Composites Part B 303 (2025) 112611

Contents lists available at ScienceDirect

Composites Part B

journal homepage: www.elsevier.com/locate/compositesb

A review on aging effects of thermoset prepregs

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ARTICLE INFO

Handling Editor: Prof. Ole Thomsen

Keywords:
Polymer matrix composites (PMC)
Carbon fiber
Processing
Curing
Mechanical properties
Recycling

ABSTRACT

Thermoset prepreg materials have emerged as the composite material of choice for demanding structural applications due to their easy processability and attainable superior part quality. However, the one-part nature of common precured thermoset resins renders prepregs susceptible to undesired premature curing, which may entail deterioration of properties. This article reviews the current state of literature on thermoset prepregs with regard to aging effects that originate before the final cure cycle of composite parts, i.e., during freezer storage and upon temporary storage or processing at room temperature. Therefore, the aging-related evolution of the physio-chemical prepreg state (conversion, T_g, water uptake, viscosity) is examined in terms of its impact on processing (forming properties, tack), applied cure cycles (temperature, dwell time, pressure) and post-cure laminate quality (porosity, mechanical performance). The economic and ecological implications of prepreg aging are discussed in conjunction with recycling strategies for out-of-spec material. Finally, approaches proposed to mitigate out-time effects of prepregs (latent curing agents, cure cycle modification, vitrimer matrices) are presented.

1. Introduction

The ever-increasing demand for high-performance lightweight materials over the last decades has driven significant progress in the development of advanced composite materials, in particular pre-impregnated reinforcement fibers (prepregs). Pioneered in the early 1960s [1], prepregs have gained widespread attention across various industries such as aerospace, automotive, wind energy, marine, and sports. Market analysts estimate the current market size for prepregs to be just over 10 billion dollars with a robust CAGR of ~10 %, which will result in a double market size in the early 2030s [2–4]. The majority of the market is dominated by a few key players, namely Hexcel, Toray, Teijin, Syensqo, Axiom, SGL Carbon, and Mitsubishi Chemical. These companies supply structural materials including a wide variety of application-specific thermoset and thermoplastic prepregs for commercial, recreational, and military markets.

Prepregs impregnated with thermoset polymer matrices are becoming increasingly popular in the composites industry due to their ability to combine ease of processing with high part quality. This advantageous combination of properties results from the convenient delivery form as high-quality composite semi-finished products made from reinforcing fibers already pre-impregnated with a partially cured resin, eliminating the need for a potentially void-inducing resin impregnation step in the manufacture of fiber-reinforced plastics (FRP) [5]. The resulting technological advantages of thermoset prepregs include:

- Easy processability via hand or automated lay-up
- Compatibility with established manufacturing methods including autoclave curing, compression molding, and vacuum bagging
- Wide variety of commercially available reinforcements (carbon, glass, aramid, etc.), textile architectures (woven, unidirectional), and matrices (epoxy, phenolic, bismaleimide (BMI), etc.)
- Straightforward realization of load-compliant laminates through lay-up of unidirectional plies
- Superior mechanical properties (high modulus/strength, impact resistance, etc.) due to:
- Absence of foreign materials, such as knitting yarns, binders, etc.
- High and constant fiber volume fraction (FVF)
- Low void content/porosity

Especially the latter quality parameters of high component FVF combined with low porosity <1 % can be achieved by autoclave cure at elevated pressures of up to 10 bar and high temperatures (mostly between 120 °C and 180 °C) or by using out-of-autoclave (OoA) prepregs in combination with vacuum-bag-only (VBO) cure. For in-depth information on composite manufacturing from OoA prepregs, comprehensive

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https://doi.org/10.1016/j.compositesb.2025.112611
Received 20 March 2025; Received in revised form 28 April 2025; Accepted 8 May 2025
Available online 8 May 2025

literature reviews by Centea et al. [6] and a more current version by Ekuase et al. [7] are recommended. Other review articles have been published on prepreg-related topics such as automated lay-up technology [8–10], quality inspection [11,12], tack [13], forming [14,15], and material composition [16,17].

1.1. Background

The depicted quality superiority of prepregs is offset by two major limitations: Firstly, the high cost of materials disqualifies prepregs for cost sensitive applications [18] while long autoclave curing cycles limit high-volume production [19]. Nevertheless, for many demanding high-performance applications, the technological advantages of prepregs, even in combination with autoclave curing, can compensate for the high material, investment, and operating costs [20]. Secondly, the major technological disadvantage of thermoset prepregs is a limited shelf life due to the hardener already being pre-mixed with the resin. Although the undesired curing reaction can be substantially slowed down by storage at low temperatures, typically −18 °C/0 °F, it cannot be brought to a complete standstill. In addition to freezer storage, the storage and processing time span at room temperature between defrosting and cure is particularly critical as cross-linking proceeds at higher rates at elevated temperatures [21]. The thawing of prepreg rolls is usually done in sealed plastic bags to avoid condensation [22] and may take multiple hours to days depending on its delivery form. Subsequent processing of prepregs, e.g. via automated lay-up technologies such as automated fiber placement (AFP) and automated tape laying (ATL) will consume additional days for large components [23]. Both time spans that are post-freezer storage and processing add up to a considerable amount of prepreg ambient exposure, which will inevitably cause changes in the physio-chemical properties of the material (Fig. 1).

For material data sheets or in-house guidelines, prepreg manufacturers and original equipment manufacturers (OEM) hence indicate critical time periods that should not be exceeded in order to maintain warranted processing and post-cure properties. Table 1 summarizes the most common expiry categories for thermoset prepregs and provides an overview of time spans for epoxy-based, phenolic, and BMI systems. These should be understood as 'typical' values for high temperature cure prepregs and may vary within a single resin type depending on the intended field of application and processing route, including snap/fast cure, OoA, latent systems, etc. However, note that no standards are applied to the expiration declaration, so the use of categories is not consistent: Some manufacturers, e.g., assign out-time restrictions for a shelf life at 4 °C/40 °F (typically 3 months) while others do so for −18 °C/0 °F (typically 12 months).

After expiration, prepregs are usually sent to land-fill or incineration [25] as an effective but ecologically questionable quality control measure. Wu et al. [26], Rybicka et al. [27], and Chadwick et al. [28] are mostly unanimous in assessing the total relative amount of disposed thermoset prepreg scrap (including trimmings) to be 40 %, 30–50 % and 30 % of the quantity initially purchased. Particularly for the aerospace industry, which is a major producer of out-of-spec prepregs, recertification of the material is too expensive [29] despite the tremendous material costs of aerospace-grade prepregs. Smith and Hubert [30] estimate the economic loss to OEMs of disposing of a carbon fiber prepreg roll (width 1.5 m, length 91.4 m) due to expiration to be close to $ 20.000 based on cost assumptions made by Centea and Nutt [31]. The potential savings from recycling are tremendous, especially in the case of recycled carbon fibers [32], which has led to the proposal of a wide range of recycling routes for outdated thermoset prepregs.

1.2. Review scope and outline

A common feature of the recycling routes discussed, however, is that they are always accompanied by a drastic loss of mechanical performance of the component as a result of fiber shortening and/or lack of economic competitiveness [33]. Meanwhile, the material manufacturers' expiry information as presented in Table 1 lacks details on the age-related development of the physio-chemical prepreg state, e.g., in terms of quantitative data on the evolution of processing and post-cure

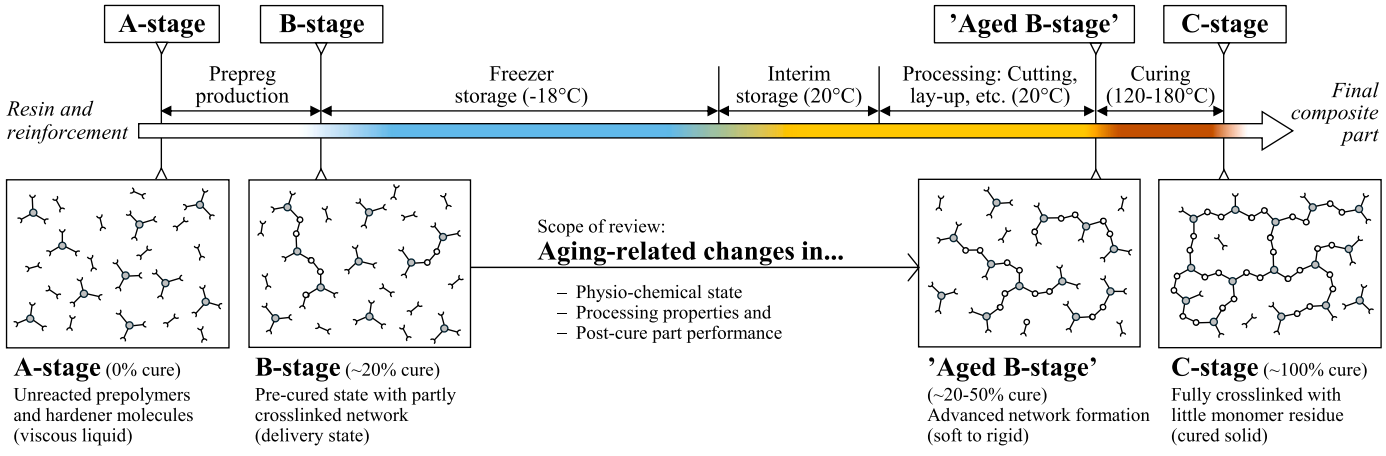

Fig. 1. Material stages of thermoset prepregs along the composite manufacturing value chain. Time spans are not in scale.

Table 1
Storage-related expiry categories and time spans of commercial thermoset prepregs.

Expiration category	Definition (according to Ref. [24])	Refers to	Typical time spans		
			Epoxy	Phenolic	BMI
Tack life	The time, at room temperature during, which prepreg retains enough tack for easy component lay-up	Processing	10 days	28 days	10–14 days
Out life	The maximum accumulated time allowed at room temperature between removal from the freezer and cure	Processing/Post-cure properties	30–42 days	30 days	20–30 days
Shelf life	The maximum storage life for prepreg, when stored continuously, in a closed moisture proof bag at −18 °C/0 °F	Post-cure properties	12 months	6–12 months	6–12 months

laminate properties. As a consequence, the composite industry has been highly motivated to explore the fundamentals of prepreg aging in order to avoid costly and environmentally critical material disposal a priori and, therefore, to use outdated prepregs as long as reasonably practicable. The issue has also received attention within the scientific community, which eventually led to a multitude of studies dealing with prepreg aging throughout the last decades. However, previous reviews on prepreg-related topics have rather been focused towards material composition, specific manufacturing processes or inspection methods, leaving a concise review on aging still lacking to date.

To address this gap, we present a condensed literature review on thermoset prepregs focusing on the effects of material aging prior to the composite part cure process. By assembling, summarizing and comparing published literature, this work for the first time highlights prepreg aging as an interdisciplinary topic between the characterization, engineering and manufacturing aspects behind thermoset prepreg materials. The article addresses a major cross-industry challenge linked to the application of prepregs and, therefore, provides valuable information for applications in aerospace, automotive, electrical, wind, and recreational industries.

In a first step, the evolution of the prepregs' physio-chemical state with respect to degree of cure, glass transition temperature, cure kinetics, and water uptake (Section 2) is reviewed. Material changes occurring in the process are eventually related to physical effects, i.e., material processing properties (Section 3) and post-cure thermo-mechanical performance (Section 4). Aging mitigation strategies in terms of latent hardeners, cure cycle modification and vitrimer matrices are reviewed together with recycling strategies in case prepregs are out-of-spec (Section 5).

