

Original Research

The Effect of the Addition of Titanium Dioxide on Silicone Self-Adhesive Tapes

Adrian Krzysztof Antosik ^{†,*}, Karolina Mozelewska [†], Marcin Bartkowiak [†]

Department of Chemical Organic Technology and Polymeric Materials, Faculty of Chemical Technology and Engineering, West Pomeranian University of Technology in Szczecin, Piastów Ave. 42, 71-065 Szczecin, Poland; E-Mails: adriankrzysztofantosik@gmail.com; karolina_mozelewska@zut.edu.pl; mbartkowiak@zut.edu.pl

[†] These authors contributed equally to this work.

* **Correspondence:** Adrian Krzysztof Antosik; E-Mail: adriankrzysztofantosik@gmail.com

Academic Editor: Fabian Ifeanyichukwu Ezema

Journal of Energy and Power Technology
2024, volume 6, issue 2
doi:10.21926/jept.2402012

Received: February 29, 2024
Accepted: May 07, 2024
Published: May 13, 2024

Abstract

The investigation aimed to delve into the intricate relationship between the presence of titanium dioxide and the resulting alterations in the adhesive's behavior under different conditions. The focus extended beyond mere observation to understand how these additives interact with the adhesive matrix, potentially imparting beneficial attributes. Notably, the study delved into the optimization of thermal stability, the extension of the adhesive's functional lifespan, and the mitigation of shrinkage tendencies. This multi-faceted approach involved comprehensive testing, analysis, and comparison of adhesive samples containing varying additions of titanium dioxide.

Keywords

Titanium dioxide; self-adhesive tapes; silicone pressure-sensitive adhesives; self-adhesive properties; adhesion



© 2024 by the author. This is an open access article distributed under the conditions of the [Creative Commons by Attribution License](https://creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium or format, provided the original work is correctly cited.

1. Introduction

The popularity of pressure-sensitive adhesives is rising due to their exceptional properties, including robust resistance to aging, easy removability from surfaces, and strong adhesion to polar substrates. Classified as a specialized category of materials, pressure-sensitive adhesives display significant adhesive forces and tack upon contact with a substrate, all without requiring a chemical reaction. Their significance in daily life cannot be understated. Over the years, researchers have been diligently working to supplant traditional joining techniques like welding and riveting with adhesives and adhesive tapes, leading to continuous advancements in this industry sector [1].

When applied as a thin layer on a carrier material (such as paper, foil, or fabric), pressure-sensitive adhesives exhibit remarkable longevity and minimal aging. They are typically applied in liquid form onto rollers and subsequently undergo solvent evaporation (in the case of solvent-based adhesives) or cooling (in the case of solvent-free adhesives) [2, 3].

In the present day, pressure-sensitive adhesives find applications across various domains. Beyond their evident uses in labeling and office supplies, they have also found intriguing applications in various other sectors. For instance, in the construction field, they serve as masking tapes and protective films to temporarily safeguard glass surfaces, building components, and automotive parts. In such applications, the ability to remove them without causing damage to the underlying surface is crucial. Pressure-sensitive adhesives, like double-sided tapes and foams, are gaining ground in microelectronics as alternatives to conventional adhesives. PSA has virtually dominated the labeling and outdoor applications market. Furthermore, their role in the medical devices industry is noteworthy, with applications ranging from one-sided and double-sided tapes for medical devices to diagnostic bags and waste management solutions. Adhesive tapes manufacture bandages, skin-electrode attachments, wound closures, and surgical dressings [4].

Due to their distinctive chemical composition, silicone adhesives stand apart from other organic polymer adhesives. The foundational structure of a silicone adhesive is primarily built upon a polysiloxane framework featuring silicon-oxygen bonds. The energy associated with such bonds considerably surpasses that of C-C bonds. Consequently, silicones manifest unparalleled attributes in terms of resilience and stability. Moreover, they possess remarkable chain flexibility due to their extended bond length and adaptable bond angle. Methyl groups are affixed to silicon atoms, offering unimpeded steric hindrance and unrestricted rotational motion. This dynamic configuration shields the primary polar -Si-O-Si- chain, permitting only limited intra- and intermolecular interactions. As a result, chemical cross-linking with a high cross-link density becomes necessary to optimize mechanical properties within polysiloxane chains [5].

The debut of silicone adhesives in the market dates back to the 1960s, and since then, they have found application across diverse industries. These adhesives have proven instrumental in various industrial tasks encompassing masking, wrapping, and bonding while also playing crucial roles in electrical engineering, electronics, healthcare, medicine, and the automotive industry. In the realm of silicone pressure-sensitive adhesives, the post-2000 era has witnessed significant exploration into novel applications, particularly in the fields of medical and industrial tapes. Conventional silicone pressure-sensitive adhesives are predominantly solvent-based. However, a mounting imperative exists to curtail VOC (Volatile Organic Compound) emissions from tape and label manufacturing processes. As a response, companies are actively pursuing the introduction of high-solids pressure-sensitive adhesives or hot-melt variants to diminish or eliminate VOC emissions. Furthermore, a

trajectory towards more energy-efficient solutions is evident in the market. Notably, there's a heightened emphasis on advancing addition-cure pressure-sensitive adhesives capable of curing around 100°C, thereby reducing the typical cross-linkers requirement of 150°C [5, 6].

