



Effects of spices-waste filler on the physical properties of silicone-based composites

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ABSTRACT

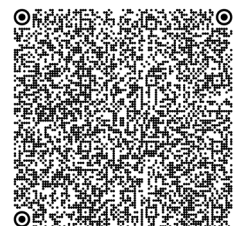
Purpose: The study aimed to examine the effect of using spices as fillers on the mechanical and physical properties of composite materials in which silicone plays the role of a filler.

Design/methodology/approach: Composite materials based on silicone were obtained by gravity casting, in which spices (garlic, paprika, pepper, and turmeric) with a mass content of 2.5, 5 and 10% of the filler. The obtained materials have been subjected to tensile test, density, hardness, and abrasion resistance.

Findings: The research was to answer whether adding spices can change the physicochemical properties of composite materials based on silicone and native silicone. Particular attention was paid to the properties that are important from the point of view of engineering applications: mechanical properties, tribological properties, hardness, and density.

Practical implications: In the face of challenges related to the growing amount of waste from the food industry, joint materials engineering tries to answer whether this waste can be used in the production of composite materials. In several publications from recent years, it has been postulated that used food industry products can be used as fillers for composite materials, as they can, on the one hand, improve specific physicochemical properties of new materials and manage food waste.

Originality/value: So far, few scientific articles discuss using food industry products as fillers in polymeric composites. Such articles usually focus on the use of natural plant or animal fibres.



Adding spices from 2.5 to 10% by mass increases the abrasion resistance compared to silicone without adding filler. The most significant decrease in abrasion resistance was recorded for 2.5 GA and 2.5 TU. A decrease in abrasive wear by over 40% was noted here relative to neat silicone.

Keywords: Composites, Mechanical properties, Organic fillers, Abrasion, Silicone

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PROPERTIES

1. Introduction

Silicone rubber is used widely in various fields, such as aerospace, automotive, and electronics [1]. It is attributed to properties such as high chemical stability, resistance to temperature changes, resistance to atmospheric conditions, and high insulation [2]. However, to give silicones new physicochemical properties, it is necessary to use them in the character of the matrix for composite materials [3]. Polymer-based composite materials are defined as materials with a polymer matrix material, with organic or inorganic particle fillers immiscible with the polymer [4-6]. Recently, composite materials have gained popularity due to the ease and economy of their manufacture [7]. Reinforcement creates a more attractive composite than pure plastic due to its low density, reduced flammability, higher tensile strength, stiffness, and increased abrasion resistance [8-12]. Newly developed materials must also lack human toxicity [13].

Industrially, polymer composites reinforced with synthetic fibres (e.g., glass, aramid, ceramic) play the most significant role and are used in many industries, such as aerospace, self-driving cars, and construction [14-16]. Composites reinforced with organic fibres play an ever-increasingly important role. They are characterised by good strength, lightness, composite stiffness, low cost, low friction coefficient, adequate resistance to abrasive and erosive wear, and, most importantly, biodegradability (including lower disposal costs) [17,18]. It aligns with growing environmental awareness and concepts of sustainability (responsible waste management and product design with recycling in mind) [19-21].

Various waste materials in agriculture, livestock farming, or the food industry produce suitable biocomposite fillers. Several papers report using biological materials like plant parts [22]. Examples of such applications include walnut shells, hazelnut shells, and sunflower seed shells to improve the stiffness of epoxy resins [21]. Orange processing waste has been used as a filler to enhance

mechanical properties [23] and sliding wear resistance [24]. Pineapple leaves [25] and peanut shells [26] have been used for insulation.