This review is limited to the material class of prepregs for advanced composite manufacturing and does not include the large number of studies exploring the hydro-/hygrothermal aging of neat and/or cured thermoset resins. Furthermore, the reexamination of scientific progress on the aging effects of composite parts that evolve after component production (in-service phase) is not the objective of the present review and can be found elsewhere [34–36]. If not stated otherwise, the compiled results refer to continuous fiber-reinforced prepregs for high-performance applications and their regular processing routes. Consequently, unsaturated polyester or vinyl ester resin-based, filled composite materials such as sheet molding compounds (SMC) and bulk molding compounds (BMC) are not subject to discussion in this article, despite being considered thermoset fiber-matrix semi-finished products. Reviews on SMC and BMC were presented in Refs. [37–39].

2. Physio-chemical prepreg state

The physio-chemical prepreg state is mainly a matter of the matrix resin considering the chemical and microbial inertness of conventional reinforcement materials such as carbon, glass or aramid. The situation may be different when applying natural fibers, which have recently gained increased interest for prepreg applications [40] but have been demonstrated to oftentimes suffer from microbial attack if the fibers are not treated chemically [41]. Fully bio-based prepregs have not been widely established yet, partly due to inadequacies in handling and processing. For example, Cheng et al. [42] prepared bio-prepregs from polylactic acid (PLA) and flax fibers which were shown to prematurely cure by more than 50 % as a result of 30 days of ambient exposure and, therefore, far beyond a maximum tolerable level for processing.

Focusing on the characterization of conventional petrochemical materials, a comprehensive overview of non-destructive testing (NDT) and evaluation methods for the quality assurance of uncured polymer matrix prepregs was provided by Pollock et al. [12]. The authors list various NDT methods from literature to study the aging-related physio-chemical state of thermoset prepregs including dielectric analysis (DEA), (fourier transform) infrared spectroscopy (FT)IR, luminescence spectroscopy (L-S), and nuclear magnetic resonance (NMR). By also including destructive and most recently employed techniques, the list can be completed by adding differential scanning calorimetry (DSC), (attenuated total reflection) FTIR, different methods of liquid chromatography (LC) including gel permeation (GPC), reverse phase (RPCL), and high-performance liquid chromatography (HPLC), photoacoustic (PA) spectroscopy, dynamic mechanical analysis (DMA) as well as broadband dielectric spectroscopy (BbDS). However, DSC was by far the technique that was most often utilized followed by FTIR. This trend can also be seen in Table 2 which summarizes the studies that experimentally investigate the conversion/degree of cure α (Section 2.1) and the glass transition temperature T_g (Section 2.2) of epoxy-based prepregs as a function of aging time. The data was obtained for aging in a room temperature range between 20 °C and 30 °C while the humidity was controlled in most studies, either in a medium-humidity range imitating workshop conditions or to replicate a dry environment. Wet aging and its influence on water uptake will be discussed later (Section 2.3).

2.1. Conversion α/degree of cure (DoC)

Fig. 2 shows an aggregation of data points from 14 different studies that experimentally determined the prepreg resin conversion as a function of ambient out-time. The investigated 60-day time span exceeds the manufacturer's recommendations for both tack life and out life, typically ≤30 days (Table 1). A moving average is calculated (k = 5 days) and fitted phenomenologically (red dotted line) to deduce a general trend from the empirical data. Some of the data points were directly read from graphs and thus may contain minor deviations from the original data. The corresponding aging conditions (temperature and relative humidity RH) are summarized in Table 2. Note that the degree of cure is not an absolute value, but rather a relative value in relation to the initial B-stage. In other words, 0 % conversion as-shown corresponds to 'fresh' prepreg as delivered, but in fact the material has undergone a substantial pre-cure.

As expected, all reviewed studies included in the graph revealed a monotonic increase in the degree of cure α as a function of out-time. Provided that the aging period has been extended to 60 days, the authors report an α between 20 % and 40 % after this time span of room temperature exposure. Relative scattering is most prominent for short aging times, which is mainly attributed to the findings of two studies [48,54] that found higher resin reactivity throughout the first days of storage. In general, scattering for this type of cross-study data comparison can be caused by multiple factors. Despite the investigated prepreg resins being all epoxy-based, formulations may still vary significantly, which would affect properties like viscosity, gelation time, reactivity,

Table 2
Studies investigating conversion/degree of cure α (Fig. 2) and glass transition temperature T_g of epoxy-based prepregs as a function aging time at room temperature.

Ref.	First author	Aging temperature	Relative Humidity	Measurement technique
[30]	Smith	20 °C	–	FTIR
[43]	Zhang	20 °C	–	DSC
[44]	Dobhal	30 °C	50 %	FTIR
[45]	Rabby	25 °C	–	DSC
[46]	Blass	25 °C	50 %	DSC
[47]	Heller	21 °C	40 %	DSC
[48]	Budelmann	21 °C	<0.1 %	DSC
[49]	Kim	21 °C	51 %	DSC
[50]	Grunenfelder	20 °C	50 %	DSC
[51]	Grunenfelder	20 °C	50 %	DSC
[52]	Frigione	18–26 °C	–	DSC
[53]	Yu	30 °C	65 %	NIR
[54]	Ahn	25 °C	–	DSC
[55]	Gu	20 °C	<10 %	DSC
[56]	Raponi	24 °C	50 %	DSC
[57]	Smith	18 °C	45 %	DSC
[58]	De Aguiar	25 °C	–	DSC

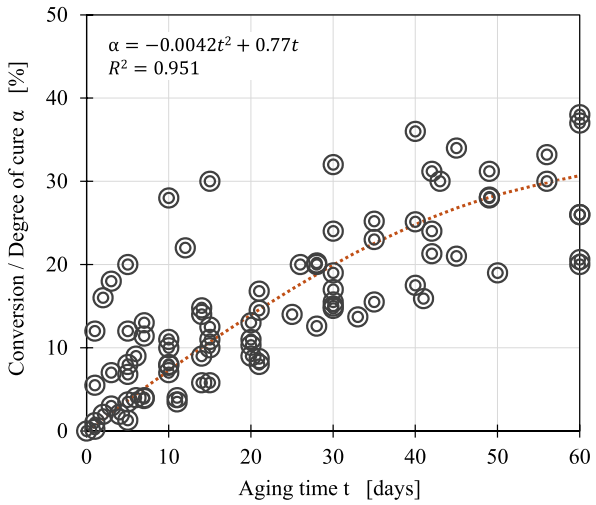

Fig. 2. Conversion of epoxy-based prepregs as a function of room temperature aging. Experimental data points collected from Refs. [30,43–55]. Further information on test and conditioning parameters is provided in Table 2.

etc. and consequently, cure progression differently. Furthermore, differences in terms of aging conditions such as temperature and RH as well as in terms of applied testing methods (Table 2) limit comparability and may lead to divergent results.

In terms of a data-derived general trend (red dotted line), the conversion progresses at a virtually constant rate of 0.7 % day-1 within the first 30–40 days, which represents the typical out-time period for most epoxy-based prepregs. After the transition region, a slowdown in conversion was observed in multiple studies, which is also reflected in the curve fit. Most authors attributed the decrease in rate to an increase in T_g and vitrification, which will be discussed in detail in the following (Section 2.2). The dependence of α on out-time is expected to reach a threshold value for infinite storage times. However, there are no experimental results on the issue, possibly due to a lack of practical relevance.

In order to gain insight into the advancing network formation during aging, changes in the apparent molecular size distribution of aged prepreg resin as shown in Fig. 3 (A) were monitored by GPC [30]. Aging progressively shifted the distribution left-hand-side tail towards higher molecular weights, while the weight fraction at the distribution peak was intensified. This finding was representative of a continuous aging-induced network formation featuring an increased number of medium-sized macromolecules in the aged B-stage. Having identified the number average molecular weight M_n and dispersity Đ as highly out-time dependent as shown in Fig. 3(B and C), the authors suggested these values to be used as potential aging metrics for prepreg inspection employing GPC.

Cole et al. [59] conducted a comprehensive study (data not included in Fig. 2) on the aging-related curing progression of carbon fiber epoxy prepregs at 22 °C and 50 % RH storage. The authors employed FTIR, FTIR-ATR, RPLC, HPLC, and DSC in an effort to investigate differences in the outcome. The authors report differences in cure rate between 0.2 % day-1 and 1.09 % day-1 depending on the measurement technique employed and evaluation parameters (peak areas, ratios, indices, etc.). The average conversion rate across all techniques amounted to 0.58 % day^{-1} which results in 34.8 % cure after 60 days and is in good agreement with the cross-study average shown in Fig. 2. Individual studies have found independence of the physio-chemical prepreg state on room temperature aging, e.g., Guo [60] for BMI-based prepregs throughout an out-time of 90 days. Epoxy prepregs made from Bisphenol A (BPA) resin and dicyandiamide (DICY) were prepared by Tao et al. [61] and showed no signs of premature cure. DICY is an extensively studied hardener for epoxy resins with latent cure behavior [62] that delays the curing reaction and will be discussed as a viable aging mitigation strategy (Section 5.1).

2.2. Glass transition temperature T_g

The glass transition temperature T_g is one of the most crucial material parameters when it comes to characterizing the mechanical and thermal responses of amorphous materials, including thermoset resins. It marks the reversible transition from a glassy into a rubbery or viscous state and is linked to changes in the viscoelastic, dilatometric, enthalpic, i.a. properties [63]. T_g is directly related to the degree of cure α (Section 2.1) as the advancement of T_g with α can be described by, e.g., the widely-used thermodynamically based Di-Benedetto equation [64] or by purely empirical relationships. The glass transition region has been the target of investigation for multiple studies on the effect of aging and storage on the physio-chemical state of prepregs as listed in Table 2. When considering the glass transition temperature, a distinction between the influence of aging on the pre-cure and post-cure T_g has to be drawn. Fig. 4 features an experimental data point accumulation showcasing the evolution of pre-cure T_g, which was measured for epoxy-based prepregs directly after specific time periods of ambient aging.

T_g of fresh uncured prepregs in the delivery state was consistently reported to range between −5 °C and 5 °C. For longer aging times, T_g continuously increases in a manner similar to the progression observed for α. Scattering, however, is more pronounced for T_g measurements which may be attributed to the glass transition region extending to several degrees and the dependence of T_g determination on test parameters (especially heating rate) and enthalpy analysis (half height, half width, inflection techniques) in DSC [65]. Authors concordantly reported a slow-down of the aging-related T_g and/or α increase (Section 2.1) for epoxy [30,48,52–54] and BMI [60,66] prepregs. The slow-down in cure progression was argued to originate from resin vitrification when T_g exceeds the cure temperature, which in case of ambient aging

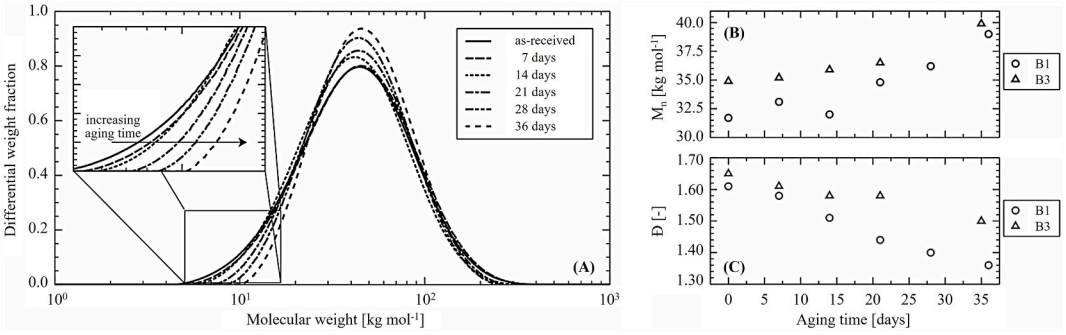

Fig. 3. Molecular weight distributions (A), number average molecular weight M_n (B) and dispersity Đ (C) of aged prepreg resin measured by GPC. Figure rearranged and axes relabeled for improved readability. Reproduced with permission, [30]. Elsevier, 2023.

