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ADHESIVE MATERIALS IN SOLAR-THERMAL COLLECTORS: RESULTS OF SEVEN YEARS FIELD EXPOURE AND ACCELERATED AGING TESTS USED TO SIMULATE 25 YEARS OF OPERATION

Abstract

The aging effects occurring in solar collectors within the adhesive material are by now not well understood. They are determined primarily by the temperature level in the collector. This temperature level has significantly increased during the last years due to the enhancement of the collector efficiency and the trend towards systems with higher solar fractions resulting in an increase in stagnation time and temperature. Furthermore, durability analyses of novel market products are needed, since only little is known about their long-term behavior.

The adhesive joint takes over multiple functions in the solar thermal collector, such as the structural bonding, the absorption of thermal stress of the transparent cover to the collector housing, and the sealing of the collector with respect to humidity, air and particle entry. Thus, it is a central component in a solar thermal collector. The accelerated aging tests represent the measured load in the solar collector at the component adhesive joint to a calculated corresponding period of 25 years.

We found that all test specimens showed a 100% cohesive fracture pattern after these stress tests. Thus, the adhesion between the transparent cover and the collector housing can be assumed to be ensured using the framework with the load depicted in the defined test cycle over 25 years. Compared to the reference value before aging, the tensile strength values change only marginally after being degraded.

The results of the mechanical properties of the small test specimens before and after seven years of exposure in maritime climate were determined and compiled. A nondestructive characterization method at the test specimens with Raman spectroscopy accompanied before the mechanical tensile testing was done.

Keywords: Components for solar collectors, adhesive, accelerated aging, thermal stress, humidity stress, tensile strength

1. Introduction

The objectives of the SpeedColl2 [1] project are the analysis of aging processes and the development of accelerated aging tests for solar thermal collectors and their components. The exposure of collectors and components in extreme climatic conditions enables the verification of these procedures and the development of degradation models.

Depending on their location and prevalent climatic conditions, the components of solar thermal collectors have to bear high climatic and mechanical stresses. High temperatures, UV-light, wind, snow, humidity or saline and corrosive atmospheres can be causes for a rapid degradation of materials and components.

This work focuses on the influence of the stress factors temperature and high humidity on the adhesive material bonding the glass cover with the frame in a solar thermal collector.

Since solar thermal technology is well-established round the globe, there is little information on the longterm behavior of collectors and solar systems. Basic statements on sealings in solar thermal collectors have been made at Sveriges Tekniska Forskningsinstitut (2013). This work was a guidance on how to define requirements on materials for seals and gaskets in solar collectors, mainly of flat plate type, potentially resulting in higher product quality and optionally also lower costs [2]. Additional work discussing the mechanical behaviour of an all-round fully adhesive supported absorber was done by Hermann Riess et al. (2013) [3].

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Of recent date are works of Shemelin et al., addressing a novel concept of unglazed solar thermal facade collector based on a commercially available metal cladding facade system. From the architectural point of view, adhesive bonding allows one to attach a hydraulic system to the absorber sheet without any visible injuries to the front of the facade collector (2022) [4]. The component adhesive materials have also appeared more frequently in connection with combining photovoltaics (PV) and solar thermal (T) technology systems in recent times. The work of Huang et al. is mentioned here as an example (2023) [5].

In this project environmental stress data such as humidity and temperature were determined. The data collection varies from tests carried out in alpine, moderate and maritime locations through to measurements in arid and tropical regions. Test stands are installed on the Zugspitze, as well as in Freiburg, Stuttgart, Gran Canaria, the Negev Desert in Israel and Kochi in India. Additionally, the solar collectors and components undergo accelerated aging tests in the laboratory. Using the collected data, we will in future validate the procedures of the aging tests, which will provide information about the collector's thermal performance over its entire lifetime.

2. Methods: Adhesive material durability testing

2.1 Adhesion and mechanical properties

Adhesion and mechanical properties of the adhesive material are determined by the use of small test specimens (H-bar, see figure 2) in tensile tests. Tensile tests are among the most frequently performed test methods in mechanical materials testing. They are used to characterize the strength and deformation behavior under uniaxial loading.

Representative material combinations were selected for this purpose. The collector manufacturers involved in the project provided the frame materials, which were then used as substrates for the H-bar with 2-component silicone in the three variants aluminum raw, aluminum powder coated and aluminum anodized.

2.2 Approach for durability testing

The determination of real load data takes place at various outdoor weathering test stands of the participating research institutes in Freiburg and Stuttgart (moderate climate), on the Zugspitze (alpine) and on Gran Canaria (maritime), as well as in India (tropical) and the Negev desert in Israel (arid). Here, the individual components adhesives, absorbers, reflectors and transparent covers as well as complete collectors are exposed.

The exposure sites are equipped with extensive stress monitoring that continuously records aging-relevant environmental conditions as well as various, microclimatic variables of materials and components. The data are used to investigate the effects of specific stress factors and combinations. The aim is to develop aging models and validated test procedures for components and collectors.

Galvanic isolation between the H-PK and the holding structure is ensured by a specially designed holder made of polytetrafluoroethylene (PTFE). In total a number of 405 test specimens were exposed.

As time transformation model the approach as by Savante Arrhenius was chosen. The derived equation of reaction kinetics, which describes the relationship between the change of a state P and the temperature, is [6]:

$$\Delta P \sim \exp\ [-E_T/RT] \qquad \text{(eq. 1)}$$

where ΔP is the chance of state, E_T is the Arrhenius activation energy expressing the temperature dependence of a thermal reaction of an absorber surface in kJ/mol, R is the ideal gas constant equal to 8.314 J/(K*mol), T is the temperature in K.

