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Global Aging and Lifetime Prediction of Polymeric Materials for Solar Thermal Systems – Part 1: Polypropylene Absorbers for pumped Systems

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Abstract

Part 1 of the paper series deals with the lifetime estimation of black-pigmented polypropylene (PP) absorber grades for pumped solar thermal hot water systems at different sites (Antalya, Mumbai, Phoenix). Absorber temperature loading profiles were calculated using the simulation tool SHW. Furthermore, global aging experiments in hot air or heat carrier fluid were carried out on specimen level for a commercial black-pigmented PP grade. The aging indicator strain-at-break was measured and endurance times at elevated temperatures (up to 135°C) were extrapolated to service relevant temperatures (maximum 105°C). The lifetime was assessed by weighting the temperature dependent endurance times with the loading profiles assuming cumulative damages. Depending on the extrapolation method, system specific parameters (i.e., collector size and storage tank volume) and installation site, lifetimes ranging from 20 to 45 years were obtained.

Keywords: Polypropylene; Solar thermal collector; Absorber; Lifetime

1. Introduction

The amount of polymers used in pumped or non-pumped solar thermal collector systems steadily increases. For polymeric solar thermal absorber materials a long-term stability in hot heat carrier fluid or hot air environment is of prime importance [Kahlen et al., 2010a, 2010b]. To meet high demands in terms of efficiency and lifetime, special attention is given to control the maximum stagnation temperature and pressure level. Based on research work in IEA SHC Task39 [Köhl et al., 2014] and collaborative projects different concepts for polymer-based collector systems have been established and successfully introduced into the market. According to Köhl et al. (2005) for solar thermal collectors a service lifetime of 20 years is required. Polymeric flat-plate collectors for pumped systems are usually operated at maximum temperatures up to 120°C and maximum pressure levels below 3 bar [Lang et al., 2013]. For overheating protection, various measures such as controlled back-cooling or venting are implemented. Novel glazed, flat-plate collector concepts allow for cost-efficient polymeric absorber materials such as black-pigmented polypropylene (PP).

At elevated temperatures some polyolefin pipe grades (e.g. polypropylene) do not exhibit the stress dependent Region II characterized by quasi-brittle failure (Ifwarson and Leijström, 1992). Hence, it is of utmost importance to investigate and characterize primarily the large scale aging and brittle failure. A main task of current research work is the lifetime assessment of these novel absorber materials for more cost-efficient and reliable collector systems. The main objective of Part 1 of the paper series is to investigate the aging behavior and to assess the lifetime of a selected PP grade for a pumped collector systems for hot water

preparation in single family houses at selected sites. Therefore, the "One World Solar Collector" of Sunlumo Technology was used.

2. Methodological approach

2.1 Materials, aging conditions and characterization

A commercial black-pigmented polypropylene grade (PP-Bα), commonly used in swimming pool absorbers, was selected for the investigations. The amount of carbon black in the investigated grade is about 2 wt.%. The PP grade is stabilized with a stabilization package commonly used in piping applications. The specific amount and type of stabilizers are described and discussed in Povacz (2014).

Based on compression molded 2 mm thick plates, 0.1 mm thick micro-sized specimens were automatically manufactured using an adapted CNC-milling technique [Grabmayer et al., 2015]. The specimens were aged in hot air and hot heat carrier fluid in a Binder FED 53 (Tuttlingen, Germany) heating chamber with forced circulation at 95, 115 and 135°C. The hot air aged specimens were freely suspended on a grid. Hot water aging was carried out in closed stainless steel vessels with integrated sample holder to prevent contact between the samples. At least three specimens were removed and characterized per aging interval.

To assess the aging behavior, tensile tests were carried out at 23°C using a screw-driven universal testing machine with a test speed of 50 mm/min. The strain-at-break values were obtained over the aging period. Embrittlement was defined and classified when strain-at-break values dropped below strain-at-yield value ($\varepsilon_b < \varepsilon_v$). The ε_v values were 8% for PP-B α

2.2 Modeling of service relevant loading conditions

Based on the study by Kaiser et al. (2013) and prevalent market potentials, three different climatic conditions (mediterranean (Antalya/Turkey), hot and dry (Phoenix/USA), hot and humid (Mumbai/India)) were taken into account for this work. Based on Meteonorm-data, relevant climatic parameters (e.g. air temperature, relative humidity, global radiation) were established on an annual basis. In a further step for all three climate zones market-based polymeric collector systems based on the Sunlumo "One World Solar Collector" for hot water preparation in single family houses were defined and evaluated. By theoretical modeling, annual time/temperature distributions for the absorber were obtained, which were significantly dependent on the location and the associated climatic conditions.

