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Preparation and Production of Acrylic Acid - Methyl Methacrylate Hydrogel and Its Antibacterial Properties

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ABSTRACT

In this study, acrylic acid-methyl methacrylate hydrogel was synthesized using a radical initiator in the presence of ethylene glycol dimethacrylate cross linker and the role of different factors such as optimum concentration of monomers, optimum initiator concentration, optimum percentage of monomers, optimum temperature, reaction time and optimum concentration of crosslinker was determined. After synthesis in addition to the water adsorption property, the antibacterial properties of hydrogel in the presence and absence of anionic and cationic surfactants were investigated by agar test. Best recorded conditions for hydrogel synthesis are as follows; (t = 60 min), ethylene glycol dimethacrylate crosslinker (EGDMA) 0.075 molar, methyl methacrylate monomer percentage (MMA = 80% 1M) and acrylic acid percentage (AA = 20% 1M), Benzoyl peroxide initiator concentration (BPO = 0.08) and optimum temperature of 70°C. On the other hand, the antibacterial effect of hydrogel on Gram-positive Bacillus was more effective than Gram-negative *E. Coli*. Hydrogel structure was confirmed by scanning electron microscope (SEM), thermal gravimetric analysis (TGA) and FT-IR (infrared spectroscopy).

INTRODUCTION

Hydrogels are three-dimensional polymeric networks with crosslinks that have the ability to absorb much water or biological fluids, even under pressure. These compounds can absorb a large amount of water without dissolution. Hydrogels are chemically or physically networked. Increasing attention to physical hydrogels is due to the relative ease of their synthesis process, while chemical hydrogels are favored because of their good mechanical strength. Also, natural hydrogels are very interesting because of the variety, abundance, cheapness, renewability, non-toxicity as well as biodegradability and biocompatibility compared to synthetic hydrogels. In the past few decades, hydrogels, because of their unique properties have been used in various industries such as food, packaging, pharmaceuticals, agriculture, biomedical and bioengineering. They have also been used in the manufacture of technical and electronic devices as well as adsorbents for the removal of pollutants for environmental applications (1-4). In selecting materials for their intended use, the special properties of the superabsorbent polymer are crucial. They have three basic properties: absorption capacity, absorption rate and swelling power of the gel that determine the actual absorption capacity. Finally, salt absorption rate is measured under load, which is stated multiple times in technical reports and inventions. There are several ways to improve gel strength, including increasing crosslinking density (resulting in fragility and low absorption of gels), surface crosslinking (using other crosslinkers for more treatment on the surface of superabsorbent polymer particles), and introducing minerals and or suitable polymers to the hydrogels for preparation of (nano) composite structures (5). In this study, the antibacterial behavior of hydrogels and their synthesis were investigated. Hydrogels may be synthesized in a number of " classical " chemical ways. These include single-step methods such as polymerization and parallel crosslinking of multifunctional monomers, as well as multi-step methods involving the synthesis of polymer molecules with their reactive groups and their subsequent crosslinks, possibly also by reacting polymers with proper crosslinking agents. A polymer engineer can design and synthesize polymer networks by controlling the structure on a molecular scale, like crosslink density, and also with appropriate properties, including biodegradation, mechanical and chemical strength and biological response to stimuli (5).

They may respond to a significant volume transfer with various types of physical and chemical stimuli. Physical stimuli include temperature, electric or magnetic field, light, pressure and sound, while chemical stimuli include pH, solvent composition, ionic strength and molecular species. The extent of swelling or de-swelling in response to the changes in the external environment of the hydrogel could be so drastic that the phenomenon is referred to as volume collapse or phase transition (6).

METHOD OF EXPERIMENT

In the first step, purification of the monomers was performed by vacuum distillation to remove the inhibitors and to recrystallize the benzoyl peroxide initiator, 2 g of benzoyl peroxide dissolved in chloroform and after filtering the solution, it was added to ice-cooled ethyl alcohol provided that the alcohol volume was ten times greater than the solution so that the benzoyl peroxide crystals would deposit. The crystals were then filtered and dried in a vacuum and during the tests, the initiators and monomers were kept in the dark at low temperatures. The total volume in the experiment was chosen to be 25 ml.

EXPERIMENT

The mixture of acrylic acid (AA) and methyl methacrylate (MMA) (1M) monomers were 0.4 ml (20%) and 1.94 ml (80%) respectively, with (0.08 M) 0.48 g Benzoyl peroxide (BPO) dissolved in 2 ml of acetone as initiator, and (0.075 M) 0.33 ml of ethylene glycol dimethacrylate (EGDMA) as cross linker as well as 23 ml of distilled water was stirred and mixed into Pyrex mixing cylinder, and then the tube was inserted into water bath was at a constant temperature of 70 ℃ and total volume of 25 ml for one hour. After a preset amount of time the filtering was done with Whatman paper. Then the sample was washed with distilled water and acetone and dried at 30℃ for 24 hours in an oven and weighed.

