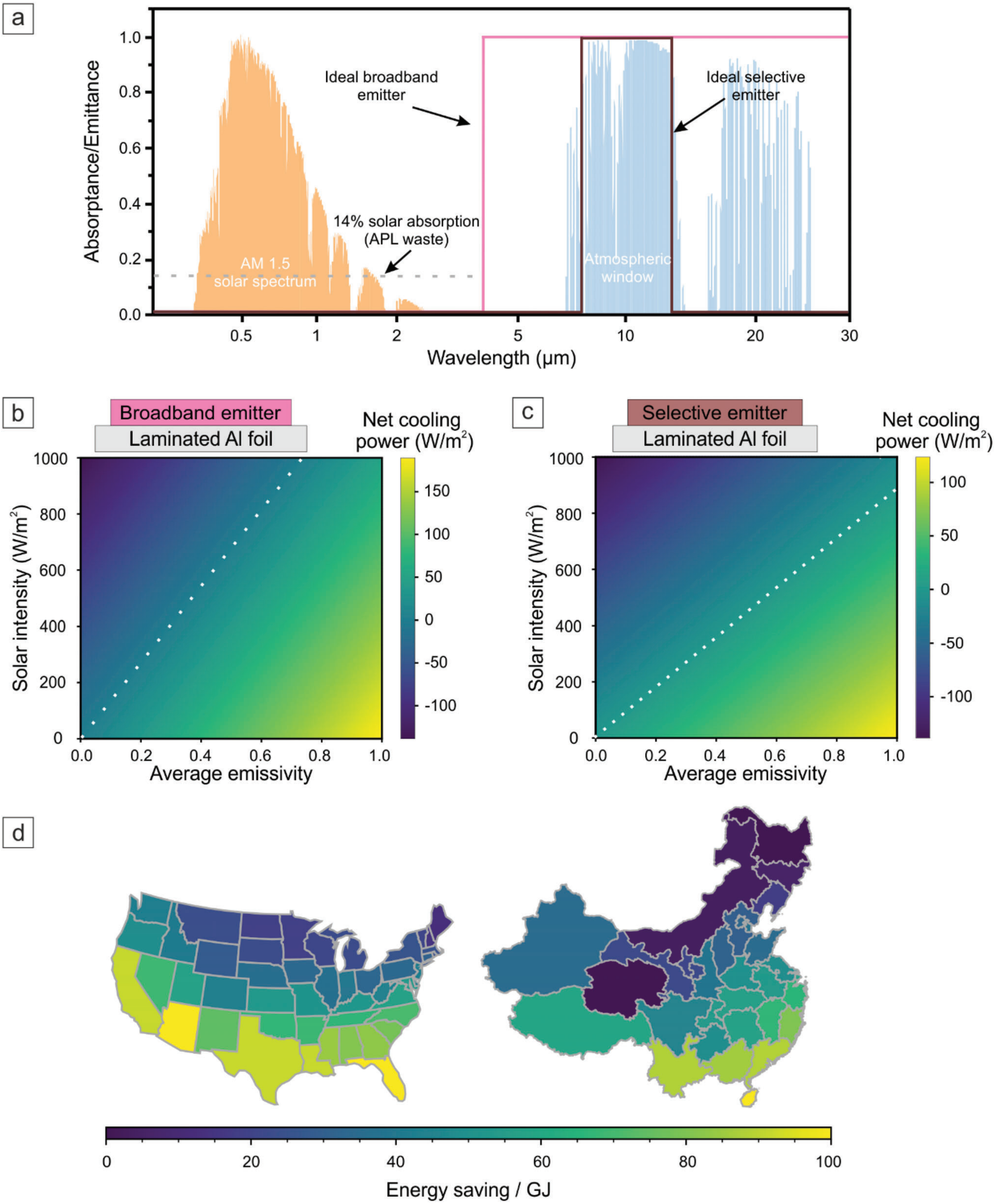

Figure 4. a) Optical properties of an ideal broadband emitter and selective emitter. The gray dotted line indicates the solar absorption of the APL foil. AM 1.5 solar spectrum[18] and atmosphere transparency[25] are plotted as background. Predicted net cooling power for APL supported b) broadband emitter and c) selective emitter with varying solar intensity and emissivity. The ambient temperature is 300 K. The average solar reflectance of the cooling foils is assumed as 0.86. The dotted lines indicate zero net cooling power. d) Predicted annual energy savings across the United States and mainland China with APL-supported broadband emitter coating on walls and roofs for a typical midrise apartment building.

