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Research, Technology and Education

Lucas F. M. da Silva · Robert D. Adams · Klaus Dilger *Editors*

2nd International Conference on Industrial Applications of Adhesives 2022

Selected Contributions of IAA 2022

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Preface

This volume of *Proceedings in Engineering Mechanics—Research, Technology and Education* contains selected papers presented at the 2nd International Conference on Industrial Applications of Adhesives 2022 (IAA 2022), held in Carvoeiro (Portugal) during March 3–4, 2022. The goal of the conference was to provide a unique opportunity to exchange information, present the latest results as well as to discuss issues relevant to industrial applications of adhesives.

This conference is held every two years. The conference is chaired by Lucas F. M. da Silva and co-chaired by R. D. Adams (University of Oxford, UK), Chiaki Sato (Tokyo Institute of Technology) and Klaus Dilger (Technische Universität Braunschweig, Germany). The focus is on applications of adhesive bonding in the industry such as automotive, aeronautic, railway, marine, energy and electronics. The idea is to bring together the adhesive makers and the adhesive users to exchange experiences and facilitate potential synergies and partnerships. Sixty-one papers were presented by researchers from 14 countries.

In order to disseminate the work presented at IAA 2022, selected papers were prepared which resulted in the present volume. A wide range of topics are covered resulting in nine chapters dealing with adhesion assessment between polymers and metals (first chapter), pressure sensitive adhesives (2 chapters), adhesive bonding process optimization (1 chapter), civil applications (2 chapters), adhesive joints in composite materials (2 chapters) and elastic adhesives (last chapter). The book is a state of the art of industrial applications of adhesives and also serves as a reference volume for researchers and graduate students working with adhesive bonding.

The organizer and editor wish to thank all the authors for their participation and cooperation, which made this volume possible. Finally, I would like to thank the

team of Springer-Verlag, especially Dr. Christoph Baumann and Ute Heuser, for the excellent cooperation during the preparation of this volume.

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Contents

A Method to Evaluate the Interfacial Adhesion Between Polymer and Metal

Achim Frick, Markus Rettenberger, and Marcel Spadaro

Abstract Up to now there is no standardized measuring method to properly quantify the interfacial adhesion between polymer and metal. This work presents a possible approach to this topic based on the quantification of the interfacial tightness of a polymer-metal hybrid. Two types of specimens, a monolithic specimen and a hybrid specimen, were fabricated by injection molding, partially thermally post-treated, and then subjected to tracer gas measurement, using helium as the tracer gas. The recording of the helium gas volume rate over the test duration enables the determination of a tight interface, which is characterized by full-faced contact between polymer and metal. Micro- or sub microscopic gaps between polymer and metal lead to a constant gas volume rate signal, while a tight interface without gaps exhibits a permeation characteristic. The determination of the gas volume rate on both specimens and a subsequent subtraction allows the separation between measurand falsifying permeation rate and actual interfacial gas volume rate. Therefore, tight interfaces without any gaps from different material combinations can be compared regarding their true adhesion performance. On the other hand, microscopic studies of the interface do not provide information on interfacial adhesion, since no interpretable visual differences in the interfaces of demonstrably tight and leaky hybrids could be detected within this study. The newly developed and presented method can enable research into compatible material combinations, effective adhesion modifications and suitable processing conditions for improving interfacial adhesion between polymer and metal.

Keywords Interfacial adhesion · Polymer-metal hybrids · Interfacial tightness · Tracer gas measurement

