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Conceptual Study of Non-Stick Cookware Coatings and the Effect of Some Corresponding Additives on Their Performance

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Abstract

In this study, the effect of some additives including Alumina (Al2O3), boron carbide (B4C), and different types of silicon carbide (SiC) on glossiness, impact resistance, pencil hardness, conical bent resistance, corrosion, humidity, and chemical resistance of non-stick coatings were studied. The results revealed that the glossiness of the coatings decrease by the addition of the additives, but the overall resistance of the coatings grow by adding these substances among which B4C additive had the best effect on the coating general performance.

Keywords: Non-stick coatings, cookware and bakeware, fluoropolymer, curing and sintering, mechanical strength

1. Introduction

Non-stick coatings are hydrophobic surfaces, which have low coefficients of friction. The most common material used in this industry is Poly Fluoro Carbon (PFC) polymers and particularly Poly Tetra Fluoro Ethylene (PTFE) because of its anti-stick characteristics, mechanical strength and low flammability could be taken into account as the best option for the non-stick industry. Besides the fluorocarbons, many other ingredients in non-stick cookware coating formulation play unique roles in the performance of the products which needs to be investigated in detail. Also, non-stick coatings are applied with different methods and the most common application methods are air spraying and roller methods. The fluorocarbon polymer coating preferably comprises a primer layer which is applied directly onto the surface of the substrate and one or more fluorocarbon polymer topcoat layers. An illustration of a non-stick coating has been shown in Figure 1. The performance of the coatings could be affected by many factors including the ingredients of the formulation, coating thickness, flash-off, and curing temperature and durations.

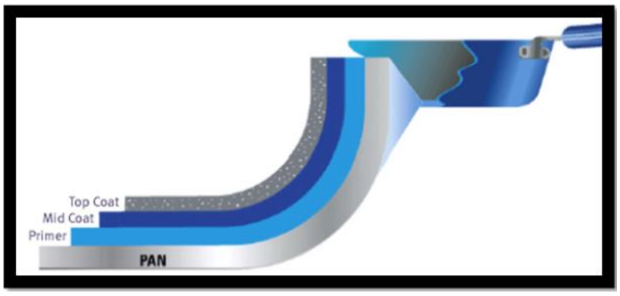


Figure 1. Coating layers of a non-stick cookware

1.1 Non-Stick Coatings

Non-stick coatings are hydrophobic surfaces, which have low coefficients of friction, high corrosion resistance, high dielectric strengths, higher temperature resistances compared to other polymers and a wide range of operational temperature. Fluoropolymers are among the oldest high-performance polymers and are of great commercial and scientific interest. Most of these are because of their anti-stick characteristics, mechanical strength, and low flammability. The most important disadvantage of PTFE is low wear resistance (Hatzikiriakos, 2012). Their most important uses are in electronics and electrical applications, chemical processing equipment, laboratory ware and tubing, material for roofing, and houseware (Ebnesajjad, 2015).

The term fluorocarbon polymer coating refers to a coating that is consist of conventional fluorocarbon polymers such as polytetrafluoroethylene (PTFE), polymers of chloro-tri-fluoroethylene (CTFE), fluorinated ethylene-propylene polymers (FEP), polyvinylidene fluoride (PVF), combinations thereof and the like. The composition of the fluorocarbon polymer coating is not critical, and a variety of fluorocarbon polymer compositions are conventionally used in the formulation of a non-stick coating. Non-stick coatings are plated with different methods like immersion, roller coating, thermal spray, curtain coating, and more are carried out in many ways. The most common coating method is the air spray method.

The PTFE with IUPAC ID: Poly (1, 1, 2, 2 tetrafluoroethylene) with the trade name of Teflon and (C2F4) n formula is a synthetic fluoropolymer compound, which was originally discovered by DuPont company in 1938. Today, Teflon™ coatings, and additives are used in paints, fabrics, carpets, home furnishings, clothing and so much. PTFE has one of the lowest coefficients of friction of any solid. It is non-reactive, partly because of the strength of carbon–fluorine bonds. PTFE application in cookware utensils has two important reasons: 1) being non-stick and 2) having a high thermal resistance. PTFE has excellent corrosion resistance, high-temperature resistance, anti-stick behavior, and low friction coefficient, widely used as an anti-corrosive material, sealing material, insulating material, non-sticky material, self-lubricating material, etc. (Lee et al., 2007; Beckford et al., 2016)

