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ORIGINAL PAPER

Impact of environmental conditions on silicone pressure-sensitive adhesives

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Abstract

Silicone pressure-sensitive adhesives are characterized by very good self-adhesive and functional properties. The first group includes: tack, adhesion and cohesion. No less important are the functional properties: maximum working temperature, shrinkage, resistance to external factors and yellowing. It is well known that silicone adhesives are materials for special applications due to their properties, namely surface tension, high UV transparency, good thermal stability, low intermolecular interactions or low surface tension. This paper attempts to investigate the influence of environmental conditions and PSA aging time on the most important functional and self-adhesive properties. This is an important aspect in terms of the use of this type of materials and their suitability for use. The tests were carried out after 1 h days and after a 6 month. To investigate the impact of environmental conditions samples (tapes and joints tapes with a steel substrate), they were conditioned in a climatic chamber simulating environmental conditions by 20 days.

Keywords Silicone pressure-sensitive adhesives · Self-adhesive tapes · Aging · Silicones

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Introduction

Adhesives are one of the most important methods for combining different elements. Our ancestors, centuries ago, also used this connection method using, for example, natural rubber. In this way, they were able to bond different materials, both wood, stone and feathers. It also suited them to produce simple tools, such as arrows. The first self-adhesive tape, obtained by Egyptian scholars more than 6000 years BC, was based on starch paste, which was applied to fabric strips. They were also able to make the first surgical bandages from the mixture of honey and fat [1, 2]. Over the centuries, the technology of obtaining adhesives did not develop significantly. Natural sticky substances were still used. The real evolution of pressure-sensitive adhesives has been developed through the use of polymers. Already in 1932, the first patent for adhesives based on acrylates was granted [3].

Pressure-sensitive adhesives are sticky substances that, when separated from the solvent, adhere to the surface in a dry state. They should be characterized by high internal strength (cohesion) as well as good adhesion to the surface (adhesion) and stickiness (tack). In addition to the above properties, high-quality self-adhesive materials are characterized by resistance to moisture, temperature and light as well as constancy of performance during application [4, 5].

The market for pressure-sensitive adhesives is growing every year. Researchers are constantly working on new products so that adhesives and self-adhesive materials made from them can replace traditional joining methods, such as welding or soldering. In 2006, the world market of PSA was worth USD 20.8 billion, while already in 2012, USD 22.7 billion. In 2012, 1700.5 kilotons were produced, while in 2018, it was to reach even 2208.2 kilotons. It is no wonder that more and more scientists and entrepreneurs are interested in the technology of obtaining adhesives and self-adhesive materials, taking into account the growing demand for these products and greater application possibilities [2, 6].

Currently, pressure-sensitive adhesives are used in practically all areas of life. In addition to obvious applications such as labels and office supplies, they also have many other interesting applications. They are used, for example, in construction as painting masking tapes and as protective foils for temporary protection of glazing, building elements and car parts. In such applications, it is important to be able to remove the tape without damaging the surface. Pressure-sensitive adhesives in the form of double-sided tapes and foams are also used in microelectronics as alternative products for conventional adhesives. The market for labeling and external applications has been practically dominated by pressure-sensitive adhesives. One cannot forget about the use of PSA in the medical devices industry, in the form of unilateral and double-sided tapes for medical devices, diagnostic bags and waste management. Bandages, attachments of skin electrodes, materials for closing wounds or surgical dressings are produced using self-adhesive tapes [7, 8].

Pressure-sensitive adhesives can be divided into a variety of ways. They could be divided because of their physical state: liquid and solid or because of the binding mechanism: physically and chemically. Physical binding can take place by

evaporation of the solvent (include water), and the chemical binding mechanism involves the formation of chemical bonds through polymerization, polyaddition and polycondensation. Pressure-sensitive adhesives should retain unchanging adhesive properties both at room temperature and in elevated temperature, which allows for further classification due to working conditions. Very often, dispersion, solvent and solvent-free adhesives are differentiated, that is, they are divided according to the process of their preparation. In dispersion adhesives, polymer particles are dispersed in water, and the curing mechanism may involve evaporation. Solvent adhesives are obtained in the presence of an organic solvent, which is used to regulate the viscosity and consistency of the adhesive and evaporates after applying the adhesive to the substrate. Solvent-free pressure-sensitive adhesives are obtained from solvent-based adhesives, after distilling off volatiles (mainly solvent) under reduced pressure [9, 10]. Very often, pressure-sensitive adhesives are divided due to the origin of the high-molecular compounds used in their production. The most important are acrylate, urethane, epoxy and silicone adhesives [11].

A very interesting segment of pressure-sensitive adhesives is silicone, due to their fascinating properties. They show low adhesion and exhibit low initial tack. In the industry, these adhesives already existed in the 1960s and their new properties are still being discovered [5, 12]. Due to their high thermal resistance, up to 300 °C, they have a very wide range of applications [13]. They are used for bonding metals and certain organic materials, for example, PTFE or polyolefins. They are very commonly used for the production of insulation of electric motors, generators, in the production of cars, as well as in the aerospace and shipbuilding industries [14].