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1 Introduction

In the automotive sector, the demands for energy-efficient and thus sustainable overall solutions have increased exponentially in recent years. In addition to the switch to alternative drive systems, the lightweight construction concept continues to represent another partial solution to this ambition [1–7]. To realize this concept, monolithic components are preferably replaced by multi-material hybrids, which offer enormous weight savings with identical mechanical load capacities [1, 3, 4]. In addition to load-bearing structures, such as front ends [8], mechatronic systems, such as connectors for signal or power transmission, are nowadays also designed as polymer-metal hybrids (PMHs) [2, 4, 7, 9]. Although the focus here is less on the mechanical load-bearing capacity of the assembly and more on media tightness, both requirements can only be realized as a result of strong adhesion in the interfaces between polymer and metal [1, 4–7, 9–11]. The term adhesion includes all interfacial forces that contribute to the mechanical cohesion between the two joined materials [1, 3, 12, 13]. Several papers have already reported possible interface failures caused by a lack of interactions at the macroscopically finite interfaces of a PMH. These detachments of the polymer component, often titled as so-called gaps, are usually shrinkage [9] or generally strain-induced [4, 14]. Consequently, these gaps indicate incompletely formed adhesion in the interface [6, 15] and therefore offer enormous potential for characterizing these critical regions.

Since the layer thicknesses of the interfaces are in the single-digit nanometer range [12], optical microscopy images are not a satisfactory characterization alternative due to the limited resolution of 0.5 µm [6, 16]. Since interfacial forces cannot be measured directly [6, 12], pull-out or pull-off tests on hybrid specimens are often used, the result of which is the bond strength correlated with the interactions [1–3, 5, 6, 10, 11, 13, 14, 17, 18]. In addition, both the simple feasibility and the high significance speak for the use of this test [3]. However, future applications of PMHs, such as use in electrical conductors [19], often require not only strong but also tight bonds, the majority of which require full-faced contact and thus full-faced adhesion of both materials at the interface [2, 7]. The smallest detachments of the polymer component from the metal surface are not statistically noticeable in the result of the bond strength and are therefore not measurable with the destructive test methods commonly used today. Especially in media-tight applications, these gaps can cause failure of the mechatronic system as a result of a short circuit caused by previous media ingress [1, 2, 4, 7, 9, 19, 20].

In addition, a methodology is to be developed which can quantify different properties between interfaces where full-faced contact is present on the nanoscale and thus offers a possibility to characterize the interfacial adhesion between different materials.

2 Materials and Methods

2.1 *Differential Tightness and Permeation Measurement (DTPM)*

Within the interfacial tightness characterization of PMHs, there is the possibility of a nondestructive tracer gas measurement [14]. The test specimen is sealed, pressurized with the tracer gas and surrounded by an evacuated detector chamber sealed against the environment. Due to the pressure and tracer gas concentration gradient between the test specimen and the detector chamber, there is an unavoidable tracer gas volume flow in the direction of the evacuated detector chamber. As the name implies, the detector chamber is connected to a corresponding detector system, which detects and records the amount of tracer gas escaping from the test specimen over the testing time, thus enabling tracer gas measurement (TGM) according to DIN EN 1779: 1999 B2.1. The test fixture used for TGM in this work is shown in Fig. 1.

Authors propose, that a tight bond between polymer and metal, must show a permeation characteristic when investigating it by means of TGM, while a local detachment of the polymer at the interface leads undoubtedly to an instantaneous tracer gas transfer without any prior dissolution and diffusion mechanism. This allows leaky and tight interfaces to be reliably detected and distinguished. In the case of a tight hybrid, where full-faced contact of the two materials must therefore be present, the time course of the gas volume curve must therefore resemble that of a known permeation curve, which initially exhibits transient behavior until the steady-state permeation level is reached [21–23]. However, assuming there is no gas volume flow through the crystalline structure of the metal [24], this permeation curve is composed of the following gas flows:

1. Permeation through the free volume of the polymer component
2. Permeation through the free volume of the sealings used
3. Permeation through the necessary sealing contacts to the polymer surface or the test fixture surface.

