A low-cost, high-efficiency, new generation material for fog harvesting fumed silica-doped polypropylene

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This study describes the development of a fog collector material for fog harvesting. Polypropylene (PP) doped with fumed silica (0–2%) was punctured at equal intervals and exposed to fog produced by a humidifier. The amount of water harvested by each sample was measured using an ultrasonic fogger. Polypropylene doped with 1% fumed silica was most effective at harvesting water, and collected almost 19–20 times more water than pure polypropylene. This improvement is due to the surface tension, which decreased from 16.754 mN/m (pure PP) to 13.512 and 9.992 mN/m (0.5% and 1% fumed silica, respectively). On the other hand, when fumed silica doping exceeded 1%, this increased the polymer’s surface tension, measured as 20.6 and 38.1 mN/m for 1.5 and 2% fumed silica doping. We therefore propose fog harvesting using 1% fumed silica-doped polypropylene as a low-cost method for collecting clean water in arid regions.

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INTRODUCTION

The boiling degree of water is 100 °C, and its freezing degree is 0 °C. Water is molecularly V-shaped. The distance between the two protons is ~104.45°. Without this angle and geometric shape, one could not talk about life. Thanks to these features, the dipole moment is 1.84 D, and the surface tension is 72.8 mN/m. Due to the high surface tension, water takes the shape of a sphere to reduce the surface energy.

Water is one of the natural resources that human beings will suffer the most from its absence in the future. Even now, water scarcity has begun in many parts of the world, and people are particularly in need of clean water. Polluted clean water resources are tried to be purified, but this increases the cost of obtaining clean water. Recently, researchers have developed engrossing and low-cost methods to meet the need for clean water. The collection of rainwater in definite reservoirs is a good example. However, the collection of rainwater only in a limited area and the fact that there is no rainfall in arid regions caused the studies in this field to cease. On the other hand, fog harvesting is quite an interesting subject. It is possible to collect water formed by sweating on the material on foggy areas and hot arid regions.

Researchers have produced many different materials for fog harvesting 1–3. Because the structure and type of fog collector materials used for fog harvesting also differ according to the region to be used. Therefore, researchers have produced new materials by examining the creatures living in different regions and collecting water even though there is no water source 4–7. Cicada, cactus, and spider webs are natural fog collectors 1–3.

Researchers have noticed that these creatures collect water through hydrophobic materials.

The lowest-cost fog collector materials known as artificial materials are the ones produced from polymers such as polypropylene (PP). However, PP cannot collect as much water as other materials. Another example is the variation of polyethylene terephthalate fibers’ ability to collect water depending on their surface and cross-sectional area 8. The materials prepared with silicone elastomers and fluoropolymers, which have superhydrophobic surfaces, have higher efficiency depending on their surface properties 9,10. For example, polycrylonitrile (PAN) is considered suitable because of its mechanical properties 11. Many studies have been examined polymers for water vapor transport. Akhtar and Peineman used block copolymer membranes 12 and Pebax®1657/Graphene oxide composite membranes 13 for water separation. Recently, very important improvements have been reported in this area with rugged structures covered with polymer on a hydrophobic surface and doping with TiO2 nanoparticles 10–12. It was discovered that convex surface, lump-like tissues increase the fog condensation rate to the maximum level 13–15. For an efficient fog harvest, micro and nano coatings alone remained insufficient, and it is understood that additional coatings are needed. According to the researchers, hydrophilic surfaces coated with a superhydrophobic substrate are needed to prevent the fog hitting the material’s surface from adhering to the surface. The interaction between the hydrophilic parts and the superhydrophobic surface must be strong to increase the liquid’s contact angle with the solid surface. To increase this interaction, researchers have started to focus on knitted structures. These braided structures are materials with different lattice structures consisting of polymers and metals 16–20. Polymers are preferred in researches because of their flexibility, high strength, and mechanical endurance. One of the best examples of them is polystyrene (PS). PS can be used as a substrate since it has glass transition temperatures around 80–100 °C and low processing temperature 21.