A very important aspect in the technology of pressure-sensitive adhesives is also the cross-linking process. PSA based on silicones contains mainly dimethylsiloxyl and diphenylsiloxy groups. Organic peroxides, such as benzoyl peroxide (BPO) or dichlorobenzoyl peroxide, are usually used for cross-linking such adhesives. The degree of cross-linking of the adhesive can be controlled by the concentration of the cross-linker added [2, 6]. Si–O–Si silicon backbone shows high flexibility, which results in very good chemical resistance, thermal stability and resistance to atmospheric factors [15, 16]. In addition, silicone adhesives are characterized by low surface tension and high UV transparency. They are hydrophobic, but nevertheless they are distinguished by good moisture permeability [17, 18].

In the case of adhesives, very important property is resistance to external factors and aging. There are many different standards regarding resistance to external factors, such as humidity or elevated temperature. European norm EN 12024 deals with this issue. According to the principle of this method, the test consists in checking the adhesive tape's behavior at elevated temperature and high humidity. The measurement lasts for 7 days and takes place with the use of a climate chamber in which the elevated temperature (40 or 65 °C) and relative humidity of 85% are maintained [19].

An interesting aspect that scientists are dealing with is the shrinkage of pressure-sensitive adhesives. This is clearly visible when applying adhesive at elevated temperatures. Usually, it applies to self-adhesive tapes and is associated with its working time. Well, often at elevated temperatures, the tapes reduce their

dimensions. The test consists in coating pressure-sensitive adhesives on PVC or PET film and placing it in the chamber for 8 weeks at a temperature of 70 °C. The result of the test is a percentage or millimeter change in the dimensions of the film. In the technology of PSA, the shrinkage should not be greater than 0.5% or 0.5 mm [20, 21].

An important property for pressure-sensitive adhesives and materials made of them is yellowing. This is impractical for many applications and is associated with product degradation associated with chemicals or exposure to light. According to the PN-EN 1244: 2000 standard regarding the color determination and/or color changes in the light-colored adhesive coatings, the color change in the test sample from clear to white to yellow can be examined [22–24].

Silicone adhesives resistant to atmospheric agents mainly high temperatures using an elastomeric polyethylene coating are known. However, they are hardly wettable, which makes them difficult to use [25]. They are also widespread laminated structures including primed fluorinated polyethylene plastics and modifications of this method by prior application of an undercoat of elastomer, to halogenated polyethylene preheated to the melting point of the plastic base [26].

This article deals with the investigation of the influence of environmental conditions and aging time of pressure-sensitive adhesives on the example of silicone pressure-sensitive adhesives. Various self-adhesive properties were tested, such as adhesion, cohesion, tack, shrinkage, maximum operating temperature, depending on the concentration of the cross-linking agent used and the type of manufacturer. The yellowness of the yellowing index was also examined. All examinations were performed after 1 day and 6 month. The contraction test, however, required more accurate observation, and it was performed more often, up to 7 days (stabilization time of shrinkage) and after 6 month. In addition, the process energy and its maximum temperature were characterized by differential scanning calorimetry (DSC). Thanks to the study, it was possible to examine the cross-linking process of silicone pressure-sensitive adhesives included in the work. To test the applicability of the adhesives obtained, flammability tests were performed, determining the rate of smoke and heat release (according to ISO 5660-1: 2015) and the heat of combustion of selected Si-PSA in accordance with PN-EN 1716: 2010.

The tests described in the article are carried out in accordance with PN and EN: ISO standards and procedures, as well as taking into account specialized standards. The organization dealing with the determination of self-adhesive materials' testing methods is FINAT (international organization of producers associated with the production and use of self-adhesive labels). As many as, 31 research methods are recommended by FINAT, and their number is constantly increasing every year, which is the result of the progress of the adhesive industry. The organization's research methods are identified by the letters FTM (from the English term FINAT Test Method) and the numbers 1–30. Usually, recipients of adhesive materials receive data on tests carried out in accordance with FTM 1-4, FTM 8-9 and FTM 12-13. Other research is often included in the so-called industry research, and generally, they are widely available only to a few important clients [3, 4]. In addition to the FINAT organization, AFERA (Association des Fabricants Européens de Rubans Auto-Adhésifs) standards are also used to test adhesive products. These standards

describe the methodology for performing basic tests, e.g., cohesive: AFERA 4012, tack: AFERA 4015 and peel adhesion: AFERA 4001 [27, 28].

When testing self-adhesive properties, both ISO standards and specialized standards (AFERA, FINAT FTM) are taken into account, so as to test the application properties of the tapes obtained.

Experimental

Materials

In the presented paper, commercial silicone adhesives products by Momentive (USA) and Dow Corning (USA) PSA 529, PSA 590 and 282, Q2-7566 were used. Dichlorobenzoyl peroxide (DClBPO) from Peroxid-Chemie (Germany) was used as a cross-linking agent.

Preparation of silicone pressure-sensitive adhesives tape

The silicone pressure-sensitive adhesive and the cross-linking agent (2,4-dichlorobenzoyl peroxide (DClBPO) according to polymer content) were mixed to obtain a homogenous organic composition containing 50 wt% polymer according to Table 1. Subsequently, the PSA was coated with a coating speed of 5 cm/s on a 50-µm-thick polyester film and dried for 10 min at 110 °C in drying channel. The self-adhesive layers prepared in this way were secured on a 36-µm-thick polyester film yielding one-side tapes. These were used for further tests, i.e., adhesion, cohesion and tack. In order to study the aging and impact of environmental conditions process on silicone pressure-sensitive adhesives, all measurements were carried out after 6 months taking into account references kept in good storage conditions.