The study employed silicone-based adhesives, typically composed of silicone polymers containing silane groups and siloxane resins. These materials are tailored for specialized applications, exhibiting remarkable performance across various temperatures, exceptional electrical characteristics, and robust resistance to chemicals and weathering. Silicone-based pressure-sensitive adhesives are distinguished by their pronounced self-adhesive attributes, although their thermal resistance often tends to be relatively modest. A similar scenario unfolds about pot life, denoting the viscosity variation over time. Unmodified silicone adhesives possess a limited pot life, rendering them suitable solely for immediate coating after mixing. Furthermore, the shrinkage exhibited by silicone pressure-sensitive adhesives is notably elevated, exceeding 1%, which proves unsuitable for the production of pressure-sensitive adhesive tapes and adhesives. The primary objective of this study was to ascertain the impact of diverse silicone additives on the characteristics of pressure-sensitive adhesives. The investigation was aimed at enhancing properties such as thermal stability, pot life, and shrinkage [5].

The fundamental objective of the study was to examine how the addition of titanium dioxide and its quantity influence the characteristics of self-adhesive adhesives. The research aimed at enhancing properties such as thermal stability, longevity, and shrinkage. By investigating the impact of titanium dioxide additives and varying concentrations, the study sought insights into the potential improvements that could be achieved in the mentioned adhesive properties [7].

The significance of this research was underscored by the pursuit of adhesive formulations that could not only address existing limitations but also elevate the performance thresholds of pressure-sensitive adhesives. The exploration of different concentrations of titanium dioxide acted as a pivotal avenue to unlock new possibilities in adhesive technology. The findings of this study could hold implications for industries reliant on advanced adhesive solutions, including electronics, automotive, and medical sectors, where optimal adhesive properties play a pivotal role in product performance and longevity.

2. Materials and Methods

2.1 Materials

The following materials were used:

- silicone adhesive from Dow Silicones Corp. (Midland, MI, USA) - resin DOWSIL™ 7358
- the crosslinking compound was Bis(2,4-dichlorobenzoyl) peroxide (NOVIPER DB50) - DCIBPO (Novichem Ltd. - Chorzów, Poland)
- solvent was toluene from Carl Roth GmbH + Co. KG (Karlsruhe, Germany)
- filler was titanium dioxide TiO₂ (Table 1) from Grupa Azoty Police Chemical Plant JSC (Police, Poland) and Nanografi Nanotechnology (Çankaya/Ankara, Turkey)

Table 1 Basic properties of selected titanium oxides.

Name	Symbol	Company	Size [μm]	Purity [%]	Rutile [%]	Oil number [g/100 g]
Tytanpol R-001	R-001		0.29	95.0	93	21
Tytanpol R-003	R-003		0.30	94.0	93	21
Tytanpol R-210	R-210		0.31	94.0	93	24
Tytanpol R-211	R-211	Grupa Azoty, Poland	0.32	92.0	91	28
Tytanpol R-213	R-213		0.35	82	80	35
Tytanpol A-11	A-11		0.31	88	84	21
Tytanpol RS	RS		0.26	98.0	95	18
P25 Titanium Dioxide	P25	Nanografi Nanotechnology, Turkey	0.02	99.0	85	N/A

Silicone resin DOWSIL 7358 is dedicated to the production of silicone PSA. It is composed mainly of polydimethylsiloxane resin diluted in xylene and toluene. Polymer content in the composition is 55-58 wt.%. The viscosity of the raw resin is in the range of 10-45 Pas, with a specific gravity of 0.98 kg/dm³.

2.2 Pot Life

Pot life, also defined as shelf life, is a property of prepared pressure-sensitive adhesive compositions related to their stability during extended storage. It is the maximum time during which the PSA composition does not change its rheological properties and may be used for coating. It is also determined by the time it takes to increase the viscosity of the mixture (from twice to four times). The tests were conducted at room temperature (23°C), and every test was started immediately after the composition had been mixed. Viscosity measurements were carried out using the spindle (Brookfield) method and IKA Rotavisc me-vi Digital Viscometer (IKA-Werke GmbH & Co. KG, Staufen im Breisgau, Germany). Spindle no. 8 was used with rotation speed of 5 rpm.