So far, Hitesh Sharma and his team researched using citrus limetta peel (CLP) as fillers in composite materials. In a 2019 publication, they examined the effect of citrus limetta peel (CLP) on the mechanical properties of epoxy resin composite materials [23]. A 2020 study examined the influence of incorporating citrus limetta peel (CLP) fillers at diverse loadings (5%, 10% and 15%) on the dynamic mechanical properties of epoxy composites [27]. Subsequent studies investigated the influence of CLP filler size on the thermal and mechanical behaviour of epoxy resin-based composites [28]. In another study, the influence of epoxy resin matrix composites with 15% CLP content on the wear behaviour of epoxy composites has been experimentally explored [29]. Research from 2023 aimed to study the potential of citrus limetta peel (fruit waste) as a novel filler in thermoplastic composites [30]. Poikelispää et al., polyacrylate rubber's dielectric and actuation properties (ACM) were studied after adding vegetable-based fillers such as potato starch, corn starch, garlic, and paprika [31]. Mrówka et al. investigated the effect of using organic waste from citrus peels, including grapefruit, lime, lemon, and orange peels, on silicone-based composites' physical properties [32].

Using silicones as a matrix for composites is a relatively new concept, with scant literature reports on the subject. In the last 10 years, there have been publications in which researchers have added ground quartz and wollastonite to silicone to create a composite used as a cable sheath [33]. Nickel was also added to silicone to test the mechanical properties of the resulting composite [34]. The mechanical properties of silicone-based composites were improved by the addition of Fe_3O_4 [35]. On the other hand, the physicochemical properties of silicone-based composites were affected by ceramic powders [36]. Adding small amounts of graphene oxide improved the resistance of

silicone-based composites to ageing under seawater conditions [37]. Silicone-based composites with added phosphors have also found applications in the manufacturing of LEDs [38]. There are literature reports regarding silicone-based composites with $ZrSi_2$ as protective coatings [39].

Such cases of using silicones as the matrix for polymer composites are based on using non-organic compounds as fillers. The paper evaluated how organic fillers affect the physical and mechanical properties of silicone matrix composite materials. The present work studied the effect of expired cooking spices (garlic, paprika, pepper, and turmeric) as fillers for silicone-based composites. The study aimed to determine how the spice wastes and their amounts affected material performance properties (tensile test, density, hardness, and abrasion resistance).

2. Materials and methods

2.1. Materials

Mold Star™ 30 silicone, produced by Smooth-On, Inc., was used as the matrix. The properties of silicone Mold Star™ 30 are shown in Table 1.

Table 1.
Properties of silicone Mold Star™ 30

Properties	Unit	Value
Ratio mixing silicone: catalyst		1:1
Pot life	min	45
Cure time	h	6
Density	g/cm^3	1.12
Viscosity	cps	12500
Tensile strength	MPa	2.9
Elongation at break	%	300-350

The spices used for the research, garlic, paprika, pepper, and turmeric, came from the kitchen of the authors of the articles. They were not spices explicitly bought to carry out this research, but old spices that had lost their properties due to the effects of time and thus became waste. The spices used for the research were a mixture of spices used at different times and from different producers. The article's authors firmly state that they are against food waste and use edible spices (and all other culinary ingredients) as fillers in polymer composite materials.

2.2. The filler's characterisation

The particle size analysis of the species was carried out using a Fritsch Analysette Vibratory Sieve Shaker equipped

with screens with the following mesh sizes: 0.5 mm, 0.25 mm, 0.18 mm, 0.125 mm, 0.09 mm, 0.045 mm, and 0.035 mm (Fritsch, Idar-Oberstein, Germany). The sieve analysis of each species lasted 45 min. Densities of the species were assessed following the ISO 3953 method using 100 cm^3 capacity cylinders. The tap density analysis of each species lasted 7 min (150 taps/min). Bulk densities were measured before and after tapping.

2.3. Composites preparation

The composites were prepared by gravity casting with filler contents of 2.5, 5, and 10 wt.%. The composites were marked as shown in Table 2. Before introducing fillers into the silicone matrix, fillers were gradually heat-treated at 80°C for 180 min until a constant weight was obtained. The silicone components were mixed with the fillers on a high-shear mixer at 500 rpm. Seventy-two hours after being poured into moulds, samples were cut by punching and subjected to mechanical and physical tests.