Methods

Pot life of silicone pressure-sensitive adhesives compositions was measurement as a difference viscosity at time. Non-cross-linked adhesive compositions were tested at an interval of 1 and 90 days from receipt of the composition. Each time viscosity of silicone pressure-sensitive adhesives composition was determined with a RM 180 rotational viscometer from Rheometric Scientific, with spindle No

Table 1 Prepared Si-PSA composition

Silicone acronym	Company	Cross-linking agent (wt%)
282	Dow Corning	2.5
PSA 529	Momentive	2.5
PSA 590	Momentive	1.5
Q2-7566	Dow Corning	1.5

3 at temperature room. As you know, pressure-sensitive adhesives should maintain a relatively constant viscosity so that it can easily be applied to the bonded substrate [29].

Reactivity studies between the cross-linking compound and silicone adhesive during the cross-linking process were carried out for selected adhesive compositions. Thermal effects of the cross-linking process at elevated temperature were tested using differential scanning calorimetry (DSC) on DSC from TA Instruments model Q 100. The measurement was carried out in a cycle heating–cooling–heating in the temperature range from − 90 to 250 °C, i.e., at standard operating temperatures of adhesives and self-adhesive materials. By using the above method, it was possible to characterize the cross-linking process of the obtained adhesives [30].

The value of peel adhesion of Si-PSA was measured in accordance with the standard 4001 developed by Association des Fabricants Européens de Rubans Auto-Adhésifs (AFERA) standard using test machine Zwick/Roell Z010 (Zwick/Roell, Germany). Strip of Si-PSA tape with a width of 2.5 cm (ca. 1 in) and length of about 17.5 cm (ca. 7 in) has been applied on a degreased steel plate horizontally providing at least 15 cm^2 contact of the plate with the tape. After applying to the plate, the contact surface was tightened with a 2-kg rubber roller to improve wettability of the tape. Thereafter, plate was mounted in the lower jaws so that the jaws formed a 90° angle with the sample and so that the not applied end was on the bottom. Not attached end of strip was mounted in upper jaws of the test machine. Lower jaws of test machine remained motionless. Upper jaws were moving at a constant speed of 300 mm/min during the test. The final result of the test was the value of the average force needed to tear off the belt from the metal plate. The values presented in the article are the average of three measurements [31].

The value of tack of Si-PSA was measured in accordance with the standard 4015 developed by Association des Fabricants Européens de Rubans Auto-Adhésifs (AFERA) standard using test machine Zwick/Roell Z010 (Zwick/Roell, Germany). A one-sided pressure-sensitive adhesive film with dimensions 175 × 25 mm was placed in the upper jaws with an adhesive side to the outside, so as to form a loop. The sample is dropped at a speed of 100 mm/min onto a steel plate placed at an angle of 90° horizontally in the machine's lower jaws. The contact surface was ca 5 cm^2 (2.5 cm × 2 cm). After the moment of contact of the strip with the plate without the participation of external forces, the tape is torn off. The result of the test is the value of the force required to detach the tape from the plate [32, 33].

The shear strength (cohesion) of Si-PSA was measured in accordance with the standard 4012 developed by Association des Fabricants Européens de Rubans Auto-Adhésifs (AFERA) standard using test machine designed at The International Laboratory for Adhesive and Self-Adhesive Materials of the West Pomeranian University of Technology in Szczecin. A one-sided adhesive film was applied to a degreased steel plate and pressed with a 2 kg rubber roller. The Si-PSA film was adhered so that contact surface was 6.25 cm^2 (2.5 cm × 2.5 cm). The plate was placed in the test machine and loaded with a 1 kg weight, such that the angle of force at the junction was 180°. The test machine automatically measured the time at which the sample of the adhesive tape would detach from the metal plate. The cohesion value was measured at 20 °C and 70 °C [32].

The value of shrinkage was measured using the cross-method. The PVC or PET foil was coated with a Si-PSA layer and cross-linked. Then, it was applied on a degreased metal plate and two cuts were made at the right angle. The width of the cuts was measured. The second measurement was made after storage of the plate for 8 weeks at 70 °C. The shrinkage is the percentage value of the ratio of width of the cuts. Shrinkage greater than 0.5% is not allowed [1, 20, 21, 34, 35].

Yellowing is one of the undesirable characteristics of pressure-sensitive adhesives. Due to aesthetic reasons, this limits the possibility of their use in many applications and is unacceptable for the consumer. The yellowness index (YI) is a measure of yellowing, which is defined as the color change in the material under the influence of atmospheric factors (natural or elevated). The reason for the yellowing of materials is the action of harmful factors such as light, treatment process or chemicals. It indicates the general degradation of the material or contamination. The most common apparatus for measuring the yellowness index is a spectrophotometer. A 3 NH NR20XE colorimeter was used to measure the value of YI. Color change from transparent or white to yellow read through spectrophotometric measurements meant the value of the yellowness index. This test was used to assess the color change in the adhesive layer caused by external factors [36–40].

