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# Effect of Glutaraldehyde Crosslinking Parameters on Mechanical and Wetting Properties of PVA/NaAlg Electrospun Mat

Ayben PAKOLPAKÇIL*[1]


## Abstract

Electrospun alginate-based materials are used in a wide range of applications, including wound dressings, tissue engineering, batteries, water treatment, bioremediation, and food packaging. However, they have low resistance to water. Crosslinking is usually used to enhance the mechanical properties of water-soluble polymers. Process parameters also play a key role in the crosslinking process. In this study, materials from sodium alginate (NaAlg) and poly (vinyl alcohol) (PVA) were prepared using the electrospinning method. To investigate the effect of the process parameters on the mechanical properties of the materials, different concentrations (1.25, 2.5 and 5 v %) and different application times (10 min, 60 min and 24 h) of the crosslinking agent were used. The wettability and mechanical properties of the electrospun mats were evaluated using a water contact angle device and a tensile strength tester, respectively. The maximum tensile strength was measured at 7 MPa which is the sample treated at 5% glutaraldehyde (GA) concentration and 60 min of application time. The sample treated with 2.5% GA concentration and 60 min of treatment time had the highest measured elongation of 11.5%. The sample treated with 2.5% GA concentration and for 10 min had the lowest water contact angle, which was measured at 27.5°. The intended usage of the materials should be considered, as the concentration of the crosslinking process and duration might affect the water-soluble polymers' mechanical and wetting properties.


**Keywords:** Crosslinking, glutaraldehyde, mechanical property, wettability, contact angle

## 1. INTRODUCTION

Biopolymers are widely used in many applications, such as in biomedical, textile, food packaging, cosmetic, energy storage and environmental applications [1]. Specifically, sodium alginate (NaAlg) as a biopolymer, is a valuable natural polymer. It is derived from brown algae. It is hydrophilic, biocompatible, and biodegradable. It has, therefore, been widely used in several applications, including paper, textiles, the food industry, pharmaceuticals and cosmetics [2, 3]. In 2020, the worldwide alginate market was valued at USD 728.4 million, with an expected increase to USD 759.8 million in 2021 [4].

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The electrospinning technique is an easy way to produce nanomaterials based on alginates. It is a versatile, simple, and inexpensive technique capable of producing continuous micro-nano fibers. Electrospun structures have superior properties, such as a high surface area and adjustable porosity; therefore, intensive research has been conducted on the use of these materials in many fields, such as textiles, packaging, and filtration. This technique, with a 100-year history, is based on the concepts of electromagnetic forces. However, over the last two decades, nanotechnology has become more sensitive to the production of nanofiber from biopolymers [5-8].