Table 2.
Composition of samples

Filler	Filler content, %	Designation
Matrix	0	REF
Garlic	2.5	2.5 GA
Garlic	5	5 GA
Garlic	10	10 GA
Paprika	2.5	2.5 PA
Paprika	5	5 PA
Paprika	10	10 PA
Pepper	2.5	2.5 PE
Pepper	5	5 PE
Pepper	10	10 PE
Turmeric	2.5	2.5 TU
Turmeric	5	5 TU
Turmeric	10	10 TU

2.4. Composites characterisation

EN ISO 527-1 performed static tensile testing using an Instron 4465 machine (Instron, Norwood, MA, USA).

Density tests were carried out by EN ISO 1183-1:2006 on an Ad-venture Pro AV264CM weighing machine (OHAUS Europe GmbH, Nänikon, Switzerland). Post measurements were performed in distilled water with a 0.997 g/cm^3 density.

Shore A hardness testing was performed by EN ISO 7619-1:2010 using a Zorn hardness tester (Zorn Instruments GmbH & Co. Hansstadt, Germany).

Abrasion wear was measured by EN ISO 4649:2007 on an APG Schopper-Schlobach apparatus (APG Germany GmbH, Friedberg, Germany).

Five measurements for tensile, density, and hardness testing were performed, from which the arithmetic mean and standard deviation were determined. For the abrasion test, the arithmetic mean of three measurements was given.

All tests were conducted at 22°C with an air humidity of 50%.

3. Results

3.1. The filler's characterisation

The densities of turmeric, garlic, pepper, and paprika are provided in Table 3. A significant difference was found between grain sizes. Table 2 shows that nearly 90 wt.% grains in turmeric were less than 0.125 mm, whereby the most abundant (48 wt.%) were grains between 0.09–0.045 mm. The particle size analysis of garlic and paprika was similar, although paprika grains <0.045 mm were not observed. For pepper, a significant number of grains >0.5 mm were identified (13 wt.%).

The particle size analyses of the tested species were similar, as demonstrated in Table 4. The lowest bulk and tap densities were paprika, 0.42 g/cm³ and 0.61 g/cm³, respectively. In contrast, the highest densities were garlic, 0.58 g/cm³ for bulk density and 0.88 for tap density.

Table 3.

Bulk and tap densities of the fillers

Filler	Bulk density, g/cm ³	Tap density, g/cm ³
Turmeric	0.50	0.70
Garlic	0.58	0.88
Pepper	0.53	0.80
Paprika	0.42	0.61

Table 4.

Particle size analyses of the fillers

Grain size, mm	Yield, %			
	Turmeric	Garlic	Pepper	Paprika
> 0.5	0.1	1.6	13.0	1.4
0.5-0.25	1.8	75.8	28.6	56.4
0.25-0.18	1.7	14.4	13.4	38.1
0.18-0.125	6.2	5.9	21.9	3.3
0.125-0.09	32.5	1.2	7.9	0.7
0.09-0.045	47.9	0.8	12.7	0.1
0.045-0.035	9.6	0.3	1.7	0.0
< 0.035	0.3	0.1	0.8	0.0

3.2. Tensile test

The results of the static tensile test for all tested materials are presented in Figure 1 (maximum load), Figure 2 (stress at break), and Figure 3 (elongation at break).