2.3 Lifetime assessment

For lifetime estimation a cumulative damage approach was used established by Wallner et al. (2016) for black-pigmented PP solar absorber materials. The main elements of the approach are the simulation of temperature loading profiles, the extrapolation of aging data and the cumulation of damages of different temperature levels. Endurance time extrapolation was carried out using a theoretical Arrhenius approach and an empirical extrapolation approach assuming a specific degradation mechanism in the temperature range. Various aging processes and changes of degradation mechanism of polymeric materials described by several authors [Gugumus, 1999; Kahlen et al., 2010a, 2010b] may result in an invalid use of the Arrhenius approach. Hence, an additional empirical extrapolation approach was used in this study which is based on an S-shaped degradation curve derived for polypropylene grades with comparable stabilizer packages by Gugumus (1999). Endurance times at service relevant temperatures were calculated by adapting and shifting aging data published by Gugumus (1999) to recorded failure times of the investigated materials. Additionally, a constant endurance time of 50 years at lower temperatures was assumed according to Leijström and Ifwarson (1998).

As endurance time limits the aging indicator time-to-ultimate-failure of the specimen was characterized by tensile testing. Expected endurance times in the service relevant temperature range from 5 to 105°C (in 5 K intervals) were determined using Arrhenius and Gugumus approach. A cumulative damage model according to ISO 13760 was used for lifetime assessment. It is assumed, that each effect caused by a constant service condition is proportional to the duration of the exposure and furthermore the damage from different service conditions can be added cumulatively. The lifetime is deduced by weighting the temperature dependent

endurance times with the frequency distribution of the temperature loading profiles (Miner's rule).

3. Results and discussion

3.1 Service relevant loading conditions

Table 1 presents the used technical parameters for the simulation of temperature loading profiles for three different locations. The collector areas vary between 3.6 m² in Mumbai and 5.4 m² in Phoenix per single household. The simulated absorber temperatures are strongly influenced by the environment and the storage system. Fig. 1 illustrates the simulated time/frequencies of the absorber temperatures in the polymeric collector system for hot water preparation in single family houses at three different locations. Maximum stagnation temperature values of 95°C for Mumbai or up to 105°C for Phoenix were deduced. However, differences between the collector systems can be observed by the frequencies at temperatures above 75°C, which occur more often for Phoenix and Mumbai. In the temperature range between 20 and 75°C all three locations show comparable curve shape with frequencies of about 400 h/a. While in the hot and humid location Mumbai the lower limit of the temperature profile is about 10°C, all other locations show minimum temperatures of 5°C with higher frequencies. The frequency maxima are in the temperature range between 10 and 30°C with a shift to higher temperatures of about 25°C in hot and humid sites such as Mumbai.

 Tab. 1: Specific parameters of pumped collector systems for hot water preparation in single family houses at different sites.

		Antalya	Mumbai	Phoenix
Collector	Area [m ²]	4.4	3.6	5.4
	Inclination [°]	36.9	19	34
	Orientation [-]	South	South	South
Storage	Volume [l]	300	150	300
	Height [m]	1.8	1.2	1.8
	Required temperature [°C]	55	45	60
	Hot water consumption [l/d]	160	120	250



Fig. 1: Time/frequencies (h/a) of the absorber temperatures in polymeric pumped solar thermal collectors at different sites.

3.2 Aging behaviour at elevated temperatures

As reported and discussed in previous research work by Povacz (2014), for the investigated PP grade a more critical aging behavior in hot air than in hot heat carrier fluid was deduced at 95, 115 and 135°C. Based on

these results only hot air aging behavior is discussed and the aging data in hot air is used for lifetime assessment in this study. Fig. 2 shows the strain-at-break values of the black-pigmented PP-B α as a function of aging time in hot air at 95,115 and 135°C. Ultimate failure with strain-at-break values below yield point is indicated with open symbols in the chart.

Unaged specimens reveal a highly ductile behavior with strain-at-break values of about 70. A significant decrease in strain-at-break values are obtained for PP-B α at 135°C with ultimate failure within 2350 h. At 115 and 95°C, an initial drop appears within 150 h, presumably related to physical aging effects such as recrystallization. At longer hot air aging times the material behavior is characterized by a plateau of about 15 and 50% with ultimate failure appearing after 8400 and 26500 h for 115 and 95°C, respectively.



Fig. 2: Strain-at-break values for 100 μm micro-sized black-pigmented PP-Bα specimen exposed in hot air at 135, 115 and 95°C as a function of aging time.