The accelerated test duration t_R corresponding to 25 years of real exposure at a constant temperature T_R is thus described by [7]:

$$t_R = 25 years \cdot \exp\left[-\frac{E_T}{R}\cdot\left(\frac{1}{T_{eff}}-\frac{1}{T_R}\right)\right] \qquad \text{(eq. 2)}$$

where t_R is the duration of the experiment in hours, E_T is the Arrhenius activation energy expressing the temperature dependence of a thermal reaction of an absorber surface in kJ/mol, R is the ideal gas constant equal to 8.314 J/(K*mol), T_{eff} is the effective mean temperature in K and T_R is the temperature for accelerated testing in K and a service life of 25 years with 8760 h per year.

2.3 Non-destructive analysis

As a complement to the tensile tests, a non-destructive analytical method was selected. A confocal Raman spectrometer (WITec, alpha 500) was used for the analysis of any molecular changes in the test specimens caused by the exposure. The excitation source is a laser with a wavelength of 532 nm and a maximum power of 75 mW. The spectrometer is equipped with a 600 lines/mm grating and a CCD camera. This analytical method was chosen to try an alternative to the classical standard tensile tests.

3. Outdoor exposure

A set of three different solar thermal collectors and small test specimens (H-bar) were exposed to outdoor weathering in different climatic regions while the climatic conditions (temperature, humidity, wind, precipitation, UV). The absorber and the adhesive joint temperature at different position were monitored continuously (figure 1).

Figure 1 Thermal solar collectors prepared with thermocouple to measure the adhesive joint temperature position top left (left) and position top middle(right) alt maritime exposition

Within the project, small test specimens (H-bar) with representative material combinations were used for the determination of real load data taking place (figure 2).

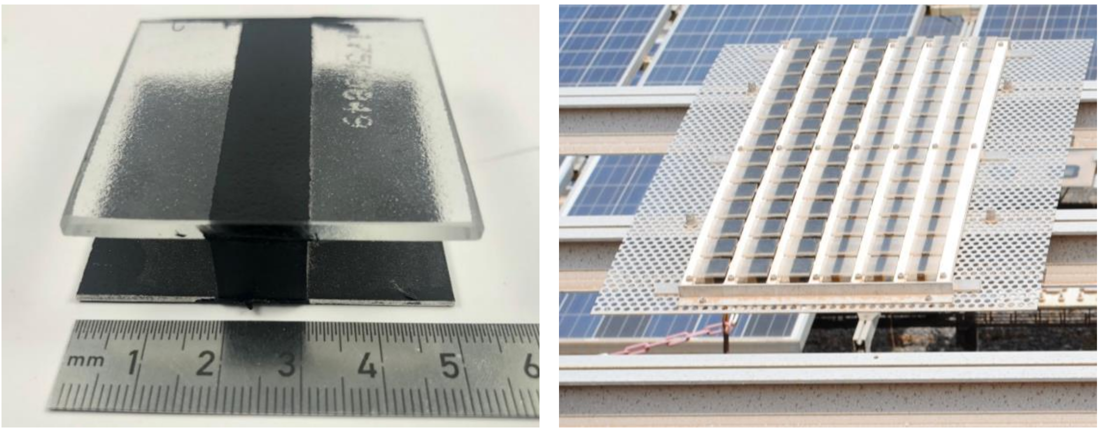


Figure 2 Test specimen (H-bar) for outdoor exposure and indoor testing, Adhesive is 12 x 12 x 50 mm³ (left) and Exposure at the maritime site Gran Canaria (right)

3.1 Quality assurance of meteorological data

A measurement system allowed all sensors read out by every minute and stored as 5-minute average values automatically. A data processing began after archiving the data in a database. The processing of meteorological parameters was done almost automatically. It started with a script-programmed pre-processing step, in which the absolute limits for each meteorological parameter are defined and existing data gaps and grossly erroneous values are identified by a plausibility check. Subsequently, the outliers were eliminated by a parameter-specific moving average and by considering the residuals. In addition, linear interpolation was applied for gaps of up to ten minutes and the data was converted to a suitable format for further manual processing. For larger gaps, data from the partner institutes at the respective sites had to be used. The missing data was then adapted accordingly by transformation using suitable models. Once this process had been completed, the data was checked for plausibility. With the release of the data, the data processing was completed. Figure 3 the shows the schematic diagram of the process [8].

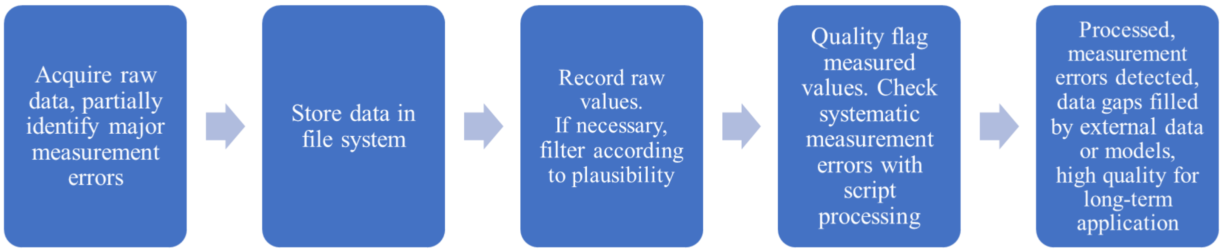


Figure 3 Schematic diagram of the process of measurement and processing of meteorological data.

