Journal of
Composites Science

Article

Effect of Silicon Nanoparticles on Moisture Absorption and Fracture Toughness of Polymethyl Methacrylate Matrix Nanocomposites

Mohammad Ali Golshokouh [1], Nima Refahati [1,*] and Pouyan Roodgar Saffari [2]

[1] Department of Mechanical Engineering, Damavand Branch, Islamic Azad University, Damavand P.O. Box 3971878911, Iran
[2] Department of Civil Engineering, Faculty of Engineering, Thammasat School of Engineering, Thammasat University, Pathumthani 12120, Thailand; poyan.safari31@gmail.com
* Correspondence: refahati@damavandiau.ac.ir or nima.refahati1978@gmail.com

Abstract: The effect of silicon nanoparticles with different percentages (2, 5, 7, and 10 wt.%) on moisture absorption in environments with different pHs (5, 6, 7, 8, 9) as well as fracture toughness of polymethyl methacrylate is discussed. The samples were prepared using pressure molding. Fracture strength was tested via the three-point bending method according to the ASTM D5045 standard and moisture absorption rate according to the absorption test according to the ASTM D570 standard. SEM images show that up to 7%, the dispersion of silica nanoparticles is acceptable, but the homogeneity is not acceptable at 10%. The results indicate that the increase in silica nanoparticles has improved the fracture toughness of the manufactured parts. The highest fracture toughness improvement is about 57% in the optimal state at 5%. Also, increasing silica nanoparticles increased the moisture absorption in the produced samples. In addition, as the acidic or base of the liquid moves to neutral, the reaction between the base polymer molecules and the test liquid decreases and, so, the moisture absorption also increases.

Keywords: PMMA; silicon nanoparticles; fracture strength; moisture absorption

Citation: Golshokouh, M.A.; Refahati, N.; Saffari, P.R. Effect of Silicon Nanoparticles on Moisture Absorption and Fracture Toughness of Polymethyl Methacrylate Matrix Nanocomposites. *J. Compos. Sci.* **2024**, *8*, 69. https://doi.org/10.3390/jcs8020069

Academic Editors: Francesco Tornabene and Thanasis Triantafillou

Received: 19 December 2023
Revised: 27 January 2024
Accepted: 8 February 2024
Published: 9 February 2024

1. Introduction

PMMA (polymethyl methacrylate) is used in medical and dental applications due to its high mechanical properties. The synthetic process is not difficult to handle and less expensive than other polymers. Also, the biocompatibility condition of PMMA is acceptable [1]. Although PMMA has lower abrasion resistance, hardness, and toughness than amalgam, due to its low cost, appearance, desirable color, allowable thermal stability, and high flexibility, it is rapidly replacing amalgam in dental applications [2–4]. Any restorative material or denture must have sufficient mechanical ability to function properly against the pressures in the oral cavity for a long time [5,6]. Fractures of teeth made of PMMA are very common and are a serious problem for their use as dental prostheses [7]. Research shows that between 63% and 68% of dentures within a few years of being made are broken by accidentally falling on a hard surface outside the mouth [8]. Most denture fractures are due to a weakness in the base polymer. Dental composites are subjected to various stresses during their operation, including compressive, tensile, and shear stresses [9]. Therefore, studying the mechanical properties of these materials to use them in clinical contexts is very important [10]. To improve the properties of PMMA, researchers have evaluated the effect of various factors, including fillers, matrices, as well as the interface between the matrix and the filler, but the results show that their properties have not changed significantly with changes in the matrix of composites. Therefore, studies have focused on fillers [11]. The properties of nanofiller-reinforced composites depend on the size, shape, type, and concentration of particles added [12,13].

In recent years, several nanoparticles, such as ZrO_2, SiO_2, TiO_2, and diamond nanoparticles, have been used in an attempt to enhance the physical and mechanical properties of PMMA [14–18].

Silicon dioxide nanoparticles, also known as silica nanoparticles or nano-silica, are one of the most common mineral fillers [19–22], and due to their molecular stability, low toxicity, and ability to bond with a wide range of molecules and polymers, as well as antimicrobial properties [23,24], are the basis of research in the field of biomedicine. Silica nanocomposites weigh less than conventional composites, and this factor has made them a suitable alternative to polymers in various applications [25,26].

Silica nanoparticles are used to improve the mechanical properties, adhesion, durability, and abrasion resistance of polymers. Silica nanoparticles generally have an amorphous and hollow structure [27,28]. In some cases, the crystalline structure of nano-silica is available, which is usually impossible to detect by X-ray tests. The proper shape for suspension silica nanoparticles is spherical [29]. This spherical shape is available by the synthesis in an ammonia medium. Most silica nanoparticles are used for industrial applications with one size (suspension) and two sizes of hydrophobic (oil and water separation).

Due to the presence of hydroxyl groups on the surface of silica nanoparticles, the agglomeration ability of these particles increases when combined with polymer composites, and this makes it difficult to prepare PMMA composites reinforced with silica nanoparticles [30]. To overcome the existing problems, many studies have considered the modification of silica nanoparticles' surfaces by chemical interaction because it leads to a much stronger interaction between modifiers and silica nanoparticles. Chemical methods include modification with modifiers or by bonding polymers. Silane agents, the most famous of which are gamma-Methacryloxypropyltrimethoxysilane, are capable of hydrolysis and can reduce hydroxyl groups on the surface of silica nanoparticles [31]. As a result, it creates a better bond between the filler and the base polymer [32]. This reduces the accumulation of silica nanoparticles when combined with the base polymer.

Many researchers have studied the mechanical behavior of PMMA and SiO_2 multilayer composites. Zhen et al. [33] and Balso et al. [34] found that with an increase in silica nanoparticles from 0 to 5 wt.%, the tensile strength and elongation in the fracture of composites have been improved, and also the flexural strength and elastic modulus have been significantly increased. Fatalla et al. [35], observed a slight increase in impact strength, transverse strength, and hardness for 5 wt. % of silica nanoparticles. Siot et al. [36] believe that, although silica nanoparticles increase the static modulus and yield strength of composites, the final properties of the composite are strongly influenced by the dispersion properties of the nanoparticles, and the better performance of the silica filler is related to its regular dispersion. Mussatto et al. [37], evaluating the mixing effect of modified and unmodified silica nanoparticles, concluded that with the addition of silica nanoparticles, the flexural strength of the composite in both modified and unmodified modes was reduced by 20 to 27%, and this reduction was not dependent on the amount of filler and the nanoparticle composition method, Also, nano-silica surface modification did not cause any major improvement in the mechanical behavior of the composites, but it seemed to improve the dispersion because the surface roughness was reduced. Salman et al. [38] showed significant improvements in impact strength, transverse strength, and hardness by adding silica nanoparticles to PMMA at 3%, 5%, and 7% wt.