In order to obtain an unambiguous permeation signal for the actual interfacial gas volume rate, this volume rate and the permeation signals just listed must be separated from each other. The so called "Differential Tightness and Permeation measurement Method "(DTPM) in which the permeation signal of both a hybrid and a geometrically identical, monolithic test specimen made of the same base polymer is recorded by means of TGM, can be used for this purpose. The actual interfacial gas volume rate then can be calculated from the both measured results according to Eq. 1.

$$\dot{q}_{\text{Interface}} = \dot{q}_{\text{Hybrid}} - \dot{q}_{\text{Permeation}} \tag{1}$$

In the equation $\dot{q}_{\text{Interface}}$ means the tracer gas volume rate through the interface, $\dot{q}_{\text{Hybrid}}$ the measured gas volume rate through the hybrid specimen and $\dot{q}_{\text{Permeation}}$ the

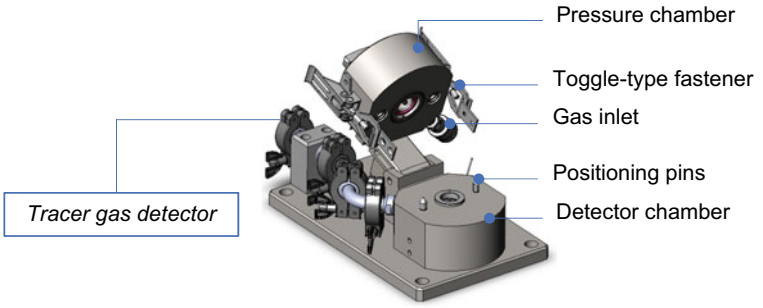

Fig. 1 Possible test fixture necessary for tracer gas measurement (TGM); presented in the opened state without test specimen

measured permeation rate through the monolithic specimen and the sealings of the test fixture. Possible test specimens necessary for the application of the method are presented in the following chapter.

2.2 Test Specimen Geometry

The polymer-metal test specimen investigated was specially developed and consists of an injection-molded polymer blank with a diameter of 30 mm and a wall thickness of 1.2 mm (z-direction), which is penetrated centrally by the metal insert, Fig. 2. The hybrid test specimen is produced in a single-cavity mold by assembly injection molding. In addition, closed, monolithic polymer blanks without metal inserts can be produced by exchangeable inserts in the injection mold, which are then used to measure the permeation rate through the polymer component used.

2.3 Metals

Deoxidized copper (Cu-PHC) with a copper content of 99.95% and pure aluminum (EN AW 1050A) with an aluminum content of 99.50% from the manufacturer Wieland-Werke AG were used as materials for the rectangular, metallic inserts with a sheet thickness of 0.8 mm. The overmolded metal inserts were produced by stamping from semi-finished products and, prior to overmolding, were placed in beakers containing propan-2-ol for 15 min and cleaned in an ultrasonic bath from the manufacturer Elma Schmidbauer GmbH (type: Elmasonic S 10 H) at a water bath temperature of 40 °C. In the following, the aluminum type will be abbreviated as “Al” and the copper type as “Cu”.

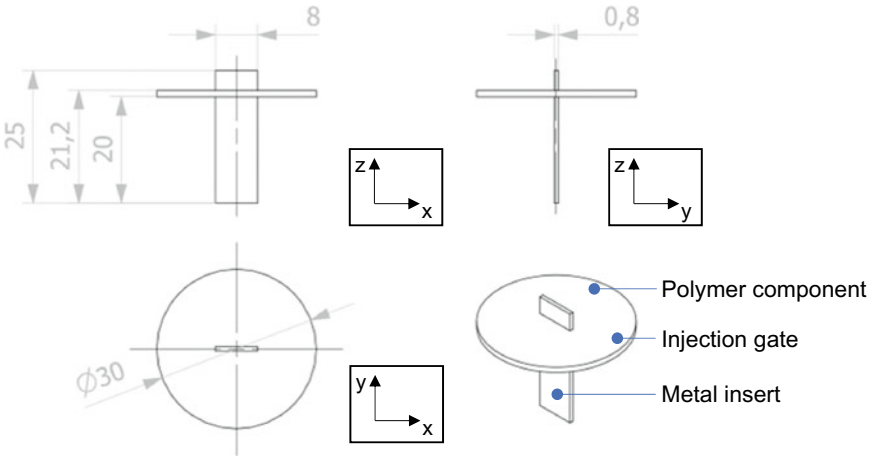

Fig. 2 Hybrid test specimen in different drawing views (dimensions in mm)