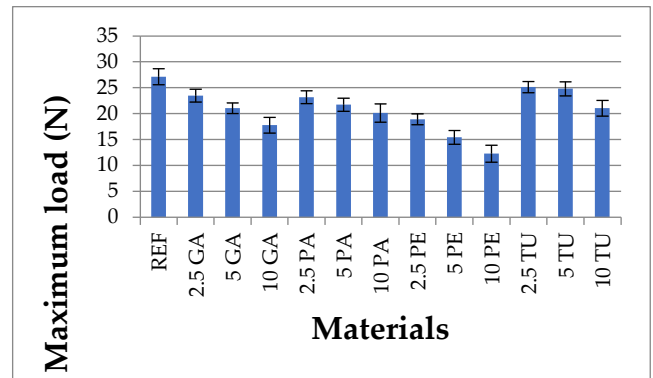


Fig. 1. Maximum loads of the tested materials

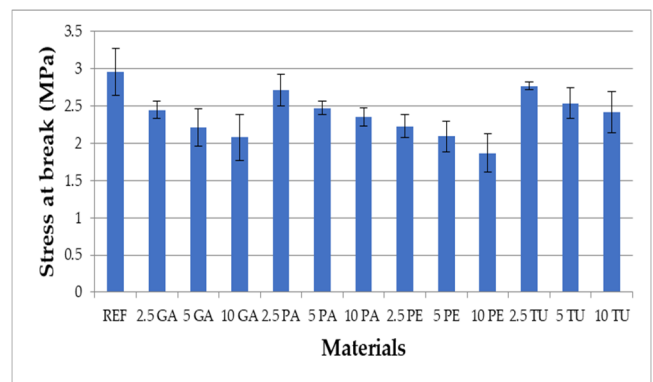


Fig. 2. Stress at break of the tested materials

The highest maximum load value was recorded for silicone without filler (REF 27.12 N). The total load results for all composite materials had lower values than REF. A concurrent trend was noted for composite materials with garlic, paprika, and pepper as fillers. As the filler mass in the composite increased, the maximum load value decreased for all three fillers. Maximum load values for material 2.5 GA (23.47 N) and 2.5 PA (23.17 N) should be considered like each other. The maximum load values for the 5 GA and 10 GA materials were 21.08 and 17.8 N, respectively. The maximum load values for the 5 PA and 10 PA materials were 21.71 and 20.15 N, respectively. For materials with pepper as filler, the maximum load values were the lowest of all materials tested. The maximum load value for the 2.5 PE material was 18.91 N, the lowest maximum load value

among all 2.5 wt.% composites. The 5 PE material had a total load value of 15.41 N, and the 10 PE material was 12.27 N. The maximum load for the 10 PE material was the lowest among all based materials, 55% softer than silicone without filler addition (REF). For composite materials where turmeric served as the filler, the maximum load values for the 2.5 TU and 5 TU materials were similar at 25.12 and 24.79 N, respectively. For the 10 TU material, the maximum load value dropped to 21.07 N.

For testing the stress at break value among all tested materials, the maximum was observed for silicone without filler addition (REF) – 2.96 MPa. All tested composite materials' stress at break value decreased as the filler mass content increased. For the garlic composite materials, the stress at break values were 2.5 GA (2.45 MPa), 5 GA (2.21 MPa), and 10 GA (2.08 N). For paprika materials, the stress at break values was 2.5 PA (2.71 MPa), 5 PA (2.47 MPa), and 10 PA (2.35 MPa). For pepper composite materials, the stress at break values was 2.5 PE (2.23 MPa), 5 PE (2.09 MPa), and 10 PE (1.87 MPa). The stress at break value for the 10 PE material was lower than any other material (37% lower than the REF material). The turmeric-filled material results were – 2.5 TU (2.77 MPa), 5 TU (2.54 MPa), and 10 TU (2.42 MPa).

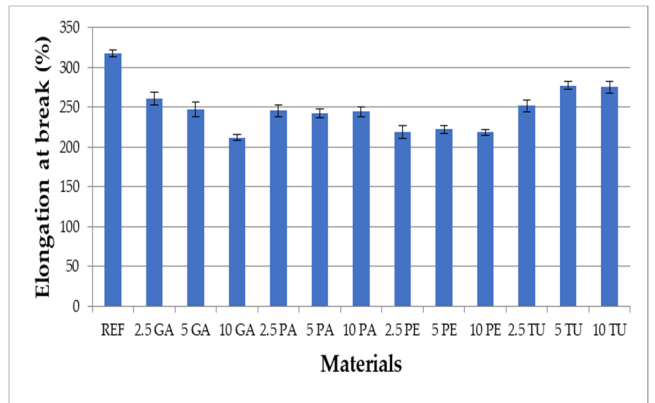


Fig. 3. Elongation at break of the tested materials

The highest elongation at break value was recorded for REF (317.45%). All other composite materials had lower elongation at break values. Garlic materials decreased the elongation at break value with increased filler levels. The value for the 2.5 GA material was 261.34%, for the 5 GA material was 247.36%, and for the 10 GA material was the lowest and 31% lower than silicone without