Electrospun alginate-based fibers have attracted a lot of attention due to their unique characteristics such as low toxicity, biocompatibility, their relatively low cost, and gelling properties under certain conditions. Many researchers have studied the development of alginate-based materials for a variety of applications, such as food packaging [9], bioremediation [10, 11] water treatment [12, 13], batteries [14], wound dressings [15, 16] and scaffolding [17, 18]. However, due to the rigid and extended chain characteristics of the alginate, the electrospinning method cannot be achieved on its own. For this reason, poly (ethylene oxide) (PEO) and poly (vinyl alcohol) (PVA) polymers are accompanied by electrospinning to produce alginate nanofibers. PVA has many desirable properties, such as thermal properties, water solubility, strength, and gas permeability [19, 20]. It is used extensively in a variety of ways, including industrial, food, packaging and medical applications, textiles, paper, antimicrobial packaging, wound dressings, and contact lenses. PVA solubility in water is high due to the significant amount of hydroxyl groups it contains; hence its stability must be increased before it can be used in aqueous environments. Both PVA and NaAlg are water-soluble polymers. Physical and chemical crosslinking have been examined as strategies for PVA-based nanofibers in the literature. Chemical crosslinking forms stable covalent bonds between polymer chains, enhancing mechanical characteristics and resulting in a water-insoluble material [21-23]. There are many chemicals for crosslinking processes such as citric acid, dextran dialdehyde, genipin, carboxylic acids, etc [19]. The use of a glutaraldehyde (GA) crosslinking agent is a widely employed method for the preparation of biopolymers. GA has high reactivity and is both inexpensive and easily available. GA is also employed in pharmaceutical sciences enzyme technology, histochemistry, biomedical applications, and chemical sterilization. It is an organic chemical compound from the group of aldehydes. It is a colorless to pale straw-colored, pungent, oily liquid. It is used for crosslinking biopolymers. A variety of factors influence the crosslinking process and crystallinity, including chemical compositions, concentrations, temperatures, and reaction times [24, 25]. Kim et al. investigated the effects of the degree of GA crosslinking time on the properties of PVA pervaporation membranes. These effects included pervaporation characteristics, contact angles, swelling characteristics and critical surfaces [26]. Mansur et al. prepared the GA crosslinked PVA hydrogels with different degrees of hydrolysis and their chemical structure was investigated by using Synchroton small-angle X-ray scattering and Fourier transform infrared spectroscopy [27]. The effects of the concentration of GA and the temperature of the crosslinking process on the thermal and chemical changes in the PVA membrane have been investigated by Figueiredo et al [28]. Ahmad et al. examined the effects of the GA crosslinking time on the pore size distribution of PVA films [29]. Qin et al. investigated the mechanical characteristics, thermal stability, water stability, and changes in the degree of crystallinity of PVA nanofibrous mats using a variety of concentrations of GA solutions [30]. Matty et al. worked on the effect of GA as a crosslinking agent with different weight ratios on the swelling behavior of PVA hydrogels [31]. Shivakumara and Demapp examined sodium alginate /PVA hydrogels which had been crosslinked with different GA concentrations; the chemical functional groups, morphology, and swelling properties of the hydrogels were investigated at different pH values, and using indifferent salts at different temperatures, and indifferent acids and bases [32]. Gadhave et al. evaluated the changes in thermal and mechanical properties caused by crosslinking different concentrations of GA on

starch and PVA blended films [33]. Musa and Hameed worked on the effect of GA as a crosslinking agent with different weight ratios on the mechanical properties of PVA/starch and PVA/polyethylene glycol blend films [34]. Nagamadhu and Kivade investigated the mechanical and dynamic mechanical analysis of PVA-GA polymers [35]. Although the effect of concentrations on mechanical properties was investigated in some studies, reaction time was not emphasized. As far as the author is aware, no study has been carried out to evaluate the mechanical and wettability effects of crosslinking agent concentrations and duration parameters on the electrospun PVA/NaAlg materials.

This work aims to present a systematic investigation of some of the physical features occurring after the crosslinking process of PVA/NaAlg electrospun mats in the presence of GA. PVA/NaAlg electrospun mats are used for various applications, such as filtration, packaging, and wound dressings, and for this purpose, it is important to control the physical features and to understand how process parameters like concentration and time influence mechanical ability and wettability. Given this, the effects of crosslinker concentration and time on the wettability and mechanical properties of PVA/NaAlg electrospun mats after the crosslinking process are investigated. In this way, it is hoped to gain a basic insight into the crosslinking processes that affect wettability and strength properties.

## 2. EXPERIMENTAL STUDY

### 2.1. Materials

The PVA polymer (a molecular weight of 85,000-124,000 g/mol with 87-89% hydrolyzed), and hydrochloric acid (HCl) (37%) came from Sigma Aldrich (Germany). NaAlg was acquired from Cargill (France). GA (50%) was supplied by Kimetsan (Turkey). Ethyl alcohol, acetone, sodium chloride, potassium chloride, sodium phosphate dibasic, and potassium phosphate monobasics were supplied from Merck (Germany). The experiments were carried out with distilled water, and all the chemicals were employed without being purified.

### 2.2. Fabrication of NaAlg/PVA electrospun mats

To prepare a PVA aqueous solution, 12 g PVA polymer was completely dissolved in 100 mL distilled water over 12 hours at 90°C with continuous stirring. To prepare a NaAlg aqueous solution, 1 g NaAlg powder was fully dissolved in 100 mL distilled water over 4 hours at 50°C with constant stirring. Following that, the PVA and NaAlg were combined in a 70:30 (v:v) ratio and mixed for 6 hours to get a homogeneous solution. A 20 mL syringe was filled with NaAlg/PVA solution, and the electrospinning of the solutions was performed in a single-nozzle electrospinning setup (Figure 1). Before starting the electrospinning equipment, the spinning parameters were set as follows: rotational speed 180 rpm/min, voltage 18 kV, spinning distance 12 cm, and extrusion speed 0.7 mL/h. The electrospun mats were collected on aluminum foil.