Researchers suggested that the water to be collected affects the hydrophobic and hydrophilic regions’ boundary and fog harvest productivity 22. Some researchers have shown that the superhydrophilic surface’s harvesting performance on the superhydrophobic surface changes depends solely on the inclination angle 23. According to these results, the modified surface’s fog harvesting performance is strongly influenced by wettabiliy changes, such as the compressive strength and the harvest conditions. These results are presented in a review by Korkmaz and Kariper 24.

When the studies are summarized, it is concluded that both hydrophilic and hydrophobic surface interactions are important in fog collecting materials, and these materials should be used together.

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Many studies have been carried out for fog harvesting using PP, which is very cost-effective. Raut et al. produced 0.2 µm porous fiber textile products with PP and polycarbonate. They proved that a nonperforated flat strip of pure PP could store 0.18 g/cm²/h of water. They found the water contact angle of the pure PP strip was 97°. Moazzam et al. proved that 0.097/cm²/h water could be stored with their polydopamine coated PP. The contact angle of the material with water was found to be ~14°, and they calculated the surface tension as 87.9 mN/m. \cite{Chin2015}. Chin et al. produced a PP membrane using the electrospun method to purify water from saline water. They announced that they managed to desorb ~30% of the saline water. Up to now, a maximum of 0.18 g/cm²/h water can be stored with PP. \cite{Raut2018}. The most interesting example is the article published by Damak and Varanasi in 2018 on electrostatic fog harvesting. Their study performed mathematical formulas that link many parameters such as voltage, wind speed, etc., to be applied for more efficient fog harvesting. \cite{Shi2021}. Shi et al. used a vertical wire system to show the system’s effect on the produced material’s fog harvesting efficiency. The study explained that the vertical wire system could store three times more water than the other wire system, and they designed a machine for fog harvesting for the first time. \cite{Moazzam2021}

In practice, the harvesting materials produced are usually stretched between two poles. Under the poles, there are water collection pipes and water collection tanks in which the pipes flow. The produced water is stored in these tanks. \cite{Batisha2015, Batisha2016}. Batisha showed the best example of using fog harvesting in 2015. Batisha talked about its applications in many countries such as Chile, Yemen, Amman, and Spain and interpreted the economic, environmental, and social dimensions of fog harvesting materials. \cite{Batisha2015, Raut2018}

Besides, this is not the first use of fumed silica (FS). Previously, Mavukkandy et al. used FS particles as porogen to prepare polyvinylidene fluoride (PVDF) membranes. In the study, they were able to add 5% of FS to PVDF. They studied the thermodynamic and kinetic aspects of membrane formation. They noticed that the emerging membrane morphology was associated with thermodynamic enhancement and FS doping rate and kinetic inhibition. They used the prepared membranes for the filtration of raw wastewater. They found that although the PVDF–FS mixed membrane showed a much higher current, they also had a high fouling tendency due to their increased hydrophobicity. They found that PVDF–FS membranes used for water filtration are suitable for water purification and pollution problem. \cite{Mavukkandy2018, Moazzam2021}

In this study, a substrate was produced by adding a hydrophilic material to a hydrophobic material and tested in a single layer. FS and molecular sieve were doped to the PP in an extruder machine at low percentage rates. The results were interesting.