Topouzi et al. [39] observed a significant increase in fracture toughness by adding modified and unmodified silica nanoparticles to PMMA but concluded that increasing the filler concentration reduces the fracture toughness, and no significant differences were reported between modified and unmodified fillers. Kundie et al. [40] investigated the effects of micro- and nano-scale particles on the mechanical properties of dental composites, concluding that the effects of nanoparticles strongly depend on factors, such as the type and mechanical properties of mineral nanofillers, uniform dispersion of nanofillers in the polymer matrix, the volume fraction of filler particles, and the type of silane used.

Although many studies have been performed on particle composites and their physical and mechanical properties, in these studies, some factors such as the percentage of filler particles and the environment to which the material is exposed have received less attention. Particularly, in the case of particulate composites used in dentistry (dental composites), little is known about the relationship between properties and structure, and the acidic or alkaline environments to which the composite is exposed for use as a denture have not been studied. According to studies, water absorbed due to the lubricating effect reduces mechanical properties, such as hardness, transverse strength, and fatigue limit [41]. Water absorption also causes volumetric expansion of the dental specimen and occlusal changes [42]. Resizing and exposing dental specimens to internal stresses cause cracks and fractures in dentures [43]. Therefore, it is important to study the environmental impact to which these samples are exposed. Karabla et al. [44] observed that the amount of water absorbed increases as the particle size of the fillers decreases. To modify the surface of silica nanoparticles, an acrylate cylinder modifier, ϒ-MPS (gamma-methacryloxy propyl trimethoxy silane), is the most widely used silane coupling agent in dental composites [45–47] and was prepared from Sigma Aldrich, Schnelldorf, Germany.

In this research, first, the samples were prepared. In the next step, by simulating the environment in which this type of material is placed, we aim to have a better understanding of their behavior. For this purpose, we prepared different solvents with different pHs, then placed the produced samples in a pH range 5 to 9 (normal pH range of human mouth) and compared the amount of moisture absorption in the samples. The aim of the current research is to find the best percentage of nano-silica to obtain better quality.

2. Experiments

2.1. Materials

In this experimental study, metamethyl acrylate polymer (MMA) from Taiwan Chemical Company with a specific gravity of 1.19 gr/cm^3 was used as the polymer substrate. This material is a product of silica nanoparticles made by Acrosun Spain with a size of 20 nm and a purity of 99.5%, used as reinforcing material. To reduce the viscosity, triethylene glycol dimethacrylate (TEGDEMA) diluting resin made by the German company Evonic was used. The molding of the samples was conducted using a two-piece mold made of aluminum with dimensions of 4 × 50 × 6 mm. Also, to prevent the samples from sticking inside the mold, TECHNOSIL acrylic separating liquid was used.

2.2. Sample Production

A vacuum mixer was used to achieve a uniform combination of silica and polymethyl methacrylate nanoparticles. Studies show that the main problem of using silica nanoparticles in dental composites is the accumulation of silica nanoparticles together when combined with polymethyl methacrylate. This leads to the formation of micrometer particles and reduces the mechanical properties of nanocomposites, so filler particles need surface modification to reduce the formation of agglomerates and better distribution of filler particles within the matrix.

In this research, the surface of silica nanoparticles is modified using an acrylate silane agent, ϒ-MPS, and hydrolysis-density method. For this purpose, a solution of water and alcohol with a weight ratio of 70.30 is used for hydrolysis of ϒ-MPS for 30 min. The pH is regulated by acetic acid at about 3–4. The nanoparticles are then added to this solution and stored at room temperature for one week. The nanoparticles are washed with alcohol, and then, in ambient air, the remaining alcohol is evaporated and the nanoparticles are dried.

To produce any desired composition of nanocomposites in all ten samples of each wt.%, first, the modified silica nanoparticles with a certain weight percentage (2, 5, 7, and 10% wt.) are mixed with monomer methyl methacrylate powder in a mixer. The milling is carried out for 12 min with a frequency of 20 Hz and a speed of 100 revolutions per minute.

To produce nanocomposite samples, a mixture of methyl methacrylate powder and milled silica is mixed with diluents to triethylene glycol dimethacrylate (TEGDMA). This is performed mechanically at ambient temperature using a small mixer. The Samples names and related SiO_2 percentage can be seen in Table 1.

Table 1. Grouping name of samples based on the percentage of nano-silica.

Sample	A	B	C	D	E
SiO_2(%)	0	2	5	7	10

Figure 1 shows the die in which the samples were produced. The mold must be prepared to prevent the dough from sticking to the mold. For this purpose, the surface of the mold is covered with a layer of acrylic separating liquid. After the mold surface is dry (this takes about 3 h), enter the paste-shaped liquid into each part of the mold and then tighten the two parts of the mold together. In the next step, to bake the dough inside the mold, we keep the mold immersed in hot water at a temperature of 80 °C for 50 min, then take out the mold and let it stand at room temperature. We then cool and take the sample out of the mold. After cooking, the samples are polished with 600 grains and then 1000 grains with the help of sandpaper.

Figure 1. Die used to produce the samples (100 × 80 mm includes 4 × Φ6 mm).

It should be noted that for each weight percentage, 10 samples were produced, and, finally, 5 healthier samples with the fewest defects such as fewer bubbles and smoother surfaces were selected theoretically. Also, ten other samples without a percentage of silica reinforcement were produced to compare with the reinforced samples, from which we selected five better samples.

2.3. SEM Tests

After producing reinforced samples, the first SEM test is performed to compare the quality of reinforced production samples with samples without modified silica nanoparticles. SEM captures images of different surfaces of the sample transversely to evaluate the chemical composition gradient on the surfaces of the samples at a distance of as little as 1 µm, allowing us to obtain a better analysis of the type and distribution of filler particles on the surface of the PMMA matrix.