In the case of coatings, the fluorocarbon polymer coating preferably comprises a primer layer and one or more fluorocarbon polymer topcoats. The primer layer comprises a blend of fluorocarbon polymers and one or more adhesion promoting high-temperature binder resins, such as polyamide-imide resins (PAI), polyethersulfone resins (PES), and polyphenylene sulfide resins (PPS). PAI is an extremely strong and stiff plastic material. It is often used in elevated temperature environments where other thermoplastics would lose their mechanical properties (Yu et al., 2018). Polyethersulfone is an amorphous, transparent, and pale amber high-performance thermoplastic and is the most temperature-resistant transparent commercially available thermoplastic resin (McKeen, 2019). PPS is an engineering plastic, commonly used today as a high-performance thermoplastic. The maximum service temperature for PPS is 218 °C. Synthetic fiber and textiles derived from this polymer resist chemical and thermal attacks (Ebnesajjad and Morgan, 2019). Cookware coating with long-term scratch resistance has always been a desirable feature for cooks. The coatings also add colour accents to the kitchen. The first component, the basic layer or primer, consists of the fluoropolymer PTFE with integrated ceramic reinforcement. The layer in direct contact with the food is a specialty high-performance polymer containing PTFE (Chemours, 2017). The primer layer is applied directly onto the surface of the substrate. The non-vitreous inorganic oxide particles in the layer of the substrate enrich the surface of the substrate with bonding sites for the binder resins in the primer, thus improving the adhesion of the applied fluorocarbon polymer coating to the substrate. After the primer layer is applied, one or more fluorocarbon polymer topcoats are typically applied by conventional wet or dry techniques and then the entire fluorocarbon polymer coating is sintered on 400 °C (Rossi et al., 2022).

The current total volume of fluoropolymers produced in the world is in excess of 200,000 tones. Based on 2007 information total volume of fluoropolymers produced atthat time was 90,000 tones valued at over US$2 billion. PTFE is by far the most widely used fluoropolymer, accounting for some 70% of the total.2 the current average annual growth is estimated to be about 4% and is predicted to be about 5%. The typical cost of fluoroplastics ranges from US$13 to 45 per kilogram. Fluoroelastomers are priced somewhat higher than fluoroplastics and specialty fluoropolymers may cost up to several thousand US dollars per kilogram. (Drobny, 2007; Drobny, 2020).

1.2 The Effects of Different Additives in Non-stick Coating

PTFE's anti-wear applications have been somewhat limited by its poor wear resistance, which has led to the failure of anti-wear parts and films. Therefore, many researchers have attempted to reinforce PTFE using various fillers (Lee et al., 2007). Additives are very important in non-stick subjects and their effect can be very helpful for the goals of any product. Beckford et al. (2014) examined the PTFE + Au composite and PTFE properties and compared them with each other to evaluate the Au additive effect. These results indicate that the PTFE film has a significantly higher surface roughness than PTFE + Au. This difference in surface roughness results from the higher degree of melting in the composite film, which allows the film to spread and produce a more even film by Au nanoparticles. Durability test results indicate that by incorporating Au nanoparticles, even at the low concentration of 0.06 wt. %, it is possible to improve the wear resistance of the PTFE film twofold. Dynamic coefficient of friction is also influenced that much. Then it is concluded that adding Au nanoparticles of approximately 15 nm diameters, at a very low concentration of 0.06 wt. %, significantly improves the tribological properties. Also, test results show that incorporating Au nanoparticles as a filler not only increased the film's wear resistance but also shows no sign of delamination or adhesive wear (Beckford et al., 2014). It is known that besides PTFE, polyamide-imide and polyether sulfone are applied for light cookware utensils too. In general, to increase abrasion and scratch resistance of Teflon, nano-sized ceramic and/or metallic-based coatings powders are mixed. Glass fibers, carbon fibers, and nonferrous metallic and ceramic powders were studied as potential fillers for the enhancement of the wear resistance of PTFE. The wear process in the composites depends mainly on three factors: thermal stability, thermal conductivity, and the characteristics of the filler materials. But it was proven that these fillers induced a large frictional coefficient and abrasion of the fractured fiber and also the glossiness of the coatings is affected by hardening additives (Khedkar et al., 2002; Tevrüz, 1998; Tevrüz, 1999; Cheng et al., 2002).