nighttime, at an emitter temperature of 26.85 °C (300 K), an increase of the average emissivity of the entire cooling foil from 0 to 1 increases the net cooling power from 0 to 215 and 132 W m^{-2} for the broadband emitter and selective emitter modified APL, respectively. During daytime, the APL cooling foil reduces the net cooling power by absorbing about 14% of solar energy. With a solar radiation of 1000 W m^{-2}, a selective emitter-modified APL cannot exhibit a net passive cooling performance. To achieve a net cooling performance with broadband emitter-modified APL, the average MIR emissivity of the emitter layer should be larger than 0.73. With an average emissivity of 1, a net cooling power of 51.5 W m^{-2} can be achieved by the cooling foil under a solar illumination of 1000 W m^{-2}. Since solar light intensity varies over the daytime and barely exceeds 900 W m^{-2}, we firmly believe that all APL-based cooling foils will ensure an appealing PDRC performance throughout the entire day. Furthermore, these calculations confirm the solar radiance-dependent measurements (Figure 1c–e) and demonstrate that a reflectance of ≈86 % of the APL waste foil is sufficient for passive daytime cooling depending on the interplay between solar radiance and emissivity of the emitter layer.

The upcycling of APL waste for the fabrication of passive daytime cooling foils will significantly alleviate the predicament of the current recycling of aluminum-laminated packing foils. In addition, the APL-based passive daytime cooling foils are low-cost, scalable, and flexible, allowing their widespread worldwide application. To predict the energy-saving potential of APL-based cooling foils on building energy efficiency, we predicted the year-round cooling energy consumption for the United States and mainland China using EnergyPlus (Version 9.6.0).[24] For the simulations, 1020 and 270 locations across the United States and mainland China, respectively, were included in the calculations. The optical properties of the traditional material (acquired from EnergyPlus) and APL-based cooling foils that are input into the model are listed in Table S1 (Supporting Information). The energy-saving is obtained by comparing the energy consumption between the traditional material and APL-based cooling foil-covered outer walls and roof of a building. An energy-saving map is created for the United States and mainland China, respectively, as shown in Figure 4d. APL-based foils allow promising energy savings in most areas, with expectedly higher energy savings in the hotter southern areas. With the upcycling of APL waste for PDRC, we believe that a substantial amount of cooling energy can be saved annually, while simultaneously avoiding the large-scale fabrication of additional APLs for this sustainable cooling technology.

3. Conclusion

This work introduced a promising strategy to turn postconsumer APL waste into high-value PDRC applications. We experimentally and theoretically verified the potential of laminated Al foils for passive daytime cooling. Due to the inherent presence of the Al layer, APLs enable a high solar reflection, up to 86%. Using the laminated Al foil as the back reflector and PDMS as an infrared emittance enhancer, a simple but effective passive daytime cooling foil is fabricated, exhibiting remarkable cooling performances. Moreover, such flexible cooling foils can be readily applied on any target surface, protecting it from harsh sunlight. We believe sustainable, low-cost, and scalable APL waste-based passive daytime cooing foils can significantly contribute to environmental protection while ensuring a substantial reduction in global energy consumption for cooling applications.

4. Experimental Section

Sample Fabrication—Laminated Al Foil: Various laminated Al packaging bags were collected from plastic waste, followed by cleaning with standard detergent and adequate water. Subsequently, the samples were rinsed with ethanol and Milli-Q water and dried for further application.

PDMS-Coated Laminated Al Foil: A prepolymer of PDMS (Sylgard 184, Dow Chemical) was mixed with a curing agent in a ratio of 10:1 (by weight) and degassed in a desiccator under a vacuum. Subsequently, films with a thickness of around 200 µm were prepared via doctor blading on the laminated Al foil. The PDMS layer was cured at room temperature for 48 h.

All the samples were cut to a size of dia. = 5 cm for the indoor and rooftop measurements.

SEM: The cross-section of a chip bag was prepared by cryo-fracturing and cutting at room temperature with a razor blade. SEM images were taken with a Zeiss Ultra plus (Carl Zeiss AG, Germany) at an operating voltage of 3 kV and with in-lens detection.

Raman Spectroscopy Measurement: A confocal WITec Alpha 300 RA+ Raman imaging system equipped with a UHTS 300 spectrometer and a back-illuminated Andor Newton 970 EMCCD camera together with the WITec Suite FIVE 5.3 software package was employed for Raman spectroscopy measurements. Typically, laser intensities of 2–5 mW and an integration time of 0.5 s (grating: 600 g mm^{-1}) were employed. Single spectra were acquired with an excitation wavelength of $\lambda = 532$ nm, and 50 measurements were accumulated for a spectrum. For line scans, a 100x objective (Zeiss EC Epiplan-Neofluar 100x, NA = 0.9) was used, and the step size was 0.2 µm. All spectra were corrected for cosmic ray spikes and subjected to a background removal routine. The WITec TrueMatch Raman spectra database software in combination with a self-created polymer database was applied to identify the polymer layer based on the obtained spectra.

