CELLULOSE TRIACETATE HONEYCOMB COMPOUNDS FOR IMPROVED FLAT-PLATE COLLECTORS: PERFORMANCE AND RELIABILITY

F. Giovannetti[1], M. Kirchner[1], G. Rockendorf[1], O. Kehl[2]

[1] Institut für Solarenergieforschung Hameln

Am Ohrberg 1, 31860 Emmerthal (Germany), ++49 (0)5151 999 501, giovannetti@isfh.de

[2] Wacotech GmbH & Co. KG, Gewerbepark Brake, Querstraße 7, 33729 (Germany)

1. Introduction

Honeycomb structures are a well known device to reduce convective and radiative heat losses in solar thermal flat-plate collectors, thus improving their efficiency at high temperature and/or low irradiance (Hollands, 1965). Despite several investigations of the last decades no suitable material has been found so far which can provide for the requested high performance and durability at reasonable costs. A commercially available modified cellulose triacetate film, already in use assembled in honeycomb compounds as transparent insulation, exhibits promising properties. The present work experimentally investigates the suitability of these compounds for solar thermal applications, focusing on their performance and long-term reliability. Beside this commercial product, other kinds of acetate films were tested. Special attention is given to the ageing effects caused by UV radiation and high temperature as well as on outgassing, which are collector specific occurrences, not considered by the technical specifications of the materials and not yet reported in the literature.

2. Honeycomb panel material and design

Transparent materials for solar energy applications have to fulfill severe requirements with regards to optical, thermal and mechanical properties, long-term weatherability as well as to cost efficiency. More than 40 years intensive research on honeycomb structures and panels for improved solar thermal collectors and transparent insulation can be summed up as follows:

- Commodity plastics (PET, PMMA, PC, etc.) exhibit good optical properties and low thermal conductivity, are cost efficient but cannot withstand temperatures above 80-110 °C or high UV irradiation level. They are usually implemented for architectural applications with limited maximum operating temperatures such as daylighting and space heating with transparent insulation or for integrated storage collector systems.
- Technical or engineering plastics (fluoropolymers, polyethersulfone, polyestercarbonate, etc.) can withstand more severe weathering, but are much more expensive (not less than 5 times more than commodity plastics), so that the superior performance cannot justify their use. Despite promising theoretical and experimental R&D results (Symon 1984; Van Orshoven et al., 1996) no product has been commercialized up to now.
- Glass is comparable to plastic with regards to optical properties and exhibits an outstanding weatherability. It has been used to manufacture small-celled capillaries both for transparent insulation and solar thermal collectors. To minimize conductive heat losses thin walls are required which results in a very fragile structure and laborious handling: The glass capillaries need to be embedded in a double glazing unit, thus increasing the weight, the manufacturing cost and reducing the optical performance of the panel. No product is currently available on the market.

In the present work modified cellulose di- and triacetate are investigated. Cellulose triacetate has been already identified as a very suitable material for the use in transparent insulation, due to its low cost (lower than PC or PMMA), its optical properties and its long-term durability (Wallner and Lang, 2005). The following materials are tested:

- Commercially available modified cellulose triacetate (CTA-I): This material is already successfully in use in architecture as well in integrated storage collectors with water (Wacotech, 2010). According to manufacturer's data, it exhibits a good temperature stability (long-term 140°C, short-term: 170°C) and UV durability, promising requirements for the use in improved flat-plate collectors. Honeycomb panels with a thickness of 7 mm were tested to improve the performance of a stationary, non evacuated CPC collector, but no information about material weatherability is reported (Pereira et al., 2003).
- Modified cellulose triacetate with reduced or no plasticizers' content (CTA-II and CTA-III): Plasticizers are usually added to soften plastic materials and increase their flexibility. At high temperatures evaporation occurs which could affect the optical performance or even the reliability of the collector. Aim of our study is to investigate their influence on outgassing as well on the ageing behavior of the films.
- Modified cellulose diacetate with thermal stabilizers (CDA-HT): According to our first calculations, significant heat loss reductions could be achieved with a compact honeycomb collector design, with a total collector thickness similar to standard flat-plate collectors. Under stagnation, temperatures between 135 and 160 °C are expected, which are close to or above the declared long-term service range of commercial cellulose triacetate sheets. Improved temperature stability can provide for prolonged durability and higher performance. No data regarding the material weatherability were available.