The effects of additives not only on fluorocarbons are investigated but also on other coatings. In a study that Abenojar et al. (2009) had done, two different B_4C particle sizes were used for reinforcing an epoxy resin: one with an average particle size of 7 mm and the other with 23 mm. The results show that the addition of B_4C did not affect the degree of conversion of the epoxy during the curing and shifted the glass transition temperature of the epoxy to lower values and also increased the abrasive wear of the material. Epoxy-B_4C composites showed excellent bending strength, increasing with B_4C content and with the smaller particles (Abenojar J, 2009). The commercial PTFE-based coatings were found to provide poor release properties due to the presence of surface micro-cracks, which allowed epoxide penetration when cured under elevated pressure and temperature (Critchlow et al., 2006). In the work of Critchlow et al. (2006), Electroless Ni/PTFE composite coatings comprise a hard nickel–phosphorus matrix containing a very fine dispersion of PTFE particles. The matrix proved sufficiently robust for industrial applications and the low friction and surface energy provided by the embedded PTFE combined with macroscopic-scale surface roughness provided efficient mold release. According to the results, the PTFE composites provide a wide water contact angle because of low friction and also provide low-energy surfaces on metal substrates. After evaluating both samples of these coatings for use with metal tooling materials, it was found that when adhesives were cured under elevated pressure and temperature whilst in contact with the coatings, the subsequent release was poor or impossible. It is proposed that the porosities present in PTFE surfaces allow penetration of the. It results in resistance against corrosion and humidity and also reducing of the glossiness. In another study investigation of the physical-mechanical properties of PTFE, which was filled with different amounts of various materials, was done. The method of study was determination of frictional thermal stability by using the friction machine. The results showed that friction of thermal stability is increased with rising additives ratio (Kutelia et al., 2015). Thomas (1998) in a study, which was done with different fluoropolymers such as PTFE, PFA and FEP, the non-stick coating sprayed on the steel without primer and at a film thickness of 13- 14 µm. The films were cured at 430 °C and then the friction properties of the film were measured. As expected, it has been seen that PTFE has the lowest coefficient of friction but poor wear and adhesion properties. PFA and FEP adhered very well to the substrate but had a higher coefficient of friction. The 50/50 blends at the beginning of the test had a coefficient of friction, which was close to that of pure PTFE showing that PTFE might stratify. As the cylinder penetrated the film, the coefficient of friction increased to a maximum value until the contact became partly metal to- metal (Thomas, 1998). As it is obvious from the literature review, the additives have very important and effective influences on the non-stick coatings. Based on these reasons, in the present study the effects of several hardening additives have been investigated.

1.3 Non-stick Coatings and Health Issue

While PTFE is stable and nontoxic at lower temperatures, it begins to get deteriorated after the temperature of cookware reaches about 260 °C and decomposes above 350°C. PTFE cookware is considered as an insignificant exposure pathway to Perfluorooctanoic acid but in recent years the use of PFOA got banned from cookware use and afterward these coatings were approved by FDA for use in food contact coatings.

2. Materials and Methods

2.1 Materials

All the materials and equipment of this work were granted by THERNA non-stick coating producer which is a brand of KANAT paints and Coatings Company. The coating used in this study was a non-stick coating consist of a primer layer which provides the adhesion to substrate and the topcoat which grant the non-stick properties to the surface. The materials to be used in this kind of coatings consist resins, solvents, pigments and additives. The materials of the primer layer were added in two main stages including co-grind (premix) and letdown stages. In co-grind stage the materials to premix were: PW, Co-solvent, Defoamer, Surfactant, Filler, and Pigment and various hardening materials including Al_2O_3, B_4C, Black and Green SiC types which was the main subject of this study. In the letdown stage following materials were added: Surfactant, Resin, pH adjuster, Rheological agent, PW. Topcoat was fixed for all of the experiments with a fixed formulation. Aluminum panels were used as the substrate of the coatings. Five groups of this study were differed with their hardening material. In the samples of first group any hardening material was not used to have a reference to general comparison of the samples. The groups labeled as Table 1.

Table 1. Five coating groups of the study

	G.1	G.2	G.3	G.4	G.5
Additive	Blank	Black SiC	B4C	Green SiC	Al2O3

2.2 Methods

The tests were performed for 5 groups of samples. These tests were glossiness, impact resistance, pencil hardness, conical bent resistance, corrosion, humidity and chemical resistance (against sulfuric acid (H_2SO_4) 10%, sodium hydroxide (NaOH) 25% and diesel).

2.2.1 The preparation of the panels

The process of application of the coatings on the Aluminum panel is involved of 7 stages including sandblasting (grinding), cleaning of the surface, primer application on the surface, a 5-minute flash off in a 150 °C oven, striking of the top-coat, curing process on 375-425 °C oven and cooling down the coated panels. The samples of the dry composite coatings on the aluminum panels were shown in Figure 2.

Figure 2. The panel samples of each composite coating group

2.2.2 Glossiness evaluation

For evaluation of the gloss effects of the additives, glossmeter was chosen and the panels were studied for hardener effects on glossiness. In Figure 3, Novo Gloss trigloss device which was used in the study and is a gloss meter is shown.

Figure 3. Glossmeter

2.2.3 Pencil hardness

To determine the hardness of the surface from soft to hard as shown below, pencils with standard hardness must be used. The hardness level would be determined when the pencil leaves a permanent mark on the surface is defined as pencil hardness (ASTM D 3363). The apparatus of the pencil hardness measurement is given in Figure 4.

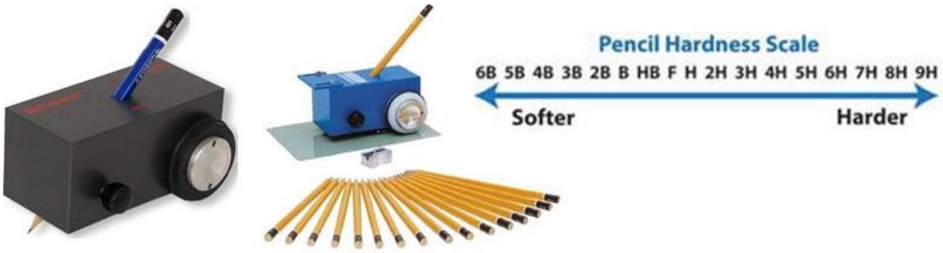


Figure 4. Pencil hardness

2.2.4 Universal impact tester

ASTM D 2794 provides a procedure for a rapid deform impact on a coating film and its substrate. If after the test some cracks were visible on the surface, then the test failed. In Figure 5, a universal impact tester (a) and a failed sample with cracks (b) have been shown.