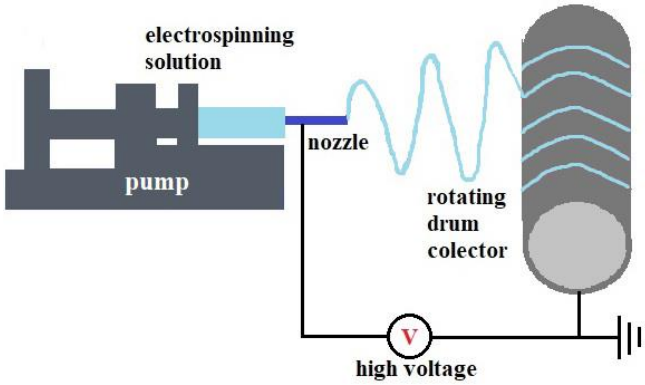


Figure 1 Illustration scheme of electrospinning setup

### 2.3. Crosslinking

The obtained electrospun PVA/NaAlg mats were soaked in a reaction solution consisting of three different (1.25, 2.5 and 5 v %) GA concentration. The recipes of the prepared solutions are given in Table 1 and a schematic diagram of the crosslinked PVA/NaAlg is shown in Figure 2. The samples were treated with the prepared solutions at room conditions (temperature = 25 ± 2 °C; relative humidity = 50 ± 5%) three separate times (10 min, 60 min, and 24 h) for chemical

crosslinking and then washed with ethyl alcohol. Subsequently, the electrospun PVA/NaAlg mats were carefully washed several times with a phosphate buffer solution to remove the remaining GA.

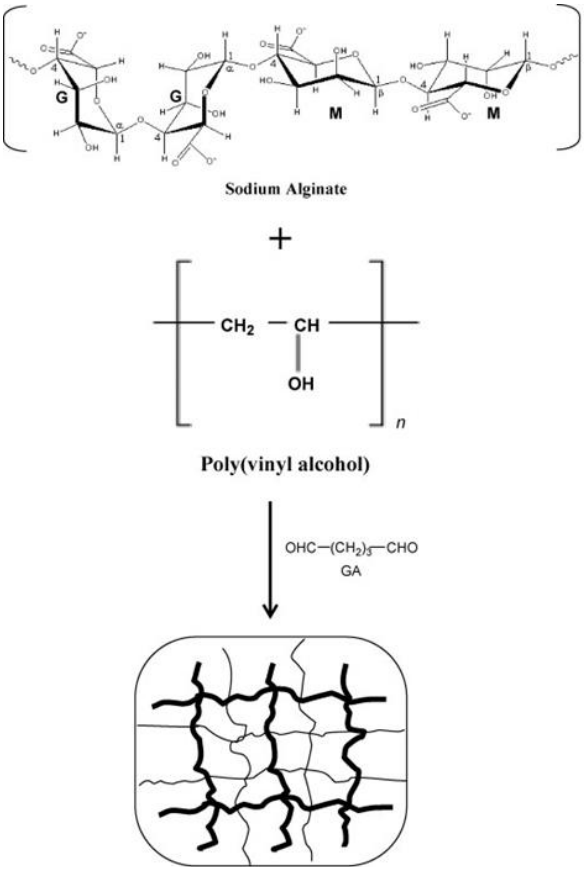


Figure 2 Schematic representation of the prepared crosslinked PVA/NaAlg [25]

Table 1 Recipes of crosslinking solution

| | Concentrations | | |
|---|---|---|---|
| Chemicals | 1.25 % | 2.5 % | 5 % |
| Glutaraldehyde (mL) | 1.25 | 2.5 | 5 |
| Hydrochloric acid (mL) | 0.5 | 0.5 | 0.5 |
| Acetone (mL) | 98.25 | 97 | 94.5 |

### 2.4. Mechanical strength

The tensile properties of the crosslinked electrospun mats were measured using the universal materials testing machine (Shimazhu AG-X plus) at room conditions (temperature = 25 ± 2 °C; relative humidity = 50 ± 5%). ISO 9073-3 Nonwovens — Test methods — Part 3: determination of tensile strength and elongation at break using the strip method was used for this study and modified according to the obtained electrospun mats. The tensile strength and elongation at break were determined using samples cut from the electrospun mats, which were 50 mm in length, 10 mm in width and at a crosshead speed of 10 mm/min. A digital micrometer was used to measure the thicknesses of the crosslinked electrospun mats (30-70 μm). The measurements were taken in triplicate, and the average values and standard deviations were determined.