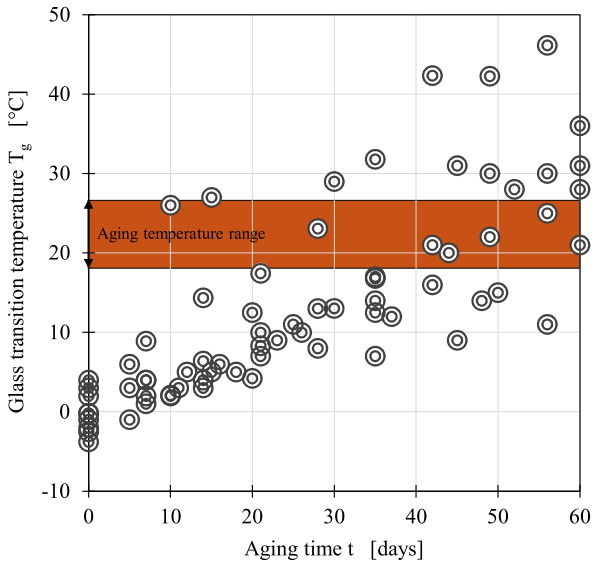

Fig. 4. Evolution of glass transition temperature T_g as a function of out-time for epoxy prepregs aged in an ambient environment (red temperature range). Experimental data points collected from Refs. [30,47–52,56–58]. Further information on test and conditioning parameters is provided in Table 2.

transpires at room temperature (Fig. 4). The transition is characterized by a change from a rubbery/viscous to a glassy state. At this point, reactivity is reduced significantly as the crosslinking reaction switches from kinetic-driven to diffusion-dominated [67] if the cure temperature is not increased further. The phase change limits the molecular mobility of the matrix, which manifests itself as a high perceived rigidity of the prepreg as soon as the prepreg T_g surpasses room temperature as a result of out-time.

The work by Salzmann et al. [68] was not included in Fig. 4 as aging trials were not conducted at room temperature but at elevated temperatures of 60 °C using a glass fiber epoxy novolak system. Within 6 h, T_g reached 41 °C (DSC) and 39 °C, respectively. For the latter measured value, a miniaturized near infrared (NIR) spectrometer was utilized. The hardware can directly be integrated in the production process, e.g., within a fiber placement head. Considering the time-consuming methods to determine the prepreg quality in a laboratory, the authors emphasize the urgent need for a fast and simple method for quality assurance directly on the production side. Other studies have also demonstrated the potential of NIRS for online cure monitoring for prepregs [69–72], however, mainly in relation to the prepreg production process. A study similar to Salzmann's work was presented by Köller et al. [73] using NMR.

In an effort to explore the influence of room temperature aging on post-cure T_g, Rabby et al. [45] found a slight T_g decrease of cured prepregs from 126.3 °C (fresh) to 122.07 °C (aged 40 days). Despite not being measured directly, the crosslinking density of composite parts made from aged prepreg was expected to be lower than for fresh material due to an aging-related increase in activation energy (84.05 vs. 106.13 kJ mol^{-1}). Miller et al. [74] used Cytec IM7/977-3 epoxy prepregs to study the impact of 60 days ambient exposure via DMA. T_g of autoclave-cured fresh material was 235 °C and 229 °C following DMA ramp rates of 0.28 and 1.11 K min^{-1} while pre-cure aging decreased T_g to 211 °C and 222 °C, respectively. No change in T_g was reported by Raponi et al. [75]. The only study known to the authors investigating the aging effect on both pre-cure and post-cure T_g was conducted by Giorgini [76]. While T_g of the B-stage prepreg increased, the corresponding T_g of the final product decreased upon DSC curing with a roughly opposite trend.

2.3. Water uptake from moisture exposition

Thermoset resins are known to be susceptible to water uptake in wet environments [77]. The vast majority of research, which has been summarized in Refs. [78–82] was focused towards hydrothermal and hygrothermal aging of cured resins and polymer composites in the C-stage and, therefore, falls out of this review's scope. During room temperature aging in B-stage, the prepreg resin is exposed to water vapor present in the air at workshop conditions and will undergo hygrothermal aging.

The water uptake of neat uncured epoxy prepreg resin was studied by Dei Sommi et al. [83] who found an upward curvature typical of a Flory-Huggins behavior [84] reaching a water saturation just over 1 %. For comparison: Cured epoxy resins are typically able to incorporate up to 7 % of water [85], which may even be increased to ~12 % with excessive hydrothermal aging if the systems show low crosslinking densities and high polarity [86]. In related research, the authors eventually used the experimental data for the model-based prediction of the thermodynamic conditions for water-generated porosity upon autoclave cure [87]. Water being absorbed in the uncured prepreg state was reported to initiate the nucleation and growth of voids, which is particularly critical if prepreg laminates are cured under high temperature and low pressure.

Another comparable study was conducted by Grunenfelder et al. [88]. The results from both studies are in good agreement in terms of experimental data as highlighted in Fig. 5. The corresponding fitting procedure carried out in Ref. [88] is based on the resin solubility parameter S ($1.24 \bullet 10^{-4}$). Multiplying by the initial relative humidity $(RH)^2$ yields the solubility that is the maximum resin moisture content.

However, in industrial practice only a limited part of the prepreg surface is usually exposed to moisture when the prepreg is either still on the roll or stacked in a laminate. As a consequence, water saturation throughout the whole laminate will most probably not been reached in most manufacturing scenarios because the surface-near absorbed water needs to diffuse through the laminate. Netzel et al. [89] stacked 1, 5 and 10 plies of prepreg and exposed the uncured laminates to 40 % and 90 % RH ambient conditions. Water absorption was quantified following the widely used procedure proposed by Chen and Springer [90] in the late 70s. The results in terms of time until full saturation, maximum weight gain and diffusion coefficient are summarized in Table 3. Considering that the percentage increase was measured in relation to the prepreg mass (including fibers), the findings are in reasonable agreement with the results shown in Fig. 5. Furthermore, similar diffusion coefficients of

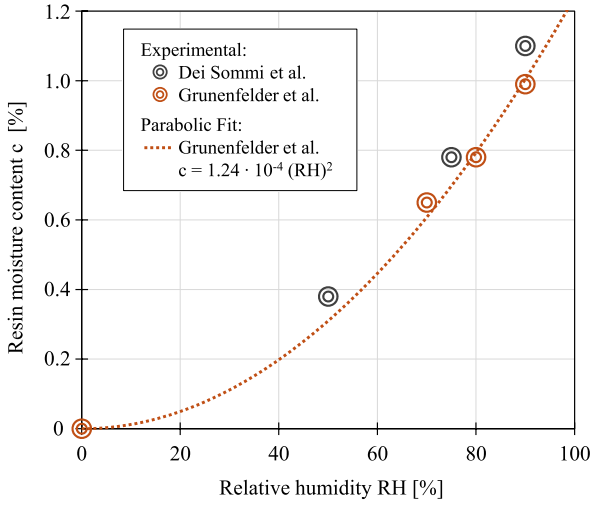

Fig. 5. Moisture content of water-saturated, epoxy prepreg resin (B-stage) as a function of relative humidity (RH) at room temperature exposure. Data adapted from Dei Sommei et al. [83], Grunenfelder and Nutt [88].

water were determined in Ref. [91].

Mohan et al. [92] aged epoxy-based prepregs for up to 308 days in a humidity-regulated environment, which was controlled using saturated salt solutions. The aged prepregs were processed in combination with a compatible epoxy adhesive layer to form co-cured composite joints. The prepregs were found to gain a 0.4 % weight gain as a result of water uptake. With the distinction being made between free and bound water, it was also shown that the prepreg stored moisture from high humidity environments as free water, while the level of bound water remained unaffected. The free water is known to act as a plasticizer within the epoxy resin [93], eventually leading to a decrease in T_g [94].

Minakuchi et al. [95] compared the water absorption (85 % RH 40 °C) and desorption (vacuum at 40 °C) for cyanate and epoxy prepregs. The epoxy matrix was observed to saturate quicker than the cyanate resin, however, both prepregs reached 0.3 % weight gain at the end of the investigated time span of 73 h. Similar trends were revealed for desorption resulting in a 0.1 % weight loss in comparison to the delivery state. An early study by Van Mele and Verdonck [96] focused on the influence of moisture on the fiber/matrix interaction in self-made prepregs from diglycidylether of bisphenol A (DGEBA) and an anhydride hardener. Strong effects of wet storage on the fiber/matrix interaction were described, which intensified if fibers with hydrophilic surfaces, in this case polyvinyl alcohol (PVA) are utilized. Hussain et al. [97] investigated the effect of geographical (Malaysia and China) and workshop conditions (humidity-controlled and as found) on the moisture uptake of prepregs during storage. Based on the findings, the authors provided practical recommendations for safe prepreg handling in different workshop environments.

Multiple studies with an ecological focus ([98–100] and others) studied the pre-cure water uptake of natural fiber-based prepregs. Water uptake was concordantly found to exceed the resins capability of moisture absorbance indicating that the reinforcement will incorporate most water due to the generally proven hydrophilicity of natural fibers [101]. Therefore, an increased awareness and, if necessary, taking countermeasures such as drying are due when exposing natural fiber-reinforced prepregs to ambient conditions.

2.4. Viscosity and viscoelastic behavior

Although no highly viscosity-dependent resin infusion process is necessary when using prepregs for composite manufacturing, the material viscoelastic properties still play a pivotal role for the success of applying prepreg technology. In general, processing properties such as handling, drape [102] (Section 3.1) and tack [103] (Section 3.2) as well as preventing void formation during cure cycles [104] (Section 4.2) heavily rely on knowledge and control of matrix viscosity. Exploration of the aging-related effects on viscosity and viscoelastic parameters was thus included in a number of publications as complementary analysis.

Kim et al. [49] compared ex-situ and in-situ measurement by applying rheology and dielectric measurement, respectively. Hereby generated data with regard to the out-time dependent minimum viscosity η_{min} and gel time t_{gel} at different dwell temperatures (blue: 93 °C, red: 121 °C) are plotted in Fig. 6. Both rheological material parameters η_{min} (A, B) and t_{gel} (C, D) were highly affected by aging. Considering the influence of the applied temperature ramp rates, the authors

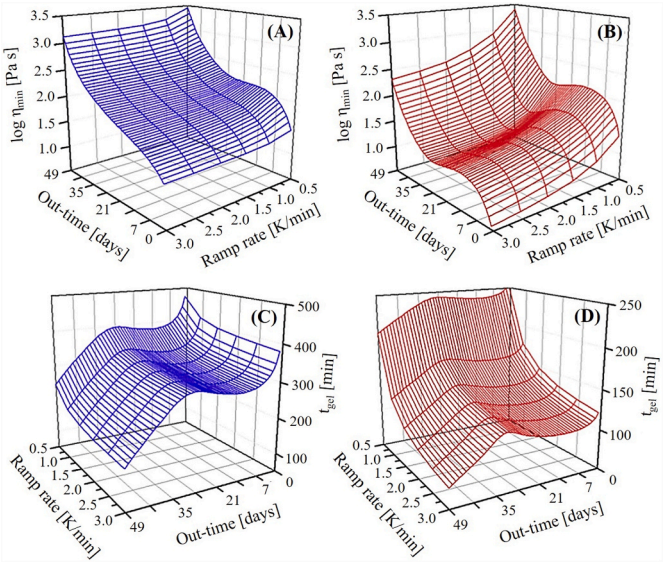

Fig. 6. Minimum viscosity η_{min} (A, B) and gelation time t_{gel} (C, D) of prepreg resin as a function of temperature ramp rate and out-time for 93 °C (blue) and 121 °C (red) isothermal dwell. Axes relabeled for improved readability. Reproduced with permission, [49]. Elsevier, 2014.

recommended prepreg subjected to long out-times to be rapidly heated to high temperatures in order to counteract long out-times. Similar out-time trends for the viscosity and gel time progression at 171 °C isothermal cure were reported in Ref. [53].