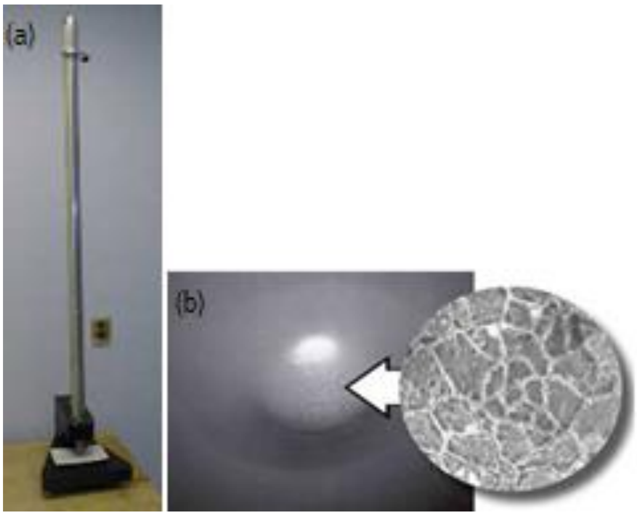


Figure 5. (a) A universal impact tester and, (b) a failed sample with cracks

2.2.5 Conical bent flexion

For evaluation of the flexibility of the samples with different hardeners, the conical bent flexion tester was chosen. It would be used to see if there are any positive or negative effects of the different additives. In Figure 6, a conical bent flexion apparatus is shown.

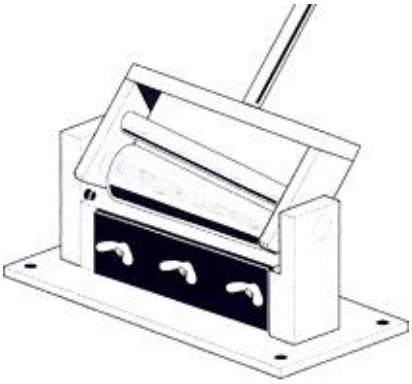


Figure 6. Conical bent flexion apparatus

2.2.6 Corrosion test (ISO 12944)

The corrosion test was done for all the five groups of the study. The method of ISO 12944 was followed in this appraisal. The samples were crossed over the middle and then placed in the corrosion cabinet. The preparation and placement of the panels in the corrosion cabinet have been shown in Figure 7.

Figure 7. Corrosion tests for the panels of the last session

"Corrosion tests in artificial atmospheres - Salt spray tests (TS EN ISO 9227)" was done on the samples. From each group, 3 repeated samples were examined. The samples were checked visually after 120 h, 240h, and 480h.

2.2.7 QCT test

The samples in 5 groups and 3 repeats were tested with a QCT test and observed after 120, 240 and 480h. Then cross-cut test was performed on the panels to see the potential effects of the humidity on the panels. In Figure 8, QCT test and the panels of the study were shown.

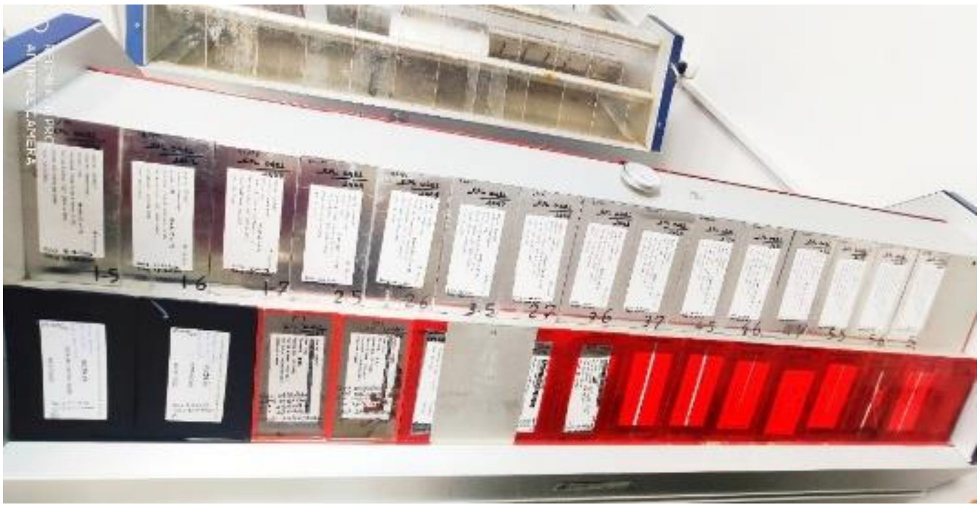


Figure 8. QCT test cabinet and the placed panels

The panels in the QCT test cabinet were placed fronted to the humidity to see the effects of the humidity on the coatings. After completion of 480h specimens were exposed to the cross-cut test.