3.2 Exposure of collectors and test specimens

A set of three collector types of similar collector design were exposed at the test sites. Table 1 documents the specifications of the collectors where η_0 is the optical efficiency [-], a_1 is the heat loss coefficient [W/(m²K)] and a_2 is the heat loss coefficient [W/(m²K²)]. The flat-plate collectors were all vented.

Tab. 1: Specification of the three collector types

type 1	type 2	type 3
solar glass cover	solar glass cover with an anti-reflective coating	solar glass cover
silicone bonding	aluminum cover strip	silicone bonding
PVD coated absorber on aluminum plate	PVD coated absorber on aluminum plate	PVD coated absorber on aluminum plate
harp-shaped copper piping ultrasonically welded	meander-shaped copper piping laser-welded	meander-shaped copper piping laser-welded
mineral wool insulation	melamine epoxy foam insulation	mineral wool insulation
aluminum frame	aluminum frame	aluminum frame
steel bottom plate with an aluminum-zinc coating	steel bottom plate with an aluminum-zinc coating	steel bottom plate with an aluminum-zinc coating
aperture area 1.94 m²	aperture area 2.33 m²	aperture area 2.35 m²
η_0= 0.739, a_1 = 3.98, a_2=0.0096	η_0= 0.793, a_1 = 3.63, a_2=0.0119	η_0= 0.835, a_1 = 3.63, a_2=0.0132

In order to gain also worst-case data, some of the test sites were located in extreme climates with very harsh conditions (Table 2).

Tab. 2: Exposure of collectors and test specimens (H-bar) at locations with extreme climates and the tilt angle

Climates	Location	Tilt angle	collectors	test specimens H-bar
tropical	Kochi, India	25°	yes	yes
alpine	Schneefernerhaus, Zugspitze, Germany	45°	yes	no
arid	Sede Boker, Negev Dessert, Israel	31°	yes	no
maritime	Pozo Izquierdo, Gran Canaria, Spain	23°	yes	yes
moderate	Stuttgart and Freiburg, Germany	45°	yes	yes

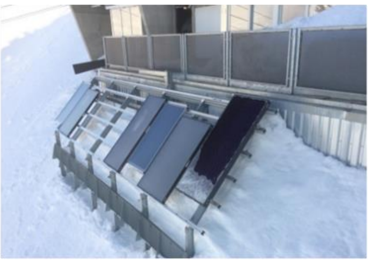
Small test specimens (H-bar) with representative material combinations were used for the determination of real load data taking place. The collector manufacturers involved in the project provided the frame materials, which were then used as substrates for the H-bar with 2-component silicone in the three variants aluminum raw, aluminum powder coated and aluminum anodized (Table 3).

Tab. 3: Specification of the three H-bar types

type 1	type 2	type 3
solar glass cover 3 mm thickness	solar glass cover 3 mm thickness	solar glass cover 3 mm thickness
silicone bonding 12 mm thickness	silicone bonding 12 mm thickness	silicone bonding 12 mm thickness
substrate aluminum row 5.0 mm thickness	substrate aluminum powder coated black 1.2 mm thickness	substrate aluminum anodized black 1.8 mm thickness

In Figure 4, the six outdoor exposure test sites are shown. The tilt angle was chosen to maximize the solar gains depending on the latitude of the different locations. All collectors were oriented to the south.

Kochi, India, tropical

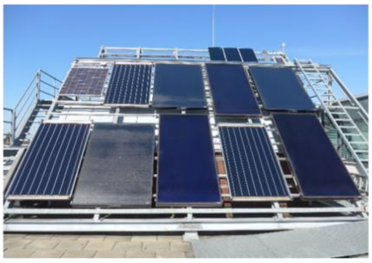


Zugspitze, Germany, alpine

Negev, Israel, arid

Gran Canaria, Spain, maritime

Stuttgart, Germany, moderate



Freiburg, Germany, moderate

Figure 4 Exposure sites for solar thermal collectors and test components at different climate conditions tropical, alpine, arid, maritime and moderate

4. Results

4.1 Outdoor exposure of solar thermal collectors and test specimens: thermal stress

A set of three different solar thermal collectors were exposed to outdoor weathering under different climatic conditions while the climatic conditions and the collector microclimate were monitored continuously.

The histogram of the absorber temperature distribution in a solar thermal collector of type 3 is shown for different climates within the project is shown in figure 5. Since the collectors were not connected hydraulically to a domestic hot water system, the operating mode took place under constant stagnation conditions in an air air-filled system. For the different exposure sites, we used the same flat-plate collector type with a transparent cover. The sides and back of the housing were insulated to reduce heat loss to the environment.