Optical Properties Characterization with UV–Vis and FTIR Spectroscopy: Solar reflectance was measured by using a UV–vis spectrometer (Cary 5000, Agilent Technologies), equipped with an integrating sphere accessory (Labspheres), with a fixed incident angle of 8°. A Spectralon diffuse reflectance standard (Labspheres) was applied as the reference. Angular reflectance measurements were carried out with samples placed inside the integrating sphere at various angles with respect to the incident light. The optical property of samples in the MIR regime was determined with an FTIR spectroscopy (Vertex 70, Bruker) equipped with a gold-coated integrating sphere accessory (A562, Bruker). A gold mirror was applied as the reference. The absorptance (emittance) was calculated with absorptance (emittance) = 1 - Reflectance. Transmission is assumed to be negligible due to the presence of the Al layer.

Indoor Measurement for Cooling Performance Characterization: For all indoor measurements, dried air was warmed up by a water bath to 40 °C and flushed in the area between the convection shield and measurement cell with a volumetric flow rate of 80 ln min^{-1}. The Al dome was cooled with liquid nitrogen to about −190 °C. Before filling the liquid nitrogen into the setup, the inner space of the dome is flushed with nitrogen to avoid water condensation. The temperature of the dome is maintained constant during the entire measurement by continuously filling liquid nitrogen into the setup. A thermocouple (type T) is used for recording the temperature, and data are collected by a digital multimeter (DAQ6510, Tektronix, Germany) every 5 s.

For the measurements with different solar intensities AM 1.5 solar light is provided by a solar simulator (AX-LAN400, ScienceTech, Canada) with an illumination area of 5 × 5 cm^2. The solar intensity was changed stepwise from 0% to 25%, 50%, 75%, and 100% of one sun (≈1000 W m^{-2}) after the sample temperature reached a steady state.

The cooling power measurement was conducted with a homemade feedback heater. Samples were placed on the feedback heater and heated to a predetermined temperature. The required heating power represents the cooling power of the sample.

Rooftop Measurement: Rooftop measurements for the plain and PDMS coated APLs were carried out on the roof of a three-floor building (18.06.2022, University of Bayreuth, Bayreuth, Germany) under a clear sky. All the test samples were each placed in identical homemade sample holders. The holders were thermally insulated by Styrofoam and covered with Mylar aluminum foil. A LDPE foil with a thickness of ≈15 μm is applied to prevent convective heat transfer. The temperatures of the samples were determined by Pt-100 temperature sensors and recorded with a digital multimeter (DAQ6510, Tektronix, Germany) every 5 s. One sample holder covered with Al foil instead of LDPE foil was used to obtain the ambient temperature. The solar irradiance data were collected from the weather station at the University Bayreuth (Ecological-Botanical Garden, 400 m away from the rooftop measurement).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

optical spectroscopy, photonic materials, polymer recycling, radiative cooling, sun shelters