2.2.8 Chemical resistance

Chemical resistance tests in this study were performed on the prepared panels. Chemical resistance tests included H_2SO_4 (10%) resistance test, NaOH (25%) resistance test and diesel resistance tests were carried out. In the following these tests have been explained in detail in order.

2.2.9 Sulfuric acid (10%)

H_2SO_4 (10%) test with immersion method was done on the panels. In this test a panel of any group was placed inside the H_2SO_4 solution. Immersing issue was done in a container which was shown in Figure 9. The samples were observed for 14 days.

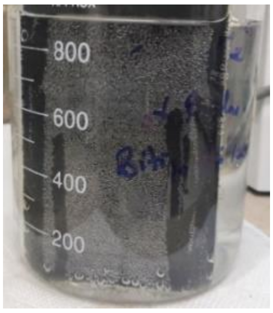


Figure 9. Resistance test against H_2SO_4 (10%) with immersion method

2.2.10 Sodium hydroxide (25%)

The chemical resistance tests were continued by with the scrubbing method which is shown in Figure 10.

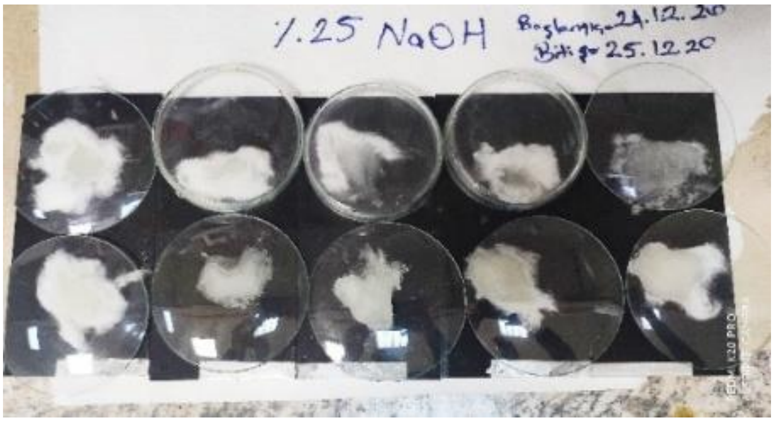


Figure 10. NaOH (25%) resistance test with the wetting method

2.2.11 Resistance against diesel

The tests of chemical resistance were continued by diesel resistance test. The method of diesel resistance test was similar to the NaOH resistance test. Saturated cotton pieces were placed upon the panels for 14 days. The panels were observed every day for 14 days.

3. Results and Discussion

At first, received sample has been analyzed to obtain a standard surface and thickness and to acquire further insights into their performance in glossiness, corrosion, QCT test, and chemical resistance tests. In this part, the results of all the tests have been given in order.

3.1 Glossiness Evaluation

The results of the glossmeter (in Table 2) upon the primers showed that addition of the additives negatively affected the gloss value of the surface and increased the opacity. The highest reduction happened in the samples containing B_4C. Reduction in glossiness of the coating samples containing hardening materials is inevitable as these materials are very small particles which embed inside of the resin molecules then these grains become exposed to the surface, scattering the light and providing matte effect. In the meanwhile, B_4C particles because of their amorphous shape and darker color influence the glossiness more.

Table 2. The results of the glossmeter of the surface of examination coatings

Gloss value	Blank	Black SiC	B_4C	Green SiC	Al_2O_3
(GU)	18.3	4.5	3.7	4.3	4.9

3.2 Pencil Hardness

The pencil hardness did not make big difference for the samples. All pieces failed on the H Pencil and only passed the F pencil. The surfaces were checked with a microscope for HB pencil test and there was no sign of scratch in the surface of the samples. The cases after the pencil hardness test are shown in Figure 11.

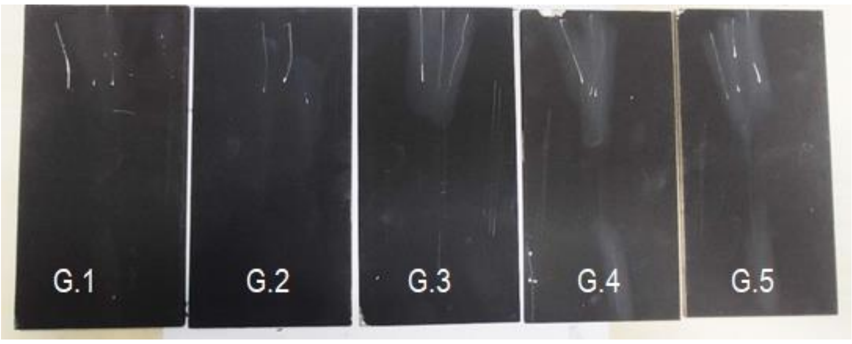


Figure 11. Results of the pencil hardness test

3.3 Universal Impact Tester

From the results of the impact tester, it could be considered that adding the hardeners did not affect negatively the flexibility and adhesion of the composite coating. The results were shown in Figure 12.