2.3 Preparation of Self-Adhesive Tapes

A mixture of silicone resin - Q2-7358, TiO₂, and a crosslinker (1.5 wt% 2-4-dichlorobenzoyl peroxide-DCIBPO) was formulated to create self-adhesive tapes. To test the effect of fillers, various amounts of filler were added to the composition: 0.1, 0.5, 1.0, and 3.0 parts per hundred (pph) based on polymer content. Subsequently, the composition was applied onto a polyester film with a 50 g/m² thickness using a semi-automatic coater, resulting in a 45 g/m² grammage. The coated adhesive film was then subjected to a drying process in a duct at 110°C for 10 minutes, promoting

the crosslinking of the adhesive film. Following this, the produced adhesive film was sandwiched between two layers of polyester film and segmented into strips, ready for subsequent testing.

2.4 Peel Adhesion

Adhesion is known as the interaction between two dissimilar substances or phases. It is closely related to the interfacial attraction on the contact surfaces of both materials. It is one of the most important relations observed in the bonding processes of materials. The adhesion tests followed the standard FINAT developed (*Fédération Internationale des Fabricants et Transformateurs d'adhésifs et thermocollants sur papiers et autres support*). Standard FTM 1, *Peel adhesion (180°) at 300 mm per minute*, has a test procedure as follows [8]. Samples of PSA tapes have to be prepared in the form of strips 25 × 175 mm and partially attached to the standardized surface (i.e. steel plate). A standard tensile testing machine measures the force required to detach the tape from the steel by peeling at a specified angle and with a defined speed. In our studies, a Zwick/Roell Z010 (Zwick/Roell GmbH & Co. KG, Ulm, Germany) tensile testing machine was used, equipped with XForce P load cell and screw grips. Measuring data was acquired using producer software “testXpert II”.

2.5 Shear Strength (Cohesion)

Cohesion, also known as shear strength, is responsible for the durability of the adhesive joint. Next to adhesion, it is one of the most essential properties of self-adhesive tapes and other adhesives. This feature is related to the internal consistency of the adhesive joint, and its value is mainly influenced by temperature, the concentration and type of crosslinking agent used, and the thickness of the adhesive layer. It is determined according to industrial standard FINAT FTM 8 entitled Resistance of shear from a standard surface, with the following test procedure [8]. A strip of PSA-coated material (25 × 175 mm) is attached to the steel plate to obtain a contact area of 25 × 25 mm. The plate with the attached tape is mounted on a rack in the thermostatic chamber. The free end of the PSA tape strip is loaded with a hanging weight of 1 kg. The measurement continues until the connection between the tape and the steel plate is broken. The measurements were carried out at the room (23°C), and elevated temperature (70°C), using a standardized testing chamber with appropriate racks, hooks, sensors, and computer unit to register testing parameters.

2.6 Tack

Tack indicates the ability of an adhesive to adhere firmly and briefly without pressure after short contact with the standard surface. It can also be defined as the force required to separate a sample from the surface immediately after contact. The tests were conducted according to the industrial standard FINAT FTM 9, *Loop Tack Measurement* [8]. Sample PSA tape is formed in a loop and mounted in the upper grip of the tensile testing machine, and the steel plate is mounted in the lower grip. The loop is contacted shortly with the plate and separated immediately, with the force measurement. The measurements were carried out using the same testing machine described for adhesion tests. The separation rate was 30 cm/min, and the contact area sample plate was 25 × 25 mm.

2.7 Thermal Stability

To check the thermal stability of prepared PSAs, the Shear Adhesion Failure Temperature tests (SAFT) were performed. The samples were prepared similarly to the procedure described for the cohesion tests. A mass of 1 kg was hung at each end of the sample and placed in the oven. The temperature slowly increased from 22 to 217°C at a heating rate of 1°C/min. The damage temperature was recorded, along with the nature of the joint break. The tests were performed with four samples for each formulation to obtain the mean value of temperature stability.

2.8 Shrinkage (Dimensional Stability)

The shrinkage test was carried out according to the standard FINAT FTM 14 entitled Dimensional stability [8]. This method was as follows: PVC foil coated with a PSA 10 cm × 10 cm adhesive film was attached to an aluminum plate using the standard 2 kg roller and kept at room temperature for 72 h. Next, the sample was trimmed to the dimension of the substrate, and vertical and horizontal incisions were made to form a cross in the center of the foil (incisions with a length of 80 mm). The sample was placed in the thermostatic chamber at 70°C for a defined time. The width of the slits formed by the cuts was examined with a magnifying glass after 10 and 30 min, respectively; 1, 3, 8, and 24 hours; 2, 3, 4, 5, 6, and 7 days.