Flammability tests were carried out for selected Si-PSAs, determining the rate of heat and smoke release using a cone calorimeter according to standard ISO 5660-1:2015 (fire test, reaction to fire, heat release, smoke release and weight loss rate). The study of the rate of heat and smoke emitted from samples in a cone calorimeter is based on the measurement of oxygen consumed, necessary for the combustion process. As a result of supported observations, it is assumed that for each 1 kg of oxygen consumed 13.1 MJ of energy is obtained. 10 cm × 10 cm horizontally arranged samples were used for the test, which were heated with a 30 kW/m^2 heat stream emitted from the conical radiator, thus imitating the first phase of fire development. The gases emitted during the decomposition of the samples ignited through the igniter. Then, the exhaust gases as a result of the flow forced by the fan are collected by the hood and flow through the chimney, where they are taken by the gas sampling probe and subjected to analysis [41].

The heat of combustion of selected Si-PSAs according to standard PN-EN 1716:2010 (reaction to fire test for construction products and determination of the heat of combustion) was also determined. Determination of heat combusting consisted of burning a given sample of adhesives film in a calorimetric bomb of a certain volume, in an atmosphere of oxygen under pressure, and measuring the increase in water temperature in the calorimetric vessel. The ignition stimulus was a glowing ignition wire [42].

Thermal silicone pressure-sensitive adhesives composition stability was assessed by thermogravimetry (TGA) using TA Instruments Inc. model 2950 TGA unit interfaced with the TA Instruments Thermal Analyst 2100 control unit. All samples about ca. 10 mg were contained in a platinum sample pan, and the TGA cell was swept with oxygen at 60 ml/min during degradation process. The temperature was ramped at 5 °C/min [43].

To investigate the impact of environmental conditions samples (tapes and joint tapes with a steel substrate), they were conditioned in a climatic chamber simulating

Table 2 Cycles of the chamber simulating the environmental conditions

Cycle	Moisture (rh)	Temperature (70 °C)
I	0	− 20.0
II	95	35.5

Table 3 Pot-life and exothermic cross-linking measurements of silicone pressure-sensitive adhesives composition

Composition	Viscosity (mPa s)		Exothermic energy (J/g)	Maximum temp. (°C)
	1 day	90 days		
282	32,250	32,250	1.51	121.3
PSA 529	2500	2500	2.48	120.9
PSA 590	8340	8340	2.31	119.4
Q2-7566	22,250	22,250	2.47	120.6

environmental conditions LHU-114 (ESPEC Corporation, Japan) according to the modified simulation of the MMM-A-132B standard. Samples were held in variable conditions in 24 h cycles simulating negative environmental conditions taking into account temperature and humidity (Table 2). In order to better investigate the impact of environmental conditions, tests were carried out both on the tapes and joint tapes with a steel substrate for adhesion and cohesion measurements which were subjected to moisture and temperature conditions. In the case of tack, simulation was carried out for tape. For all measurements, references were made—samples kept in a climatic chamber to avoid aging processes and the impact of the environment on the tested properties. The simulation was carried out for 20 days (10 days per cycle).

Results and discussion

The first stage of the study was to produce single-sided self-adhesive tapes to obtain the material for testing. Selected compositions were characterized by constant viscosity (pot-life study—90 days); the cross-linking process was confirmed by DSC testing where exothermic energy of the process and its maximum temperature were determined The peak associated with the cross-linked process was visible only at the first heating, which confirms the cross-linking of the adhesive films (Table 3).

To better characterize the adhesive tapes obtained, flammability, combustion and thermogravimetric tests were carried out. The highest temperature of the onset of thermal decomposition was obtained for 282 adhesives composition (337 °C) and the lowest for PSA 590 adhesives (254 °C). The temperatures of the beginning of thermal decomposition of the remaining adhesives did not differ significantly. None of the tested materials was decomposed in half the weight of tested samples. The 282 composition burned in the largest part—50.23% of the residue remained, and in the smallest part PSA 590—59.29% of the residue remained. The temperature at the beginning of thermal decomposition decreased

Table 4 Results of thermogravimetric studies of the selected Si-PSA compositions

Adhesives composition	Temperature of start thermal decomposition (°C)	Maximum temperature of the weight loss rate (°C)	Maximum mass loss rate (%/min)	Mass of residue (%)
282	337	469	10.8	50.2
Q2-7566	289	455	3.1	55.2
PSA 529	284	480	3.9	58.1
PSA 590	254	486	3.2	59.3

Table 5 Results of testing the selected Si-PSA compositions using a conical calorimeter

Adhesives composition	282	Q2-7566	PSA 529	PSA 590
HRR_{max} (kW/m^2)	101	119	139	76
MAHRE (kW/m^2)	38	13	62	14
THR (kW/m^2)	1.5	1.8	2.1	1.6
The total amount of smoke (m^3)	0.8	1.1	0.8	0.7
CO emissions (mg/g)	21.8	17.7	19.3	21.4
CO_2 emissions (mg/g)	118.3	98.1	109.4	116.8
Ignition time (t_{ig},s)	16	22	36	20

with the increase in the amount of residue after analysis, for samples without filler. The faster the onset of thermal decomposition, the less the glue was burned. This could indicate the earlier formation of a coating insulating the combustible material in the form of a layer of porous coke and thus a smaller amount of burned material. The results of thermogravimetric tests are collected in Table 4.