RESULTS

Fourier transform infrared (FTIR) analyses

FTIR analyses (200–600 cm⁻¹) are given in Fig. 1a–e. FTIR analyses were performed in the wavelength range of 200–600 cm⁻¹ to see both PP and Si–O vibrations. The region above 600 cm⁻¹ was not included because the 700 cm⁻¹ regions are already known, and doped PP and Si’s vibration peaks will be better seen under 700 cm⁻¹. As expected, out of plane deformation of Si–O at 450–460 cm⁻¹, which is not seen much in pure PP, became more apparent as doped Si increases. \cite{Moazzam2021} –CH₂ and bending –CH vibrations (at 450 and 240–250 cm⁻¹, respectively), which are very weak in pure, undoped PP, are further enhanced by Si–O vibrations’ contribution. Besides, –CH vibrations at 279 cm⁻¹ disappeared within the SiO₂ crystal vibrations. \cite{Moazzam2021} Si–O vibrations in the structure become dominant with 2% FS doping. When the doping exceeds 2% by weight, deterioration occurs in the polymer structure, and the extrusion machine fails to produce strips from the polymer. FTIR analyses (450–4000 cm⁻¹) are given in Fig. 1f–j. Aliphatic –CH vibration peaks are observed at 2800–2900 cm⁻¹. Si–O–Si at 1165 cm⁻¹, Si–O at 1372–1453 cm⁻¹, and Si–OH vibration peak at 969 cm⁻¹. Characteristic vibrations of terminal unsaturated-CH₂ groups present in isotactic PP are identified at 843–808 and 1001 cm⁻¹. \cite{Moazzam2021, Moazzam2021}. 

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According to the Zisman method, a solid’s surface tension is calculated from the slope of the line drawn by plotting the contact angle with the produced fog harvesting materials. The water’s contact angle with the fog harvesting materials is measured as 103° for pure PP, 97° for 0.5% Si-doped PP, 92° for 1% Si-doped PP, 100° for 1.5% Si-doped PP, and 102° for 2% Si-doped PP. If the water droplet spreads by wetting a large area of the surface, the contact angle is less than 90°, and the surface is considered hydrophilic. In our study, the hydrophilic properties of the materials have a very fine line. The highest efficiency obtained from 1% Si-doped PP also has the lowest contact angle of 92°, which is quite close to the hydrophilic and hydrophobic limit. In this study, the highest fog harvesting efficiency was achieved when water’s contact angle with the material surface is below 100°.

The literature often reported that the water to be collected depends on the surface tension. In Fig. 4, the samples’ surface tension, measured by using the Zisman method, is shown. Water, diiodomethane, ethylene glycol, and formamide with known surface tension values were used. After measuring these liquids’ contact angles with the surface, these contact angles were plotted versus the surface tension. Our samples’ surface tensions were calculated from the slope of the line obtained, and the results are presented in Fig. 5. While the surface tension of pure PP was 16.754 mN/m, the surface tension decreased to 13.512 and 9.992 mN/m when 0.5 and 1% FS was doped, respectively. However, the 1.5 and 2% FS-doped polymer’s surface tension increased again to 20.6 and 38.1 mN/m.

Figure 6 shows the average amount of water stored on the materials’ surface in an hour. Pure PP was used as a reference in this study. The materials with a surface area of 50 cm² were weighted. The amount of water that can be collected per 1 g of material is given in Fig. 7. As the researchers have stated before, the materials’ surface tension is effective in collecting water. The amount increased up to 0.267 g/g-material/cm² with 1% FS doping, whereas this value decreased to the amount of the undoped PP for 2% FS-doped PP.

In Fig. 8, the relationship between surface tension and the amount of collected water is given. The only data that disrupts this curve is undoped PP; thus, it has been removed from this chart. FS is one of the best humectants. It is hydrophilic. FS increases its weight in just a few minutes by absorbing up to 0.5% moisture, and then it can desorb very easily. Therefore, an increase of 5% in the structure up to a certain level indicates an increase in FS. The more FS, the more water will be absorbed into the structure. As shown in Fig. 8, there is also a relation such as surface tension = 5.0648 (deposition water) − 0.452, although it is not a very compatible graphic.

**DISCUSSION**

Regarding the studies involving undoped PP, around 0.05 g of water can be collected with an angle of 90° between the sample and the fogger (in a 60–65% humidity)45. In their study, Raut et al. stated that cellulose fibers could store five times more water than pure PP. Among the studies about water collection in the literature, Kim et al. produced ZnO and Ag nanostructured materials; they reported collecting up to 0.3 g/cm² water with a machine that fogged water at a speed of 120 mL/h, at ~1150° angle (70% humidity)46. Although Almasian et al. stated that they could collect 300 mg of water in a 70° humidity environment with fluorinated-PAN nanofibers, they did not.