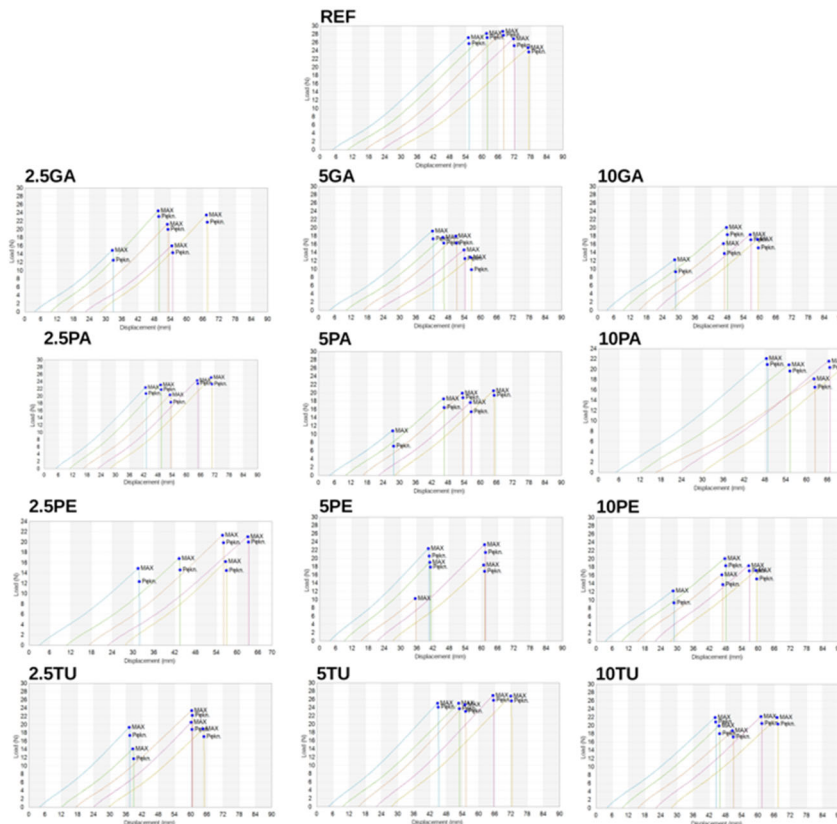


Fig. 4. The load-deformation curves of the tested materials

filler additives. Paprika composite materials had similar elongation at break values regardless of the mass content (2.5 PA, 245.67%; 5 PA, 242.55%; and 10 PA, 244.48%). Similar tendencies were observed for materials pepper fillings (2.5 PE, 219.33%; 5 PE, 222.28%; and 10 PE, 218.54%). Materials with 5 and 10% fillings for turmeric samples had similar elongation at break values (277.64 and 275.29%) with a lower break for 2.5 TU (252.23%). The load–deformation curves of the tested materials are presented in Figure 4.

3.3. Density test

The density measurement results for all tested materials are presented in Figure 5.