Table 5 shows the results of testing adhesive compositions using a cone calorimeter. The highest heat excretion rate (HRR_{max}) and average heat excretion rate (MAHRE) as well as the amount of heat excreted (THR) were found in the composition of PSA 529, while the lowest were found in composition Q2-7566 for HRR_{max} and THR and the value of MAHRE was similar to the composition of PSA 590. This value significantly affected the development of fire. Considering the measurement of the total amount of smoke (m^3), the most smoke 1.1 m^3 was emitted by the Si-PSA Q2-7566 composition, and the remaining ones resulted in almost identical amounts of 0.7–0.8 m^3. The highest average CO and CO_2 emissions were observed for composition 282, and slightly lower for PSA 529 compositions, which contained a higher amount of DClBPO compared to other compositions.

The results of the heat of combustion study are shown in Fig. 1. The lowest average value of the heat of combustion was obtained for the composition Si-PSA 4 (18.0 kJ/g), and the highest value was obtained for the composition PSA 529 (19.3 kJ/g). A similar relationship was obtained in the cone calorimeter test by analyzing the total heat released from the burned samples (THR). The differences in the obtained values were relatively small and did not exceed 1.5 kJ/g (Fig. 1).

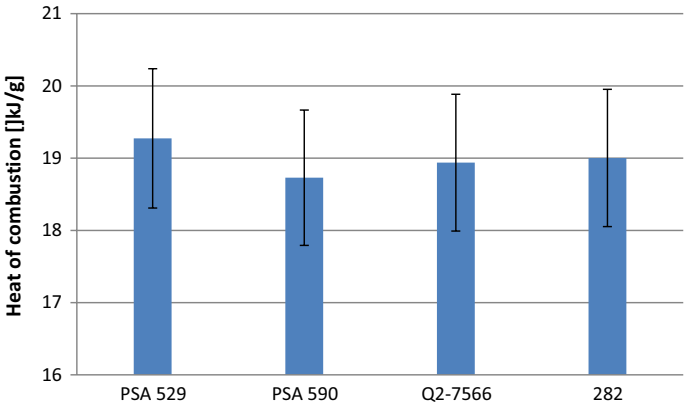

Fig. 1 Combustion heat of the selected Si-PSA composition

To define the impact of environmental conditions, the best physical properties were selected, such as tack, adhesion, shrinkage, shear strength at different temperatures, maximum work temperature and yellowness index. The results of self-adhesive tape tests, immediately after receipt, are presented in Table 6 [15, 32, 44]. All adhesives are characterized by good self-adhesive and functional properties.

The adhesive from which the material is made influences the aging of the adhesive materials to a large extent. It is known from the literature reports that aging usually causes a decrease in the properties of pressure-sensitive adhesives [44, 45].

Table 7 presents the results of the tested properties for the self-adhesive tapes obtained, after aging tests, lasting 6 months. This test was performed under conditions stimulating the storage of self-adhesive tapes, i.e., they were exposed to changing environmental conditions, similar to those resulting from inadequate storage (changes in humidity, temperature and insolation). Adhesives that do not contain unsaturated bonds in their structure show higher resistance to aging and thermal stability. It is known from the literature that the aging of the adhesive affects the self-adhesive properties of silicone tapes, and this usually appears in the fall of adhesion and tack [44, 47] like similarly in this case. In addition, an increase in yellowness

Table 6 Useful typical properties of silicone pressure-sensitive adhesives tape

Sample	Shrinkage[a] (mm)	Adhesion (N/25 mm)	Cohesion (h)		Maximum work temperature (°C)	YI	Tack (N)
			20 °C	70 °C			
282	0.40	12.2	> 72	> 72	108.8	0.345	8.4
PSA 529	0.35	15.4	> 72	> 72	110.1	0.652	8.2
PSA 590	0.35	13.7	> 72	> 72	154.2	0.526	8.3
Q2-7566	0.40	12.5	> 72	> 72	133.2	0.330	9.6

[a]In the case of contraction of pressure-sensitive adhesives, the tests were carried out until day 7 when samples stabilized

Table 7 Influence of time on the useful properties of silicone pressure-sensitive adhesive after 6 months

Sample	Shrinkage (mm)	Adhesion (N/25 mm)	Cohesion (h)		YI	Tack (N)
			20 °C	70 °C		
282	0 ↔	3.6 ↓	0 ↔	0 ↔	3.230 ↑	1.2 ↓
PSA 529	0 ↔	4.5 ↓	0 ↔	0 ↔	3.042 ↑	0.5 ↓
PSA 590	0 ↔	3.7 ↓	0 ↔	0 ↔	3.017 ↑	0.3 ↓
Q2-7566	0 ↔	3.2 ↓	0 ↔	0 ↔	3.141 ↑	1.7 ↓

↑—increased; ↓—decreased; ↔—no change; the data presented are the difference value from the data collected in Table 2

Table 8 Influence simulation environmental conditions on cohesion silicone pressure-sensitive adhesive in room temperature