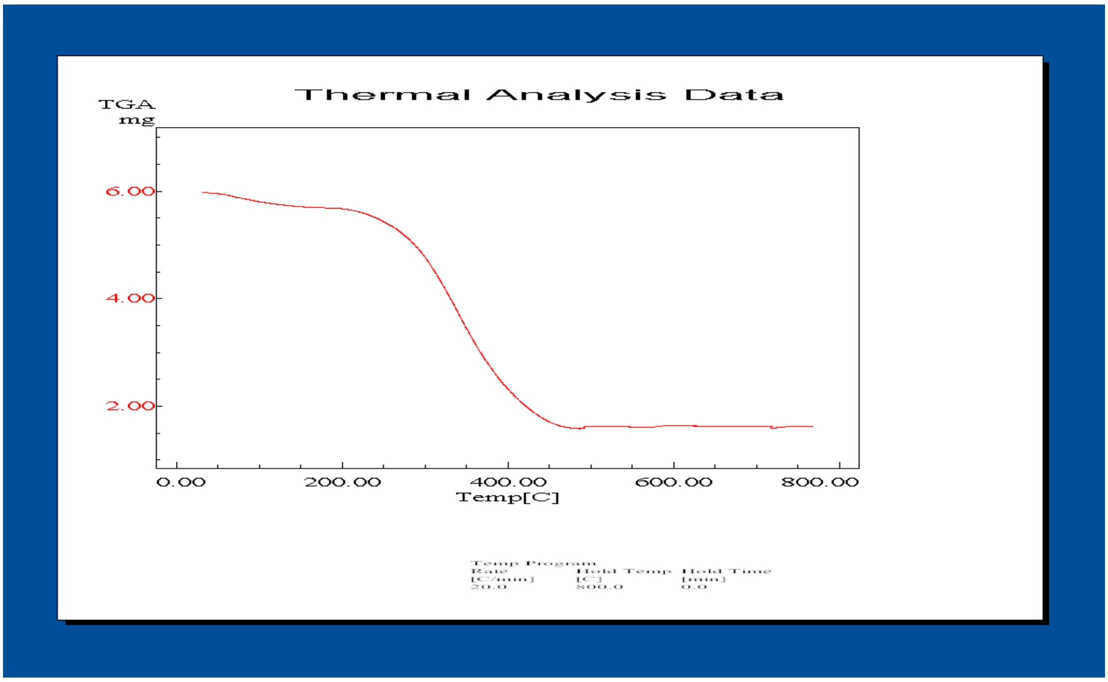
RESULTS

Best recorded conditions for hydrogel synthesis are as follows; (t = 60 min), ethylene glycol dimethacrylate crosslinker (EGDMA) 0.075 molar, methyl methacrylate monomer percentage (MMA = 80% 1M) and acrylic acid percentage (AA = 20% 1M), Benzoyl peroxide initiator concentration (BPO = 0.08) and optimum temperature of 70°C.

Tests for the confirmation of Hydrogels

Thermogravimetric Analysis (TGA)

Heating characteristics of 20 °C / min is relatively stable. Thermal cracking from about 210 °C which is a relatively high stability for hydrogel.



Graph 1: Thermogravimetric Analysis (TGA)

Scanning Electron Microscope

As shown in Figures 1 through 10, the hydrogel surfaces do not have impurities at different magnifications, and their transparency indicates that the hydrogel is pure.