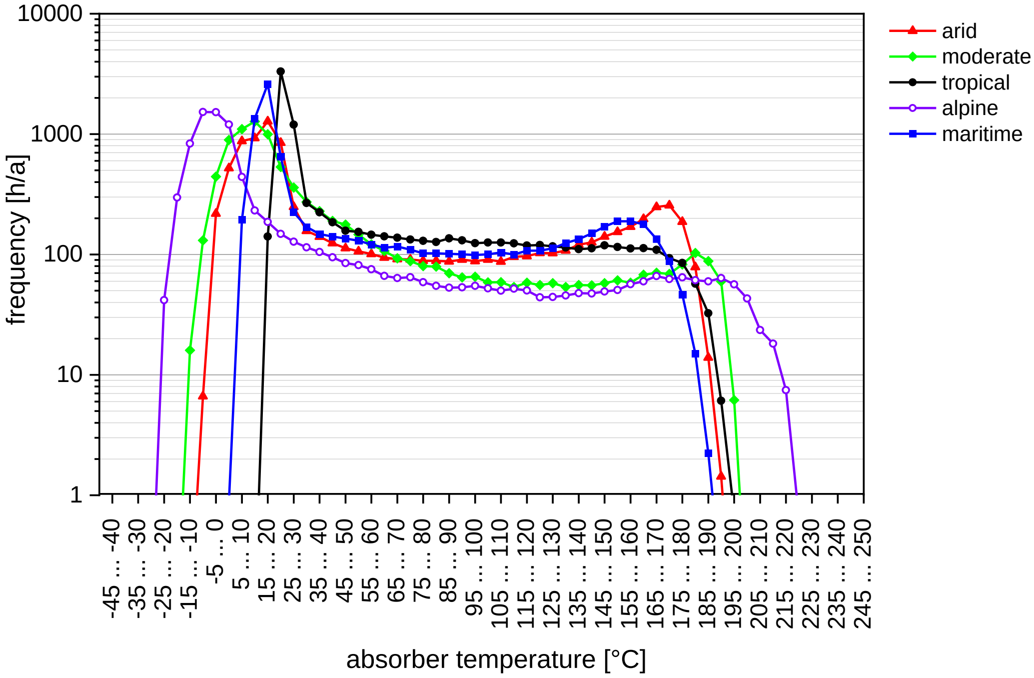


Figure 5 Absorber temperature distribution in a solar thermal collector under stagnation condition for different climates

The highest absorber temperatures, up to 225 °C, were measured at the alpine exposure site. This unexpected result is explained by the albedo effect of reflection from glacier and snow surfaces and by the relatively low pollution levels of the air at the alpine exposure site compared to all other exposure sites [8].

In figure 6 the ambient- and adhesive joint temperature and irradiance in collector plane at the arid exposition site at Negev desert in Israel for one typical day in September is shown.

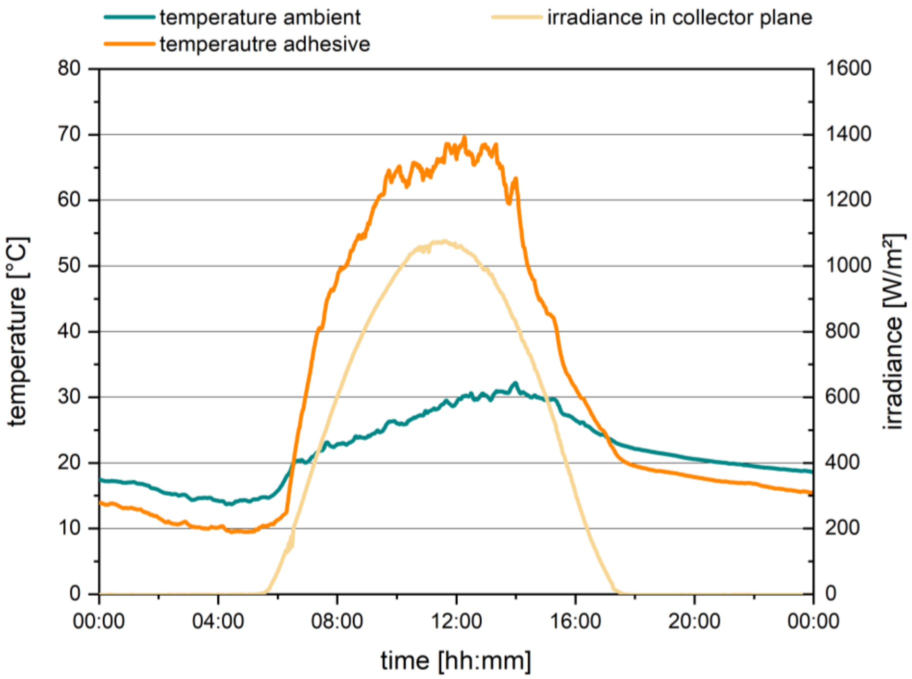


Figure 6 Ambient- and adhesive joint temperature one day in °C and irradiance in collector plane at arid exposition at Negev desert, Israel

In comparison to the other locations the alpine exposure site reaches the highest absorber temperature, which is consequently associated with a higher temperature within the collector and therefore the highest thermal load on the adhesive joint can be assumed in this case. Therefore, the thermal stress load of the alpine exposition site was chosen for the further for deriving the service life modeling. In figure 7 the frequency in hours in the period of one year of the adhesive joint- and ambient temperature at alpine exposition site is shown.