Sample	Cohesion (h)			
	Tape		Joint	
	Simulation	Reference[a]	Simulation	Reference[a]
282	> 72	> 72	> 72	> 72
PSA 529	> 72	> 72	> 72	> 72
PSA 590	> 72	> 72	> 72	> 72
Q2-7566	> 72	> 72	> 72	> 72

[a]Sample (tape or joint) kept in a climatic chamber to avoid aging

Table 9 Influence simulation environmental conditions on cohesion silicone pressure-sensitive adhesive at 70 °C

Sample	Cohesion (h)			
	Tape		Joint	
	Simulation	Reference[a]	Simulation	Reference[a]
282	6.5	2.0	6.5	2.0
PSA 529	> 72	5.5	6.5	> 72
PSA 590	6.5	> 72	> 72	5.5
Q2-7566	> 72	> 72	> 72	> 72

[a]Sample (tape or joint) kept in a climatic chamber to avoid aging

was observed for all tested samples, which is not a positive aspect in the perspective of the implementation of such a product. Shrinkage of pressure-sensitive adhesives was characterized by complete resistance to aging. The longer storage time of this type of adhesives does not affect the above parameter. The same applies to internal strength (cohesion). These tests are consistent with the previous article, in which the measurements were carried out in a shorter time [44]. As results from the conducted research, silicone pressure-sensitive adhesives show high resistance to aging in conditions of inadequate storage, maintaining stable self-adhesive properties.

Cohesion values at room temperature and elevated for the tapes obtained in the conducted simulation showed no effect on changing environmental conditions (Tables 8, 9). Table 8 presents the cohesion results for metal plates connected with

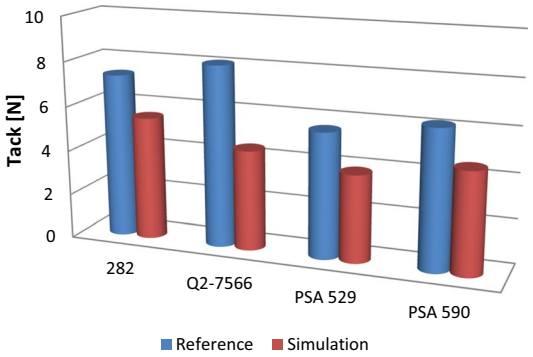

Fig. 2 Influence simulation environmental conditions on tack silicone pressure-sensitive adhesive

the produced strip. Silicone pressure-sensitive adhesives based on PSA 529 and 282 resin showed a decrease in cohesion at elevated temperatures (Table 9). This may be due to the relatively lower resistance to high temperatures compared to other samples tested, which may be more susceptible to lowering immunity caused by external factors [45].

Cohesion tests confirmed the influence of environmental conditions on self-adhesive tapes and their adhesive joints. The next measurement was tack (Fig. 2). All tested silicone bands were characterized by a relatively high tray, for reference samples (stored under storage conditions, at the same time as the samples subjected to stimulation). However, in the case of materials present in the climatic chamber, the tack is lower. The biggest decrease (almost by half the value) was characterized by resin Q2-7566. This may be due to the extreme conditions prevailing in the climatic chamber on which the tape samples are not completely resistant [46].

In the tack and cohesion results, small changes in the values for the reference and stimulated samples were noticed. In the case of adhesion, these changes are also visible (Fig. 3). In the case of self-adhesive tapes based on resin adhesive 282 and

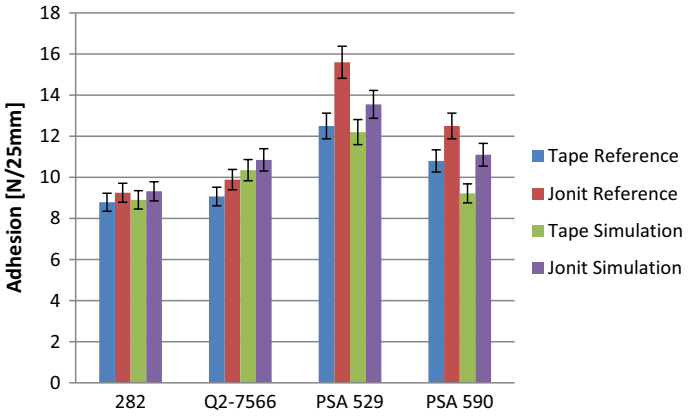

Fig. 3 Influence simulation environmental conditions on adhesion silicone pressure-sensitive adhesive

PSA 529, no environmental impact on adhesion is observed. On the other hand, the adhesive containing the Q2-7566 resin improved its adhesion value, which shows that it has a higher adhesion to the substrate. This may be due to the cross-linking of the adhesive at elevated temperature, which results in an improvement of this value [47]. The results for the PSA 590 resin adhesive in turn show the negative impact of the presence of the samples in the climatic chamber. These tapes were characterized by inferior surface adhesion, like the reference sample. This may be due to the lack of resistance of the sample to the conditions prevailing in the chamber [46].

The adhesive joints showed practically in all cases higher adhesion values compared to self-adhesive tapes. Adhesion of adhesive joints for adhesives 282 and Q2-7566 only shows slight differences for the reference joints and those tested in the chamber. On the other hand, in the case of silicone adhesives based on resin 529 and 590, a decrease in adhesion to the surface after the sample in the chamber is observed. Considering the conditions present in the chamber, such results were expected. The adhesive joint, for a long period of time, was exposed to variable humidity conditions, or high insolation, and this could cause a decrease in this self-adhesive property [46].