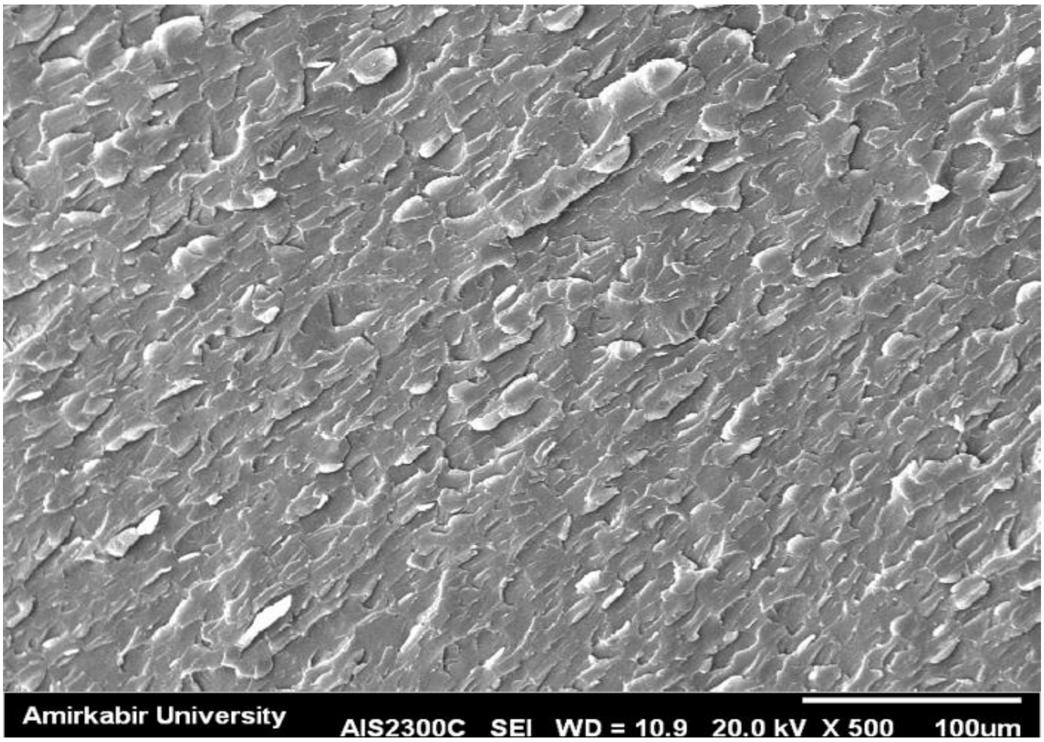


Figure No. 1: SEM WD = 10.9 20.0 kV * 500

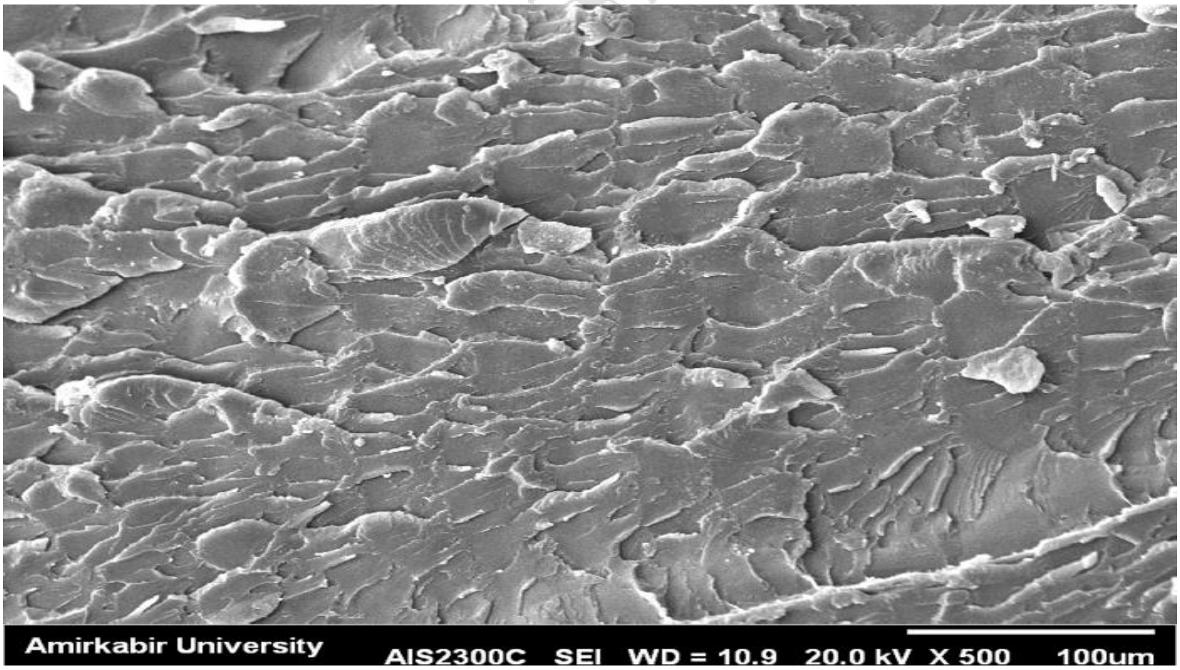


Figure No. 2: SEM WD = 10.9 20.0 kV * 500

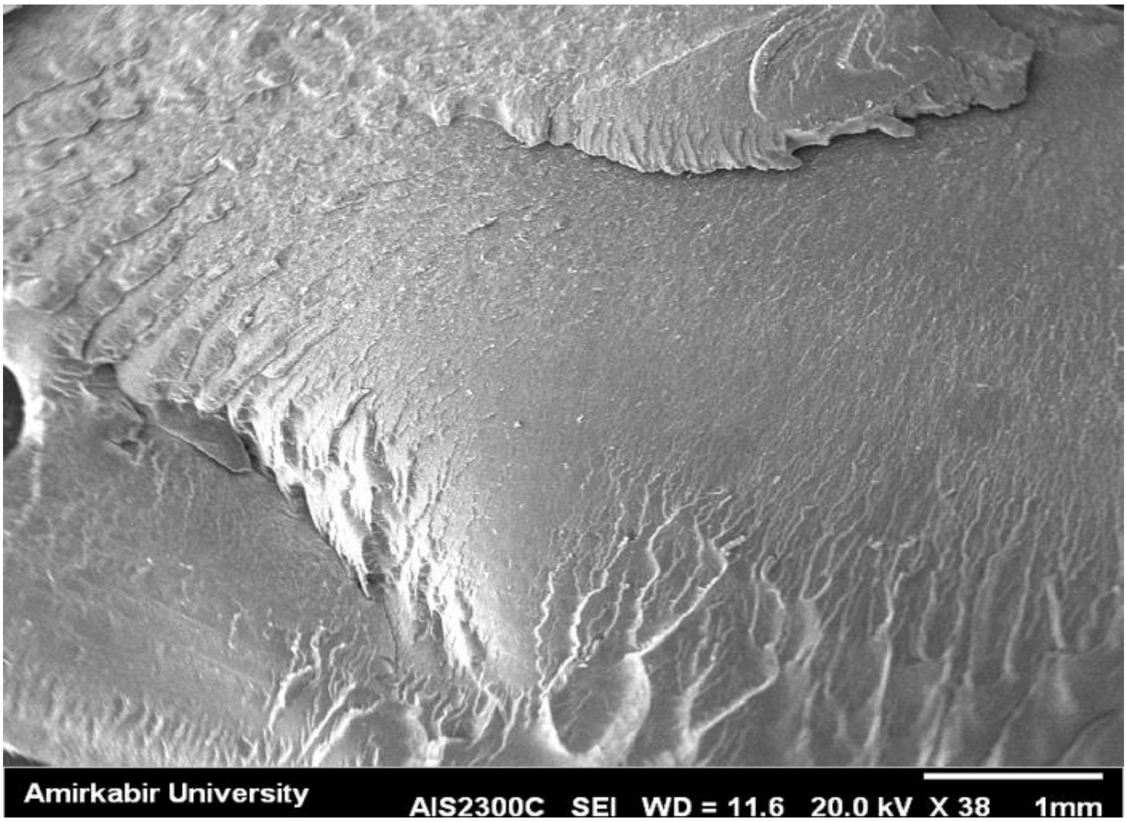


Figure No. 3: SEM WD = 11.6 20.0 kV * 38

Figure No. 4: SEM WD = 10.8 20.0 kV * 100

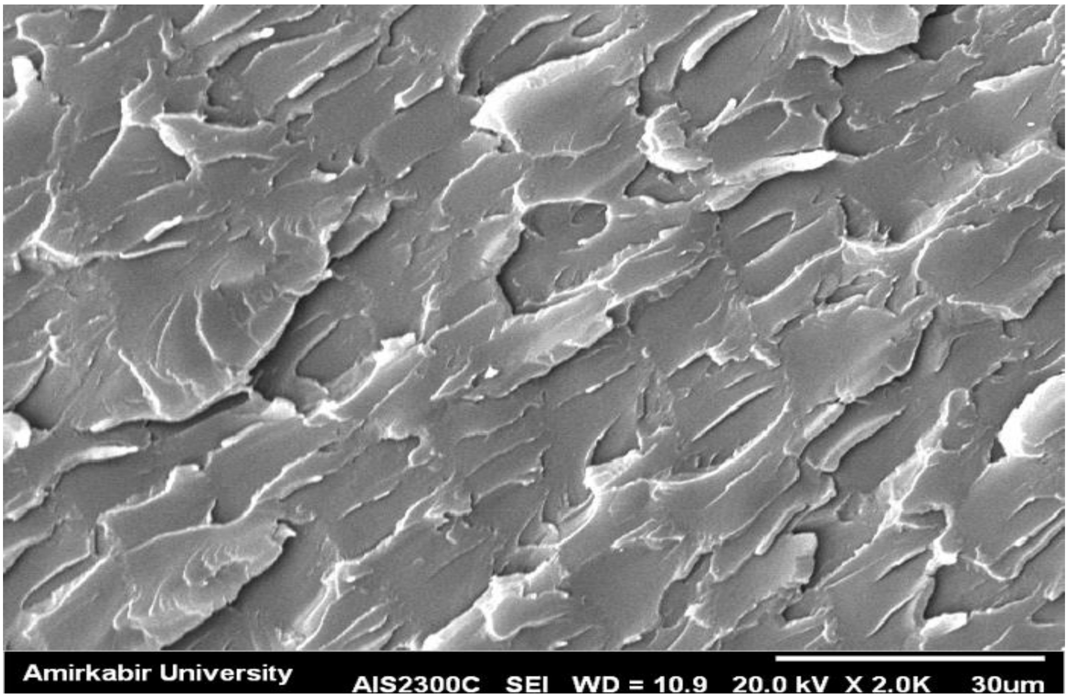


Figure No. 5: SEM WD = 10.9   20.0 kV   * 2.0K

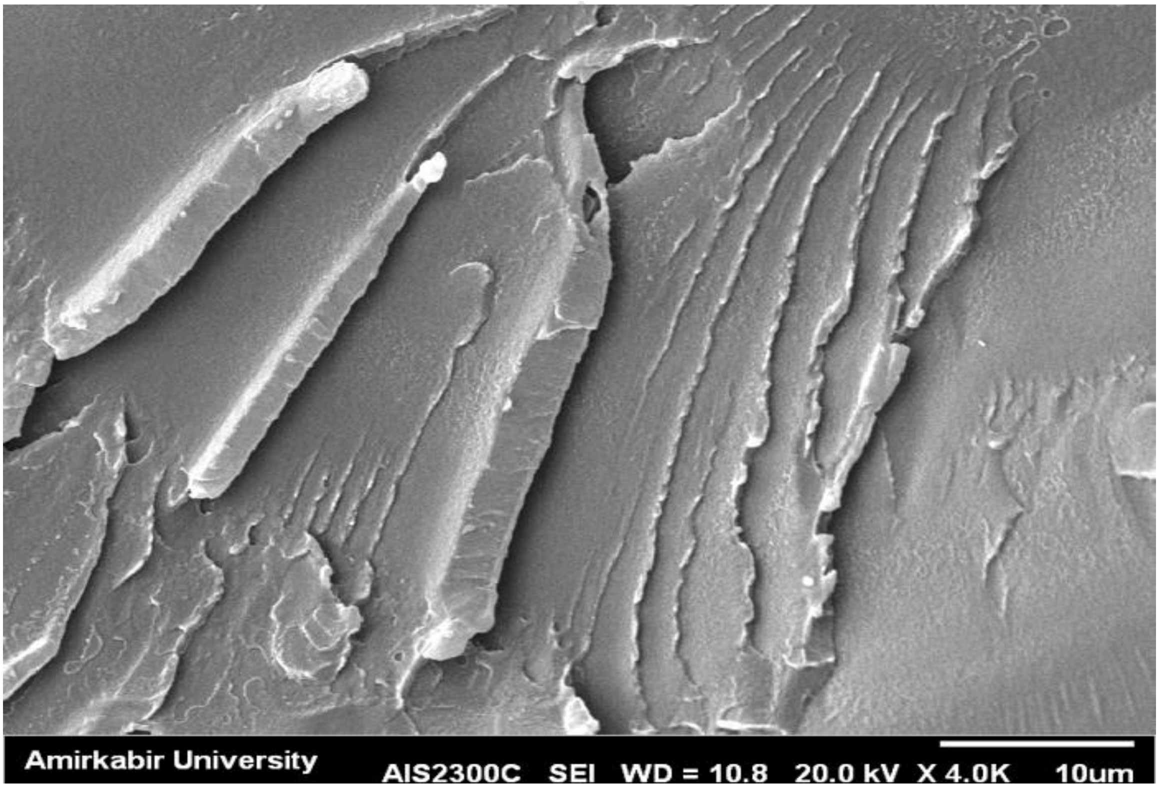