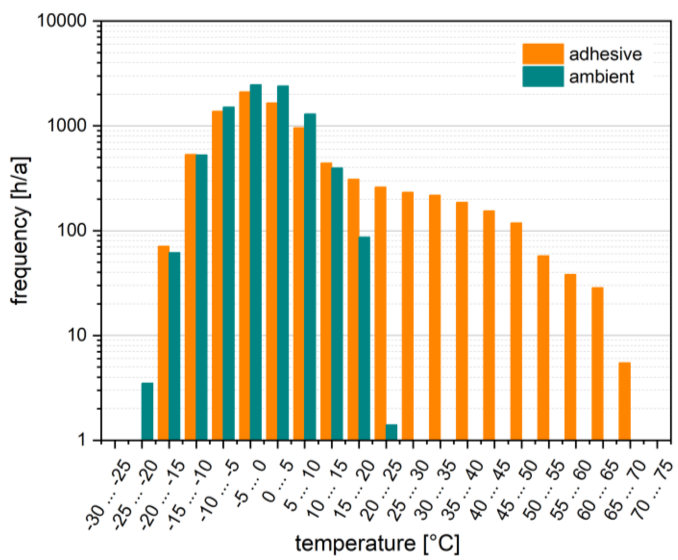


Figure 7 Frequency in hours in the period of one year of adhesive joint- and ambient temperature in °C at alpine exposition at Zugspitze, Germany

The maximum measured temperature of adhesive joint reaches 70 ° C. This value is over 45 K higher than the maximum measured ambient temperature.

4.2 Outdoor exposure of solar thermal collectors and test specimens: humidity stress

In order to answer the question of the combination of temperature and humidity stress regarding the adhesive material, a measurement was carried out on collectors in a cooled operating state at the reference site in Stuttgart. For this purpose, the collectors were hydraulically connected to a corresponding heat sink. The temperature and relative humidity inside the collector in the air gap between the absorber and the transparent cover was measured with a combined temperature and humidity sensor. The sensor was inserted through the back wall of the solar collectors and through their absorbers into the space between the absorber and the transparent cover. The preparation of the collector was done during the production at the solar collector manufacturer. The positioning in the collector plane was centered to the absorber length and width.

In figure 8 the temperature of the adhesive and relative humidity in the collector between absorber and inner glass surface is shown.

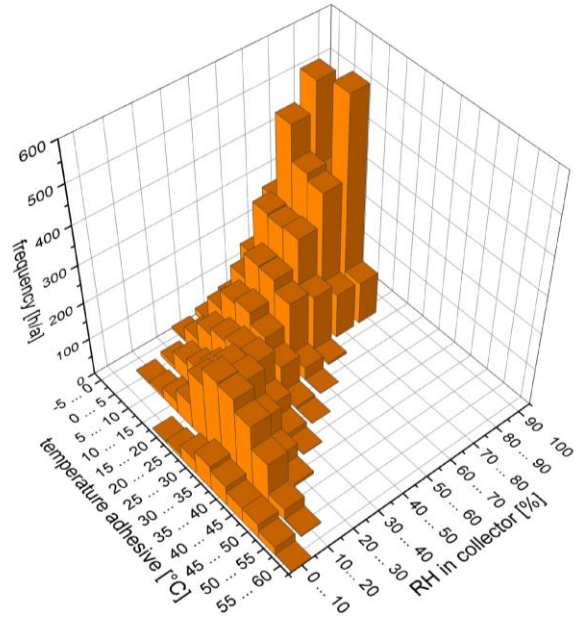


Figure 8 Temperature adhesive and relative humidity in the collector between absorber and inner glass surface at moderate exposition site, Stuttgart, Germany

A simultaneous occurrence of high temperature and relative humidity cannot be observed. in solar collectors during operation conditions. Therefore, a combined test with high temperature and humidity is not considered to be useful, since this stress situation for the adhesive joint in solar collectors does not occur in operation.

4.3 Adhesive material durability testing: Modelling

These measurement data are used to define the test conditions and times. As a model for the kinetics of the degradation processes, the approach of a time transformation according to Arrhenius was chosen (eq. 2). As a corresponding time period for a service life estimation, 25 years were used, corresponding to the expected period of service life for a thermal solar collector. In figure 9 the testing time for adhesive materials for thermal stress equivalent to a lifetime of 25 years with thermal loads taken from the alpine exposition site for three different testing temperature 75 °C, 85 °C and 95 °C, as function of the activation energy are shown.

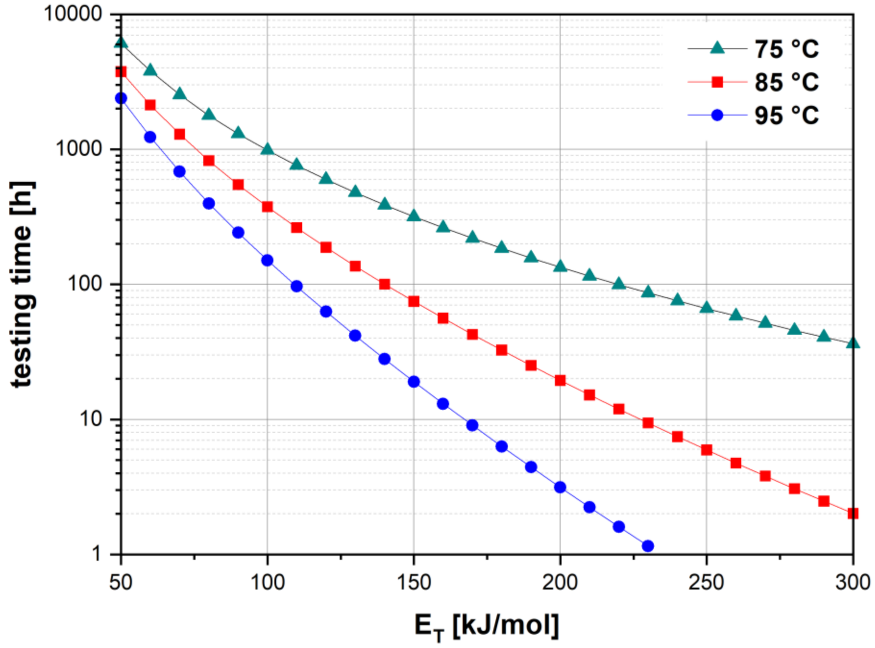


Figure 9 Testing time for adhesive material for thermal stress equivalent to a lifetime of 25 years with thermal loads taken from Zugspitze (alpine) in the German Alps for 75 °C, 85 °C and 95 °C testing temperature, as function of the activation energy

Based on the temperature stresses measured at the site and the accelerated aging model, we propose to perform the following tests on H-test specimens. The relevant stress factors of temperature and humidity will be considered. In addition, the adhesion properties between transparent cover and substrate will be tested. For this purpose, the adhesive joints are statically stretched in tension during aging (figure 10).