**Fig. 1** Fourier transform infrared spectroscopy spectrums of the samples. a Fourier transform infrared spectroscopy analysis (200–600 cm⁻¹) of the pure polypropylene. b Fourier transform infrared spectroscopy analysis (200–600 cm⁻¹) of the 0.5% fumed silica-doped polypropylene. c Fourier transform infrared spectroscopy analysis (200–600 cm⁻¹) of the 1% fumed silica-doped polypropylene. d Fourier transform infrared spectroscopy analysis (200–600 cm⁻¹) of the 2% fumed silica-doped polypropylene. e Fourier transform infrared spectroscopy analysis (200–600 cm⁻¹) of the 2% fumed silica-doped polypropylene. f Fourier transform infrared spectroscopy analysis (450–4000 cm⁻¹) of the 1.5% fumed silica-doped polypropylene. g Fourier transform infrared spectroscopy analysis (450–4000 cm⁻¹) of the 1% fumed silica-doped polypropylene. h Fourier transform infrared spectroscopy analysis (450–4000 cm⁻¹) of the 0.5% fumed silica-doped polypropylene. i Fourier transform infrared spectroscopy analysis (450–4000 cm⁻¹) of the pure polypropylene. Si–O at 1372–1453 cm⁻¹ and Si–OH vibration peaks are seen at 969 cm⁻¹.

Scanning electron microscope (SEM) and energy dispersive X-ray (EDX) analysis

SEM and EDX analysis results are given in Figs. 2 and 3. In Fig. 2a, the TEM image of pure PP without doping shows a flat ground except for a small agglomeration due to the extruder machine’s principle pull. Figure 2b shows large SiO₂ particles around 200 nm identified when 0.5% Si is added. The particle size in Fig. 2c is 200 nm and below, and in Fig. 2d, there is an agglomerated coarse particle. In Fig. 2e, the particles are agglomerated more in certain regions as the doped amount increases, which indicates why no more than 2% can be added to PP. Because the agglomerated particles disconnect from the polymer and cause holes and ruptures on the surface during extraction. Even though FS is in nano size, it is added from the extruder machine’s reservoir. FS begins to agglomerate due to the environment’s humidity at this temperature, which is ~200 °C. When it enters into the structure of the molten PP, it can agglomerate in certain areas. However, when the added amount is small and added to the extruder machine simultaneously, there is very little agglomeration. However, in the case of excessive agglomeration, ruptures may be seen in the structure of PP. In the experiments, critical ruptures were observed in the polymer when 3% or more of FS were added to PP.

According to the EDX analysis given in Fig. 3, when 0.5% of FS is added, the very low amount of silicon on the polymer surface increases as the doping increases. The amount of silicon in the structure increased from 0.07 to 0.76% with doping. However, when 1.5 and 2% FS is doped into PP, the strip-form polymer structure deteriorated due to the reasons above. In other studies in the literature, the FS doped to the polymers was also agglomerated after exceeding a certain amount.48

Surface tension measurements

According to the Zisman method, a solid’s surface tension is calculated from the slope of the line drawn by plotting the contact angle (cosθ) between the solid and the liquid dropped on that solid versus these liquids’ surface tension (σ). The slope of that line gives the surface tension of that solid. Moreover, when looking at liquids that are standardly dropped onto the solid surface, they are generally polar molecules. In other words, by adding a hydrophilic material, the angle of contact will decrease. The decrease in the contact angle causes the slope of the intended line to be low. Therefore, as the doping of a hydrophilic material increases, the surface tension is expected to increase, which can be seen very clearly from Zisman’s mathematical formula.