Figure No. 6: SEM WD = 10.8   20.0 kV * 4.0K

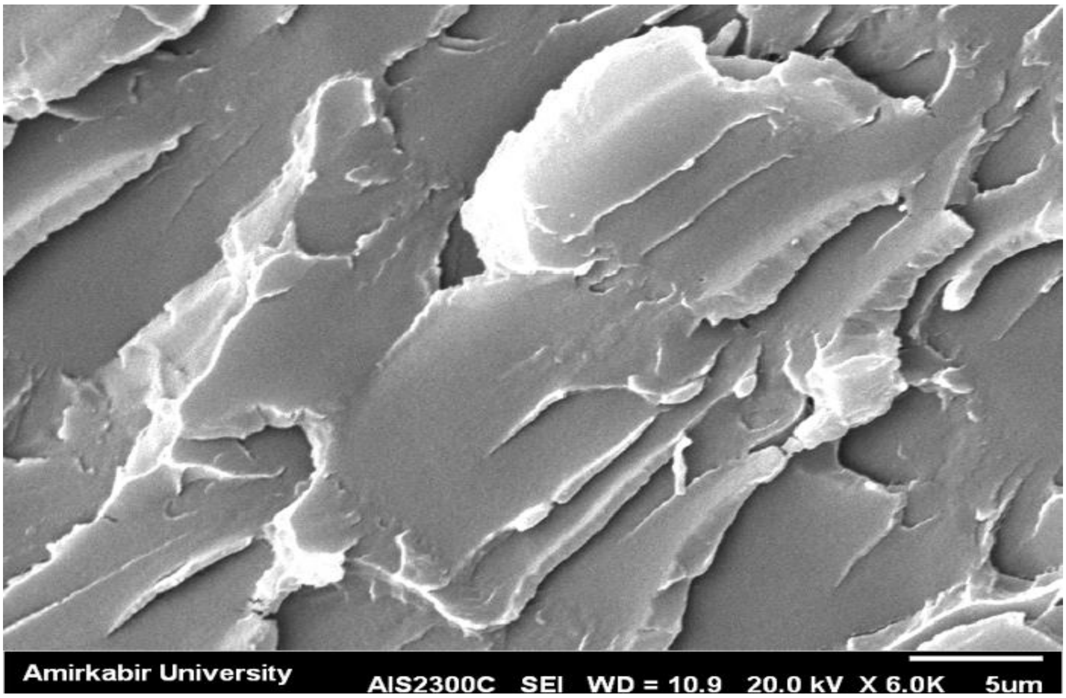


Figure No. 7: SEM WD = 10.9 20.0 kV * 6.0K

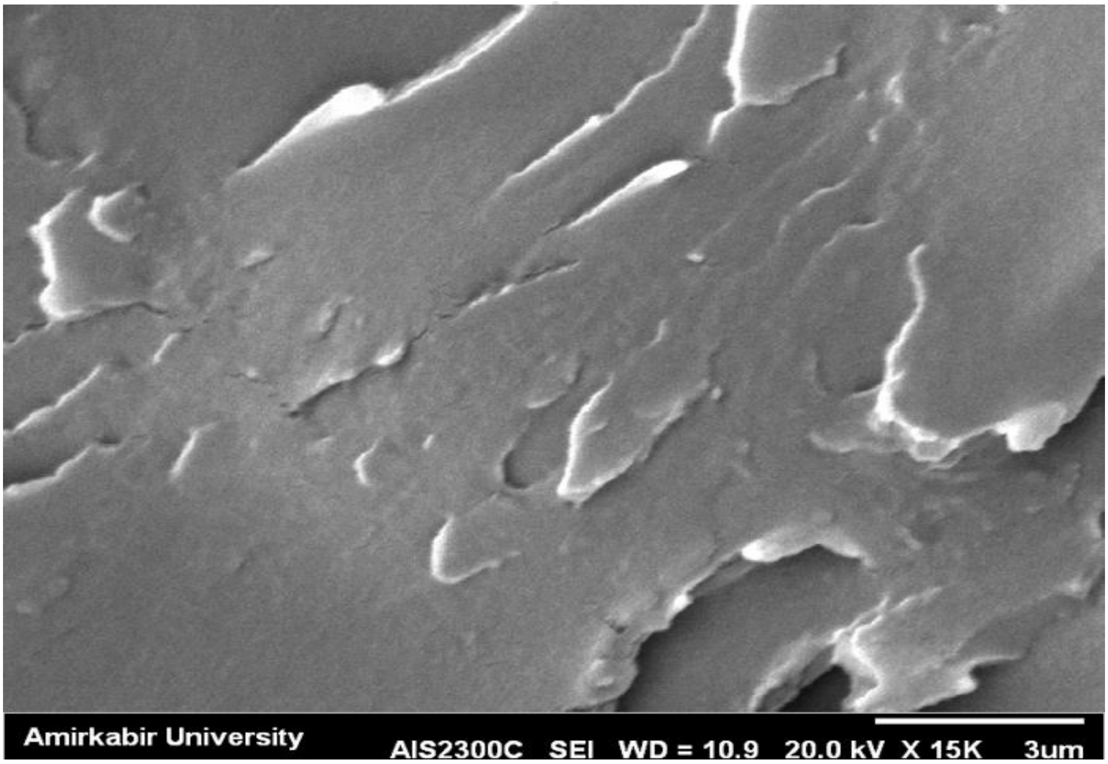


Figure No. 8: SEM WD = 10.9 20.0 kV * 15K

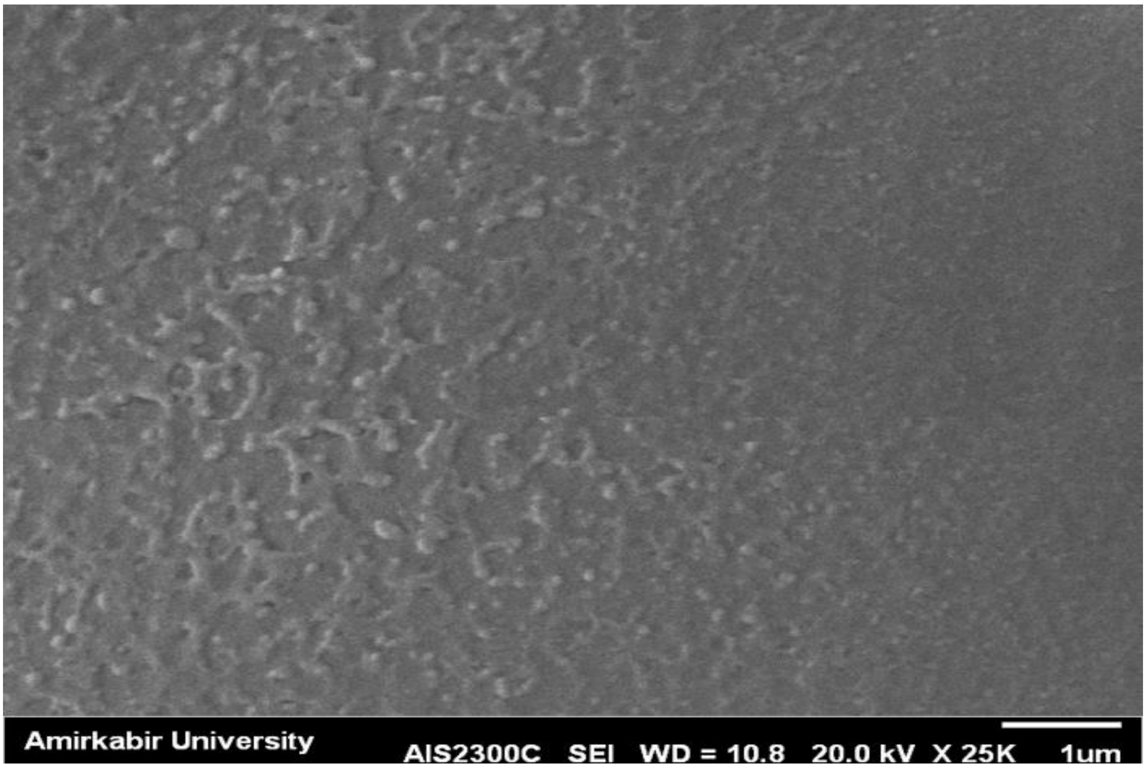


Figure No. 9: SEM WD = 10.8 20.0 kV * 25K

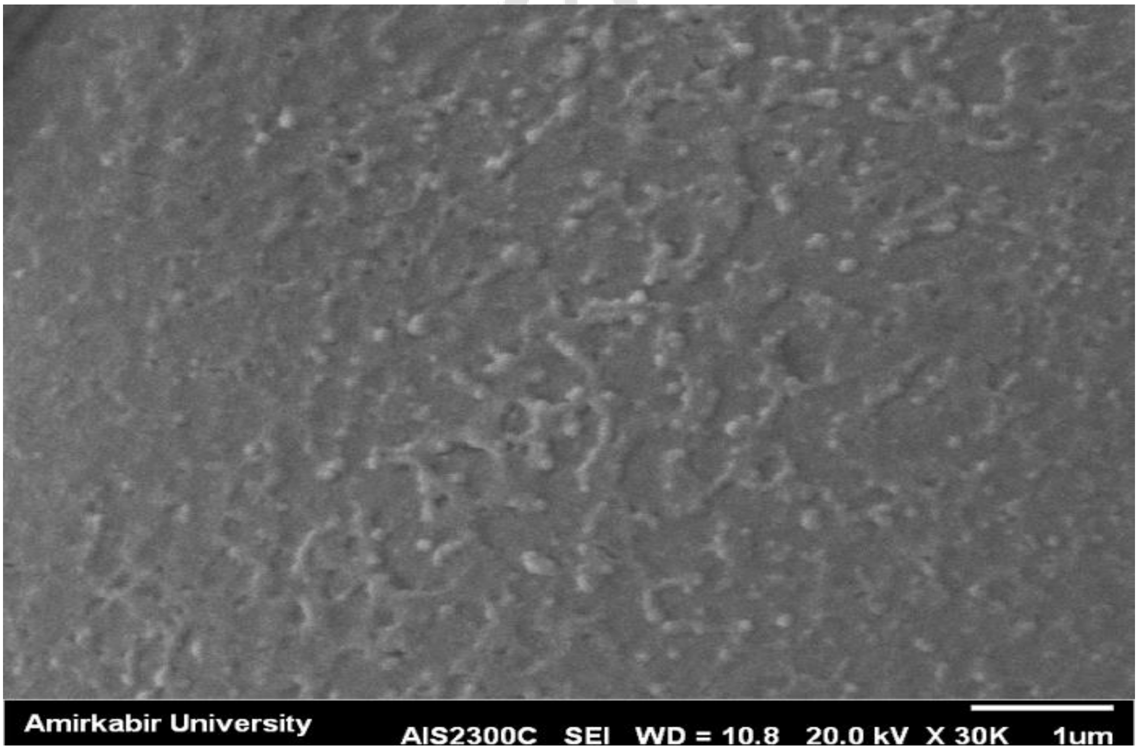


Figure No. 10: SEM WD = 10.8 20.0 kV * 30K

FT-IR Infrared Spectroscopy of Synthesized Hydrogel