2.4. Moisture Absorption Test

The moisture absorption test is performed under the ASTM D570 standard. Since the pH range of a healthy adult's mouth is between 5 and 9, we performed moisture absorption tests in this interval. To increase the contact surface of the samples with the liquid and also the effect of the environment on the pH of the test liquid, we used closed containers, placing each sample in a container and identifying the samples by numbering the containers.

The recommended duration for the moisture absorption test according to ASTM D570 is 24 h. The amount of moisture absorption is determined by measuring the weight of the samples before and after placing them in the liquid bath. It is worth noting that during the experiments, the liquid bath at a temperature of 20 ± 2 °C is in temperature equilibrium with the environment, and a digital scale with an accuracy of 0.001 g is used to weigh the samples.

If M1 is the primary mass of the sample and M2 is the second mass of the sample, then the amount of moisture absorption is determined using Equation (1).

$$\frac{\Delta w}{W} = \frac{M2 - M1}{M1} \tag{1}$$

2.5. Fracture Toughness Test

The fracture toughness test of nanocomposites was performed according to the ASTM D5045 standard. In this test, a general test device with a speed of 0.1 mm/min was used. The loading of the composite parts made to determine the fracture toughness is schematically shown in Figure 2, in which a rectangular piece with an edge crack is subjected to a three-point bending load with a support distance of s2.

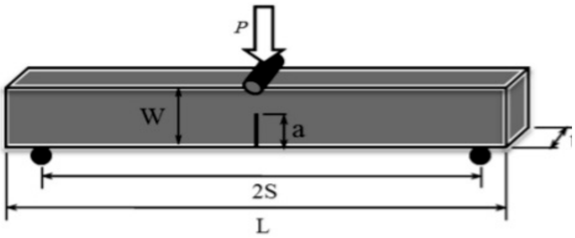


Figure 2. Schematic of the test sample.

Before performing the fracture toughness test to create initial cracks in the samples, the rectangular pieces were cut using a thin saw blade to a thickness of 0.25 mm. The tips of the created notches were sharpened by pressing a razor blade.

In this way, the grooves created in the samples can be assumed with acceptable accuracy as a sharp crack to perform the fracture toughness test. The length of the final edge crack created in the middle of each specimen is 3 mm. As a result, the crack length ratio without the a/w dimension in all experiments of this study is a constant value of 0.5. Before performing the test, the samples are stored for 24 h in standard conditions of a laboratory environment (temperature 23 ± 2 °C and humidity 50 ± 5%). Then, the cracked specimens are placed inside the three-point bending fixture supports (with a support distance of 2 s = 40 mm) in the testing machine and loaded monotonically at a constant rate of 1 mm/min. The loading continues until the complete failure of the parts and the growth of the crack from the tip of the initial crack, and, finally, the load–displacement curve of each sample is obtained (Figure 3).

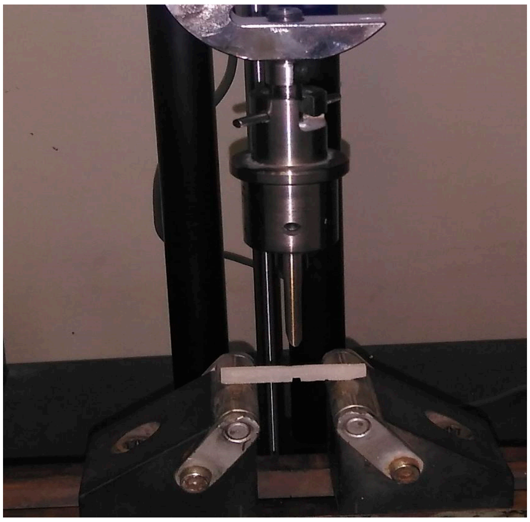


Figure 3. Sample tested in bending tester.

Using the critical load proportional to the breaking moment of each specimen (F_S), the K_{IC} fracture toughness of dental composite specimens was calculated from Equation (2).

$$K_{IC} = f(a/w)\frac{F_s}{t\sqrt{w}}MPam^{\frac{1}{2}} \tag{2}$$

where f (a/w) is a geometric coefficient that depends on the type of part and the load applied to it. This coefficient is calculated by modeling the finite element method of the tested part for conditions s/w = 3.3 and a/w = 0.5. Also, t is the thickness (in mm) and w is the width (in mm).

$$f\left(\frac{a}{w}\right) = \frac{\left(2+\frac{a}{w}\right)\left(0.886+4.64\frac{a}{w}-13.32\frac{a^2}{w^2}+14.72\frac{a^3}{w^3}-5.6\frac{a^4}{w^4}\right)}{\left(1-\frac{a}{w}\right)^{\frac{1}{2}}} \tag{3}$$

3. Results and Discussion

3.1. SEM Test

Figure 4 shows SEM data of TESCAN VEGA CZECH taken from the surface of the sample without filler, while in Figure 5, samples with different weight percentages can be seen. Figure 4 can be used for evaluation of the volume fraction of the additive, but for more detailed information about additive roll, XRD can be used. SEM images show that the higher the filler percentage, the greater the accumulation of filler particles. The highest number of agglomerates is in samples with 10% wt., where the formation of agglomerates is due to the presence of hydroxyl groups on the surface of silica nanoparticles, which prevents proper bonding of the base polymer (PMMA) with silica nanoparticles.

Although the surface of silica nanoparticles has been modified, the accumulation of nanoparticles and the formation of agglomerates can still be seen in some parts of the composite surface.