The honeycomb panel is assembled with a cost efficient technology by welding plastic strips with the desired dimensions of the honeycomb structure. The resulting squared-cell has a rather high lateral size of 9 mm (s. Figure 1): Large-celled honeycomb panels provide for material and cost saving and can at the same time effectively reduce heat losses if combined with a spectrally selective solar absorber (Hollands et al., 1992), which is the case of the application under investigation.

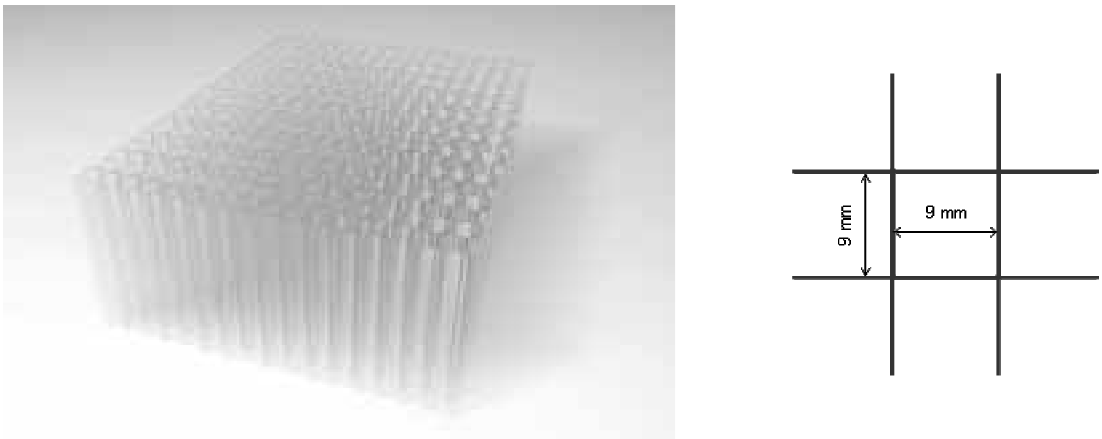


Figure 1: Panel assembly (left, photo credits to Wacotech GmbH) and simplified cell geometry of the honeycomb structure under investigation (right), specific weight 16 kg/m³

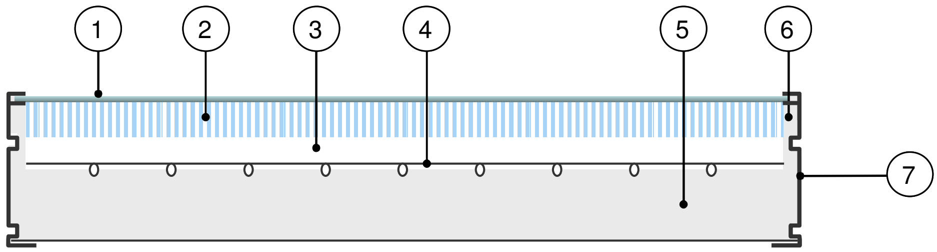
The small specimens for laboratory tests manufactured with all mentioned materials as well as the large-sized panels for experimental testing in solar collectors manufactured with materials CTA-I and CTA-III were supplied by the German company Wacotech GmbH & Co. KG, Bielefeld.

3. Collector design and performance

Most of the previous studies on honeycomb structures for solar thermal applications aimed at the development of a product which could equal the performance of an evacuated tube collector und made use of thick panels up to 100 mm. Our main goal is to improve the efficiency of common flat-plate collectors with a compact design, thus simplifying the architectural integration in the building envelope and at the same time reducing the maximum operating temperature of the plastic honeycomb. The overall collector thickness was constrained to about 100 mm, which is still in the range of standard flat plate collectors.

Our investigations have been carried out on a modified flat-plate collector: The honeycomb structure is glued to the inner side of the front glass cover, thus avoiding a second glass pane and providing for a minimal increase of the collector weight (Rommel and Wagner, 1992). A customized aluminum frame enables the integration of honeycomb panels and insulations of different thicknesses as well as the adjustment of the air gap size between cover and absorber plate (s. Figure 2). Following design parameters have been varied, whereas not all possible combinations were tested:

- Honeycomb compound: The large-celled panels were manufactured with a thickness of 15, 20, 25 and 30 mm (corresponding to aspect ratios of 1.7, 2.2, 2.8 and 3.3).
- Glass cover: Panes with different iron content as well as with additional antireflective layers (τ_s between 0.91 and 0.96) were used. Different panes are not only responsible for a different optical efficiency, but also for a different transmittance of UV radiation, which could significantly affect the durability of plastic materials.
- Gluing technique: Gluing the honeycomb panel to the glass pane reduces the collector performance due to additional solar absorption and reflection of the glue layer. Rommel and Wagner (1992) estimate 6 points reduction in conversion factor with a similar collector using strips of transparent silicone rubber. Care has been taken to choose a weatherable type of glue with high solar transmittance. Full gluing is compared with a strip gluing.
- Air gap between absorber plate and honeycomb panel: The gap is introduced to minimize the coupling effect between conductive and radiative heat transfer (Hollands et al., 1985) and to reduce the maximum temperatures of the lower surface of the honeycomb panel. Even though theoretical studies identify 10 mm as optimum to suppress heat losses, larger air gaps (15, 20, 25 and 30 mm) are chosen in order to prevent the plastic honeycomb panel to come into contact with the hot absorber plate under the weight of the cover or under typical environmental mechanical loads (snow, wind, etc.).
- Insulation: All prototypes were equipped with 10 mm side insulation consisting of temperature resistant melamine resin foam. For the back insulation 30 or 50 mm of the same material have been implemented.



1. Glass pane: 3 mm low iron, with/without AR layer (τ_s = 0.91…0.96)
2. Honeycomb panel: 15…..30 mm
3. Air gap: 15…..30 mm
4. Solar aborber: selective copper plate (ε = 0.05 ± 0.02, α = 0.94 ± 0.02)
5. Back insulation: resin foam, 30….50 mm
6. Side insulation: resin foam, 10 mm
7. Adjustable aluminium frame

Fig. 2: Design of the investigated honeycomb flat-plate collector

We investigated the collector performance by means of different tests and corresponding facilities. Heat losses have been extensively investigated on collector prototypes without irradiation, according to a well known calorimetric, time and cost saving procedure. Results are corrected taking the effect of irradiation into account. Standard indoor and outdoor tests according to EN 12975-2 (2006) were carried out afterwards on selected collector designs. The results of our experiments can be summed up as follows:

- A heat loss coefficient a_{60} (referred to a temperature difference between fluid and ambient air of 60 K) between 2.9 and 3.6 W/m²K was calculated from calorimetric tests without irradiation (s. Figure 3) and confirmed by standard measurements according to EN 12975-2. The results attest the possibility to reduce the losses of a standard flat-plate collector by about 30%. The gap size between absorber plate and honeycomb panel minimally influences the overall heat losses (max. 3%) thus playing a negligible role as performance design parameter.

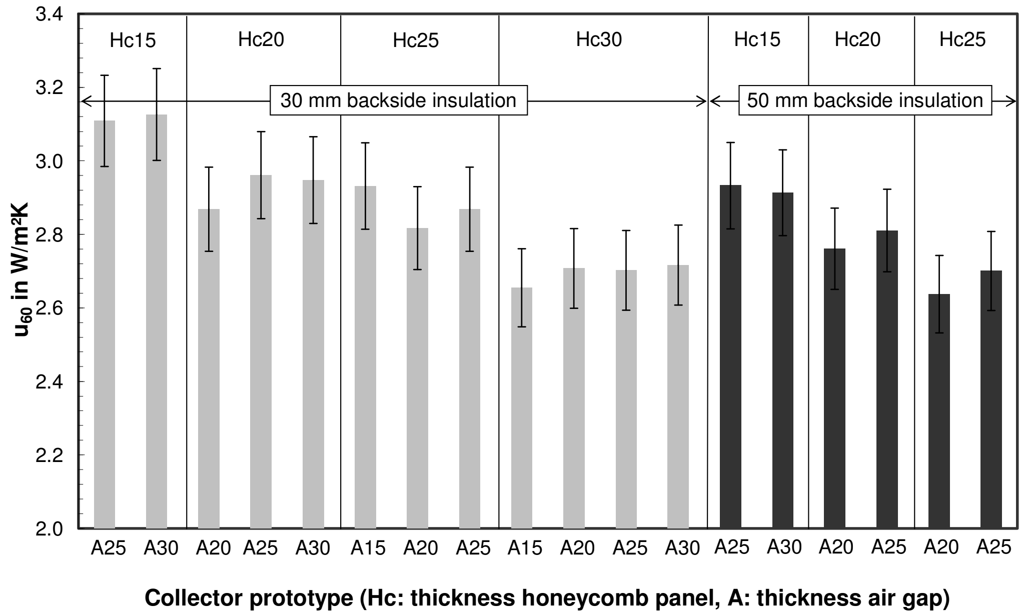


Figure 3: Heat losses of the collector prototypes under investigation at a temperature difference between fluid and ambient air of 60 K. Experimental results from calorimetric measurements without irradiation. Corresponding heat loss coefficients a_{60} under standard reference conditions (EN 12975-2) are estimated to be between 0.3 and 0.4 W/m²K higher, depending on the collector design