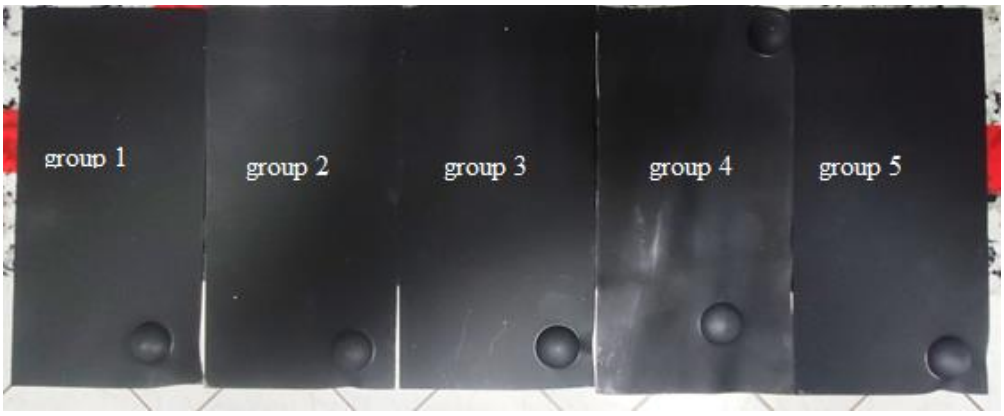


Figure 12. Results of the universal impact test

3.4 Conical Bent Flexion

The results of this test also showed no crack of peeling sign for the samples with or without the additives. This could be because of the high flexibility characteristics of the PTFE polymer structure which is the dominant resin in the primer and topcoat formulations. The results of the test were shown in Figure 13.

Figure 13. Results of the conical bent flexion test for the different groups of the study

3.5 Corrosion

After 120h the results did not show any sign of corrosion. In the time of 240h also the process didn't show significant corrosion sign but after 480h some white corrosion stains were observed. The results of the corrosion test after 480h were shown in Figure 14.

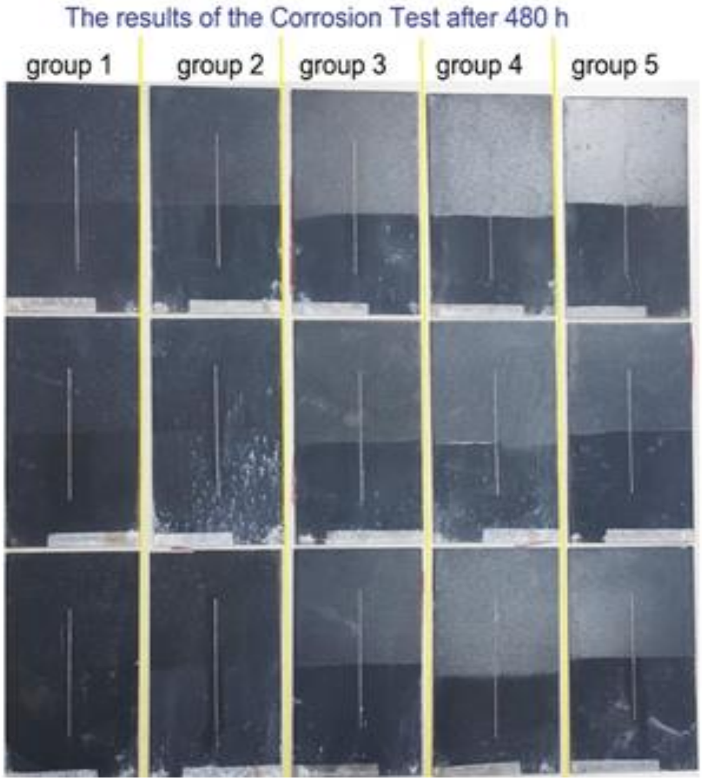


Figure 14. The panels of the 5 group after 480h corrosion test

The panels were analyzed according to TS EN ISO 4628-3 and ASTM D 714 standards. Corrosion test results, according to TS EN ISO 4628-3 and ASTM D 714 standards, are shown in Table 3.