Conclusions

The described work presents the impact of environmental conditions on self-adhesive tapes and adhesive joints (application of the obtained tapes). It is generally accepted that one-sided adhesive tape must meet basic properties after production like adhesion > 10 N/25 mm, tack > 8 N and cohesion > 72 h [15]. For the composition of self-adhesive Si-PSA adhesives, during the storage and holding of self-adhesive tapes, small deviations for adhesion and stickiness are acceptable from general accepted standards due to the specificity of this group of adhesives. The compositions tested showed excellent properties and very good stability in time (aging resistance). Longer study (6 month) confirmed aging effect of the Si-PSA which exhibit low adhesion and less tack; slightly increasing yellowness index; increasing shrinkage within the first few days and after that time stabilization occurred described in the article [44]. There was no effect of aging on the cohesion of adhesives films at 20 °C and 70 °C. Impact of environmental conditions (changes in temperature and humidity) degraded the properties of self-adhesive tapes and significantly affected the tape connections with the steel substrate. Tapes having been exposed to a simulation environmental condition still showed comparatively good performance considering that the storage conditions were not intended for storing tapes-based Si-PSA. In addition, flammability and combustion tests were carried out to check their applicability as a coating to isolate combustible material. As a result of the tests, it was possible to obtain a tape with a thermal decomposition temperature over 330 °C, which is a very good result compared to other self-adhesive materials.

References

1. Czech Z, Kowalczyk A, Świderska J (2011) Pressure-sensitive adhesives for medical applications. Wide Spectra Qual Control 17:309–332. https://doi.org/10.15406/japlr.2018.07.00251
2. Antosik AK, Czech Z (2016) Pressure-sensitive adhesives (PSA) based on silicone. Adv Mater Interfaces 7:249–274. https://doi.org/10.1002/9781119242604.ch7
3. Czech Z (2002) Sieciowanie samoprzylepnych poliakrylanowych wodnych klejów dyspersyjnych. Polimery 47:234–240
4. Monte S (2009) The adhesive advantage: strong joints, low costs. Mach Des 20:42–44
5. Czech Z, Kowalczyk A, Madejska E, Głuch U, Ortyl J (2011) Adhesion of pressure-sensitive adhesives (PSAs). Coating 5:32–35
6. Czech Z, Milker R (2005) Development trends in pressure-sensitive adhesive systems. Mater Sci Poland 23:1015–1022
7. Basedow A, Kura F, Seeger K (1998) C.A. Patent No. 2260844 A1. Lohmann KG
8. Tan H, Pfister W (1999) Pressure-sensitive adhesives for transdermal drug delivery systems. Pharm Sci Technol Today 2:60–69
9. Polkowski M (2015) Types of adhesives used in the printing industry. Acta Poligraphica 6:19–28
10. Antosik AK, Czech Z (2015) Klasyfikacja klejów samoprzylepnych ze względu na proces otrzymywania. Wiad Chem 69:111–119
11. Creton C (2003) Pressure-sensitive adhesives: an introductory course. MRS Bull 28(6):434–439. https://doi.org/10.1557/mrs2003.124
12. Fischer SCL, Kruttwig K, Bandmann V, Hensel R, Arzt E (2017) Adhesion and cellular compatibility of silicone-based skin adhesives. Macromol Mater Eng. https://doi.org/10.1002/mame.201600526
13. Lin SB (1996) Silicone pressure-sensitive adhesives with selective adhesion characteristics. J Adhes Sci Technol 10(6):559–571. https://doi.org/10.1163/156856196X00580
14. Min H, Qiuyu Z, Jiying G (2011) Synthesis and characterization of silicone based pressure sensitive adhesive. Adv Mater Res 306–307:1773–1778. https://doi.org/10.4028/www.scientific.net/AMR.306-307.1773
15. Antosik AK, Czech Z (2016) Double-side tape based on silicone pressure-sensitive adhesive. Chemik 70:369–374
16. Zhang MY, Wang GH, Yu P, Wang Y (2012) The study of high heat resistance of nano-modified silicone pressure sensitive adhesive. Adv Mater Res 403–408:1106–1112. https://doi.org/10.4028/www.scientific.net/AMR.403-408.1106
17. Mecham S, Sentman A, Sambasivam M (2010) Amphiphilic silicone copolymers for pressure sensitive adhesive applications. J Appl Polym Sci 116:3265–3270. https://doi.org/10.1002/app.31752
18. Sun F, Hu Y, Du H-G (2012) Synthesis and characterization of MQ silicone resins. J Appl Polym Sci 125:3532–3536. https://doi.org/10.1002/app.35194
19. PN-EN 12024:1996 E Taśmy samoprzylepne-pomiar odporności na podwyższoną temperaturę i wilgotność
20. Czech Z (2004) Development in the area of UV-crosslinkable solvent-based pressure-sensitive adhesives with excellent shrinkage resistance. Euro Polym J 40:2221–2227. https://doi.org/10.1016/j.eurpolymj.2004.05.012
21. Singh AK, Niyogi UK, Sabharwal S, Kowalczyk A, Czech Z, Mehra DS (2013) Shrinkage studies in electron beam curable polyurethane pressure-sensitive adhesive. J Adhes Sci Technol 27:1511–1524. https://doi.org/10.1080/01694243.2012.746160
22. PN-EN 1244:2000 Oznaczanie barwy i/lub zmian barwy powłok klejowych pod wpływem kleju
23. Grzesiak E, Woźniak B (2005) Wpływ środków pomocniczych na zmiany barwy materiałów obuwniczych i metody obiektywnej oceny zmian. Barwniki. Środki Pomocnicze 3:113–125
24. PN-EN. 12705:2002 Kleje do skóry i materiałów obuwniczych. Oznaczanie zmian barwy powierzchni skór białych i jasnych wskutek migracji
25. Krohn RC (2000) C.A. Patent No. 2396978C. Allied Photochemical Inc
26. Chu WK (2000) US Patent No. 6482878B1. Tarrytown, Henkel IP and Holding GmbH
27. Benedek I (2006) Pressure—sensitive design and formulation, application, theoretical aspects. CRC Press, Boston
28. Jack A (2001) FINAT technical handbook 6th edition, Haga