2.4 Polymers

The polymers used for the production of the test specimens described are, on the one hand, polypropylene (PP) from the manufacturer Ineos Holdings Ltd (type: 100-GA12), whereby two sample batches were adhesion-modified with 20 wt% of a PP grafted with maleic anhydride (PP-g-MAH) from the manufacturer Mitsui Chemicals Europe GmbH (type: Admer 3115E). On the other hand, a commercially available, black colored polyamide 6 (PA6) from the manufacturer LANXESS Deutschland GmbH (type: Durethan B30S) was used, which was dried for 6 h at 80 °C immediately before processing using a dry air dryer from the manufacturer Bierther GmbH (type: DR202 MT), in order to be able to exclude hydrolytic degradation of the material.

2.5 Specimen Production

While the monolithic specimens were merely injection molded, the hybrid ones were produced by assembly injection molding. Both specimen variants were produced employing an injection molding machine from the manufacturer Arburg GmbH & Co. KG (type: 220 S Allrounder 150–30) with a screw diameter of 15 mm using a single-cavity mold with spot gating. In order not to produce a different morphology of the two variants within one polymer type with regard to permeation behavior, the injection molding processing conditions were always identical for both specimen variants within one polymer type, Table 1.

The insertion of the metal inserts and the removal of the hybrid specimens were performed without a handling system using nitrile gloves.

Table 1 Injection molding processing conditions for the production of the investigated test specimens

Parameter	Unit	PP	PP + PP-g-MAH	PA6
Nozzle temperature	[°C]	240	240	265
Mold temperature	[°C]	60	60	100
Injection speed	[cm^3/s]	30	30	12
Switchover melt pressure	[bar]	700	700	550
Holding pressure	[bar]	500	500	500
Holding pressure time	[s]	7.5	7.5	9

2.6 Thermal Post-Treatment

In this study, thermal post-treatment means that in an additional process step after assembly injection molding of the PMHs, the areas of the polymer component close to the interface are re-melted by induction heating of the metallic joining partner. On the one hand, this allows the number of intermolecular interactions between the joining partners to be increased [25, 26], and on the other hand, any nanoscale pores present on the metal surface can be better infiltrated by the polymer melt. Both potentially contribute to an increased adhesion effect, regardless of physiochemical [25, 27] or mechanical nature, which should lead to a full-faced contact at the interface of the two materials.

Re-melting of the regions close to the interface was carried out immediately after the injection molding process by means of a mobile induction heating device from the manufacturer EFD Induction GmbH (type: Minac 6/10) with a maximum output power of 10 kW at a frequency range of 10–40 kHz. The temperature of the metal insert during thermal post-treatment was determined by a miniature sheath thermocouple from the manufacturer TC Mess- and Regeltechnik GmbH, and the temperature distribution on the surface of the polymer component was determined using an infrared camera from the manufacturer Optris GmbH. The temperature profile over time was recorded digitally. Possible measurement errors due to a slight inclination of the thermal camera were not investigated further, Fig. 3.

To ensure a high reproducibility of the measurements, the hybrid test specimens were placed and aligned in a holder manufactured for this purpose, consisting of two pneumatically controlled clamping jaws. The material Kelutherm 800 M from the manufacturer KELUX Hochleistungswerkstoffe GmbH, from which the clamping jaws were made, has a low thermal conductivity of only 0.26 W m^{-1} K^{-1}. The thermal conductivity of copper is about 399 W m^{-1} K^{-1}, that of aluminum 220 W m^{-1} K^{-1}, and those of PP and PA6 are 0.22 W m^{-1} $K^{-1}$and 0.31 W m^{-1} K^{-1}, respectively.

During thermal post-treatment, which causes partial melting of the polymer component in the regions near the interface, a joining pressure is generated due to the local volume expansion of the polymer in the region of this zone, which is largely dependent on the expansion coefficient of the respective polymer and its volume increase during the phase transition from the solid to the liquid aggregate state. In