3. Results and Discussion

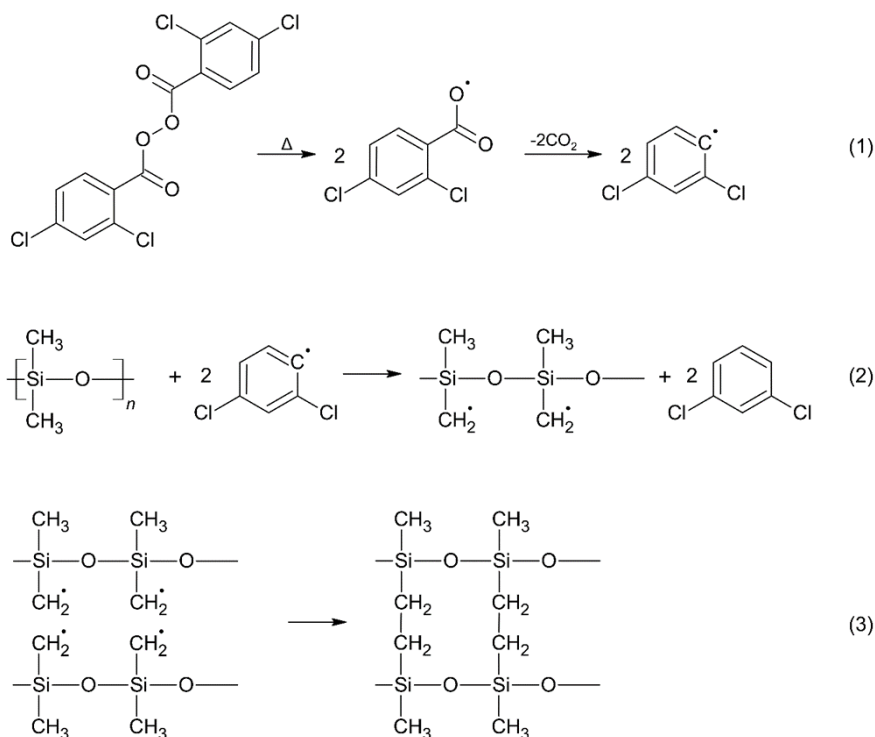
Q2-7358 resin was selected to study titanium dioxide's influence on silicone pressure-sensitive adhesives' properties. Based on previous experience and tests, a resin modified with 1.5 pph of DCIBPO was selected as a pure composition (not modified with fillers). This composition showed the best physicochemical and functional properties (Table 2), especially a very good adhesion-cohesion balance. This allowed us to assume that physical modification with fillers should bring the desired effects with minor changes in adhesion and stickiness. This assumption allowed us to expect new self-adhesive tapes for special applications.

Table 2 Basic properties of pure resin - silicone pressure-sensitive adhesive.

Silicone resin	Adhesion [N/25mm]	Tack [N]	Cohesion [h]		SAFT [°C]	Shrinkage after 72 h [%]
			23°C	70°C		
Q2-7358	8.67	13.0	>72	>72	150	1.33

Crosslinking agent used for the studies was DCLBPO, also known in literature as DCIBP. It is Bis(2,4-dichlorobenzoyl) peroxide, which, together with its basic form DBP (dibenzoyl peroxide), belongs to the most commonly used thermal initiators for crosslinking reactions.

The general mechanism of crosslinking reaction between polydimethylsiloxane resin and DCIBPO can be proposed as shown below in Scheme 1.



Scheme 1 Crosslinking of polydimethylsiloxane with DCIBPO.

Table 3 summarizes the results of viscosity measurements over time of prepared samples containing various degrees of titanium dioxide filling. In the case of most of the tested fillers, the viscosity increased evenly after addition until gelation approximately on the 10th day after receiving them (i.e. the samples doubled their viscosity or reached such a high that they did not allow for sealing with self-adhesive foil). The values of most of the tested samples were similar and behaved analogously. The phenomenon of silicone self-adhesive compositions gelling after modification with a filler is quite widely described in the literature [9]. However, the composition with titanium oxide RS already showed a viscosity value approximately one and a half times higher than its counterparts in the other compositions on the first day. Additionally, the gelling process took place around 5 and, with a high degree of filling, 3 days after receiving the composition. Titanium oxide with the symbol RS was characterized by the lowest oil number (18 g/100 g) and the highest Rutil content (95%) compared to the other fillers used. Thanks to this, the addition of a larger amount of filler could take on the consistency of a paste much faster and thus accelerate the gelling process of the entire composition, whereas, in the case of other fillers, it took about 10 days. Additionally, the smallest average particle size could have an additional stimulating effect on gelation [10].

Table 3 Viscosity changes of the prepared silicone compositions with various concentrations of titanium dioxides.