Zisman found that \( \cos \theta = a - b \sigma_i = 1 - \beta (\sigma - \sigma_i) \) (1)

\( \sigma_i \) represents a solid’s critical surface tension, which is a common characteristic property for all solids. \( \sigma > \sigma_i \) in liquids that wet the surface. This method is commonly used to measure the polymers’ surface tension. The contact angle between the liquid and the solid (cosθ) is plotted versus the surface tension (σ) of these liquids, and the line’s slope is obtained. The line here is \( y = ax + b \). The polymer’s surface tension is calculated from the slope \( a_{\text{Zisman}} \). Figure 4 shows the curves drawn by the Zisman method and the water’s contact angle with the produced fog harvesting materials. The water’s contact angle with the fog harvesting materials is measured as 103° for pure PP, 97° for 0.5% Si-doped PP, 92° for 1% Si-doped PP, 100° for 1.5% Si-doped PP, and 102° for 2% Si-doped PP. If the water droplet spreads by wetting a large area of the surface, the contact angle is less than 90°, and the surface is considered hydrophilic. In our study, the hydrophilic properties of the materials have a very fine line. The highest efficiency obtained from 1% Si-doped PP also has the lowest contact angle of 92°, which is quite close to the hydrophilic and hydrophobic limit. In this study, the highest fog harvesting efficiency was achieved when water’s contact angle with the material surface is below 100°.
give information such as ultrasonic power fogger and the amount of water produced per hour. Zhong et al. stated that they could collect 0.3 g/600 mm² water with the materials they produce from copper and titanium oxide; they used a powerful ultrasonic fogger of 252 mL/h and 70 cm/s, at 70% humidity.

On the other hand, Gürsoy et al. showed that 107 g/m² water was collected with a 30 L/h fogger using natural fibers, and 58 g/m² of water can be collected with the polyethylene polymer they used as a reference. Twice as much water can be collected compared to a normal polymer. White et al. tested different materials they prepared with polytetrafluoroethylene (PTFE), aluminum, titanium, and carbon nanotubes in a closed container with a fogger of 420 mL/h. They could collect 0.7 g of water with PTFE, and they increased this value to 0.8 g with the material they made from aluminum.

As can be seen, it is not easy to compare the results of this study with the literature since each researcher created and used his/her parameters. The literature review showed that researchers not mentioned here have generally performed their experiments in the humidity chamber, in a high humidity environment, and using high-performance foggers. It would be much better to consider studies that use the same material as a reference. In some studies, the humidity of the environment was almost three times higher than our study, and the speed of the foggers was 6–100 times higher than ours. Therefore, it is more logical to look at studies such as that of Raut et al., White et al., and Gürsoy et al. Among these, only Raut et al. compared the material they have produced with a base material such as PP and stated that they could collect up to five times more water. It has been proven that up to 19–20 times more water can be collected with 1% FS-doped PP than pure PP, with a fogger of 20 mL/h, without humidity chamber and at 20% humidity at room conditions. No material in the known literature can collect so much water. This material also has its disadvantages. Besides, when 3% or more of FS was added to PP, deterioration (ruptures) was observed in the sample structure. Also, when 2% or
more of FS was added to PP, there was a decrease in the fog harvest. This is because FS is also a good moisturizer and tries to keep water in its structure by swelling. Also, with the increase of agglomeration due to excessive doping, the nonhomogeneous distribution of FS in PP reduces the efficiency.

PP was used as a reference in this study. Hydrophilic FS was doped to a hydrophobic material (PP) by 0.5–1–2%. One percent of FS-doped PP was found to collect almost 19–20 times more water than pure PP. However, one of the main findings of the study is the surface tension. In this study, the material with the lowest surface tension collected the maximum amount of water. However, it was not easy to compare the obtained results with the literature. Since the parameters that researchers specified vary in each study, the comparison was made according to the reference material. In terms of cost, we produced almost 50–100 times cheaper materials than those known in the literature, which were claimed to be good in collecting water.