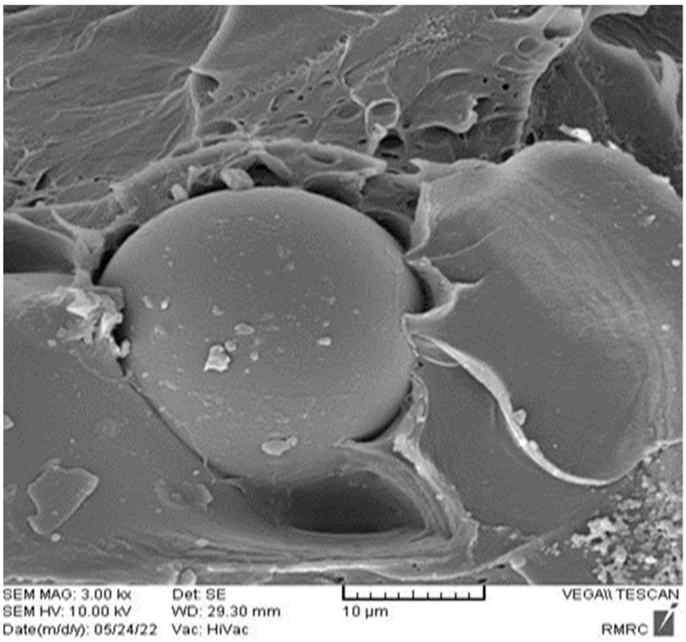
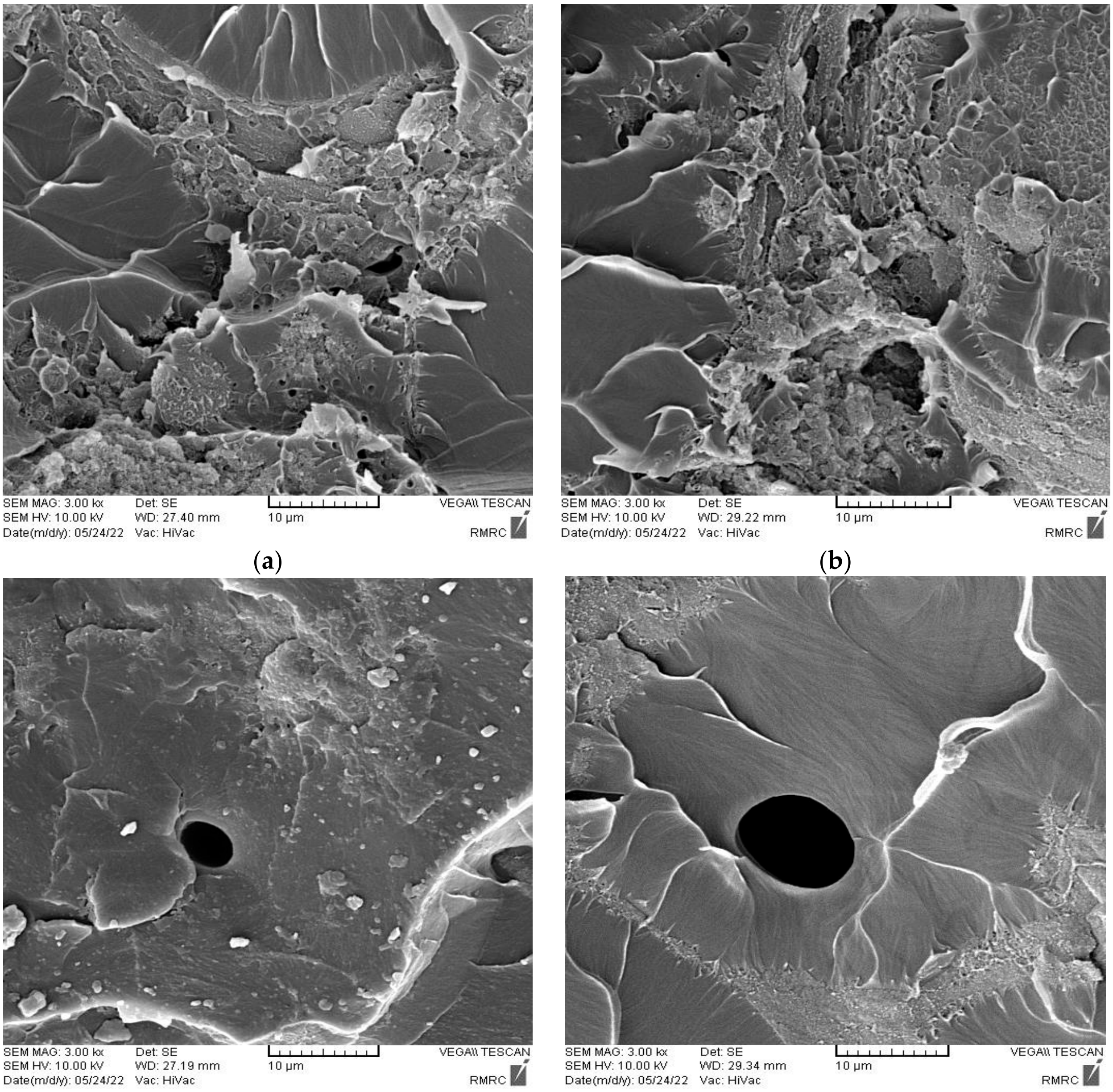


Figure 4. SEM of a sample without silica filler.



(**a**) (**b**)

(**c**) (**d**)

Figure 5. SEM of samples and modified silica filler with different percentages: (**a**) 2% by weight of filler; (**b**) 5% by weight of filler; (**c**) 7% by weight of filler; (**d**) 10% by weight of filler.

3.2. Moisture Absorption

Polymer composites filled with hard mineral particles in micro or nano dimensions show different behaviors with increasing particle percentages. The adsorption of water in the polymer is the result of the molecular polarity of the polymer, the unsaturated bonds of the molecules, or the unbalanced intermolecular forces in the polymers. According to ISO 1567, water absorption for thermal and self-cooking materials should not exceed 32 micrograms per cubic millimeter. The amount of moisture absorption in the samples (Table 2) shows that with increasing the percentage of nano-silica, the amount of moisture absorption in the samples decreases. The reason for this may be related to the amorphous structure of the underlying polymer. The distance between PMMA molecules is greater in the absence of silica filler particles than in the case in which nanoparticles are added. Silica nanoparticles fill the gaps between molecules so that as the amount of nano-silica in the base polymer increases, the distance between the molecules decreases, the polymer structure becomes more regular, and, as a result, the amount of moisture absorption decreases. It shows that in 10% by weight of nano-silica, we have the lowest amount of moisture absorption.

Table 2. Moisture absorption rate of samples.