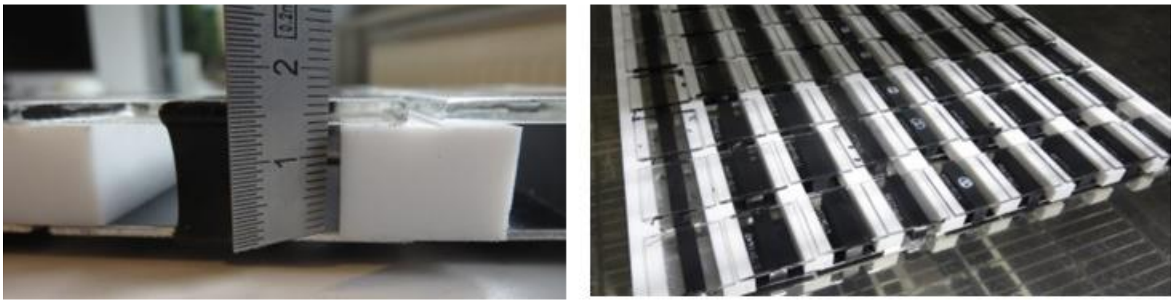


Figure 10 Test specimen (H-bar) for indoor Part C mechanical load via static elongation (12,5%) (left) and set in climate cabinet

The tests represent the measured load in the solar collector at the component adhesive joint to a calculated corresponding period of 25 years. For the activation energy, a value of 90 kJ/mol was assumed, at which the hydrolysis of covalent siloxane bonds takes place [9]. A maximum testing temperature of 85 °C was specified for the service life test. Using the approach described above, a value of approx. 600 h test time can be derived for the temperature stress factor in order to achieve an accelerated aging of 25 years. The test sequence consists of three parts addressing temperature and moisture stability as well as a combination of moisture stability and mechanical load via static elongation shown in table 4.

Tab. 4: Testing Part A, B, C

Testing	**Temperature**	**Humidity**	**Duration**
Part A temperature stability	85 ° C	RH <10%	600 h
Part B moisture stability	40 ° C	RH 95%	600 h
Part C moisture stability and mechanical load via static elongation (12,5%)	40 ° C	RH 95%	600 h

4.4 Adhesive material durability testing: results

In table 5 the results of the test sequence part A, part B and part C for one H-bar test specimen are shown. The results of the mechanical properties of the small test specimens before and after seven years of exposure are being determined and compiled. For statistical reasons, five test specimens were used for each test. The values shown are therefore mean values from a sample of n=5. In order to capture the time dependence of the accelerated aging on the test specimens, the following time steps were chosen: 150 h, 300 h, and 600 h.

Tab. 5: Testing results for Part A, B,

Part A temperature stability				
Mean value from 5 H-bars	tensile strength [N/mm²]	elongation F_{max}. [%]	tensile strength at 10% elongation [N/mm²]	tensile strength at 50% elongation [N/mm²]
Reference	0,58	115	0,12	0,30
150 h	0,62	141	0,11	0,29
300 h	0,52	131	0,10	0,25
600 h	0,52	134	0,09	0,24
fracture pattern	**100% cohesive fracture each (n=20)**			