29. Czech Z, Milker R, Butwin A, Herko E (2008) Removal of organic solvents for the purpose of manufacturing of solventfree pressure-sensitive adhesives. Pol J Chem Technol 10(1):37–40
30. Lu D, Wong CP (2000) Effects of shrinkage on conductivity of isotropic conductive adhesives. Int J Adhes Adhes 20(3):189–193
31. Wilpiszewska K, Czech Z (2014) Citric acid modified potato starch films containing microcrystalline cellulose reinforcement—properties and application. Starch 66:660–667. https://doi.org/10.1002/star.201300093
32. Antosik AK, Czech Z (2015) Effect of the dye amount on physical properties of silicone pressure sensitive adhesives. Przem Chem 94:41–42. https://doi.org/10.15199/62.2015.1.3
33. Antosik AK, Bednarczyk P, Czech Z (2015) Jednostronnie klejące taśmy na bazie samoprzylepnych klejów silikonowych-dobór najlepszej kompozycji. Chemik 69:95–97
34. Czech Z, Kowalczyk A, Kabatc J, Świderska J (2012) Photoreactive UV-crosslinkable solvent-free acrylic pressuresensitive adhesives containing copolymerizable photoinitiators based on benzophenones. Euro Polym J 48:1446–1454. https://doi.org/10.1016/j.eurpolymj.2012.05.010
35. Czech Z, Kowalczyk A, Sowa D, Kowalska J, Ragańska P (2013) Copolymerizable photoinitiators based on benzophenones and their influence on shrinkage of photoreactive acrylic PSAs. Int Lett Chem Phys Astron 8:49–56
36. Garcia-Castaneda C, Benavides R, Martinez-Pardo ME (2010) Crosslinking of rigid PVC by ionizing radiation to improve its thermal properties. Rad Phys Chem 79:335–338. https://doi.org/10.1016/j.radphyschem.2009.08.004
37. Matuana LM (2009) Rigid PVC/(layered silicate) nanocomposites produced through a novel melt-blending approach. J Vinyl Addit Technol 15:77–87. https://doi.org/10.1002/vnl.20183
38. Pimentel Real LE, Ferraria AM, Botelho do Rego AM (2008) Comparison of different photo-oxidation conditions of poly(vinyl chloride) for outdoor applications. Polym Test 27:743–751. https://doi.org/10.1016/j.polymertesting.2008.05.009
39. Sterky K, Hjertberg T, Jacobsen H (2009) Effect of montmorillonite treatment on the thermal stability of poly(vinyl chloride) nanocomposites. Polym Deg Stab 94:1564–1570. https://doi.org/10.1016/j.polymdegradstab.2009.04.036
40. Strait CA, Tabor RL, Lancaster GM (1990) US Patent No. 07229078. U.S. Patent and Trademark Office, Washington, DC
41. ISO 5660-1:2015, Badanie ogniowe. Reakcja na ogień. Wydzielanie ciepła, wydzielanie dymu i szybkość ubytku masy
42. PN-EN 1716:2010, Badanie reakcji na ogień wyrobów budowlanych. Określenie ciepła spalania
43. PN-EN ISO 11358-1:2014, Tworzywa sztuczne. Termograwimetria (TG) polimerów
44. Antosik AK, Bednarczyk P, Czech Z (2018) Aging of silicone pressure-sensitive adhesives. Polym Bull 75:1141–1147. https://doi.org/10.1007/s00289-017-2083-2
45. Czech Z, Kowalczyk A (2014) Fotoreaktywne termotopliwe kleje samoprzylepne na bazie poliakrylanów. Chemik 68:53–60
46. Shuangyan Xu, Dillard DA, Dillard JG (2003) Environmental aging effects on the durability of electrically conductive adhesive joints. Int J Adhes Adhes 23:235–250. https://doi.org/10.1016/S0143-7496(03)00027-7
47. Zhou Z, Engle LP (2001) US Patent No. 6777079B2. 3M Innovative Properties Company, St. Paul

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