Symbol	Filler content [pph]	Viscosity [Pa·s]				
		1 day	2 days	3 days	5 days	7 days
R-001	0.1	14.6	17.4	19.5	25.4	31.1
	0.5	15.6	16.4	18	21.3	26.9
	1.0	16.1	17.0	18.8	22.6	30.2
	3.0	18.3	19.1	21.4	25.9	33.0

R-003	0.1	15.5	16.4	18.7	24.5	29.3
	0.5	16.1	16.9	18.5	22.1	27.1
	1.0	16.8	17.7	19.2	23.2	28.9
	3.0	17.1	18.2	20.0	23.9	28.7
R-210	0.1	17.4	18.5	21.5	26.6	34.5
	0.5	18.3	19.3	22.5	27.7	35.2
	1.0	18.6	19.5	22.7	28.0	36.0
	3.0	18.6	19.4	22.7	28.2	37.0
R-211	0.1	17.6	18.4	21.4	26.7	35.0
	0.5	17.4	18.3	21.2	26.1	32.0
	1.0	17.8	18.8	22.0	27.3	37.0
	3.0	18.3	19.4	22.6	27.3	35.0
R-213	0.1	17.4	18.2	20.9	25.5	32.5
	0.5	17.5	18.4	21.4	26.6	35.0
	1.0	17.7	18.6	21.6	26.6	35.1
	3.0	18.5	19.4	22.7	28.3	37.0
R-A11	0.1	16.6	16.4	18.1	24.0	30.5
	0.5	17.0	17.9	19.7	23.1	28.5
	1.0	18.1	18.9	20.7	23.9	31.5
	3.0	18.7	19.7	22.0	26.1	34.0
RS	0.1	25.1	27.3	30.3	45.6	gel
	0.5	32.3	35.2	39.1	47.9	gel
	1.0	39.8	42.3	45.4	gel	gel
	3.0	43.9	47.2	49.5	gel	gel
P25	0.1	14.6	17.0	19.0	25.3	31.4
	0.5	15.5	16.3	17.7	21.6	25.6
	1.0	15.6	16.5	18.1	21.9	28.5
	3.0	17.5	18.9	20.6	25.9	32.6

The first drawing (Figure 1) shows the peel adhesion values of tapes made of Q2-7358 silicone resin modified with titanium dioxides with different concentrations. In most cases, an initial increase in the adhesion value was noted, followed by a decrease, although in none of the cases did the value for the 3 pph filling reach the value of the pure composition. The existing process is most likely due to the compatibility of the filler in the matrix, its miscibility, and its tendency to agglomerate [10]. This behavior is quite challenging to interpret. It may result from a certain orientation and arrangement of the chains in the emerging adhesive layer forced by adding the filler, which is then eliminated when adding the filler is increased due to its agglomeration. However, in the case of samples R-001 and R-003, the increase as mentioned above was not recorded, while with increasing filling, a minimal decrease in the adhesion value was recorded, which would suggest that the addition of fillers increases the internal cohesion of the adhesive film (cohesion) and slightly shifts the adhesion-cohesion balance [11, 12]. The failure pattern observed for all tested samples was the adhesion failure. No adhesive remains were observed on the specimen surface after the peel adhesion test.

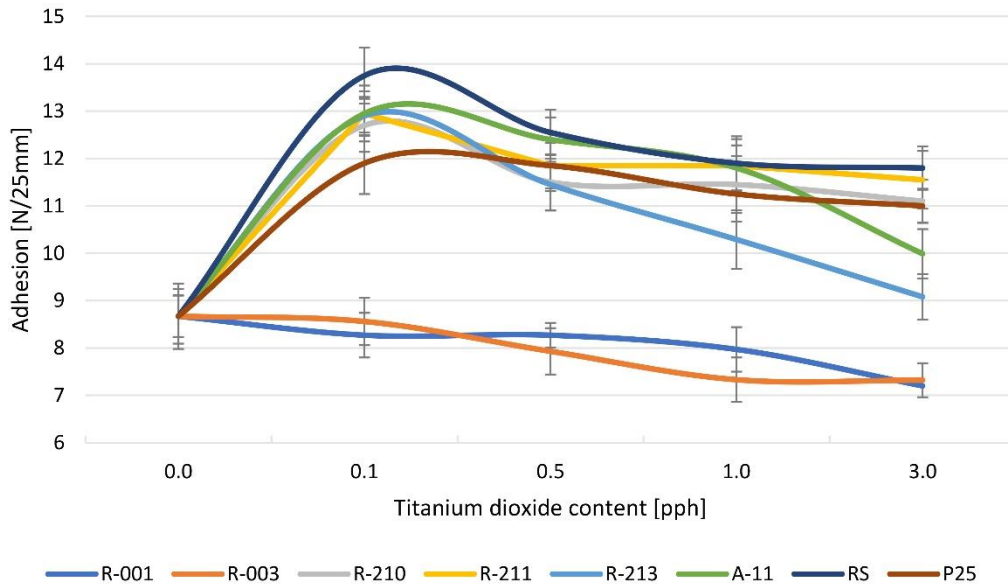


Figure 1 Effect of the addition of titanium dioxides on the peel adhesion of silicones pressure-sensitive adhesives.