Szpoganicz [105] prepared prepregs from a silica-filled phenolic resin in an effort to investigate the dependence of manufacturing properties on environmental factors including aging. The filled resin viscosity was shown to increase by two magnitudes for all testing temperatures 25 °C (10^5–10^7 mPa s), 50 °C (10^4–10^6 mPa s) and 75 °C (10^3–10^5 mPa s) induced by 150 days of aging. A similarly large increase in viscosity was reported within 56 days out-time for epoxy-based OoA prepregs elsewhere [51]. Here, the difference in modeled viscosity remained virtually constant throughout the whole curing cycle. Apicella et al. [106] also modeled the rheological behavior and found a shift of viscosity towards longer cure times when applying a dynamic temperature profile. The results match the reported aging-related shifts for cure progression as reviewed later in this review (Section 3.3)

By employing DMA temperature sweeps, Dobhal et al. [44] investigated the variation of viscoelastic parameters storage modulus, loss modulus and tan δ for cured composites that were produced from aged prepregs. Over an aging span of 80 days, the storage modulus decreased by 20 % while the loss modulus increased by up to 30 %. The dependencies were assigned to changes in the cure-related molecular flexibility. The corresponding gain in tan δ was claimed to be an indicator of improved damping behavior.

3. Processing properties

In a workshop environment, out-of-spec prepregs are perceived to

Table 3
Prepreg moisture absorption at 21 °C after drying [89].

Relative Humidity	Layers	Time until saturation	Maximum weight gain	Diffusion coefficient
40 %	1	25 h	0.18 % ± 0.003 %	1.28E-12 m^2s^{-1}
	5	625 h		1.88E-12 m^2s^{-1}
	10	1225 h		3.87E-12 m^2s^{-1}
90 %	1	25 h	0.33 % ± 0.007 %	1.20E-12 m^2s^{-1}
	5	625 h		1.72E-12 m^2s^{-1}
	10	1225 h		3.43E-12 m^2s^{-1}

have turned rigid and non-tacky making defect-free processing more challenging. This subjective perception was scientifically substantiated in literature, which dealt with the exploration of aging-induced changes in processing properties. In the following, the most impactful research on forming properties (Section 3.1), tackiness (Section 3.2) and the adaptation of cure cycles (Section 3.3) are reviewed.

3.1. Forming properties

Whenever the geometric complexity of components exceeds flat panels, the manufacturing of FRP from prepregs involves a (pre)forming stage, in which uncured stacked flat prepreg plies are transferred into desired three-dimensional geometries. For established forming, molding and lay-up processes, the major involved deformation mechanisms are intraply/in-plane shear, interply shear/friction and out-of-plane bending [107]. The parameters have extensively been studied for fresh prepreg material. Superordinate research topics include but are not limited to experimental determination [108–110], testing methods [111,112] and the use for draping [113,114] and process models [115–117].

A contribution to the far less comprehensive state of research for aged prepregs was recently made by Zhang et al. [43] who investigated resin aging, forming temperature and prepreg ply orientation on the interply friction. Interply friction of aged material was reduced by 26.5 % if tested at room temperature. For room temperature forming (Fig. 7 upper row) an aging-related pre-cure state was observed to mitigate wrinkling defects and to improve the component quality of L-shaped panels. Slippage for this parameter set resembled more of a dry friction behavior, which became more viscous if the forming temperature was raised or if fresh prepregs were processed. The friction behavior was shown to be closely related to the resin viscosity, which in turn is a function of out-time (Section 2.4). In terms of tool-ply friction, multiple studies on fresh prepregs [118–120] have shown that increasing forming temperatures favor lower tool-ply friction due to lower viscosity. The increased resin viscosity of aged prepregs will, therefore, most likely hinder the forming process as a result of high tool-ply friction. In conclusion, the authors emphasized the need to consider the complex interaction between forming temperature, ply angles and out-time in order to establish robust forming processing routes in future.

Banks et al. [121] investigated the bending behavior of self-made pre-cured prepregs by determining the overhang length of one-sided clamped prepreg strips according to ASTM D1388. As expected, the apparent flexural rigidity increased as a function of level of cure. A steeper increase was observed for cure levels exceeding the typical B-staging level of ~30 %, which represents aging. An aging-induced rise in the degree of cure from A-stage to 60 % lead to quadrupling in flexural rigidity. At the same time, the intraply shear stiffness measured via bias extension tests increased sharply for the highly aged prepregs. Note that all experiments were carried out at room temperature with no variations in temperature made.

Heller and different coworkers conducted a series of studies investigating the changes in processing properties in the context of AFP. In a first study featuring applied research with AFP trials [122] it was shown that the overall lay-up behavior worsened once the material clearly exceeds its tack life of 14 days. Within this timespan, the occurring defect type during steering changed as in-plane fiber waviness converted into out of plane tape buckling. Based on a comparison of testing methods for intraply shear properties of uncured prepreg tapes [123], the out-time effects on material processing properties were eventually studied in detail in Ref. [47]. A correlation coefficient of ~0.4 between out-time and the two processing-related mechanical model input parameters, namely transverse tensile modulus and in-plane shear modulus, was found. Two weeks of room temperature aging greatly increased both parameters. The dependence, however, was virtually lost if tests were carried out at an elevated temperature of 40 °C. The described interdependences between material properties, test parameters and out-time would serve as guiding input parameters for defect prediction models to make the automated processing of prepregs more robust to aging effects.

3.2. Tack

Tack is a processing property that exclusively applies to thermoset rather than thermoplastic prepregs and refers to the ability of the material to adhere to either itself or other surfaces upon the application of light pressure. Tack can be defined and quantified as a measure of mechanical resistance that must be overcome in order to separate the created adhesive interface [13]. For composite manufacturing, tack is a crucial aspect in the built phase of a laminate either by hand [124] or via automated lay-up technology [9]. If adjusted properly, it enables precise lay-up and makes sure the material stays in place throughout the manufacturing process. Prepreg tack has been in the focus of a multitude of studies investigating the effects of different influencing factors including aging. By applying experimental methods such as probe tack testing or peel testing, multiple studies confirmed the subjectively perceived loss of tackiness discussed in the beginning of the section.

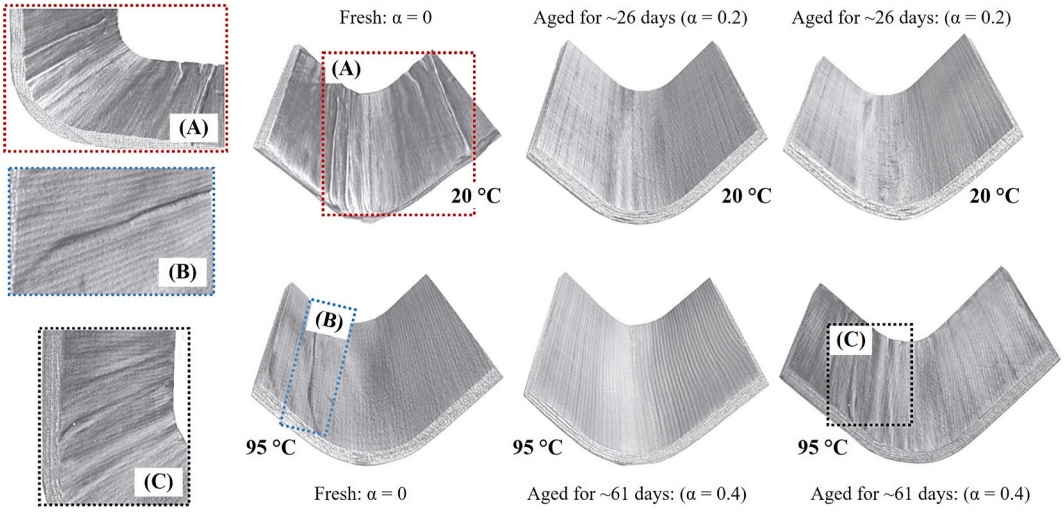

Fig. 7. Micro-CT scans showcasing the defects of preforms for compression molding made from fresh, 26 and 61 days aged thermoset prepregs at different preforming temperatures (bold temperatures 20 and 95 °C). Axes relabeled for improved readability. Reproduced with permission, [43]. Wiley, 2024.

Cole et al. [125] conducted qualitative tack testing on aged prepregs, which failed the test after 25 days of ambient out-time. Dobhal et al. [44] employed 180° peel test to quantify tack at room temperature. The measured release force gradually decreased from 16 N to 2 N in the course of 80 days. A similar but less marked decrease of was reported in Ref. [126]. Considering the results from cure monitoring presented in Section 2.1 and Section 2.2, the small extent in tack loss in this study is to be expected as the investigated out-time of two days was rather small. Shi et al. [127] characterized an Arrhenius-based decay law of prepreg tack for different aging temperatures and reported faster decay rates for higher aging temperatures as a result of accelerated cure. The authors also proposed a new statistical unit for prepreg tack (handling life unit), which was meant to establish a relationship between the tack of prepreg and its remaining storage time. Nguyen and Krombholz [128] reported an onset of decline in tack after 11 days of out-time while Raponi et al. [56] found an exponential decay of tackiness reaching virtually zero after 15 days of ambient exposure.

The presented studies have in common that the employed tack measurement techniques including prepreg application and debonding was carried out at room temperature. If tack is measured at different test temperatures, results may vary as shown in the following and more elaborate studies on tack testing. Heller et al. employed probe tack testing and peel testing on [47] and were able to demonstrate that the data plots of tack vs. aging time are highly dependent on temperature, measurement technique and other test parameters. Closely related to the research of Section 2.2, Ahn et al. [54] observed that maximum tack could be found at a relatively constant test temperature T_t above T_g (ΔT = 20–25 °C) even if T_g increases as a function of aging. Still, the prepreg tackiness in terms of absolute value (energy of separation) was almost halved from 16 kPa to 7 kPa as a result of 46 months freezer storage. A similarly constant differential between T_t and T_g (ΔT = 40 °C) was reported for room temperature aging [48]. In this study however, prepreg material, which had exceeded tack life according to the data sheet, was found to be even tackier than fresh material (+65 %) when being processed at elevated temperatures.

In an effort to account for the out-time effects on prepreg tack in AFP, Smith et al. [57] constructed process maps using tack master curves from the testing procedure similar to the recently established standard for tack testing ASTM D8336 [129,130]. The work was based on preliminary work of Endruweit et al. [131] and was designed to inform process parameters for automated lay-up while achieving desirable tack levels. According to the authors, this type of tailored process control is anticipated to improve resource utilization when manufacturing large preforms that take multiple days to complete. The process map of Fig. 8 shows the normalized estimated tackiness that can be obtained at different prepreg out-times by variation of the lay-up temperature. In order to account for out-time effects and maintain maximum tackiness (100 % line), the deposition temperature has to be raised linearly from 50 °C for fresh prepregs up to 75 °C for 35 days old prepregs. If tack is not controlled properly through process parameter adjustment, processors run the risk of defect formation during laminating [132]. For this instance, Wang et al. [133] presented a comprehensive modeling framework for the automated fiber placement process that is to include aging responses additionally to the already implemented influences of temperature, pressure and contact time.