### 2.5. Wettability

Contact angle measurements were performed using the sessile drop method with an Attension Theta (Biolin Scientific Inc.) system and distilled water. The crosslinked electrospun samples measuring 3 x 1 $cm^2$ were employed for the contact angle analysis. Water droplets (5 μL) were softly deposited on the surface, and the angle formed between the drop and the surface was determined by image analysis via software. Three different times (0 s, 10 s, and 30 s) were monitored as the droplets of water were released onto the electrospun mats. Three measurements were taken, and the mean values and standard deviations were determined by taking both the left-side and right-side angles into account.

## 3. RESULTS AND DISCUSSION

### 3.1. Mechanical strength of the electrospun mats

One of the most important and widely estimated properties of materials used in many applications is their ability to resist breaking under tensile stress [36]. This characterization was performed to study the effect of the condition of crosslinking duration and concentration on the tensile strength

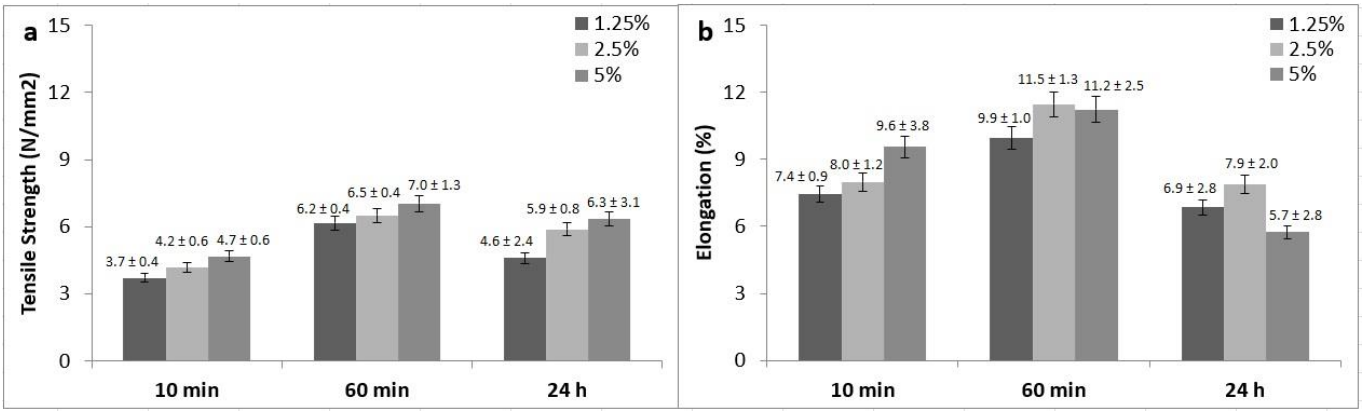


Figure 3 a) Influence of GA toward tensile strength and b) Influence of GA toward elongation

behavior of the electrospun mats. Figure 3 shows the mechanical properties of the electrospun mats using different concentrations (1.25, 2.5 and 5 v %) and application times (10 min, 60 min, and 24 h) of GA. With the increasing exposure time of GA, the tensile strength and elongation at break of the electrospun mats increased steadily at first but then decreased gradually when the application time of GA was 24 h. The improved tensile strength and elongation properties were attributed to an increase in acetal bonds between the aldehyde groups of crosslinking agents and the interaction of the hydrogen bond between the hydrophilic PVA and NaAlg. However, when the application duration approached 24 h, free hydroxyl and crosslinking points tended to become saturated. Excess GA tangled with the NaAlg/PVA molecular chains, meanwhile reducing the strength of the electrospun mats. The efficiency of GA on the crosslinking of NaAlg and PVA is due to the strong reactivity of the aldehyde groups, which quickly form imine bonds (Schiff's base) with amino groups and acetal bonds with hydroxyl groups [37]. The formation of a crosslinked network of electrospun mats, which limited the mobility of the molecular chains, was related to the improvement in tensile strength and the decrease in elongation at break. The highest tensile strength was 7.0 MPa for 5 % GA at 60 min. The high strength is usually related to high crystallinity [23]. It is thought that the maximum possible crystallinity for the samples had been reached under these operating conditions. The highest elongation was 11.5 % for 2.5 % GA at 60 min. This study showed us that although the increase in crosslinker concentrations initially gave an increase in elongation, at a certain point, higher crosslinking density protected the fiber from elongation and it became harder thus, elongation decreased. Because of the presence of two extremely reactive alpha protons the GA become more reactive and acidic in nature [33]. Due to the greater reactivity of GA, particularly at higher doses, the H-bond between the PVA chains is reduced. Mansur et al. showed that when PVA hydrogen was prepared with different GA concentrations in FTIR spectroscopy, the reaction of the PVA with the GA resulted in a significant reduction in the intensity of O-H peaks at higher GA concentrations [27].