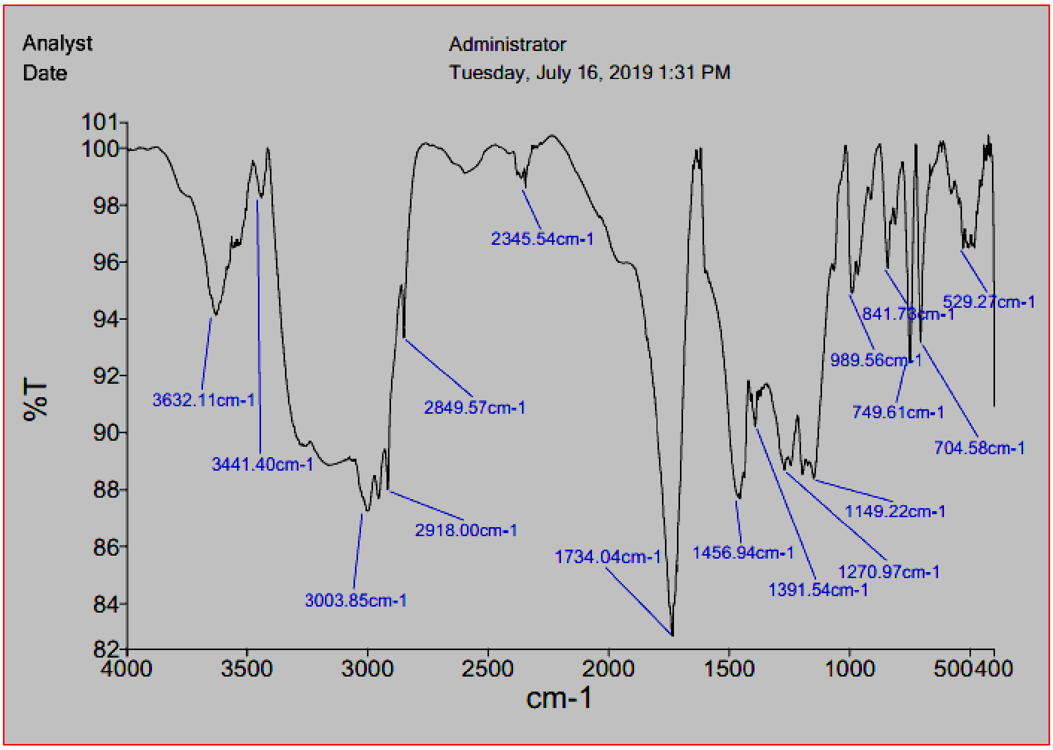
1. The sharp peak in the area of 3632.1 cm^{-1} is related to the O-H alcohol stretch.

2. The wide peak corresponds to the OH stretch of the carboxylic acid group ranging from 2849.57 cm^{-1} to 3441.40 cm^{-1}

3. The sharp peak corresponding to C=O strong stretch is related to the ester group at 1734.04 cm^{-1}.

4. The average and weak peak with multiple bonds corresponds to C=C at 1456.94 cm^{-1}.

5. Small peaks in the range of 1391.54 cm^{-1} and 1270.97 cm^{-1} are related to C-O stretch.



Graph 2: FT-IR Infrared Spectroscopy

Water Absorption by Hydrogel in Different pH Levels

We weighed the hydrogel and obtained the final weight and calculated the water absorption percentage using the following equation.

Water absorption percentage= final weight – initial weight/ initial weight * 100

$$pH: 3/16 \rightarrow \text{water absorption percentage} = \frac{0/230 - 0/2}{0/2} * 100 = 15$$

$$pH: 5/5 \rightarrow \text{water absorbtion percentage} = \frac{0/221 - 0/2}{0/2} * 100 = 10/5$$

$$pH: 8/62 \rightarrow \text{water absorption percentage} = \frac{0/319 - 0/2}{0/2} * 100 = 59/5$$

Based on the above calculations, we conclude that the water absorption rate of the hydrogel at alkaline pH is higher.