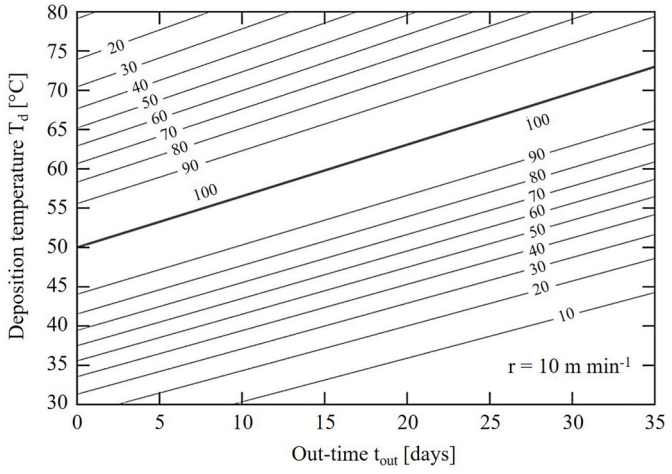

Fig. 8. Tack process map showing normalized tack isolines as a function of material out-time t_{out} and deposition temperature T_d for a prepreg lay-up rate r of 10 m min^{-1} via automated fiber placement (AFP). Axes relabeled for improved readability. Reproduced with permission, [57]. Elsevier, 2020.

3.3. Curing cycles

As outlined before (Section 2.1), out-time at ambient conditions and long-term freezer storage entail progressive curing of thermoset prepreg matrices. Under these circumstances it stands to reason that cure cycles, which are employed to transfer B-stage prepregs into the C-stage upon final part production, are affected in a similar manner. Multiple studies addressed this challenge through experimental and kinetic modeling approaches over the last decades as reviewed in the following.

The thermokinetic effects of aging were studied by Frigione and Kenny [52] by comparing fresh to 21 and 33 days out-time epoxy prepregs and, three years later, for BMI-based prepregs [66]. For epoxy, it was derived from experimental and kinetic model data that the aging of prepregs for prolonged times did not only reduce the overall resin reactivity (cf. literature review of Section 2.1) but also caused a delay in the crosslinking reaction: When applying temperature ramps, both the reactivity maximum and the degree of conversion were shifted towards higher temperatures. The total heat as well as the maximum degree of reaction meanwhile reached similar values, irrespective to the aging time. Still, higher isothermal cure temperatures were needed for complete curing of 33 days aged material. Similar general trends were described by Kim et al. [134] which can be as shown in Fig. 9. Here, a predictive phenomenological cure kinetics model (red line) was modified by using two weight factors and an out-time dependent initial degree of cure α_0 to capture aging effects. The determined α_0 vs. out-time curve runs through the experimental data of Fig. 2 but values will overestimate α_0 for long out-times due to nature of the applied second-order polynomial fit. The ex-situ measured DSC data was eventually compared to in-situ data from dielectric analysis in a related cure monitoring study [49] by the research group. Later on, the curing model was extended to take account of moisture absorption in process conditions [135]. While investigating cure kinetics by using a modified nth-order kinetic model, Guo [60] found similar limitations of attainable conversions α_{max} when applying isothermal cure cycles to BMI prepregs (Table 4). The correlations between α_{max} and cure temperature were observed to be linear while ambient exposure shifted α_{max} to higher temperatures. Raising the cure temperature well above 200 °C was shown to omit the aging effect by providing high α.

Also in connection with cure modeling, the activation energy E_a of tetraglycidyl diaminodiphenyl methane (TGDDM)/DDS-based prepregs was found to increase from 67 to 107 kJ mol^{-1} within 22 days of ambient exposure, which was integrated into model for the prediction of the rheological behavior during processing by Apicella et al. [106]. An additionally run numerical simulation revealed how the out-time history of prepregs affects the process behavior and confirmed the necessity for a adapted temperature ramp in order to obtain a homogeneously cured laminate. A rise in E_a for aged prepregs was also reported in Ref. [45] amounting to an increase from 84.05 to 106.13 kJ mol^{-1} (Kissinger method) and 86.65–107.64 kJ mol^{-1} (Ozawa method), respectively.

In an effort to mark the reaction beginning for aged prepregs, Raponi et al. [56] successfully fitted experimental data of DSC onset temperature T_{onset} vs. out-time by a linear regression. T_{onset} gradually increased as a function of out-time while the enthalpy peak temperature remained

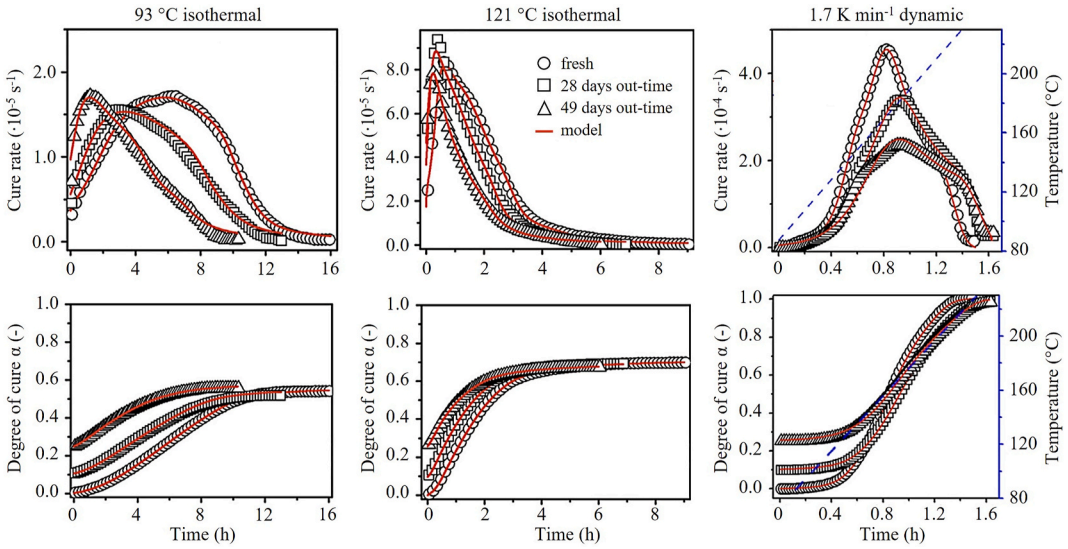

Fig. 9. Isothermal and dynamic cure kinetics measurement and model prediction of the cure rate and degree of cure for fresh and aged prepregs. Figure rearranged and axes relabeled for improved readability. Reproduced with permission, [134]. Elsevier, 2014.

Table 4
Maximum degree of cure α_{max} for isothermal cure of fresh and aged BMI prepregs [60].

Isothermal cure temperature	Maximum degree of cure α_{max}			
	Fresh	10 days	20 days	39 days
170	0.320	–	–	–
180	0.610	0.581	0.524	0.492
190	0.690	0.661	0.661	0.562
200	0.830	0.820	0.781	0.724
210	0.931	0.918	0.901	0.884
220	0.996	–	–	–

constant. In a related study [75] the experiments were extended by employing DMA. Blass et al. [46] split the DSC curing peaks into two Bi-Gaussian fit curves with peak locations at 250 °Cand 285 °C, which were assigned to different group reactions. The fact that aging affected both peaks in different manner led to the assumption that one reaction type (pre-crosslinking at low temperature) is more affected by aging than the other one. To summarize, it can be stated that the gained in-depth insight into thermokinetic behavior of aged thermoset prepregs can be transferred into informed cure cycle adaptation to meet aging effects in advanced composite manufacturing.

4. Post-cure quality

Load-bearing characteristics are usually the main basis of valuation for the viability of polymer composite usage in lightweight applications [136]. In literature, the changes in the physio-chemical prepreg state discussed in earlier sections of this review have been hypothesized to influence post-cure properties in a similar manner to pre-cure characteristics. This presumption applies to aged material that is to meant to be processed into the same laminate stacks by identical processing routes or use of machinery, respectively. This way, comparability is ensured and the influence of prepreg aging on post-cure properties can be isolated. Following this approach, a reasonable number of studies has been published to investigate differences in mechanical performance between aged and fresh prepregs-based FRP. The component porosity as one of the main drivers for deteriorated mechanical performance is often explored concomitantly and will be reviewed in Section 4.2.

4.1. Mechanical properties

Table 5 compiles the relevant literature on the evolution of different mechanical properties as a result of room temperature aging. Information on applied aging cycles regarding temperature, RH and time span is displayed, along with the utilized prepreg systems and curing method. It is indicated whether a significant influence of aging variation was observed and if so, the relative change in properties after are given for the maximum of out-time. In general, a certain inconsistency in results can be observed within the state of research. The majority of investigated properties (64 %) was found to be affected by room temperature aging with differences in extent caused by the prepreg system, curing method, aging time, temperature and others. The studies demonstrating a decline in performance predominate and average a decrease of −23.5 %, however, showcasing a wide range between −6.3 % and −73.6 %. Three dependencies can be derived from the data collated in Table 5.

Firstly, a comparative view on the experimental data suggests that OoA prepregs, which have been cured in a vacuum bag tend to be more sensitive to ambient exposure than systems, which have been developed for autoclave cure. However, the significance of cross-study comparison is limited due to divergences regarding aging conditions and assessed mechanical properties. The only study known to the authors, which directly investigated the differences between OoA and autoclaved prepregs in terms of out-time effects was presented by Sutter et al. [137]. The proposed general trend was confirmed: The autoclave systems preserved their mechanical performance after 45 days in out-life conditions. While the OoA composites had comparable mechanical and thermal performance after processing fresh prepreg, the OOA prepregs thermal and mechanical performance (interlaminar shear strength (ILSS), compressive strength, open hole compression) was reduced after curing the aged prepreg. The quantitative data was substantiated by void exploration and SEM-based investigation of fracture surfaces. The findings are in general agreement with the repeatedly studied and well-understood effect of increased autoclave consolidation pressures on mechanical performance, e.g. studied in Refs. [138–142]. Apparently, the low VBO cure pressure is not able to sufficiently make up for aging-related increases in viscosity (Section 2.4), which hinders void-filling mechanisms and eventually leads to reduced mechanical composite performance.

Secondly, matrix-dominated properties of composites, i.e., transverse tensile strength and modulus as well as compressive properties appear to be more affected than fiber-dominated properties if prepregs

Table 5
Overview of studies exploring the influence of room temperature aging of thermoset prepregs on post-cure mechanical properties.