In summary, the electrospun PVA/NaAlg mats may require different strength and flexibility properties depending on their use. Therefore, the process parameters should be optimized before crosslinking.

### 3.2. Water contact angle measurements of the electrospun mats

Wetting is a natural phenomenon with significant consequences for both nature and human existence. When a liquid interacts with a solid, it generates a liquid coating that spreads across the solid's surface. Materials have varying wetting qualities due to the different intermolecular interactions between the liquid and the solid. The contact angle is a numerical representation of the sample's wetting features. The contact angle of water has a considerable influence on the wetting of micro-sized droplets on solid surfaces [38]. Figure 4 shows the water contact angles of the

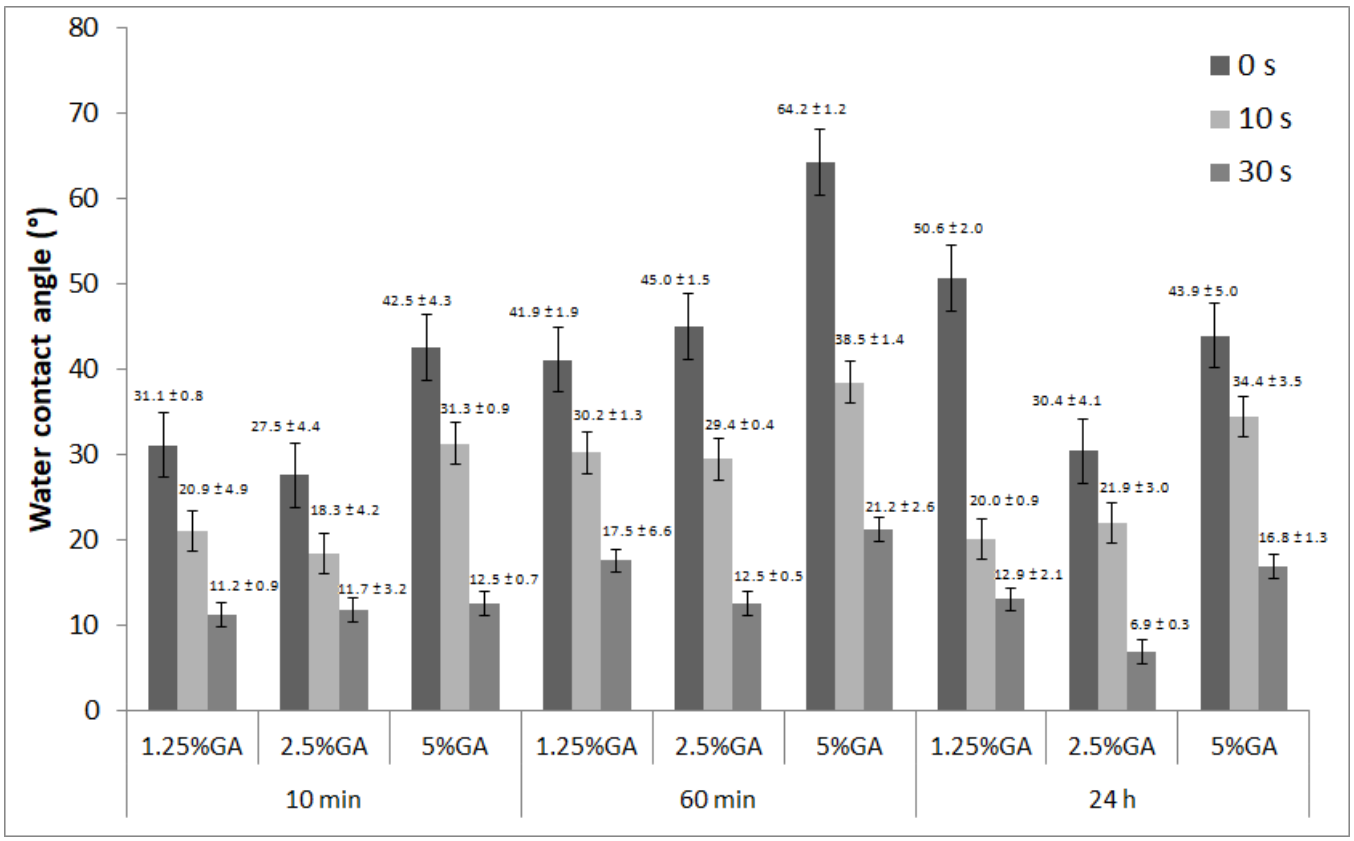


Figure 4 Water contact angles values of PVA/NaAlg electrospun mats at 0 s, 10 s and 30 s

electrospun mats at 0 s, 10 s and 30 s, with different concentrations and application times of GA. As shown in Figure 4, the electrospun mats demonstrated low water contact angles (27-64°). The water contact angle values were found to be consistent with those reported by Pakolpakçıl et al, where the GA crosslinked red cabbage extract loaded Alg/PVA contact angle value was ~46° [13].

The application time of GA was 10 min, it was observed that the electrospun mats exhibited the hydrophilic surfaces characteristics of more low contact angles (27-42°) when compared to the other application times. PVA and NaAlg's hydroxyl groups are hydrophilic, which means they can form hydrogen bonds with water and absorb a lot of it. This results in a decreased degree of water contact angle.

The highest contact angle value of 64° was obtained at 5% GA concentration and 60 min treatment time. The finding can be explained by increasing the crosslinking of the network; in other words the concentration of GA. The highest results were relatively consistent with the strength measurement at the same concentration and the time of application. It is thought that the water molecules are unable to find any spaces to enter, as the attraction force between the polymer molecules is high and the sample is too compressed [39]. This results in an increased degree of water contact angle. Pakolpakçıl and Draczynski showed that the water contact angle value of the EDC/NHS crosslinked PVA/alginate nanofiber mat increased as the EDC/NHS concentration increased [8].