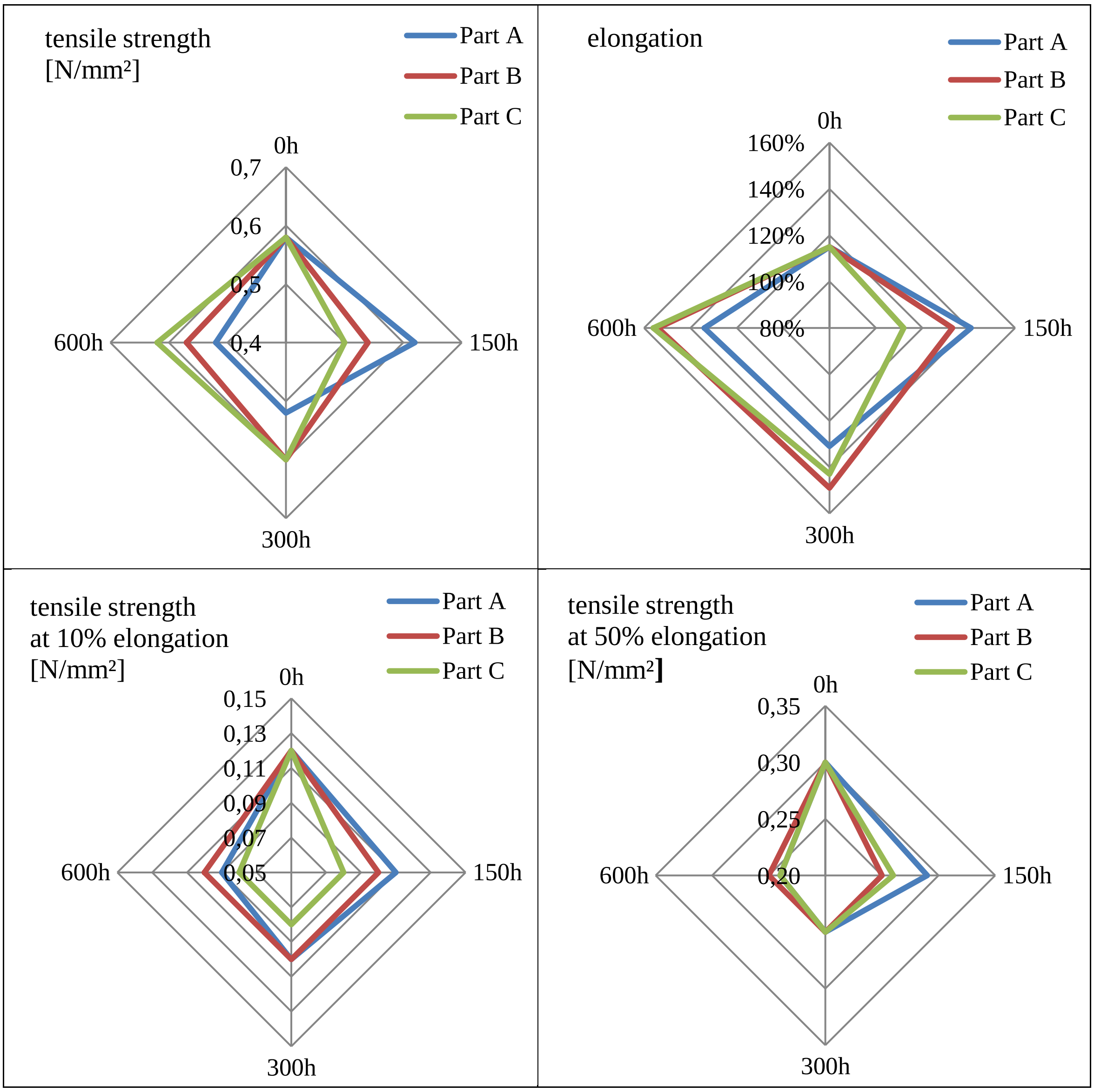
Part B moisture stability				
Mean value from 5 H-bars	tensile strength [N/mm²]	elongation F_{max}. [%]	tensile strength at 10% elongation [N/mm²]	tensile strength at 50% elongation [N/mm²]
Reference	0,58	115	0,12	0,30
150 h	0,54	133	0,10	0,25
300 h	0,60	149	0,10	0,25
600 h	0,57	154	0,10	0,25
fracture pattern	**100% cohesive fracture each (n=20)**			

Tab. 5: Testing results for Part C

Part C moisture stability and mechanical load via static elongation (12,5%)				
Mean value from 5 H-bars	tensile strength [N/mm²]	elongation $F_{max.}$ [%]	tensile strength at 10% elongation [N/mm²]	tensile strength at 50% elongation [N/mm²]
Reference	0,58	115	0,12	0,30
150 h	0,50	112	0,08	0,26
300 h	0,60	143	0,08	0,25
600 h	0,62	156	0,08	0,24
fracture pattern	**100% cohesive fracture each (n=20)**			

In order to present the changes in mechanical properties more clearly, the results were also presented graphically (table 6).

Tab. 6: Testing results for Part A, B, C



Performed tensile tests after aging according to part A, B or C showed 100% cohesive failure for all samples regardless of the chosen testing period up to 600 h. Therefore, the mechanical properties with respect to the adhesion of the transparent cover to the collector housing can be assumed to be ensured within the scope of the load depicted in the defined test cycle for more than 25 years. Compared to the reference values of the tensile strength, the values change only slightly after the service life test (-14% to 7%).

4.5 Outdoor exposure of test specimens at maritime exposition site

At the maritime site we had the facilities to keep a complete set of test specimens in sum 7.8 years continuously in exposure. In figure 11 an unexposed (reference) and an exposed test specimen are shown.

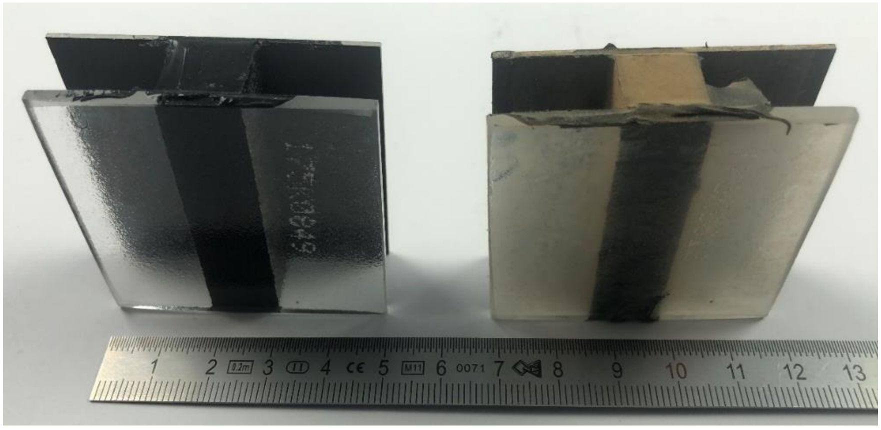


Figure 11 Test specimen (H-bar) Adhesive is 12 x 12 x 50 mm³left: reference, right after 7.8a Gran Canaria

Raman spectra were previously recorded on these test specimens before the mechanical tensile tests. In figure 12 Raman spectra and the associated molecule group vibrations for the reference sample and for the 7.8 years exposed sample at the maritime test site as measured and after a background subtraction are shown.