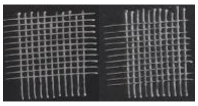
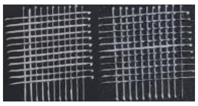
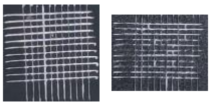
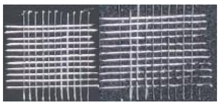
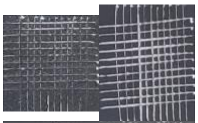
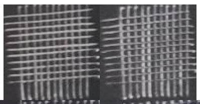
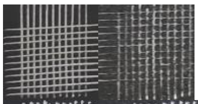
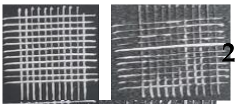
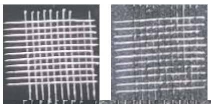
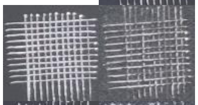
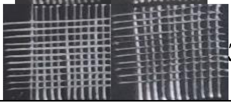
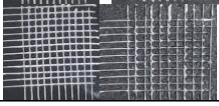
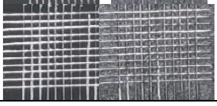
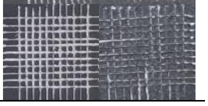
Table 3. Corrosion test results based on TS EN ISO 4628-3 and ASTM D 714 standards

	Blank			**Black SiC**			**B_4C**			**Green SiC**			**Al_2O_3**	
1	No8. D Ri3	No8. D Ri0	**1**	No8. M Ri3	No8. M Ri0	**1**	No8. M Ri2	No8. M Ri0	**1**	No8. M Ri0	No8. D Ri0	**1**	No6. F Ri2	No6. F Ri0
2	No6. M Ri1	No6. M Ri0	**2**	No8. D Ri5	No8. D Ri2	**2**	No8. M Ri1	No8. D Ri0	**2**	No8. D Ri3	No8. D Ri0	**2**	No6. M Ri2	No6. M Ri0
3	No8. D Ri3	No8. D Ri0	**3**	No8. F Ri2	No8. F Ri0	**3**	No8. M Ri1	No8. M Ri0	**3**	No8. M Ri2	No8. M Ri0	**3**	No6. F Ri2	No6. F Ri0

3.6 QCT Test

The results showed no sign of blister or corrosion on the panels, and it indicates that humidity does not affect these coatings. Evaluation of the cross-cut tests also showed no difference between the fresh panels and the panels which exposed to QCT test. In Table 4, cross-cut test results for fresh panels (left panel in every cell) and panels which exposed to QCT test (right panel in every cell) have been shown.

Table 4. Cross-cut test results for fresh panels (left panel in every cell) and panels which were exposed to QCT test (right panel in every cell)

				QCT test					
	Blank		**Black SiC**		**B_4C**		**Green SiC**		**Al_2O_3**
1		1		1		1		1	
2		2		2		2		2	
3		3		3		3		3	

3.7 Chemical Resistance

At first days the surface resisted thoroughly against the acid solution but after 14 days the results showed some defects on most of the panels with primer coatings. The results are shown in Figure 15.

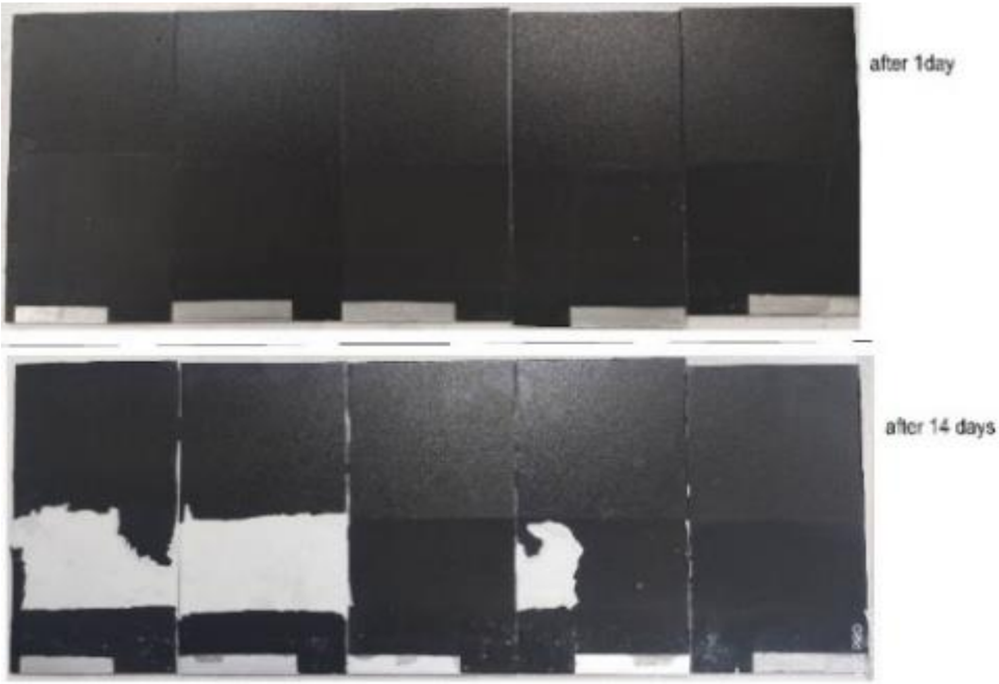


Figure 15. The results of the H_2SO_4 (10%) resistance test

In the case of NaOH (25%) resistance test, the panels were checked out after first days and also 14 days which can be seen in Figure 16.