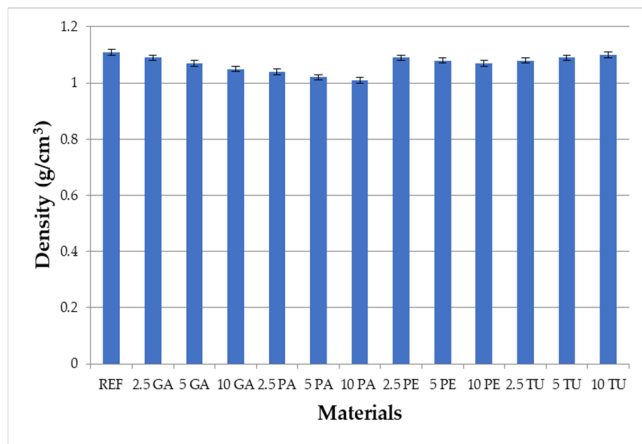


Fig. 5. Density of tested materials

The highest density was the silicone used as the matrix of composite materials (1.11 g/cm³). Lower densities than REF characterised other composite materials. The densities of garlic, paprika, and pepper materials decreased with increased filler levels in the composite. For garlic, the values were – 2.5 GA, 1.09 g/cm³, 5 GA, 1.07 g/cm³, and 10 GA, 1.05 g/cm³. For paprika, the values were – 2.5 PA, 1.04 g/cm³, 5 PA, 1.02 g/cm³, and 10 PA, 1.01 g/cm³. The density for 10 PA was lower than any other material, 9% lower than the highest measured density for the REF material. For pepper, the values were – 2.5 PE, 1.09 g/cm³, 5 PE, 1.08 g/cm³, and 10 PE, 1.07 g/cm³. An opposite trend was observed for turmeric; as the filler content increased, so did the density. Those values were – 2.5 TU, 1.08 g/cm³, 5 TU, 1.09 g/cm³, and 10 TU, 1.1 g/cm³.

3.4. Hardness test

The hardness results for all materials are presented in Figure 6.

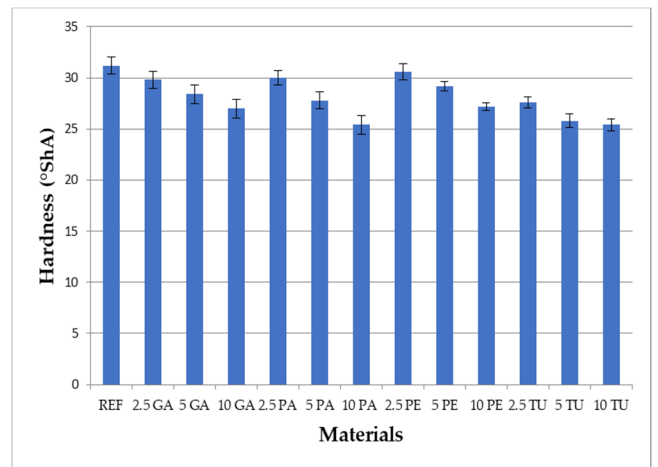


Fig. 6. Hardness of tested materials

The highest hardness was recorded for silicone (REF, 31.2°ShA). All other composite materials had lower hardness compared to REF. A convergent tendency was observed for all tested composite materials: hardness decreased with increasing filler content. For garlic, the values were 2.5 GA – 29.8°ShA, 5 GA – 28.4°ShA, and 10 GA – 27°ShA. For paprika, the values were 2.5 PA – 30°ShA, 5 PA – 27.8°ShA, and 10 PA – 25.4°ShA. For pepper, the values were 2.5 PE – 30.6°ShA, 5 PE – 29.2°ShA, and 10 PE – 27.2°ShA. For turmeric, the values were 2.5 TU – 27.6°ShA, 5 TU – 25.8°ShA, and 10 TU – 25.4°ShA. Material 10 TU had the lowest hardness among all tested materials, almost 20% lower than REF.

3.5. Abrasive wear test

Abrasive wear values for all tested materials are presented in Figure 7.