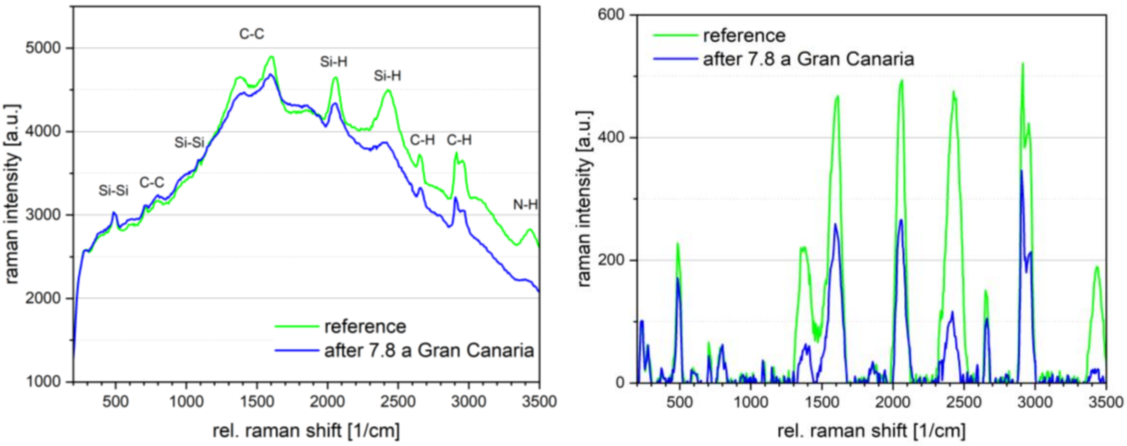


Figure 12 Raman spectra and associated molecule group vibrations reference and after 7.8a Gran Canaria as measured (left), after background subtraction (right)

It remains to be noted that no new Raman bands were caused by the exposure. It can therefore be concluded that the material has probably not changed chemically. The results of the tensile test are summarized in Table 7. A glass breakage was detected on one of the test specimens. Thus, mechanical tensile tests could only be performed on 4 H-bars for the type 1 material combination.

7.8 years exposure at maritime test site, Gran Canaria H-bars type 1, type 2, type 3				
Mean value	tensile strength [N/mm²]	elongation F_{max}. [%]	tensile strength at 10% elongation [N/mm²]	tensile strength at 50% elongation [N/mm²]
type 1 Reference from 5 H-bars	0,45	101	0,10	0,26
type 1 7.8a maritime from 4 H-bars	0,58	50	0,10	0,57
type 2 Reference from 5 H-bars	0,76	67	0,25	0,66
type 2 7.8a maritime from 5 H-bars	0,58	50	0,20	0,57
type 3 Reference from 5 H-bars	0,98	57	0,34	0,91
type 3 7.8a maritime from 5 H-bars	0,55	31	0,28	-
fracture pattern	**100% cohesive fracture each (n=29)**			

As a result of the comparison of the values of the tensile strength before and after exposure, changes from -44% to 45% can be observed. For all samples a 100% cohesive failure pattern was found.

5. Conclusion

Different solar thermal collectors and components we exposed in various extreme climates within the speedcoll2 project. Continuously monitoring the outdoor climatic conditions and the micro-climate inside the collector was performed. A complex quality assurance of the measured data was successfully defined and implemented.

The maximum measured temperature of adhesive joint in a solar thermal collector under stagnation conditions reaches 70 °C at the alpine exposition site. This value is over 45 K higher than the maximum measured ambient temperature. Based on the temperature stresses measured and the accelerated aging model, a proposal for service life test with three parts was developed. The relevant stress factors of temperature (Part A) and humidity (Part B) will be considered as well as the adhesion properties between transparent cover and the collector frame. For this purpose, the adhesive joints are statically stretched in tension during the aging tests (Part C).

Performed tensile tests after aging according to part A, B and C showed 100% cohesive fracture pattern for all samples regardless of the chosen testing period up to 600 h. Therefore, the mechanical properties with respect to the adhesion of the transparent cover to the collector housing can be assumed to be ensured within the scope of the load depicted in the defined test cycle for more than 25 years. Compared to the reference values of the tensile strength, the values change only slightly after the service life test (-14% to 7%).

The accelerated aging tests represent the measured load in the solar collector at the component adhesive joint to a calculated corresponding period of 25 years.

At the maritime site we had the facilities to keep a complete set of test specimens in sum 7.8 years continuously in exposure.

Tensile tests of the unexposed and the exposed test samples showed 100% cohesive fracture pattern for all samples. As a result of the comparison of the values of the tensile strength before and after exposure, changes from -44% to 45% and a 100% cohesive failure pattern can be observed.

A confocal Raman spectrometer was used as a non-destructive analytical method as a complement to the tensile tests. This allowed the analysis of any molecular changes in the test specimens caused by the exposure. No new Raman bands were caused by the exposure. It can therefore be concluded that the material has probably not changed chemically. This statement is consistent with the measurement results of the tensile test and the suitability of the method could be proven.

6. Acknowledgements

The authors would like to thank the industrial partners and the German Federal Ministry for Economic Affairs and Energy (FKz. 0325969A) for their support and funding.

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