**METHODS**

**Preparation of fog harvesting materials**

In this study, fog harvesting materials (PP and FS-doped PP (Si-PP)) were produced via extruder machine. FS (Si) was purchased from Sigma Aldrich. SiO₂ molecular weight was 60.08 g/mol. Physical properties were: surface area: 200 m²/g (±25 m²/g), bulk density: 2.3 lb/cu.ft, and average particle size: 0.2–0.3 µm. Different proportions of FS solid powder (0.5–1–2% Si) were added to PP via extruder machine (Gülnar Machine). Pure PP was used as a reference in this study. In the extruder machine (16 mm double screw, L/D: 40), the inlet temperature was 50 °C while doping the polymer, and the feed temperatures were set to 180, 190, 200, 200, and 190 °C. Strips of 113.5 cm in length, 6.1 cm width, and micron-size thickness were produced from 6.75 g of polyethylene. Concentrated glycerin, iodo-methane, deionized water, and ethylene glycol were used for contact angle measurements. The test liquids' surface tension was defined using the pendant drop method (KSV CAM200, KSV Instruments, Finland). All measurements were performed at 21 °C.

The polymers produced from the extruder in the form of film strips were cut rectangularly in an area of 50 cm². Holes were opened on the samples with a pin tip to facilitate the airflow (Fig. 9). These samples were then hung one by one at a distance of 10 cm opposite a small fogger machine (Lemon Humidifier, working voltage: DC5V, water capacity: 180 mL spray amount: 20 mL/h) (Fig. 7). The same procedures were applied for all samples. The experiment was carried out under normal room conditions, at 20% humidity, and in an open room closed to the airflow (Fig. 10). Figure 10a shows the water droplets collected on the sample in 1 min. In Fig. 10b, the diameter of the holes drilled on the materials was taken through different measurements. Accordingly, the average hole's diameter on the samples was 450.6 µm. The smallest hole diameter was 295 µm, and the largest one was 550 µm. No holes with a smaller or larger diameter than these values were found.

**Fig. 3 Energy dispersive X-ray spectrometer analysis of the samples. a Undoped, pure polypropylene. b 0.5% fumed silica-doped polypropylene. c 1% fumed silica-doped polypropylene. d 1.5% fumed silica-doped polypropylene. e 2% fumed silica-doped polypropylene.**
Fig. 4  Zisman plot and water drop on the surface of the samples. a Pure polypropylene Zisman plot and water drop on the surface. b 0.5% fumed silica-doped polypropylene Zisman plot, and water drop on the surface. c 1% fumed silica-doped polypropylene Zisman plot and water drop on the surface. d 1.5% fumed silica-doped polypropylene Zisman plot and water drop on the surface. e 2% fumed silica-doped polypropylene Zisman plot and water drop on the surface. The water's contact angle with the fog harvesting materials is measured as 103° for pure polypropylene, 97° for 0.5% fumed silica-doped PP, 92° for 1% fumed silica-doped PP, 100° for 1.5% fumed silica-doped PP, and 102° for 2% fumed silica-doped PP. $\cos \theta$ is drop angle on the surface and $\gamma$ is surface tension of the test liquids.
The materials’ surface properties were examined using an EVO40-LEO computer-controlled digital SEM. Quantitative elemental analysis was performed with an EDX spectrometer attached to SEM (EDAX Octane).

The surface tensions were measured using KSV CAM200, KSV Instruments, at room temperature, under room condition, and controlled airflow. An analytical balance weighed the water collected on the sample surface. FTIR device (Bruker Alpha, having a resolution of 4 cm⁻¹; equipped with a DTGS detector, performing ten scans for each spectrum) was used to record vibration peaks.

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**Analysis**

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Fig. 10  Water collected in 1 min, and the diameter of the holes on the sample surfaces, scanning electron microscope images. a The water droplets collected on the sample in 1 min. b The diameter of the holes drilled on the materials was taken through different measurements. The experiment was carried out under normal room conditions, at 20% humidity, and in an open room closed to the airflow.

DATA AVAILABILITY
The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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ADDITIONAL INFORMATION

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