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[1] O. Horodytska, F. J. Valdes, A. Fullana, *Waste Manag.* **2018**, *77*, 413.
[2] M. Lamberti, F. Escher, *Food Rev. Int.* **2007**, *23*, 407.
[3] a) J. N. Hahladakis, E. Iacovidou, *Sci. Total Environ.* **2018**, *630*, 1394; b) A. S. Bauer, M. Tacker, I. Uysal-Unalan, R. M. S. Cruz, T. Varzakas, V. Krauter, *Foods* **2021**, *10*, 2702; c) G. Robertson, *Recycling* **2021**, *6*, 20.
[4] a) A. Ahamed, A. Veksha, A. Giannis, G. Lisak, *Curr. Opin. Chem. Eng.* **2021**, *32*, 100684; b) A. Farrukh, S. Mathrani, A. Sajjad, *Sustainability* **2022**, *14*, 4737.
[5] a) S. F. Zhang, L. L. Zhang, K. Luo, Z. X. Sun, X. X. Mei, *Waste Manag. Res.* **2014**, *32*, 317; b) T. W. Walker, N. Frelka, Z. Shen, A. K. Chew, J. Banick, S. Grey, M. S. Kim, J. A. Dumesic, R. C. Van Lehn, G. W. Huber, *Sci. Adv.* **2020**, *6*, eaba7599.
[6] a) A. Korkmaz, J. Yanik, M. Brebu, C. Vasile, *Waste Manag.* **2009**, *29*, 2836; b) J. Haydary, D. Susa, J. Dudas, *Waste Manag.* **2013**, *33*, 1136.
[7] S. Yin, R. Rajarao, B. Gong, Y. Wang, C. Kong, V. Sahajwalla, *J. Clean Prod.* **2019**, *211*, 321.
[8] K. Kremser, P. Gerl, A. B. Borrás, D. R. Espinosa, B. M. Martínez, G. M. Guebitz, A. Pellis, *Resour. Conserv. Recy.* **2022**, *185*, 106444.
[9] European Commission, https://www.europarc.org/wp-content/uploads/2018/01/Eu-plastics-strategy-brochure.pdf (accessed: June 2022).
[10] L. Al-Ghussain, *Environ. Prog. Sustainable Energy* **2019**, *38*, 13.
[11] a) V. Thompson, A. T. Kennedy-Asser, E. Vosper, Y. T. E. Lo, C. Huntingford, O. Andrews, M. Collins, G. C. Hegerl, D. Mitchell, *Sci. Adv.* **2022**, *8*, eabm6860; b) G. A. Meehl, C. Tebaldi, *Science* **2004**, *305*, 994; c) S. E. Perkins-Kirkpatrick, S. C. Lewis, *Nat. Commun.* **2020**, *11*, 3357.
[12] a) X. Yin, R. Yang, G. Tan, S. Fan, *Science* **2020**, *370*, 786; b) M. M. Hossain, M. Gu, *Adv. Sci.* **2016**, *3*, 1500360.
[13] a) Y. Zhu, Y. Ye, D. Wang, Y. Cao, *Osa Contin.* **2021**, *4*, 416; b) L. Zhou, H. Song, J. Liang, M. Singer, M. Zhou, E. Stegenburgs, N. Zhang, C. Xu, T. Ng, Z. Yu, B. Ooi, Q. Gan, *Nat. Sustain.* **2019**, *2*, 718.
[14] X. Yu, J. Chan, C. Chen, *Nano Energy* **2021**, *88*, 106259.
[15] a) J. Mandal, Y. Fu, A. C. Overvig, M. Jia, K. Sun, N. N. Shi, H. Zhou, X. Xiao, N. Yu, Y. Yang, *Science* **2018**, *362*, 315; b) D. Li, X. Liu, W. Li, Z. Lin, B. Zhu, Z. Li, J. Li, B. Li, S. Fan, J. Xie, J. Zhu, *Nat. Nanotechnol.* **2021**, *16*, 153.
[16] a) X. Li, J. Peoples, Z. Huang, Z. Zhao, J. Qiu, X. Ruan, *Cell Rep. Phys. Sci.* **2020**, *1*, 100221; b) J. Song, W. Zhang, Z. Sun, M. Pan, F. Tian, X. Li, M. Ye, X. Deng, *Nat. Commun.* **2022**, *13*, 4805.
[17] A. Gopanna, R. N. Mandapati, S. P. Thomas, K. Rajan, M. Chavali, *Polym. Bull.* **2018**, *76*, 4259.
[18] Solar Spectral Irradiance: Air Mass 1.5. http://rredc.nrel.gov/solar/spectra/am1.5 (accessed: 15 June, **2022**).
[19] K. Herrmann, T. Lauster, Q. Song, M. Retsch, *Adv. Energy Sustainable Res.* **2021**, *3*, 2100166.
[20] Q. Song, T. Tran, K. Herrmann, T. Lauster, M. Breitenbach, M. Retsch, *Cell Rep. Phys. Sci.* **2022**, *3*, 100986.
[21] D. Grewell, A. Benatar, *Int. Polym. Process.* **2007**, *22*, 43.
[22] K. Lin, L. Chao, T. C. Ho, C. Lin, S. Chen, Y. Du, B. Huang, C. Y. Tso, *Energy Build.* **2021**, *252*, 111400.
[23] B. Zhao, M. Hu, X. Ao, N. Chen, G. Pei, *Appl. Energy* **2019**, *236*, 489.
[24] a) X. Li, B. Sun, C. Sui, A. Nandi, H. Fang, Y. Peng, G. Tan, P. C. Hsu, *Nat. Commun.* **2020**, *11*, 6101; b) Y. C. Peng, L. L. Fan, W. L. Jin, Y. S. Ye, Z. J. Huang, S. Zhai, X. Luo, Y. X. Ma, J. Tang, J. W. Zhou, L. C. Greenburg, A. Majumdar, S. H. Fan, Y. Cui, *Nat. Sustainable* **2022**, *5*, 339; c) S. Wang, T. Jiang, Y. Meng, R. Yang, G. Tan, Y. Long, *Science* **2021**, *374*, 1501.
[25] Spectral atmospheric transmittance. https://www.gemini.edu/observing/telescopes-and-sites/sites#Transmission (accessed: 15 June, **2022**).