- Collector efficiency: Depending on collector design we determine a conversion factor η_0 between 0.76 and 0.80. These values are about 2-3 points lower than those of flat-plate collectors with a corresponding glass pane but no honeycomb panel. A fully glued panel leads to a further efficiency reduction of 2.3 points on a collector with low iron glass pane. Higher reductions are expected with antireflective glass panes. At high temperatures or low irradiation levels (η at $\Delta T/G = 0.12$ Km²/W) the honeycomb collector can achieve an efficiency of about 0.40, almost 70% better than a standard flat-plate collector.
- Incident angle modifier (IAM): The honeycomb panel geometry, the specular reflection and the reduced thickness of the cell walls are designed to achieve an optimum solar transmittance even at high incident angles. Due to absorption in the structure optical losses cannot be avoided. We measured an incident angle modifier coefficient b_0 of 0.17 on a collector with a low iron glass pane. As comparison standard flat-plate collectors with a corresponding cover show b_0 coefficients of about 0.12-0.13. That results in a lower transmittance for diffuse radiation, adversely affecting the performance of this kind of collectors especially at low irradiation levels.

Based on these results the annual yield of two honeycomb collectors representing the performance range of the investigated prototypes is compared to that of a high efficiency, state-of-the-art flat-plate collector by means of simulations with the program TRNSYS. The prototype Honeycomb_{MIN} consists of 15 mm panel and 30 mm backside insulation, the prototype Honeycomb_{MAX} features 25 mm panel and 50 mm backside insulation. Prototype and reference collectors are equipped with an antireflective glass cover with low iron content. Corresponding performance coefficients are reported in Table 1.

Tab. 1: Performance coefficients used for the calculation of the annual yield of the selected solar collectors with TRNSYS

Collector Type	η_0	a_1	a_2	a_{60}
Reference FPC	0.83	3.95	0.0094	4.52
$Honeycomb_{MIN}$	0.80	2.99	0.0096	3.57
$Honeycomb_{MAX}$	0.80	2.31	0.0106	2.94

The annual collector yield is calculated in dependence of the inlet collector temperature, which is assumed to be constant over the year. The results are displayed in Figure 4.

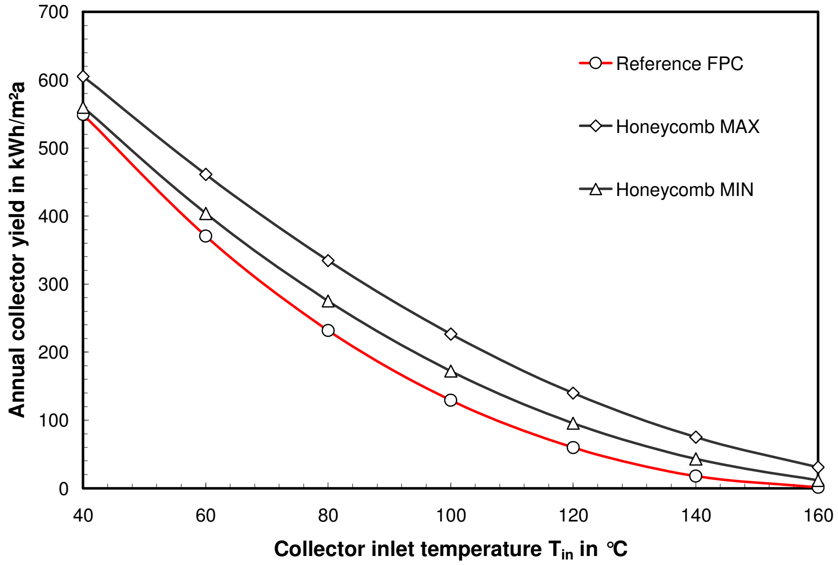


Figure 4: Annual collector yield of two prototype honeycomb collectors compared to a high-efficiency, state-of-the-art flat-plate collector. Collector $Honeycomb_{MIN}$ consists of 15 mm honeycomb panel and 30 mm backside insulation, collector $Honeycomb_{MAX}$ features 25 mm honeycomb panel and 50 mm backside insulation. All collectors are equipped with antireflective glass cover with low iron content. Simulation parameter: South orientation, 45° slope, Zürich

The efficiency improvement is especially noticeable at higher temperatures: At 80°C inlet temperature the honeycomb collectors could increase the annual yield of the reference collector by 20 and 44%, respectively. The same annual gain of the reference collector at 80°C could be achieved by the prototypes at temperatures of 88 and 100°C, thus attesting their potential in extending the operating range of a standard flat-plate collector.