Sample \ pH	5	6	7	8	9
A	0.0174	0.0239	0.0644	0.0196	0.0177
B	0.0152	0.0195	0.0310	0.0095	0.0075
C	0.0146	0.0189	0.0189	0.0092	0.0070
D	0.0123	0.0188	0.0167	0.0087	0.0066
E	0.0023	0.0014	0.0059	0.0046	0.0033

On the other hand, by comparing the samples that are located at different pHs, we find that the slower the pH of the liquid bath moves to the neutral, the higher the absorption rate. The maximum amount of moisture absorption for different pHs is pH = 7. The chemical structure of PMMA shows that the haplomer atoms do not tend to react with acidic and alkaline solvents, and, as a result, by being in these media, their adsorption decreases. On the other hand, silica nanoparticles based on the base polymer do not tend to react with acids and bases. As a result, we see that the more acidic and alkaline the liquid, the lower the absorption (Figure 6).

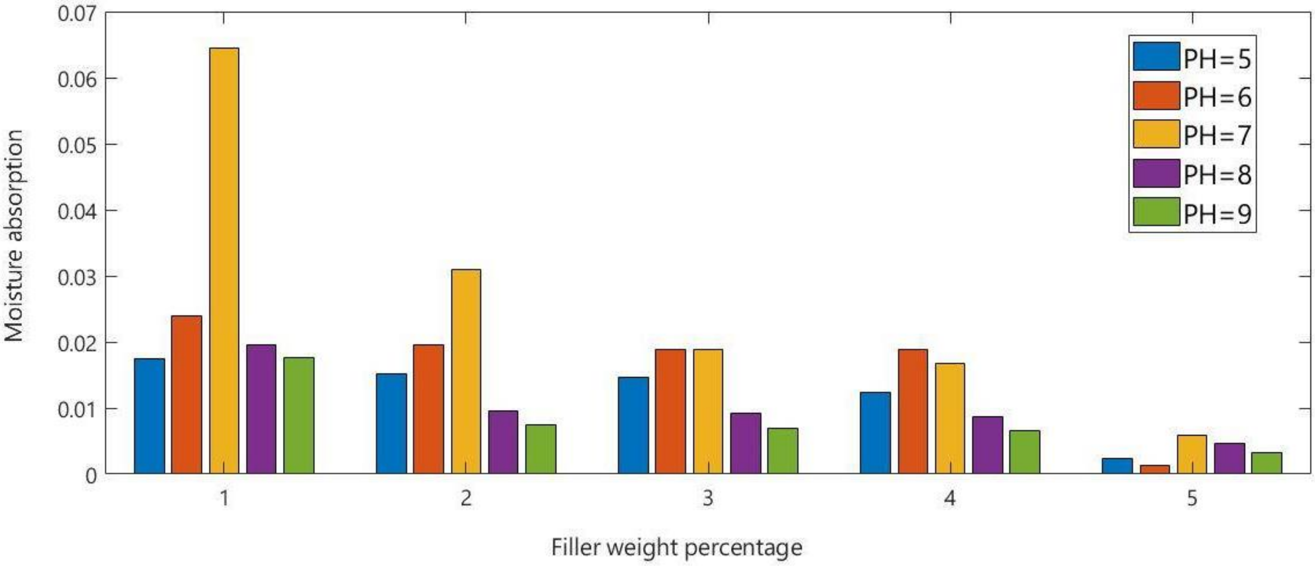


Figure 6. Comparison of moisture absorption for samples at different pHs.

3.3. Fracture Toughness

The fracture toughness of a material indicates the material's resistance to crack propagation when subjected to loading and is directly related to the total energy absorbed to deform the plastic. The cross-sectional areas of the specimen after loading are shown (Figure 7).

Table 3 shows the results of sample loading. Examination of the test results shows that increasing the amount of silica filler increased the fracture toughness of the samples, but when the amount of filler exceeds 5% by weight, not only does the fracture toughness not improve but it also decreases, so that in the case of 10, the weight percentage of silica and the fracture toughness of the samples are less than the samples without silica filler. Of course, fracture toughness in composites is a complex process, and its increase or decrease cannot be considered only due to the number of fillers. The study of the failure mechanism in composites shows that the type of filler, fracture process, protrusion and thickness of the crack tip, the yield strength of the matrix plastic, the structure of the composite, the separation of the joint particle, the matrix, and, especially, the aggregation of filler particles considers other factors, which can affect the fracture toughness test results of composites. Because polymers have an amorphous and irregular structure, the increase in filler particles reduces the distance between the particles, and this factor reduces the crack growth rate; the long crack growth path leads to an increase in the fracture toughness of the composite. Researchers attribute the increase in fracture toughness of composites due to the addition of filler to the interaction of the crack tip with the filler phase. On the other hand, increasing the amount of filler, the accumulation of particles increases and causes the formation of agglomerates, which accelerates the growth of cracks and, consequently, reduces fracture toughness, as can be seen in Figure 8. Agglomerates act as weak regions in the polymer matrix and collapse in the presence of the applied load, which results in the initial failure of the material and, thus, a reduction in fracture toughness. In addition, the presence of agglomerates in the polymer matrix leads to contraction of the contact area and the formation of defects. This phenomenon prevents the effective transfer of the applied load from the matrix to the filler nanoparticles, which leads to an inevitable reduction in the fracture toughness of the composite material.

Although in this study, we tried to create a uniform distribution of silica nanoparticles on the surface of the base polymer by modifying the filler particles, at high weight percentages, the accumulation of nanoparticles forms agglomerates.

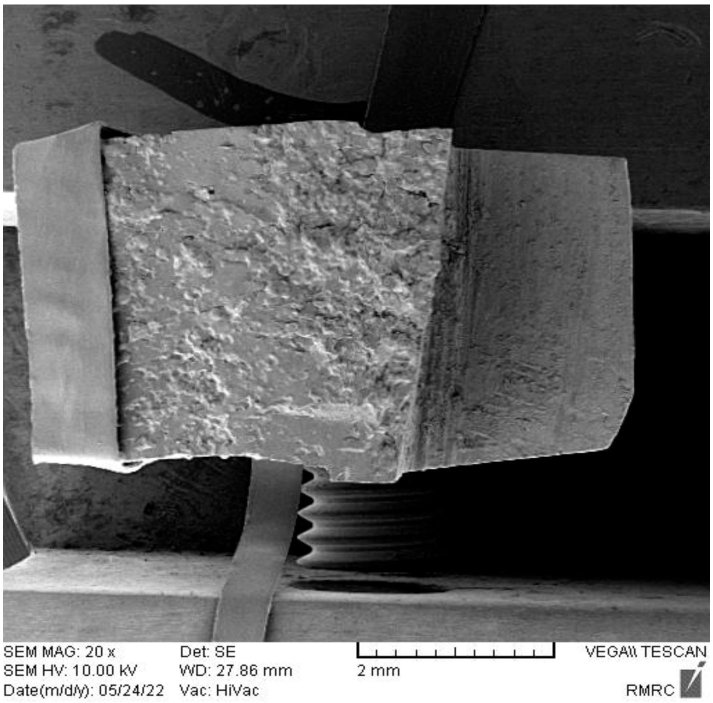


Figure 7. Sample failure cross-section.