The behavior of the samples modified with titanium dioxide described above is reflected in the tack of some tested samples - R-001, R-210 and R-003 - however, in most cases, the greater the addition of filler to the Q2-7358 resin, the lower the tack value for the adhesive film was obtained (the lowest value tested was approximately 4 N for the RS-modified composition). Figure 2 visualizes the effect of adding various titanium dioxides to the tack of the tested silicone compositions. The reduction in the tack value, despite previous better correlations in terms of adhesion, may be due to the high bulk density of the filler, which, despite the excellent compatibility of titanium dioxide with the matrix, contains so much of it by volume that it lowers the tack value [7, 10].

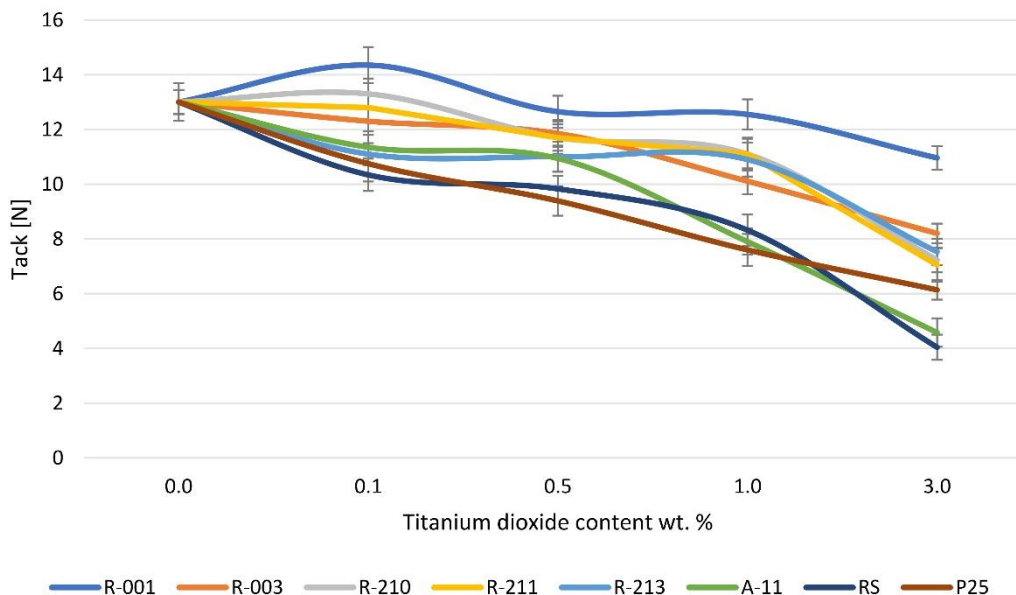


Figure 2 Effect of the addition of titanium dioxides on the tack of silicones pressure-sensitive adhesives.

Table 4 shows the cohesion results at room temperature (23°C), elevated temperature (70°C), and thermal stability determined using the SAFT test. In the case of a temperature of 20°C, only Q2-7358 resin compositions modified with titanium dioxide RS did not retain values higher than those required for an industrially produced tape (above 72 h), which may indicate a shift in the adhesion-cohesion balance towards cohesion [11, 13].

Table 4 Cohesion and thermal stability of prepared silicones PSA with titanium oxides.

Symbols	Titanium oxide content [pph]			
	0.1	0.5	1.0	3.0
Cohesion at 23°C				
R-001	>72	>72	>72	>72
R-003	>72	>72	>72	>72
R-210	>72	>72	>72	>72
R-211	>72	>72	>72	>72
R-213	>72	>72	>72	>72
A-11	>72	>72	>72	>72
RS	>72	37.2	26.9	2.8
P25	>72	>72	>72	>72
Cohesion at 70°C				
R-001	57.8	>72	>72	>72
R-003	>72	>72	>72	>72
R-210	>72	>72	>72	11.6
R-211	12.8	19.8	>72	>72
R-213	>72	>72	>72	>72
A-11	>72	>72	>72	>72
RS	5.5	6.0	6.5	12.0
P25	>72	>72	>72	>72
Thermal stability				
R-001	>225	>225	>225	212
R-003	206	217	>225	>225
R-210	>225	>225	>225	219
R-211	>>225	>225	219	216
R-213	>225	>225	219	216
A-11	>225	>225	>225	199
RS	>225	>225	>225	>225
P25	>225	>225	>225	208

Cohesion tests at a temperature of 70°C confirmed the lack of thermal stability of the obtained tapes with the addition of RS. In addition, small additions of fillers R-001 and R-211 also reduced the cohesion value, which may indicate that in their case, small amounts of filler acted as a hindrance and did not support chain alignment. Additionally, for the modified R-210 resin, a decrease in the cohesion value was noted when filling at 3 pph, which may result from too many spatial lumps and agglomeration of the filler [14, 15].