4. Collector reliability

To investigate the long term temperature and UV radiation stability of the honeycomb structures for the use in flat-plate collectors we carried out extensive indoor and outdoor experiments on the different types of modified cellulose di- and triacetate films (s. Figure 5):

- Short- and long-term temperature stability tests on small honeycomb panels manufactured with all four supplied materials (150 x 150 mm) have been carried out in a hot cabinet equipped with a cooled receiver to collect possible outgassing products. The specimens were exposed to temperatures between 80 and 170 °C over a time up to 450 hours.
- Long-term temperature stability tests on selected large-sized collector covers (honeycomb panels manufactured with CTA-I and CTA-III glued to a low-iron glass pane, 1844 x 1125 mm) have been performed with a self-developed test facility, consisting in a climate chamber which enables to simulate the temperature distribution on the warm and the cold side of the collector cover under operating conditions. The specimens were exposed to temperatures of 130, 140, 150 and 160 °C over a time of

about 450 h.

- Laboratory weathering tests combining high temperature with UV irradiation were carried out on small collector covers (honeycomb panels manufactured with CTA-I and CTA-III glued to an antireflective low-iron glass pane, 200 x 100 mm). The specimens were exposed to a constant UV irradiance of about 70 W/m² using filtered metal halide lamps with an emitting spectrum similar to AM 1.0 and to temperatures ranging from 80 to 170°C over a time up to 300 h. The exposure time was defined on the basis of the observed material degradation.

- In addition two stagnating collector prototypes (honeycomb panels manufactured with CTA-I and CTA-III glued to an antireflective, low-iron glass pane, 1844 x 1125 mm) were exposed outdoor over a period of about 2 months in summer at the ISFH test roof (orientation: south, slope: 38°), in order to compare natural and artificial weathering and to better understand the degradation mechanisms of the materials.

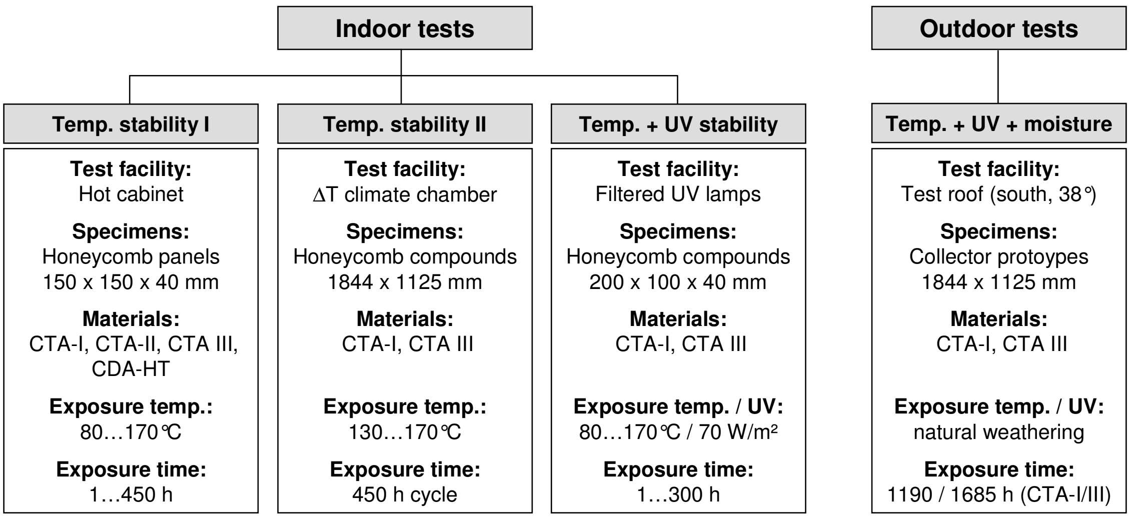


Figure 5: Overview of the reliability tests carried out on honeycomb panels and collector prototypes under investigation

Ageing effects are evaluated by means of visual inspection at different exposure time, focusing on the changing of the mechanical and optical properties of the structures (embrittlement, shrinking, yellowing), with following results:

- Temperature stability: modified cellulose triacetate honeycomb specimens (CTA-I) show outgassing already after 5 hours exposure at 80°C temperature, due to the evaporation of the plasticizers. Similar occurrences on CTA but at higher temperatures are already reported by the literature (Wallner and Lang, 2005). This effect gradually increases at higher temperatures and with prolonged exposure. After 450 hours ageing at 140°C a reduction of about 3 points of the optical coefficient η_0 was measured in an indoor test, thus proving the adverse effect of outgassing not only on the appearance but also on the performance of the collector. Noticeable changing in the mechanical properties was observed starting from temperatures above 140 °C, in agreement with manufacturer's data. Beside embrittlement, also heat shrinking occurs: a change in lateral dimensions of the honeycomb composite of about 5% was measured at 140°C. As a result compression resistance of the honeycomb structure dramatically decreases. Yellowing is observed as well at temperatures above 140°C.

  The presence and the content of plasticizers in cellulose triacetate films do affect as expected the material behavior under high temperature exposure. Only light outgassing has been detected on specimen and CTA-III and not below 140 °C. After prolonged exposure, similar yellowing as material CTA-I but more severe embrittlement was observed, which can be related to the missing plasticizers and the consequent reduced flexibility of the film.

  CDA-HT samples exhibit outgassing effects at low temperatures as well, even though different kinds of outgassing products could be detected, due to the different chemical composition of this film (plasticizers, thermal stabilizers or other additives). At higher temperatures we observed similar embrittlement and yellowing as with CTA-I samples. Against our expectations no improvement in the temperature stability of the material could be assessed.

- UV radiation stability: Combined stress of UV radiation and high temperature accelerates and intensifies the ageing processes of cellulose triacetate. Embrittlement could be already detected at temperatures above 100°C on CTA-I specimens and above 80°C on CTA-III specimens, whereas a much more severe degradation was detected on the latter samples. At higher temperatures the mechanical resistance of CTA-III films dramatically decreases and the honeycomb structure gets destroyed under minimum load. In spite of the positive influence in mitigating outgassing, the lack of plasticizers proves to adversely affect both temperature and UV radiation durability of cellulose triacetate films.

  Outdoor tests on stagnating collector prototypes qualitatively confirm our laboratory results, even though more intense degradation occurs, most probably due to the prolonged exposure and to the effect of additional weathering agents like moisture. Degradation level is strongly dependent on the temperature distribution over the honeycomb panel: At the warmest spots, where stagnation temperatures over 150 °C were measured, the beginning of a melting process of the honeycomb cells could be observed, below the declared short-term maximum temperature. This effect is assumed to be partially induced by UV radiation.

On the basis of our reliability tests none of the investigated materials proves to be suitable for the use in improved flat-plate collectors. The results attest the more severe operation conditions encountered by plastic materials in solar thermal collectors if compared to those of standard material test procedures.

5. Conclusion and outlook

We experimentally investigated honeycomb-compounds manufactured with modified cellulose di- and triacetate films for the use in high efficiency flat-plate solar thermal collectors. Aim of the work was to reduce the heat losses of a standard-collector of about 30% with a compact and light design, taking the temperature stability of these promising materials into consideration. Performance results confirm the expectations and show that a honeycomb-collector with an overall thickness of about 100 mm could achieve an optical efficiency of 0.80 and a loss coefficient a_{60} of about 3 W/m². At high temperatures or low irradiation level (η at $\Delta T/G = 0.12$ Km²/W) the collector efficiency is about 70% better than that of a standard collector.

Extensive reliability tests with regards to UV-radiation and high temperature durability have been carried out both on honeycomb-compounds and the collectors. The results show that none of the investigated materials is suitable for applications in improved flat-plate collectors. Depending on the selected material yellowing and embrittlement are already observed at temperatures above 100°C, clearly lower than expected according to manufacturer’s data. Outgassing, affecting the optical efficiency as well as the appearance of the collector, may occur at temperatures below 100 °C. The lack of plasticizers could mitigate outgassing, but drastically reduces the material durability. For applications at lower operating temperatures like integrated storage collectors the use of a glass pane with a moderate transmittance in ultraviolet spectral range is suggested to extend the long-term durability.

The project results confirm the difficulties to find a suitable plastic material for solar thermal applications at operating temperatures above 100°C. Considering the promising performance as well the attractive light design of similar collectors, further efforts are worthwhile to find new durable and cost efficient products or to develop simple strategies to control the maximum temperature level of collector components.

6. Acknowledgment

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