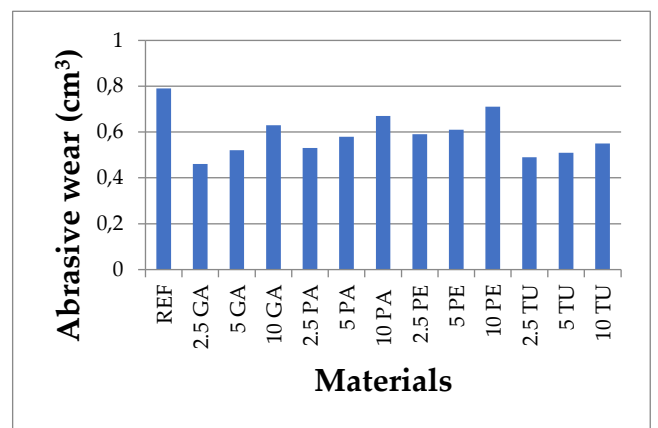


Fig. 7. Abrasive wear of tested materials

The abrasive wear was maximised with the REF material (0.79 cm^3). All composite materials had lower abrasive wear values compared to silicone. For all composite materials, abrasive wear tended to increase with added filler in the composite. The lowest abrasive wear value was recorded for 2.5 GA (0.46 cm^3 , <40% lower than REF). The abrasive wear values for 5 GA and 10 GA materials were 0.52 cm^3 and 0.63 cm^3 , respectively. For paprika, the values were 2.5 PA, 0.53 cm^3 , 5 PA, 0.58 cm^3 , and 10 PA, 0.67 cm^3 . For pepper, the values were: 2.5 PE – 0.59 cm^3 , 5 PE – 0.61 cm^3 , and 10 PE – 0.71 cm^3 . For turmeric, the values were 2.5 TU, 0.49 cm^3 , 5 TU, 0.51 cm^3 , and 10 TU, 0.55 cm^3 . Figure 8 shows photos of the samples before and after the abrasive wear test using the Schopper-Schlobach apparatus.

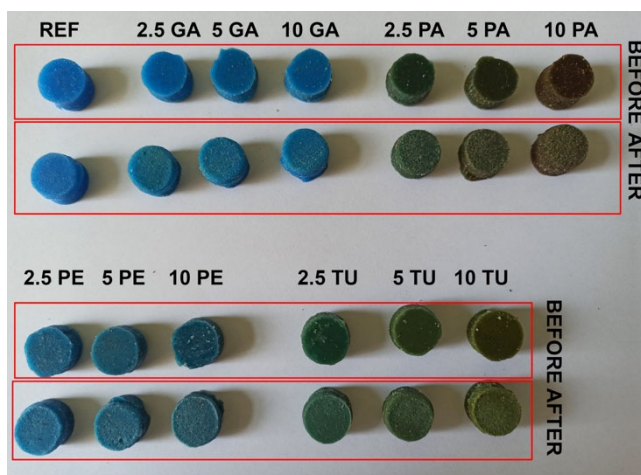


Fig. 8. Samples before and after the abrasive wear test

4. Discussion

The article's authors focused on the use of spices commonly used as fillers in composite materials. The studies are not intended to waste food and call for using spices in producing engineering materials instead of in preparing meals. The spices used were collected in the authors' homes, remained in open packages for an extended period, and lost their original taste and aroma.

The maximum load and stress at break measurements decreased with added filler for the tensile test. A similar trend was observed for the hardness measurement. With added filler, the hardness of the material decreased. The abrasive wear tests also showed a tendency in which the most minor abrasive wear occurred with materials containing 2.5% of the filler. With higher filler mass fractions in the composite, the abrasive wear of the composite material increased. Among all tested composites, those incorporating turmeric had the best mechanical

properties and the lowest abrasive wear (except for the 2.5 GA sample). It was due to the fineness of turmeric grains, which had the smallest diameter grains. This positively affected the turmeric filler properties relative to other composites.

The authors believe it is because agglomerates may form in the composite material as the filler content increases, reducing their abrasion resistance during abrasion wear. It is assumed that the abrasive wear of a composite material in which the filler grains are evenly distributed is lower than that in which the filler grains form agglomerates or, more importantly, irregular structures.

5. Conclusions

The tests sought to demonstrate whether adding spices in gastronomy could improve composite materials' physical and mechanical properties with silicone as the matrix. All composites showed lower mechanical property values relative to the silicone matrix. Adding waste spices also reduced the hardness and density of the composite materials close to the silicone used as the matrix. Composite materials increased resilience concerning neat silicone, except in the 2.5 PE material, whose resilience equalled the reference sample. All composite materials increased resistance to abrasive wear by the Schopper-Schlobach method. Adding spices from 2.5 to 10% by mass increased the abrasion resistance compared to silicone without adding filler. The most significant decrease in abrasion resistance was recorded for 2.5 GA and 2.5 TU. A decrease in abrasive wear by over 40% was noted here relative to neat silicone.

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Additional information

The authors declare no conflict of interest. All authors contributed equally to this study as the co-first authors. All the co-authors have approved the final version of the manuscript.

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