The obtained self-adhesive tapes were tested for thermal stability using the so-called SAFT test to determine their maximum operating temperature. All tested tapes showed increased thermal stability; most reached the test limit ($>225^{\circ}\text{C}$). Interestingly, the high thermal stability obtained by adding various titanium dioxides did not always confirm long-term cohesion at both room and elevated temperatures. This proves that titanium dioxide is somewhat universal in this respect.

The effect of different amounts of titanium dioxide addition on the shrinkage of the silicone self-adhesive glue is presented in Table 5. The following tendency was observed for the tested resin - the lower the shrinkage, the higher the filler content in the polymer matrix (the shrinkage stabilized around the 7th day of the test). This may be due to a better arrangement of the polymer mesh or a more compact internal structure of the self-adhesive film, and is quite a common phenomenon [13, 16]. Moreover, the amount of contraction was individual for each tested filler. The lowest values were obtained for compositions modified with R-210 and the highest for R-211. Comparing the results in each case, the best values were obtained with a high degree of filling (3 pph), regardless of the titanium dioxide used for the modification.

Table 5 Shrinkage of silicone pressure-sensitive adhesives with titanium dioxides addition.

Shrinkage (%)												
Cont.of filler [pph]	10 min	30 min	1 h	3 h	8 h	24 h	2 Days	3 Days	4 Days	5 Days	6 Days	7 Days
R-001												
0.1	0.23	0.27	0.30	0.35	0.43	0.47	0.51	0.57	0.61	0.65	0.65	0.65
0.5	0.13	0.16	0.21	0.22	0.29	0.36	0.40	0.45	0.50	0.52	0.54	0.54
1.0	0.09	0.15	0.17	0.20	0.26	0.30	0.33	0.40	0.40	0.44	0.46	0.46
3.0	0.14	0.17	0.18	0.20	0.22	0.26	0.30	0.31	0.36	0.39	0.41	0.41
R-003												
0.1	0.20	0.25	0.28	0.33	0.38	0.41	0.46	0.52	0.57	0.60	0.60	0.60
0.5	0.10	0.12	0.20	0.28	0.36	0.39	0.43	0.46	0.55	0.56	0.57	0.57
1.0	0.09	0.12	0.19	0.20	0.25	0.34	0.40	0.40	0.46	0.49	0.50	0.50
3.0	0.08	0.10	0.15	0.17	0.20	0.29	0.38	0.40	0.40	0.41	0.44	0.44
R-210												
0.1	0.20	0.24	0.26	0.28	0.30	0.32	0.35	0.36	0.37	0.40	0.40	0.40
0.5	0.14	0.16	0.18	0.20	0.24	0.26	0.28	0.30	0.32	0.34	0.34	0.34
1.0	0.10	0.11	0.13	0.14	0.15	0.17	0.18	0.19	0.20	0.20	0.20	0.20
3.0	0.09	0.09	0.10	0.12	0.12	0.13	0.13	0.14	0.15	0.17	0.17	0.17
R-211												
0.1	0.22	0.27	0.30	0.37	0.43	0.47	0.51	0.55	0.61	0.65	0.65	0.65
0.5	0.13	0.16	0.20	0.22	0.28	0.36	0.39	0.45	0.50	0.51	0.54	0.54
1.0	0.09	0.15	0.17	0.20	0.26	0.30	0.33	0.40	0.41	0.44	0.47	0.47
3.0	0.14	0.17	0.18	0.20	0.22	0.26	0.30	0.31	0.36	0.39	0.41	0.41
R-213												
0.1	0.09	0.10	0.15	0.19	0.24	0.26	0.32	0.34	0.39	0.41	0.41	0.41
0.5	0.08	0.09	0.10	0.18	0.21	0.25	0.30	0.32	0.35	0.38	0.40	0.40
1.0	0.08	0.08	0.10	0.15	0.20	0.22	0.29	0.31	0.34	0.36	0.36	0.36

3.0	0.06	0.09	0.10	0.11	0.13	0.15	0.16	0.18	0.18	0.18	0.18	0.18
A-11												
0.1	0.18	0.22	0.28	0.31	0.33	0.35	0.37	0.40	0.43	0.45	0.45	0.45
0.5	0.16	0.18	0.21	0.24	0.28	0.30	0.33	0.35	0.40	0.42	0.42	0.42
1.0	0.11	0.12	0.14	0.16	0.17	0.21	0.25	0.28	0.30	0.30	0.30	0.30
3.0	0.10	0.11	0.13	0.14	0.16	0.17	0.21	0.23	0.24	0.24	0.24	0.24
RS												
0.1	0.20	0.28	0.32	0.37	0.42	0.47	0.51	0.54	0.57	0.57	0.57	0.57
0.5	0.19	0.28	0.31	0.39	0.41	0.44	0.45	0.47	0.49	0.52	0.56	0.56
1.0	0.15	0.21	0.31	0.40	0.44	0.46	0.48	0.50	0.52	0.52	0.52	0.52
3.0	0.18	0.24	0.28	0.30	0.32	0.35	0.37	0.41	0.41	0.41	0.41	0.41
P25												
0.1	0.16	0.26	0.30	0.33	0.45	0.47	0.50	0.54	0.58	0.60	0.60	0.60
0.5	0.08	0.12	0.14	0.26	0.38	0.41	0.45	0.49	0.54	0.55	0.55	0.55
1.0	0.07	0.10	0.15	0.19	0.29	0.36	0.40	0.41	0.47	0.49	0.50	0.50
3.0	0.06	0.09	0.13	0.15	0.24	0.30	0.37	0.39	0.43	0.46	0.46	0.46