Ref.	First author	Used prepregs	Aging conditions			Cure method	Mechanical property	Significant influence?	Rel. change in property
			Temperature	RH	Time span				
[143]	Akay	EP CF UD	18 °C	55 %	39 days	Autoclave	Compressive strength	No	
							Flexural strength	No	
							Flexural modulus	No	
							Impact strength	No	
							ILSS	No	
[46]	Blass	EP CF UD	23 °C	50 %	120 days	Autoclave	Tensile strength 0°	No	
							Tensile strength 90°	Yes	−40 %
							Flexural strength	No	
							ILSS	Yes	−9 %
							G_{IC}	No	
[144]	Chandrakala	EP GF WV	n/a	n/a	45 days	Vacuum bag	Tensile strength	Yes	−33 %
							ILSS	Yes	−7.6 %
[125]	Cole	EP CF WV	22 °C	50 %	66 days	Autoclave	Tensile strength	No	
							Tensile modulus	No	
							Elongation	No	
							Compressive strength	Yes	−17.5 %
							Compressive modulus	Yes	−13.3 %
							ILSS	Yes	−25.5 %
[44]	Dobhal	EP GF WV	30 °C	50 %	80 days	n/a	Flexural strength	Yes	−16.2 %
							Flexural modulus	Yes	−35.7 %
							ILSS	Yes	−40.8 %
[145]	Hübner.	EP CF UD	23 °C	50 %	90 days	Autoclave	ILSS	Yes	−18.5 %
							G_{IC}	Yes	+63.1 %
[146]	Ji	EP GF WV	25 °C	50 %	196 days	Comp. molding	Tensile strength	Yes	−21 %
							Tensile modulus	Yes	−14 %
							Flexural strength	Yes	−36.6 %
							Flexural modulus	Yes	−21.4 %
							Elongation	Yes	−10.2 %
[147]	Jones	EP CF WV	n/a	n/a	60 days	Vacuum oven	ILSS	Yes	−11 %
[148]	Nakagawa	EP CF[a]	23 °C	50 %	84 days	Comp. molding	Tensile strength	Yes	+24.8 %
							Tensile modulus	No	
							Compressive strength	Yes	+27.7 %
							Compressive modulus	No	
							Shear strength	Yes	+24.2 %
							Shear modulus	No	
[149]	Nandini	EP CF WV	n/a	n/a	10 days	Comp. molding	Tensile strength 45°	Yes	−22 %
[45]	Rabby	EP GF UD	25 °C	n/a	40 days	Comp. molding	Tensile strength 45°	Yes	−23.9 %
							Flexural strength 45°	Yes	−28 %
							Shear modulus	Yes	−23 %
[56]	Raponi	EP CF UD	24 °C	50 %	60 days	Autoclave	ILSS	No	
[150]	Schmidt	EP CF UD	20 °C	35 %	27 days	Vacuum oven	Flexural strength 0°	No	
							Flexural modulus 0°	No	
							Flexural strength 90°	Yes	−20 %
[151]	Scola	EP CF UD	n/a	n/a	140 days	n/a	Shear strength	No	
[137]	Sutter	EP CF UD	n/a	n/a	45 days	Autoclave	ILSS	Yes	−13.2 %
							Open hole compression	No	
							Compressive strength	No	
[137]	Sutter	EP CF UD	n/a	n/a	35 days	Vacuum bag	ILSS	Yes	−73.6 %
							Open hole compression	Yes	−50.2 %
							Compressive strength	No	−55.1 %
[152]	Wang	EP CF UD	20 °C	30 %	60 days	Autoclave	Tensile strength	Yes	−10.6 %
							Tensile modulus	No	
							Compressive strength	Yes	−38.0 %
							Compressive modulus	Yes	−10.8 %
							Flexural strength	Yes	−8.7 %
							Flexural modulus	No	
							ILSS	Yes	−29.3 %
[43]	Zhang	EP CF UD	20 °C	n/a	100 days	Comp. molding	Tensile strength 0°	Yes	−13.7 %
							Tensile strength 90°	Yes	−19.9 %
							ILSS	Yes	−6.4 %
							Flexural strength 90°	Yes	−6.3 %

Abbreviations: CF: carbon Fiber, GF: glass fiber, EP: epoxy, UD: unidirectional, WV: woven, ILSS: interlaminar shear stress, n/a: not specified.
[a] Prepreg scrap processed into discontinuous CFRP.

have experienced relevant pre-cure out-time at room temperature. A study was presented by Blass et al. [46] that focused on the direct comparability between both matrix- (90° tensile and flexural strength) and fiber-dominated (0° tensile strength) characteristics. Although the above-mentioned relationships were confirmed, the authors trivialized the findings for industrial practice by arguing that that no CFRP part is designed on its matrix strength. Still, some studies revealed a decline of fiber-dominated properties concomitant with prolonged out-times. Although aging generally does not affect the mostly chemically resistant carbon fiber bulk itself [153], load transfer at the fiber/matrix interface could be dependent on the pre-cure and eventually influence fiber-dominated properties. In fact, Gibhardt et al. [154] demonstrated that the aging of glass fiber sizing can shorten the composite fatigue life and reduce strength by up to 20 % as a result of aging-related

degradation of the fiber/matrix interphase. Otogo Be et al. [155] observed differences in aging effects depending on whether a reactive or a non-reactive sizing was applied to the glass fiber reinforcement. A comprehensive study conducted by Plonka et al. [156] on aging effects on sized glass fibers revealed a strong dependency of the interfacial strength on the chemical nature of the fiber surface and on surface roughness. The findings were assisted by complimentary analysis including eta potential, inverse gas chromatography (IGC), atomic force microscopy (AFM), and wetting techniques. Generally, similar effects may occur during prepreg out-time. However, the presented studies explored aging of the reinforcement fibers and the fiber-matrix interface for cured composites under hydrothermal and hygrothermal aging so that findings are not readily transferrable. Prepreg aging takes place in pre-cured composite materials on different time and temperature scales, respectively. Further research is required to reveal the mechanisms involved in the fiber and fiber-matrix aging during out-time and long-time freezer storage of thermoset prepregs.

Thirdly, almost all studies that included ILSS measurement discovered an aging-induced restriction of interlaminar ply bonding. Advanced cure propagation appears to hinder the formation of a load bearing interface with bulk properties of the matrix component. In an effort to explore the morphological impact of aging, Dobhal et al. [44] investigated the surface of Au-sputtered prepreg surfaces utilizing a scanning electron microscope (SEM). SEM images of fresh and aged (80 days) prepreg surfaces are displayed in Fig. 10.

While the surface of fresh prepreg is smooth and uniformly covered with resin (A), continuous cure progression led to the formation of surface-near defects such as voids and cracks (B-D). These irregularities are potentially not fully healed in the course of consolidation part cure and lead to insufficient interlaminar bonding. In this context, an exclusive but very interesting approach of investigation was pursued by Rabby et al. [45]. The herein presented aging method involved post-lay-up exposure of prepreg stacks to ambient conditions for time-spans up to 40 days. The results from mechanical testing after curing were compared to composites, which were made from prepregs that have been aged prior to lay-up. The negative impact of out-time exposure was shown to be mitigated if the aging was performed on an already consolidated yet uncured laminate. Possible reasons given were that the aging-related curing reaction at the interface of the stacked laminate favors interply crosslinking.

Despite the considerable amount of study outcomes with no effect of aging (Table 5), there are individual authors who reported an increase in FRP component performance. Hübner et al. [145] prepared industrial quality prepregs from HT carbon fibers, DGEBA, DICY, and an urea

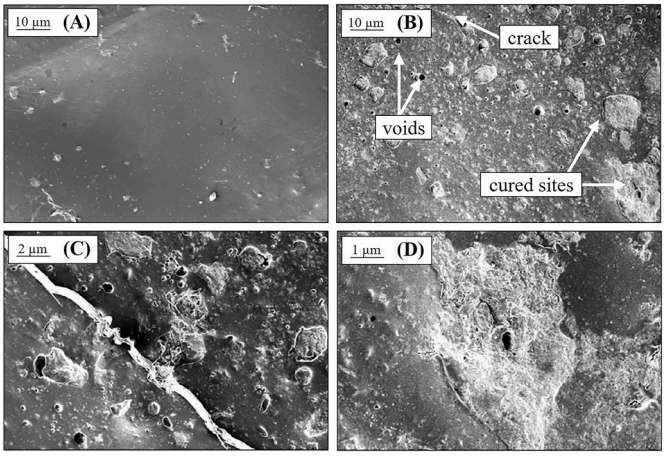

Fig. 10. SEM images of fresh (A) and aged (B–D) prepreg surfaces. Figure rearranged and relabeled for improved readability. Reproduced with permission [44]. Wiley, 2024.

accelerator and exposed them to ambient conditions for 90 days total. While the ILSS dropped by up to almost 20 %, the interlaminar energy release rate in mode I (G_{IC}) was increased substantially. The unexpected behavior was explained by as a result of the aging-induced secondary crack growth, which is usually not tolerated for G_{IC} testing. Another study revealing part performance-enhancing prepreg aging was conducted by Nakagawa et al. [148] who aged prepregs to three times the out-life equaling 84 days. The aged prepregs were not processed as delivered (continuous unidirectional) but rather repurposed by compression molding into discontinuous fiber composites (DFC). The laminate stiffness remained unaltered whereas tensile, compressive, and shear strength increased. The increase was attributed to the discontinuous nature of the produced specimens and a different effect of matrix plasticization compared to continuous FRP.

In terms of the influence of prepreg freezer storage on mechanical post-cure properties, a limited number of studies has yet been published. The outcomes are displayed in Table 6 following the same presentation structure as in Table 5. The investigated time spans range from 2 to 7 years, which is roughly 1–2 magnitudes longer than the variation interval for the explored ambient out-times (10–196 days). Analogously, either no or a negative influence were reported. The negative effect on mechanical properties, however, was less pronounced and results lacked runaway values exceeding a 20 % decline.

4.2. Void formation and porosity

Void formation leading to porosity in fiber reinforced plastics is considered one of the primary processing issues in composite manufacturing making voids the most extensively studied manufacturing defect [5]. Porosity is closely linked to the mechanical performance of composites (Section 4.1) and still remains an active topic of today's research. For prepreg technology, porosity has been targeted early in conjunction with autoclave cure. First systematic experimental studies and model proposals investigating void initiation, growth and reduction in autoclave cured prepreg laminates date back to the early 1980s [161–163]. More recently, a lot of research effort was put into out-of-autoclave prepregs - most likely due to the criticality of pores as a result of both the partly impregnation for air evacuation [164] and the reduced consolidation pressure of $\leq$0.1 MPa. In this context and with regard to material aging, Centea et al. [165] studied the effect of out-time and cure cycle on the consolidation behavior of OoA prepregs but found no influence of aging on the material microstructure by x-ray microtomography. The resin viscosity, however, was reported to increase, which was eventually implemented in an analytical model [166] to predict the impregnation behavior upon OoA cure. The developed models were used for a parametric study to determine the impact of fiber architecture, cure cycle temperature and resin initial degree of cure on porosity. The aging-relevant parameter 'resin initial degree of cure' α_0 was discretized into intervals of 5 % cure at 0.01, 0.05, 0.10, 0.15, 0.20, 0.25, and 0.30, which appears reasonable in comparison to the data of Fig. 2. Run simulations estimated the duration that is needed to reach zero-porosity/full impregnation ($\beta = 1$) when applying different cure cycles (Fig. 11).

Actually, the initial degree of cure was identified as the most important effect on impregnation times. An $\alpha_0 > 25$ % could not be mitigated by long cure cycles for any investigated material. The findings underline the importance of knowing and accounting for the out-time exposure of OoA prepregs prior to cure. The analytical model was validated in Ref. [51]. The model was shown to slightly underestimate the void content for high out-times longer than 25 days (model: 26 % vs. exp.: 17 % after 56 days) but replicated the general trend accurately. Interestingly, the onset of void formation coincided with the out-life specified in the data sheet (21 days) while the results show a stabilization of the detrimental aging effect on porosity after 40 days, reaching a plateau. Corresponding micrographs of composites made from room temperature aged carbon fiber epoxy prepregs are collated on the

Table 6
Studies exploring the influence of long-term freezer storage on post-cure mechanical properties.