Table 3. Results of the three-point bending test.

Sample	F (N)	Bending Strain	K_{IC} (Mpa/$\sqrt{}$m)
A	140.97	0.06	66.25
B	151.83	0.06	70.97
C	221.97	0.07	104.3
D	175.16	0.06	82.32
E	119.9	0.05	56.35

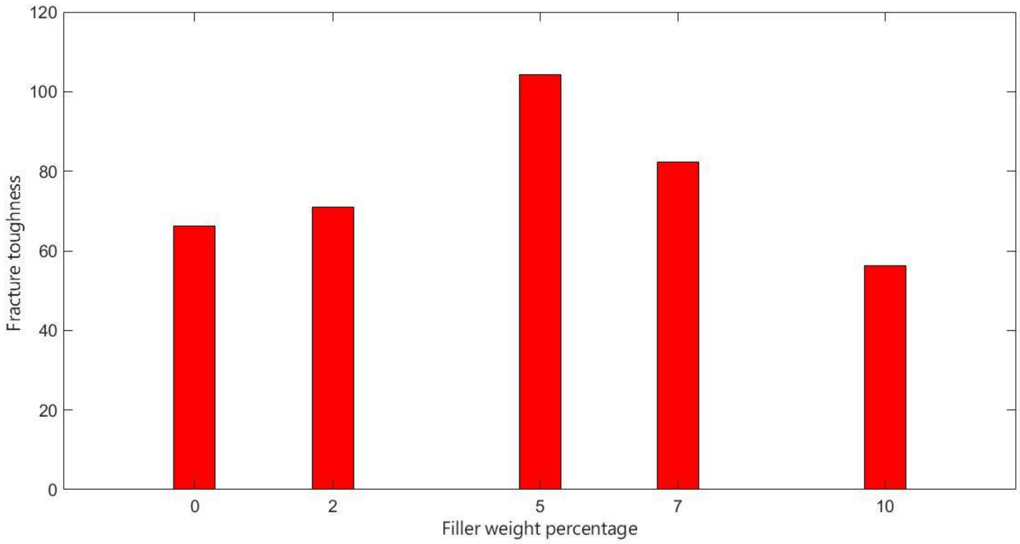


Figure 8. Fracture toughness (Mpa/$\sqrt{}$m) for samples with different filler percentages.

4. Conclusions

In this research, we investigated the effect of adding silica nanoparticles on the mechanical properties and moisture absorption of PMMA-based dental samples. After performing experiments and analyzing the obtained values, it was found that increasing the filler percentage caused the accumulation of nano-silica particles and subsequent agglomerate formation, so that the highest number of agglomerates is observed in composites with 10% wt. nano-silica. Also, increasing the amount of silica filler improves the fracture toughness, but there is an optimal limit for this increase so that increasing the filler weight by more than 5% negates the slope of the fracture toughness diagram. And, after the optimal state (5% wt. of filler), increasing the silica filler reduces the fracture toughness of the composites. So, at 10% wt., not only is the toughness unimproved but it is less than the case where PMMA lacks a silicon filler. Increasing the filler reduces moisture absorption. The tested samples react to acidic and alkaline environments so that as the amount of acid or liquid base becomes neutral, the absorption of moisture in dental samples increases. Also, in samples less than 10% wt., the rate of moisture absorption in acidic media is higher than in alkaline media, but at 10% wt., the rate of moisture absorption in alkaline media is higher than in acidic media.

Author Contributions: Conceptualization, N.R; methodology, M.A.G. and N.R; Validation, N.R., P.R.S. and M.A.G.; Investigation, P.R.S., N.R. and M.A.G.; Resources, P.R.S.; data curation, N.R. and P.R.S.; writing—original draft preparation, M.A.G. and N.R.; Writing—review and editing, N.R. and P.R.S.; Visualization, N.R. and M.A.G.; project administration, N.R. and M.A.G. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Data sharing is applicable to this article by requesting corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Hata, K.; Ikeda, H.; Nagamatsu, Y.; Masaki, C.; Hosokawa, R.; Shimizu, H. Dental Poly (methyl methacrylate)-Based Resin Containing a *Nanoporous silica* Filler. *J. Funct. Biomater.* **2022**, *13*, 32. [CrossRef]
2. Mousavi, A.; Aliha, M.R.; Imani, D.M. Effects of biocompatible Nanofillers on mixed-mode I and II fracture toughness of PMMA base dentures. *J. Mech. Behav. Biomed. Mater.* **2020**, *103*, 103566. [CrossRef]
3. Abutalib, M.M.; Rajeh, A. Influence of MWCNTs/Li-doped TiO_2 nanoparticles on the structural, thermal, electrical and mechanical properties of poly (ethylene oxide)/poly (methylmethacrylate) composite. *J. Organomet. Chem.* **2020**, *918*, 121309. [CrossRef]