3.3 Extrapolated endurance times and estimated lifetimes for micro-sized PP specimen

Fig. 3 illustrates the reciprocal temperature (K^{-1}) over the derived endurance limits for the investigated PP grade. The experimental embrittlement times at elevated temperatures were fitted using an Arrhenius approach (top layer) as well as the Gugumus approach (bottom layer). All extrapolated curves exhibit sufficient high coefficient of determination. The dashed vertical line depicts the assumed 50 years cut-off limit. Experimental data examined and published by Gugumus (1999) are shown in the bottom layer of Fig. 3. Two different antioxidant stabilizer packages (AO-1 and AO-7) are illustrated to show the S-shaped curve of endurance limits. In this study, average values of these curves are considered to establish endurance time limits at service relevant temperatures.

For PP-B α the linear Arrhenius approach results in endurance limits of about 4 years (1500 days) at 90°C and of about 13 years (4650 days) at 75°C. By using the Gugumus approach, the endurance limits at 90°C are in the same range of about 4 years. The obtained endurance limits of about 5 years (1790 days) at 75°C are significantly lower compared to the Arrhenius extrapolation approach. Hence, the Gugumus approach results in more conservative behavior due to the S-shaped curve. Compared to the homopolymer grades investigated by Gugumus (1999) a much better behavior is obtained for the black copolymer grades of this study.

Lifetime estimation is deduced for the investigated black-pigmented specimens according to an optimistic upper bound scenario (Arrhenius approach) and a conservative scenario (Gugumus approach) for three different climate zones. Table 2 shows the calculated lifetimes which are ranging from 20 to 45 years for the investigated black-pigmented PP grade. Highest lifetimes are achieved for Antalya due to the mild mediterannean climate. Using the Arrhenius approach, the lifetimes are significantly affected by the constant endurance time of 50 years that is assumed for temperatures below 60°C. Due to the S-shaped curve of the Gugumus approach, this effect is even less pronounced and particularly shifted to shorter endurance times.

Hence, for all climate locations the predicted lifetimes using the Gugumus approach are much lower, ranging from 20 to 24 years. The estimated lifetime is reduced due to harsher climate conditions (hot and humid) in Mumbai to about 20 years.



Fig. 3: Experimental failure times for micro-sized black-pigmented PP-Bα specimens and extrapolated endurance times at service relevant temperatures using an Arrhenius approach (top) and the Gugumus approach (bottom).

Tab. 2: Estimated lifetimes for PP-Bα absorber materials in pumped flat-plate collector systems at three different sites for the Arrhenius and Gugumus extrapolation approach.

Location	Climate zone	Estimated lifetimes in years		
		Arrhenius	Gugumus	
Antalya	mediterranean	45	24	
Mumbai	hot and humid	41	20	
Phoenix	hot and dry	41	23	

4. Summary and conclusion

To assess the lifetime of a black-pigmented PP absorber grade for pumped solar thermal hot water systems, temperature loading profiles for three different climate zones were established based on climatic input data. Strain-at-break values as experimental failure data were determined after exposure to hot air and hot heat carrier fluid at elevated temperatures ranging from 95 to 135° C of micro-sized specimen with a thickness of 100 µm. Due to the pronounced aging behavior in hot air, these data were used for lifetime estimation. Furthermore, endurance times at service relevant temperatures (5 to 105° C) were extrapolated using a linear Arrhenius approach (optimistic upper boundary) and a S-shaped Gugumus approach (conservative scenario). Subsequently a model of cumulative damages (Miner's rule) was applied to assess lifetimes for the investigated black-pigmented PP absorber materials.

Depending on the installation site and the system type, annual time/frequency curves over the collector temperature were simulated for polymeric hot water systems in single family houses. The absorber temperature was ranging between 5 and 105°C for the investigated climate zones with comparable curve shape between 20 and 75°C at frequencies of about 400 h/a. However, differences between the three systems were observed with pronounced frequencies at low temperatures (< 20°C) for Antalya (mediterranean) and Phoenix (hot and dry) as well as at high temperatures (> 75°C) for Mumbai (hot and humid) and Phoenix (hot and dry).

The unaged specimens exhibited ductile material behavior with strain-at-break values of about 70% for PP-B α . Hot air aging at 135°C resulted in a steady decrease of strain-at-break values till ultimate failure appeared within 2350 h. The estimated lifetimes for the PP absorber material was ranging from 20 years (Mumbai, Gugumus approach) to 45 years (Antalya, Arrhenius approach). The calculated lifetimes were significantly depending on the used extrapolation method. As an optimistic upper boundary scenario, the linear Arrhenius approach showed estimated lifetimes higher than 41 years for all climate zones, strongly affected by the endurance cut-off limit of 50 years. Contrary, the non-linear Gugumus approach as a conservative lifetime assessment resulted in lifetimes ranging from 20 to 24 years.

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