Ref.	First author	Used prepregs	Aging conditions			Cure method	Mechanical property	Significant influence?	Rel. change in property
			Temperature	RH	Time span				
[157]	Amare	EP CF UD	−18 °C	n/a	84 months	Autoclave	Tensile strength	No	
							Tensile modulus	No	
							ILSS	Yes	−20.4 %
							Bending strain	No	
[158]	Amare	EP CF UD	−18 °C	n/a	84 months	Autoclave	Tensile 45°	Yes	−7 %
[159]	Miller	EP GF UD	−18 °C	n/a	22 months	Autoclave	Tensile strength 0°	No	
							Tensile modulus 0°	No	
							Tensile strength 90°	No	
							Tensile modulus 90°	No	
							Compressive strength 0°	No	
							Compressive modulus 0°	No	
							Compressive strength 90°	Yes	−8.9 %
							Compressive modulus 90°	No	
							In-plane shear strength	Yes	−7.7 %
[160]	Rao	EP CF UD	−18 °C	n/a	24 months	Vacuum bag	Tensile strength	Yes	−13 %
							Tensile modulus	Yes	−15 %

Abbreviations: CF: carbon Fiber, EP: epoxy, UD: unidirectional, ILSS: (Apparent) interlaminar shear stress, n/a: not specified.

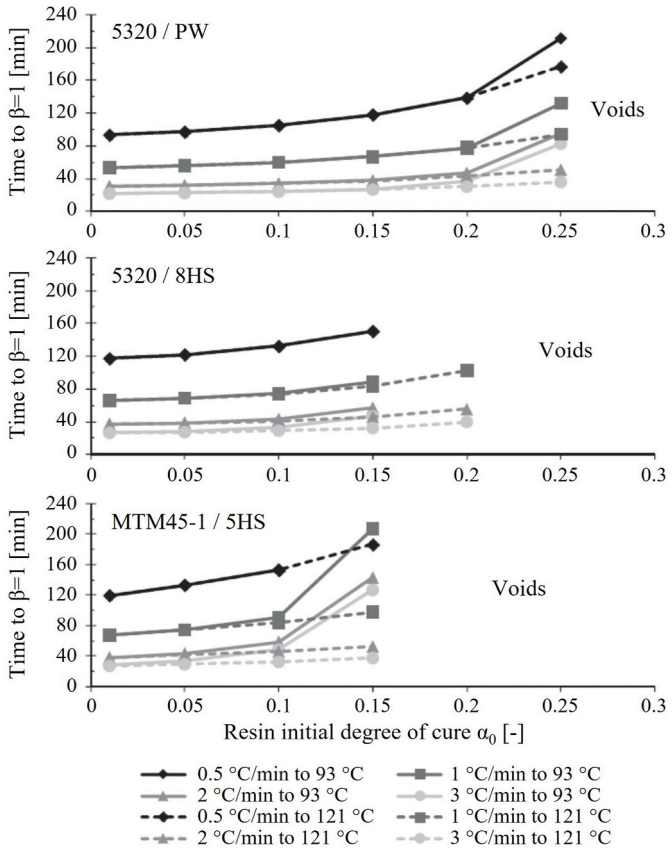

Fig. 11. Model-based prediction of the necessary time for void-free composite manufacturing (β = 1) as a function of aging-related pre-cure. The data was calculated for three OoA prepregs and different applied cure cycles. Areas labelled 'voids' indicate conditions in which β = 1 is not reached. Figure relabeled for improved readability. Reproduced with permission [166]. Elsevier, 2012.

left-hand side of Fig. 12. For comparison, cross-sections of composite laminates that were manufactured by Amare et al. [158] from fresh and nine years freezer-stored prepregs are shown on the right. Storage induced intraply porosity increased the 10-ply composite thickness from 1.86 to 2.01 mm. Furthermore, the toughener located at the ply interfaces of the interleaf prepreg seems to be affected by storage.

In the context of void formation and porosity, Putnam and Seferis [167] explored prepreg gas permeation as a function of aging time to quantitatively describe the ability of prepreg to vent off volatiles during consolidation and curing. Results showed that interlaminar permeation increases greatly with aging time while intralaminar permeation increases only slightly. Both findings could be explained by surface resin hardening of the prepreg samples similar to the surface changes pictured in Fig. 10. Other following studies [168–170] experimentally exploring or modeling gas transport through prepregs did not study the aging effects directly, but linked permeability to resin viscosity, which is affected by aging-related pre-cure (Section 2.). Umer et al. [171] did similar research measuring creep compaction and X-ray microtomography (μCT)-based permeability of aerospace-grade prepregs that were outdated by six months.

5. Aging mitigation strategies and recycling approaches

5.1. Aging mitigation strategies

In addition to storage at low temperatures, minimizing aging effects can also be achieved by adapting the resin formulation as part of pursuing a latent curing strategy. Latent curing strategies aim at prolonging out-time and processability by slowing or even completely preventing the resin reactivity at room temperature. Three latent curing strategies can be harnessed according to Kim et al. [172], namely, latent curing agent such as DICY or diaminodiphenyl sulfone (DDS), encapsulation and phosphorus-based complex compound. When exposed to external stimulation, e.g., heating above specific temperatures or photo-irradiation, the curing process is triggered an will continuously progress from this point forward [173]. In the context of prepreg technology, however, most studies focused on the application of latent (high-temperature) curing agents as reviewed in the following.

Pouladvand et al. [174] introduced a dual-curable epoxy prepreg formulation based on DGEBA cured with a combination of DICY as latent curing agent and diethylenetriamine (DETA) as a low-temperature curing agent. A 7 % increase in conversion was measured after ambient aging over 21 days, which is about half the reactivity of commercial systems (Section 2.1). Prepreg processing properties in terms of tack and flexural rigidity could mostly be maintained during this rather short period. The separation in curing stages was argued to cause to the removal of prepreg thermal history and hence leads to a high storage stability. A similar study was presented by Hübner et al. [145] who also used DGEBA and DICY but in combination with a semi-latent urea-based catalyst for creating a fast-curing potential of prepregs. Despite being able to fully cure the system within 15 min, the authors reported constant tack and resin flow over 92 days and a limited increase of T_g

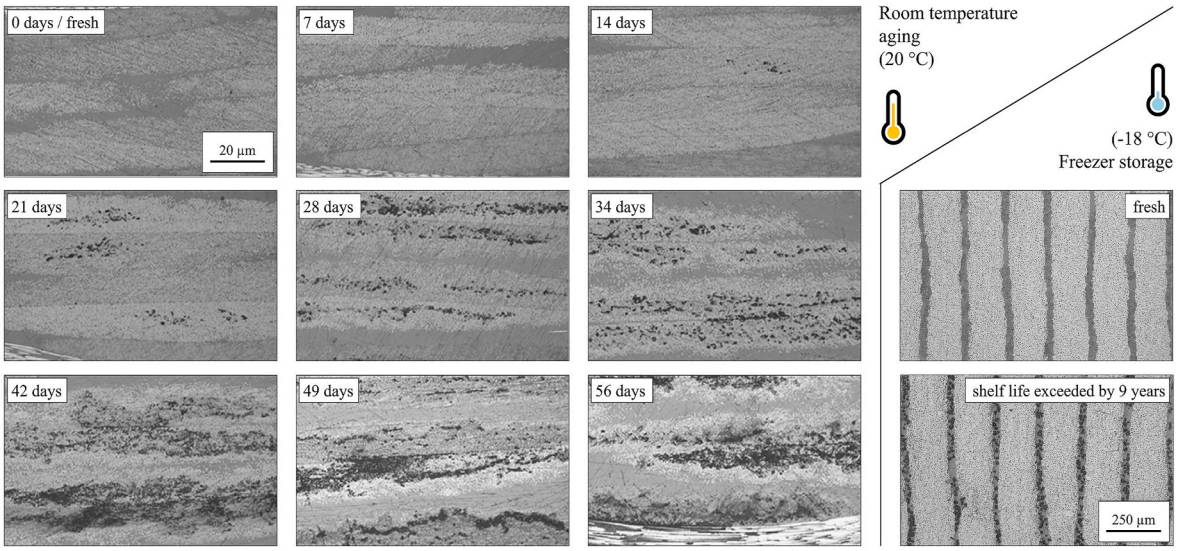

Fig. 12. Influence of room temperature aging (left, [51]) and freezer storage (right, [158]) on the porosity of cured CFRP parts made from thermoset prepregs. Figure rearranged and relabeled for improved readability. Reproduced with permission. Elsevier, 2013 and IOP Publishing, 2023.

(uncured) by 5.2 °C. The same combination of resin and hardener with three different accelerators, namely a methylene bis (phenyl dimethyl urea), a cycloaliphatic substituted urea and a modified polyamine was used for prepreg production by Dalle Vacche et al. [175]. Significant differences in shelf life, which was defined as the time to reach double initial viscosity at room temperature, were reported. The cycloaliphatic substituted urea performed best in terms of independence of out-time reaching a shelf life of more than 70 weeks. Long term stability over 8 months was confirmed for two out of the three investigated formulations by DSC. Similar observations were made in Ref. [61] where both the epoxy absorption peak at 915 cm^{-1} in a FTIR spectrum as well as exothermic DSC enthalpies remained unchanged over 60 days of room temperature aging. Niazi and Beheshty [176] studied the potential of their newly proposed accelerator for DGEBA/DICY systems in terms of shelf life extension by quantifying the resin pot life via viscosity measurement. Compared to two liquid commercial accelerators, the prepreg shelf life could be increased fivefold.

Kim et al. [172] added organic and inorganic deactivators, namely fumed silica, boric acid, benzoic acid, and triethyl borate, to an epoxy/amine system to subsequently produce prepregs thereof. Especially the incorporation of triethyl borate induced latent curing behaviors of the epoxy resin matrix delaying immature cure progression at low temperatures. Carbon fiber prepregs made from methanol-ester and isopropyl-ester monomer solutions were comparatively investigated by Aston et al. [177]. Room temperature storage of the novel isopropyl ester system was found to increase the prepreg shelf life by at least an order of magnitude while maintaining composite properties. Rabby et al. [178] demonstrated that a plasma surface treatment can mitigate aging effects by raising the surface free energy of uncured prepregs. Enhanced bonding properties in the course of this led to composite parts (from 30 days aged prepregs) that could be subjected to 52 % higher tensile and 20 % higher flexural loads compared to non-treated samples. In terms of tensile strength, the full load-bearing capacity of fresh prepregs was restored.

For short-term industrial implementation, however, practical strategies are needed for commercial out-of-spec prepreg where resin chemistry change is not feasible. In pursuit of this objective, Kim and Nutt [179] developed a mitigation strategy for aged prepregs through cure cycle modification. Composite parts with less than 1 % voids as desired by the aerospace industry were produced by VBO processing. The flow-optimized cure cycle allowed the use of long-term ambient aged prepreg materials by extending the boundary of the manufacturer's specified out-life for OoA prepregs by 175 %.

Another promising approach to mitigate aging effects is to use vitrimers as the matrix material. Vitrimers like thermosets form polymer networks upon curing, however, their networks feature reversible covalent bonds also known as covalent adaptable networks (CAN) that can either be dissociative or associative [180]. By replacing thermoset matrices with vitrimer counterparts, new capabilities emerge such as healability, reprocessability and recyclability of cured components if heated above a specific temperature [181]. The potential of the new polymer class was quickly explored for composites in general and for prepregs in particular, the latter most recently i.a. in Refs. [182–184].

With respect to out-time, Gómez Sánchez et al. [185] tried to prepare non-expiring carbon fiber/epoxy prepregs based on a vitrimeric matrix cured with 2-aminophenyl disulfide. The term 'non-expiring' may be partly misleading as after 30 days of out-time, the prepregs showed the same general changes in terms of processing characteristics after B-staging as outlined before (Section 3), e.g., loss of formability and tackiness. This will most likely effect lay-up processes such as AFP and ATL in a crucial way. Still, the authors found no negative effect on the mechanical properties of cured parts made from aged vitrimer prepregs. Note that the investigated time spans were rather short (30 days) and processing properties were not assessed directly, which leaves room for further investigation. Zamani et al. [186] also investigated the favorable properties of vitrimer prepregs but at the same time were able to achieve a long shelf life. Compared to commercial epoxy-diamine cured epoxy, the vitrimer system based DGEBA and a synthesized multifunctional secondary amine cured 11 times slower at room temperature.