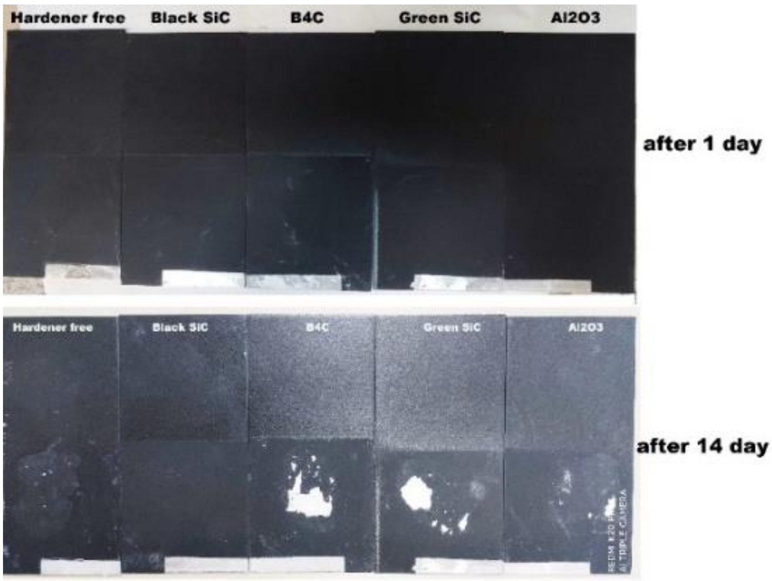


Figure 16. The results of the NaOH (25%) resistance test

These results showed no effect of the NaOH on topcoats of any groups but the last three groups have shown a sign of defection on their primer surfaces.

Diesel resistance examination of the coatings were accomplished and no sign of any defection was detected after this time. The resulted panels are shown in Figure 17.

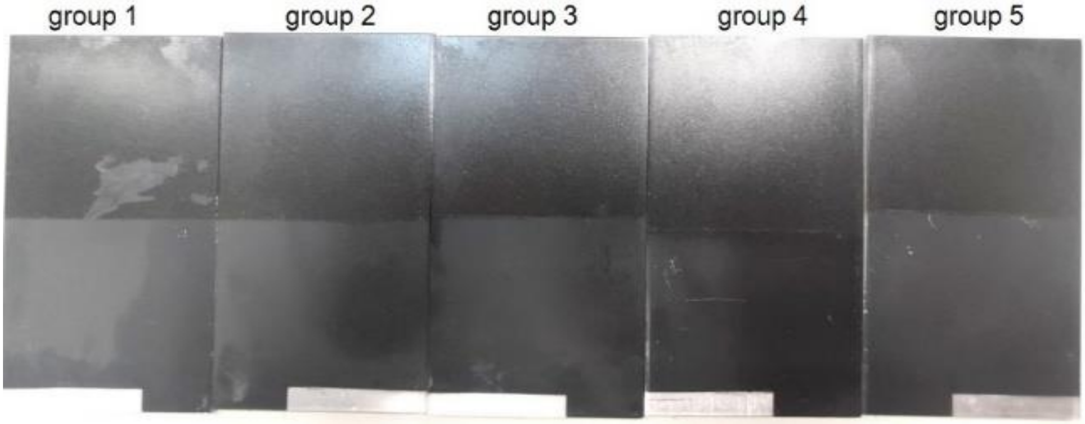


Figure 17. The results of the resistance test against diesel after 14 days

4. Conclusions

A non-stick surface is a surface engineered to reduce the ability of other materials to stick to it. Non-stick cookware is a common application, where the coatings allow food to brown without sticking to the pan. Most non-stick pans are coated with polytetrafluoroethylene.

In this study the effect of some additives known as hardening materials were studied in the subjects of glossiness, impact, hardness, conical bent, corrosion, humidity and chemical resistance. The results indicated that the samples containing B_4C were the best in overall performance following by black and green SiC . Although samples containing Al_2O_3 performed significantly better than samples lacking any hardener material, these samples were far behind the ones containing B_4C hardeners.

The samples with aluminum oxide was a head and shoulders behind these hard materials but samples including Al_2O_3 also was far better than the samples without any hardener material (blank sample). Glossiness of the samples containing hardener materials reduced when the additives were added to the coating formulation. Some other tests did not show significant difference among the samples.

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Author Statement

The authors confirm contribution to the paper as follows: study conception and design: Bikem ÖVEZ, Jabir ISMAEILI, Jigneshkumar PATEL; data collection: Jigneshkumar PATEL, Jabir ISMAEILI; analysis and interpretation of results: Bikem ÖVEZ, Jabir ISMAEILI; draft manuscript preparation: Jabir ISMAEILI. All authors reviewed the results and approved the final version of the manuscript.

Conflict of Interest

The authors declare no conflict of interest.

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