4. Conclusions

The result of modifying the silicone adhesive composition with various titanium dioxide fillers was to obtain new silicone pressure-sensitive adhesives. The tests carried out on composite materials confirmed, in most cases, the maintenance of high cohesion values and the improvement of heat-resistant properties and shrinkage while at the same time slightly (or improving them) lowering the adhesion and viscosity values of the modified tapes compared to the tape without filler in its composition.

The obtained self-adhesive tapes can be used, among others, for the automotive, heating, and heavy industries. The physical modifications carried out using fillers, confirmed by tests, also create application potential for the adhesive compositions themselves, which can be used in a form other than the one presented in the article, e.g., for applying metal supports and attaching them in selected places.

Author Contributions

Authors contributed equally to this work.

Funding

The research was carried out as part of the Leader Program project of the National Center for Research and Development no. LIDER/9/0028/L-11/19/NCBR/2020.

Competing Interests

The authors have declared that no competing interests exist.

References

1. Benedek I, Feldstein MM. Technology of pressure-sensitive adhesives and products. Boca Raton, FL: CRC Press; 2009.
2. Sun S, Li M, Liu A. A review on mechanical properties of pressure sensitive adhesives. *Int J Adhes Adhes.* 2013; 41: 98-106.
3. Czech Z. Developments in crosslinking of solvent-based acrylics. In: *Developments in pressure-sensitive products.* 2nd ed. Boca Raton, FL: CRC Press; 2005. pp. 225-272.
4. Satas D. *Handbook of pressure sensitive adhesive technology.* Boston, MA: Springer; 1989.
5. Lin SB, Durfee LD, Ekeland RA, McVie J, Schalaus GK. Recent advances in silicone pressure-sensitive adhesives. *J Adhes Sci Technol.* 2007; 21: 605-623.
6. Mecham S, Sentman A, Sambasivam M. Amphiphilic silicone copolymers for pressure sensitive adhesive applications. *J Appl Polym Sci.* 2010; 116: 3265-3270.
7. Sanghvi MR, Tambare OH, More AP. Performance of various fillers in adhesives applications: A review. *Polym Bull.* 2022; 79: 10491-10553.
8. FINAT. *FINAT Technical Handbook Test Methods.* 10th ed. The Hague, The Netherlands: FINAT; 2019.
9. Barnes HA. A review of the rheology of filled viscoelastic systems. *Rheol Rev.* 2003; 15: 1-36.

10. Ashraf MA, Peng W, Zare Y, Rhee KY. Effects of size and aggregation/agglomeration of nanoparticles on the interfacial/interphase properties and tensile strength of polymer nanocomposites. *Nanoscale Res Lett.* 2018; 13: 214.
11. Wolf C, Angellier-Coussy H, Gontard N, Doghieri F, Guillard V. How the shape of fillers affects the barrier properties of polymer/non-porous particles nanocomposites: A review. *J Membr Sci.* 2018; 556: 393-418.
12. Czech Z, Arabczyk W, Hełminiak A, Kowalczyk A. Influence of iron carbide filler in carbon matrix on the adhesive properties of acrylic pressure-sensitive adhesives. *Int J Adhes Adhes.* 2013; 40: 210-214.
13. Salasinska K, Polka M, Gloc M, Ryszkowska J. Natural fiber composites: The effect of the kind and content of filler on the dimensional and fire stability of polyolefin-based composites. *Polimery.* 2016; 61: 255-265.
14. Galimberti M. *Rubber-clay nanocomposites: Science, technology, and applications.* Hoboken, NJ: John Wiley & Sons; 2011.
15. Clauß S, Dijkstra DJ, Gabriel J, Karbach A, Matner M, Meckel W, et al. Influence of the filler material on the thermal stability of one-component moisture-curing polyurethane adhesives. *J Appl Polym Sci.* 2012; 124: 3641-3649.
16. Giachetti L, Russo DS, Bambi C, Grandini R. A review of polymerization shrinkage stress: Current techniques for posterior direct resin restorations. *J Contemp Dent Pract.* 2008; 7: 79-88.