In the general case of using vitrimers as prepreg matrices, two scenarios have to be distinguished: If vitrimeric prepregs are intentionally being transferred to the C-stage after impregnation to realize a fully cured delivery form [187], freezer storage becomes redundant and room temperature a non-factor leading to the avoidance of waste as pictured in Fig. 13. Some authors in this context speak of composite manufacturing from 'enduring prepregs' [188–192], which resemble thermoplastic organo sheets in terms of processing. Aging effects will be mitigated in a sense that the prepreg will remain reformable at high temperatures, e.g., via compression molding or hot forming processes. If vitrimeric prepregs are otherwise provided in the B-stage, which allows the possibility of low temperature forming such as AFP or ATL, aging effects similar to those of conventional thermoset prepregs can be expected as reviewed.

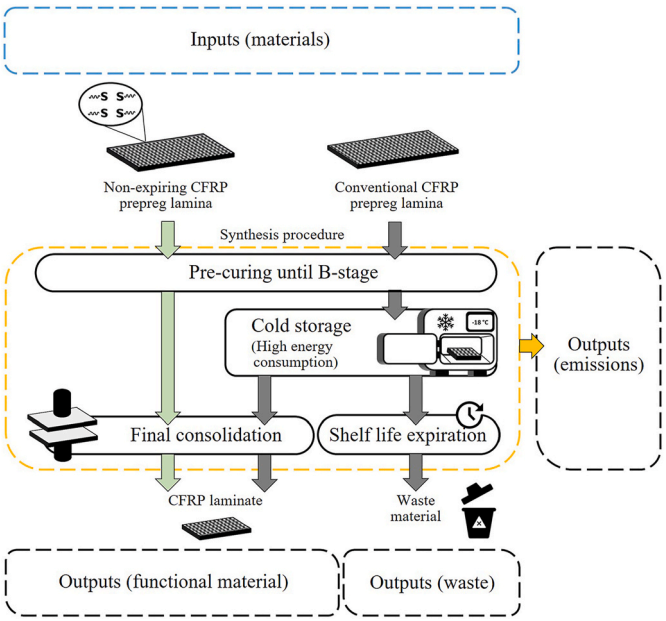

Fig. 13. Schematic illustration comparing the non-expiring vitrimeric and the conventional thermoset prepreg life cycles. Figure relabeled for improved readability. Reproduced with permission [185]. Elsevier, 2024.

5.2. Recycling of out-of-spec prepregs

Beyond commercial interest, a transnational commitment towards sustainability and climate protection is driving the advancement of eco-friendly materials and recycling routes to accompany established composite production chains. In this context, multiple approaches were proposed to re-use outdated thermoset prepregs as feedstock for recycled composite materials in an environmentally sensitive manner. Fiber recovery strategies by thermal or chemical procedures are not reviewed at this point but can be found in numerous review articles on composite recycling [193–199].

For direct re-use of aged prepregs, Chadwick et al. [28] demonstrated the production of chips using sheet-molding compounding (SMC) equipment for subsequent conversion into composite parts by compression molding. Here, the SMC processing route required a certain amount of pre-cure but higher mechanical properties could be achieved compared to conventional glass-reinforced SMC. The same recycling strategy was followed earlier by Sultana et al. [200]. The authors argued that the production of recycled composites without the addition of additional resin and the absence of any effect of prepreg age, would demonstrate the potential of converting out-of-spec prepregs with unknown thermal history into high-performance composites using existing SMC techniques.

Wu et al. [26] aged epoxy prepreg scraps for 14 and 28 days at ambient conditions before hot-pressing parts with random fiber distribution. Surprisingly, increasing aging times favored the production of low-porosity composites, which was attributed to a uniform initial chip distribution and at the same time enabled chip re-arrangement and nesting during consolidation. Part production from discontinuous prepreg scrap was also investigated in detail in consecutive studies by Feraboli et al. [201–204]. Autoclave-cure was applied to thermoset prepreg waste by Souza et al. [205,206] who demonstrated feasibility of the production route to manufacture composite parts for non-critical structural applications. Smith and Hubert [207] set up a recycling framework, which employs comprehensive material characterization and targeted elevated temperature staging to produce a recyclate with tailorable processing characteristics. Further studies on recycling strategies of composite materials, which can potentially be sourced from outdated thermoset prepregs, were conducted by Mansour et al. [208], Giorgini et al. [209], Nilakantan and Nutt [210], Bao et al. [211], Martinez et al. [212], Turner et al. [213] and others. Alongside academic research, there are ongoing industrial initiatives to put feasible recycling strategies into use. Owing to the increasing amount of carbon fiber prepreg waste, both established material suppliers and newly formed startups like Elevated Materials, US and Fairmat, FR are industrializing processes for direct re-use of out-of-spec materials.

Instead of direct forming or molding of thermoset prepreg waste, mechanical recycling routes have been proposed that are based on using out-of-spec prepregs as fillers to be mixed into either virgin or likewise recycled thermoplastic matrices. Cured waste carbon fiber prepregs were milled and processed in the recent research of Irez and Yakar [214] to produce chopped carbon fiber-based composites with a r-polypropylene matrix. A similar approach was followed by Gröning et al. for glass fiber prepregs in combination with polypropylene and polyamide 6 as well as Butenegro et al. [215] for bio-based polyamide 11.

Ajam et al. [216] employed a series of chemical and thermal treatments to reshape and re-strengthen prepreg rolls that were cured over the shelf. The recovered strength and modulus of the composite parts made from recycled prepreg reached 65 % of the original properties.

6. Conclusion and future perspectives

Prepregs impregnated with thermoset resin are known to be temperature and moisture sensitive precursor materials for manufacturing high-performance components from fiber reinforced plastics. The aging process in the course of both freezer storage and ambient exposure has hence raised major scientific interest in the field of advanced composite manufacturing. In the presented literature review, a strong focus on epoxy-based prepregs was identified while only individual studies on phenolic and BMI prepregs were conducted. In general, aging effects seem to be more pronounced whenever OoA prepregs are used instead of conventional prepregs and autoclave cure. For the latter, processors can to a certain extent rely on the high applied pressures to neutralize lay-up defects and compensate for material factors, such as absorbed moisture, air entrapment and out-time.

The main findings from the presented literature review can be concluded as follows. Given values are to be understood as averages or general trends derived from comparing different studies from literature and may vary depending on the investigated materials, testing techniques and aging conditions.

Physio-chemical prepreg state

- Cure progression monitoring during room temperature aging reveals an average conversion rate of 0.7 % per day within the first 60 days of ambient exposure.
- The glass transition temperature T_g rises accordingly while the rates of α and T_g increase are reported to slow down after out-time induced resin vitrification.
- Similar changes in the prepreg state are observed for long-term freezer storage but on a much larger time scale that is 1–2 magnitudes slower rates compared to ambient aging.
- Water uptake from moisture exposure during aging can reach up to ~1 % for neat resin equaling 0.3–0.4 % weight gain for prepregs, which can lead to porosity in the cured FRP part.
- Aging-related cure progression is linked to an increase in viscosity implicating earlier resin gelation and a shift in viscoelastic behavior towards the elastic component.

Processing

- Out-time increases the bending-stiffness of thermoset prepregs and effects inter- and intraply shear, which has to be considered in forming, molding, and lay-up processes.

- For room temperature processing, prepreg tack fades and is eventually lost completely with increasing out-times. It can be restored if processing parameters are adapted to the prepreg state, e.g., in terms of lay-up velocity, compaction pressure and in particular temperature.
- Cure cycles for aged prepregs must be adapted to the prevalent precure state in order to produce highly or even fully cured FRP components, e.g., by higher cure temperatures at isothermal plateau stages or extended dwell times.

Post-cure quality

- Mechanical properties suffer an average decrease of more than 20 % after weeks of ambient exposure, however, the cross-study results show high variation.
- Matrix-dominated properties such as ILSS, transverse tensile strength/modulus and compression strength/modulus are more strongly influenced by aging than fiber-dominated characteristics.
- Void formation and resulting part porosity are enhanced if prepregs have suffered substantial ambient exposure and/or freezer storage due to hindered air and volatile release. OoA prepregs are more vulnerable than autoclave-cured prepregs.

For future use of thermoset prepregs, meeting the growing pressure to increase sustainability and to reduce the carbon footprint of prepreg-based composites requires a departure from landfilling activities for out-of-spec material. The intrinsic characteristics of thermoset prepregs are one of the reasons why the use of thermoplastic carbon fiber prepregs, e.g., as an alternative for structural aircraft parts manufactured via thermoplastic in-situ AFP, is currently a topic of high interest, both industrially and academically. Still, a turn away from thermoset prepregs in a short and mid-term perspective is unlikely so that the problem of out-of-spec material will persist. That is why practitioners still seek short-term solutions to prolong both shelf life and out-time or to make their process more robust to aging effects, respectively. The latter requires a yet to be gained in-depth knowledge on the complex interdependency between prepreg state, processing, and part quality. Simple and rapid detection techniques need to be made available for workshop operations to assess how much shelf life and out-time is left. This way, changes in prepreg state can quantitatively be tracked enabling processes to be adapted to out-time effects and minimizing waste. For the case that conventional thermoset prepregs are no longer processable in their intended processing routes due to excessive aging, recycling strategies for out-of-spec materials have been proposed. Some of these strategies go beyond conventional thermal or chemical recycling by further processing out-of-spec material, however, a certain degree of downcycling due to fiber shortening is inevitable. First companies have picked up on the challenge by showcasing that recycling out-of-spec carbon fiber prepregs can be economically feasible if transferred into the right applications. Facing the persistent growth of prepreg waste, especially in the aerospace industry, capabilities will have to be ramped up substantially in the future.

For future use, new thermoset prepreg resin formulations based on latent curing strategies were proposed in an effort to mitigate the pictured aging-related challenges for future materials. Looking forward, the results show potential to reduce or fully suppress the impact of ambient out-time on the evolution of the resin physio-chemical state and hence on prepreg processing and post-cure properties. Furthermore, first studies on vitrimer for the usage in prepregs have been conducted since their introduction in 2011. Although research has mostly been limited to academic and lab-scale settings, vitrimeric materials show high potential for establishing enduring prepregs for the composites industry. Due to the nascent stage of the new material class, knowledge gaps remain in terms of formulation, processing and thermo-mechanical performance, especially for the prepreg manufacturing route. Also, the boundaries of vitrimer-specific, thermoplastic-like properties of enduring prepregs including formability and weldability are yet to be fully explored and understood.

The industry has been and still is trending towards the generation of big data in composites production environments for monitoring and controlling manufacturing processes in operational situations. While individual or less complex phenomena can be modeled accurately by physics-based models as shown in this review, the complexity of production processes can quickly push these models to their limits, especially if additional material changes are considered. Data-driven methods based on big data analytics, machine learning (ML), and artificial intelligence (AI), however, are prospects to enable informed process adaption to changed production and material conditions such as aging-affected thermoset prepregs. Different directions of ML-based property prediction have already been taken by the scientific composite community [217–220] but data driven methods to account for prepreg aging in the B-stage are yet to be presented.

CRediT authorship contribution statement

D. Budelmann: Writing – review & editing, Writing – original draft, Visualization, Funding acquisition, Data curation, Conceptualization. **D. Gibhardt:** Writing – review & editing, Project administration. **B. Fiedler:** Writing – review & editing, Supervision, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors are grateful for the financial support by Deutsche Forschungsgemeinschaft (DFG – German Research Foundation) granted for the research project ‘FurPreg’ (project number 559990233).

Data availability

Data will be made available on request.

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