4. Earar, K.; Solomon, O.; Topor, G.; Constantin, I.; Beznea, A.; Ciprian, D.; Anghel, L.; Ștefănescu, V.; Fotea, S.L.; Feier, R.D.; et al. Comparison study on the influence of Al_2O_3Nanoparticle size and ternary hybrid on several properties of a PMMA denture composite. *Mater. Plast.* **2021**, *58*, 119–129. [CrossRef]
5. Gad, M.M.; Abualsaud, R.; Khan, S.Q. Hydrophobicity of denture base resins: A systematic review and meta-analysis. *J. Int. Soc. Prev. Community Dent.* **2022**, *12*, 139. [CrossRef]
6. Chladek, G.; Pakieła, K.; Pakieła, W.; Żmudzki, J.; Adamiak, M.; Krawczyk, C. Effect of antibacterial silver-releasing filler on the physicochemical properties of poly (methyl methacrylate) denture base material. *Materials* **2019**, *12*, 4146. [CrossRef] [PubMed]
7. Alhotan, A.; Yates, J.; Zidan, S.; Haider, J.; Silikas, N. Assessing Fracture Toughness and Impact Strength of PMMA Reinforced with Nano-Particles and Fibre as Advanced Denture Base Materials. *Materials* **2021**, *14*, 4127. [CrossRef]
8. Salih, S.I.; Oleiwi, J.K.; Mohamed, A.S. Investigation of mechanical properties of PMMA composite reinforced with different types of natural powders. *ARPN J. Eng. Appl. Sci.* **2018**, *13*, 8889–8900.
9. Kumar, V.; Kumar, L.; Sehgal, K.; Datta, K.; Pal, B. A comparative evaluation of effect of reinforced autopolymerizing resin on the flexural strength of repaired heat-polymerized denture base resin before and after thermocycling. *J. Int. Soc. Prev. Community Dent.* **2017**, *7* (Suppl. S2), S99. [CrossRef]
10. Naji, S.A.; Behroozibakhsh, M.; Kashi, T.S.; Eslami, H.; Masaeli, R.; Mahgoli, H.; Tahriri, M.; Lahiji, M.G.; Rakhshan, V. Effects of incorporation of 2.5 and 5 wt% TiO_2 nanotubes on fracture toughness, flexural strength, and microhardness of denture base poly methyl methacrylate (PMMA). *J. Adv. Prosthodont.* **2018**, *10*, 113–121. [CrossRef]
11. Zafar, M.S. Prosthodontic applications of polymethyl methacrylate (PMMA): An update. *Polymers* **2020**, *12*, 2299. [CrossRef]
12. Gad, M.M.; Fouda, S.M.; Al-Harbi, F.A.; Näpänkangas, R.; Raustia, A. PMMA denture base material enhancement: A review of fiber, filler, and nanofiller addition. *Int. J. Nanomed.* **2017**, *12*, 3801. [CrossRef]
13. Jiangkongkho, P.; Arksornnukit, M.; Takahashi, H. The synthesis, modification, and application of nanosilica in polymethyl methacrylate denture base. *Dent. Mater. J.* **2018**, *37*, 582–591. [CrossRef]
14. Tzetzis, D.; Tsongas, K.; Mansour, G. Determination of the mechanical properties of epoxy silica nanocomposites through FEA-supported evaluation of ball indentation test results. *Mater. Res.* **2017**, *20*, 1571–1578. [CrossRef]
15. Ergun, G.; Sahin, Z.; Ataol, A.S. The effects of adding various ratios of zirconium oxide nanoparticles to poly (methyl methacrylate) on physical and mechanical properties. *J. Oral Sci.* **2018**, *60*, 304–315. [CrossRef]
16. Elshereksi, N.W.; Muchtar, A.; Azhari, C.H. Effects of nanobarium titanate on physical and mechanical properties of poly (methyl methacrylate) denture base nanocomposites. *Polym. Polym. Compos.* **2021**, *29*, 484–496. [CrossRef]
17. Slane, J.; Vivanco, J.; Meyer, J.; Ploeg, H.L.; Squire, M. Modification of acrylic bone cement with mesoporous silica nanoparticles: Effects on mechanical, fatigue and absorption properties. *J. Mech. Behav. Biomed. Mater.* **2014**, *29*, 451–461. [CrossRef] [PubMed]
18. Tommasini, F.J.; Ferreira, L.D.; Tienne, L.G.; Aguiar, V.D.; Silva, M.H.; Rocha, L.F.; Marques, M.D. Poly (methyl methacrylate)-SiC nanocomposites prepared through in situ polymerization. *Mater. Res.* **2018**, *21*, e20180086. [CrossRef]
19. Cevik, P.; Yildirim-Bicer, A.Z. The effect of silica and prepolymer nanoparticles on the mechanical properties of denture base acrylic resin. *J. Prosthodont.* **2018**, *27*, 763–770. [CrossRef]
20. Canché-Escamilla, G.; Duarte-Aranda, S.; Toledano, M. Synthesis and characterization of hybrid silica/PMMA nanoparticles and their use as filler in dental composites. *Mater. Sci. Eng. C* **2014**, *42*, 161–167. [CrossRef] [PubMed]
21. Ikeda, H.; Nagamatsu, Y.; Shimizu, H. Preparation of silica–poly (methyl methacrylate) composite with a nanoscale dual-network structure and hardness comparable to human enamel. *Dent. Mater.* **2019**, *35*, 893–899. [CrossRef]
22. Silva, E.D.; Ribeiro, L.A.; Nascimento, M.C.; Ito, E.N. Rheological and mechanical characterization of poly (methyl methacrylate)/silica (PMMA/SiO_2) composites. *Mater. Res.* **2014**, *17*, 926–932. [CrossRef]
23. Gad, M.M.; Bahgat, H.A.; Edrees, M.F.; Alhumaidan, A.; Khan, S.Q.; Ayad, N.M. Antifungal activities and some surface characteristics of denture soft liners containing silicon dioxide nanoparticles. *J. Int. Soc. Prev. Community Dent.* **2022**, *12*, 109. [CrossRef]
24. Bai, X.; Lin, C.; Wang, Y.; Ma, J.; Wang, X.; Yao, X.; Tang, B. Preparation of Zn doped mesoporous silica nanoparticles (Zn-MSNs) for the improvement of mechanical and antibacterial properties of dental resin composites. *Dent. Mater.* **2020**, *36*, 794–807. [CrossRef]
25. Thongchom, C.; Refahati, N.; Roodgar Saffari, P.; Roudgar Saffari, P.; Niyaraki, M.N.; Sirimontree, S.; Keawsawasvong, S. An experimental study on the effect of nanomaterials and fibers on the mechanical properties of polymer composites. *Buildings* **2021**, *12*, 7. [CrossRef]
26. Rende, D.; Schadler, L.S.; Ozisik, R. Controlling foam morphology of poly (methyl methacrylate) via surface chemistry and concentration of silica nanoparticles and supercritical carbon dioxide process parameters. *J. Chem.* **2013**, *2013*, 864926. [CrossRef]
27. Kaseem, M.; Ur Rehman, Z.; Hossain, S.; Singh, A.K.; Dikici, B. A Review on Synthesis, Properties, and Applications of Polylactic Acid/Silica Composites. *Polymers* **2021**, *13*, 3036. [CrossRef] [PubMed]
28. Ali, U.; Karim, K.J.; Buang, N.A. A review of the properties and applications of poly (methyl methacrylate)(PMMA). *Polym. Rev.* **2015**, *55*, 678–705. [CrossRef]
29. Lee, D.W.; Yoo, B.R. Advanced silica/polymer composites: Materials and applications. *J. Ind. Eng. Chem.* **2016**, *38*, 1–2. [CrossRef]
30. Kim, G.H.; Hwang, S.W.; Jung, B.N.; Kang, D.; Shim, J.K.; Seo, K.H. Effect of PMMA/Silica hybrid particles on interfacial adhesion and crystallization properties of Poly (Lactic Acid)/Block acrylic elastomer composites. *Polymers* **2020**, *12*, 2231. [CrossRef] [PubMed]