The results show that the crosslinking concentration and time influenced the water contact angle of the electrospun mats, which can be attributed to the chemical interaction of the polymers and the crosslinker.

## 4. CONCLUSION

The PVA and NaAlg structures both include hydroxyl groups, which makes the reaction with GA possible. In this study, the PVA/NaAlg electrospun mats were fabricated successfully by

the electrospinning method. To investigate the effect of the concentration and application time of GA crosslinking agent on the electrospun PVA/NaAlg mats, different concentrations (1.25, 2.5 and 5 v %) and duration times (10 min, 60 min and 24 h) of the GA were applied. According to the findings, the longest application time (24 h) has a negative effect on elongation. The highest strength values were obtained at all concentrations at 60 min of application time. In general, the highest contact angle values were observed at a concentration of 5%. The results of the investigation revealed that the crosslinking process parameter, the application time and the concentration influenced the development of the PVA/NaAlg electrospun mats' mechanical and wetting properties.

The end-use of the materials should be considered, where the choice of concentration and the duration of the crosslinking process have an impact on the mechanical and wetting ability of the water-soluble polymers. Mechanically strong fibers are generally preferred in many areas of use. However, since high water absorbance is required in applications such as wound dressings, wettability may be at the forefront.

### *The Declaration of Conflict of Interest/ Common Interest*

No conflict of interest or common interest has been declared by the author.

### *The Declaration of Ethics Committee Approval*

This study does not require ethics committee permission or any special permission.

### *The Declaration of Research and Publication Ethics*

The author of the paper declare that they comply with the scientific, ethical and quotation rules of SAUJS in all processes of the paper and that they do not make any falsification on the data collected. In addition, they declare that Sakarya University Journal of Science and its editorial board have no responsibility for any ethical violations that may be encountered, and that this study has not been evaluated in any academic publication environment other than Sakarya University Journal of Science.

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