31. Zou, H.; Wu, S.; Shen, J. Polymer/silica nanocomposites: Preparation, characterization, properties, and applications. *Chem. Rev.* **2008**, *108*, 3893–3957. [CrossRef]
32. Ali Sabri, B.; Satgunam, M.; Abreeza, N.M.; N Abed, A. A review on enhancements of PMMA denture base material with different nano-fillers. *Cogent Eng.* **2021**, *8*, 1875968. [CrossRef]
33. Zhen, X.; Zhang, L.; Shi, M.; Li, L.; Cheng, L.; Jiao, Z.; Yang, W.; Ding, Y. Mechanical behavior of PMMA/SiO_2 multilayer nanocomposites: Experiments and molecular dynamics simulation. *Macromol. Res.* **2020**, *28*, 266–274. [CrossRef]
34. Balos, S.; Puskar, T.; Potran, M.; Milekic, B.; Djurovic Koprivica, D.; Laban Terzija, J.; Gusic, I. Modulus, strength and cytotoxicity of PMMA-silica nanocomposites. *Coatings* **2020**, *10*, 583. [CrossRef]
35. Fatalla, A.A.; Tukmachi, M.S.; Jani, G.H. Assessment of some mechanical properties of PMMA/silica/zirconia nanocomposite as a denture base material. *IOP Conf. Ser. Mater. Sci. Eng.* **2020**, *987*, 012031. [CrossRef]
36. Siot, A.; Léger, R.; Longuet, C.; Otazaghine, B.; Caro-Bretelle, A.S.; Azéma, N. Dispersion control of raw and modified silica particles in PMMA. Impact on mechanical properties, from experiments to modelling. *Compos. Part B Eng.* **2019**, *157*, 163–172. [CrossRef]
37. Mussatto, C.M.; Oliveira, E.M.; Subramani, K.; Papaléo, R.M.; Mota, E.G. Effect of silica nanoparticles on mechanical properties of self-cured acrylic resin. *J. Nanopart. Res.* **2020**, *22*, 317. [CrossRef]
38. Salman, A.D.; Jani, G.H.; Fatalla, A.A. Comparative study of the effect of incorporating SiO_2 nano-particles on properties of poly methyl methacrylate denture bases. *Biomed. Pharmacol. J.* **2017**, *10*, 1525–1535. [CrossRef]
39. Topouzi, M.; Kontonasaki, E.; Bikiaris, D.; Papadopoulou, L.; Paraskevopoulos, K.M.; Koidis, P. Reinforcement of a PMMA resin for interim fixed prostheses with silica nanoparticles. *J. Mech. Behav. Biomed. Mater.* **2017**, *69*, 213–222. [CrossRef] [PubMed]
40. Kundie, F.; Azhari, C.H.; Muchtar, A.; Ahmad, Z.A. Effects of filler size on the mechanical properties of polymer-filled dental composites: A review of recent developments. *J. Phys. Sci.* **2018**, *29*, 141–165. [CrossRef]
41. Zirak, M.; Vojdani, M.; Mohammadi, S.; Khaledi, A.A. Comparison of the water sorption and solubility of four reline acrylic resins after immersion in food-simulating agents. *J. Int. Soc. Prev. Community Dent.* **2019**, *9*, 40. [PubMed]
42. Ristic, B.; Carr, L. Water sorption by denture acrylic resin and consequent changes in vertical dimension. *J. Prosthet. Dent.* **1987**, *58*, 689–693. [CrossRef] [PubMed]
43. Rajaee, N.; Vojdani, M.; Adibi, S. Effect of food simulating agents on the flexural strength and surface hardness of denture base acrylic resins. *Oral Health Dent Manag.* **2014**, *15*, 1041–1047.
44. Karabela, M.M.; Sideridou, I.D. Synthesis and study of properties of dental resin composites with different nanosilica particles size. *Dent. Mater.* **2011**, *27*, 825–835. [CrossRef] [PubMed]
45. Karkanis, S.; Nikolaidis, A.K.; Koulaouzidou, E.A.; Achilias, D.S. Effect of Silica Nanoparticles Silanized by Functional/Functional or Functional/Non-Functional Silanes on the Physicochemical and Mechanical Properties of Dental Nanocomposite Resins. *Appl. Sci.* **2021**, *12*, 159. [CrossRef]
46. Kundie, F.; Azhari, C.H.; Ahmad, Z.A. Effect of nano-and micro-alumina fillers on some properties of poly(methyl methacrylate) denture base composites. *J. Serbian Chem. Soc.* **2018**, *83*, 75–91. [CrossRef]
47. de Menezes, B.R.; da Graça Sampaio, A.; da Silva, D.M.; do Amaral Montanheiro, T.L.; Koga-Ito, C.Y.; Thim, G.P. $AgVO_3$ nanorods silanized with γ-MPS: An alternative for effective dispersion of $AgVO_3$ in dental acrylic resins improving the mechanical properties. *Appl. Surf. Sci.* **2021**, *543*, 148830. [CrossRef]