The Results of Microbial Culture and the Inhibitory Effect of Hydrogel on *Escherichia Coli* (E.coli) and Bacillus Bacteria

The results of microbial culture and the effect of hydrogel inhibition on Bacillus and *Escherichia coli* show that the protective and inhibitory effect in the presence of hydrogel was effective for Bacillus and figures 11 to 14 illustrate this claim. However, the microbial growth of *E.coli* has changed little in the presence of hydrogel. Therefore, the above synthesized hydrogel can be studied in industrial scale and on other bacteria.

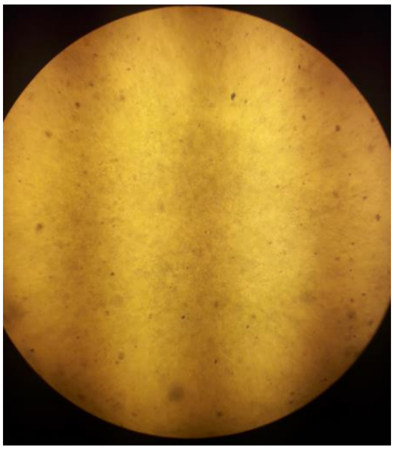


Figure No. 11: control sample of *E.coli*

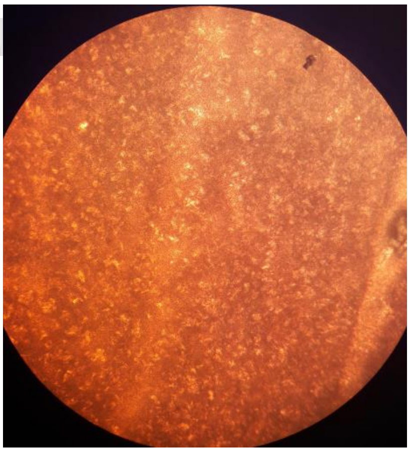


Figure No. 12: control sample of Bacillus

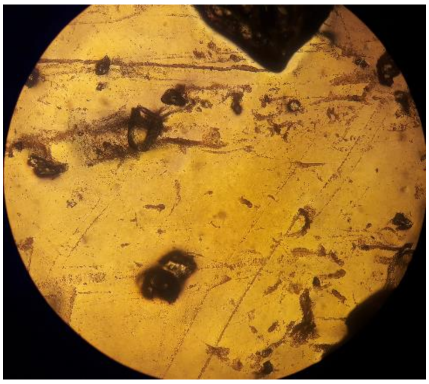


Figure No. 13: Bacillus sample

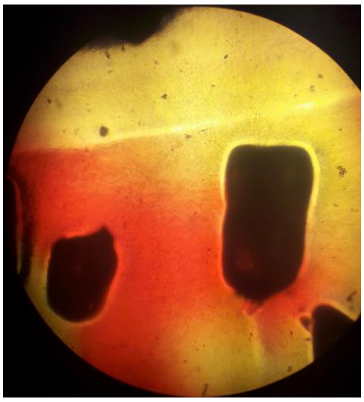


Figure No. 14: *E.coli* sample

Microbial Test Confirmation

To confirm the microbial test, samples of optimal hydrogel concentration (0.25 g, 0.5 g, 0.75 g) were placed on the plate surface area and compared with the control sample. Then, under the optimum conditions of the above inhibition in the presence of hydrogel synthesized with Cetyl Trimethyl Ammonium Bromide (CTAB) and Sodium Dodecyl Sulfate (SDS), agar test was performed for comparison and its effective role was investigated.

Effects of Sodium Dodecyl Sulfate Anionic Surfactant (SDS) on the Growth of *Escherichia Coli (E.coli)* and Bacillus Bacteria

Due to the anionic surfactant of sodium dodecyl sulfate (SDS = 2%), the amount of hydrogel was approximately equal to 1.85 g and the effect of its antibacterial properties on Escherichia coli and Bacillus bacteria was evaluated under optimal conditions. As can be seen in the presence of SDS surfactant, the inhibitory effect is higher in Figures 15 to 20 than in the surfactant-free state. The presence of SDS anionic surfactant has improved the antibacterial effect.

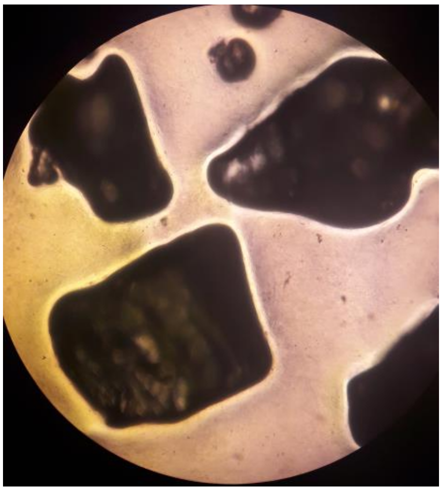


Figure No. 15: 0.25 g SDS E. coli sample

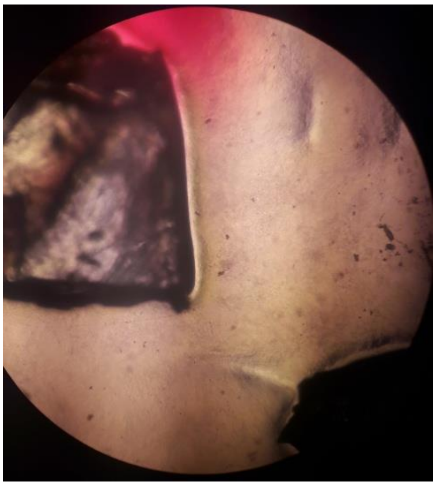


Figure No. 16: 0.5 g SDS E. coli sample

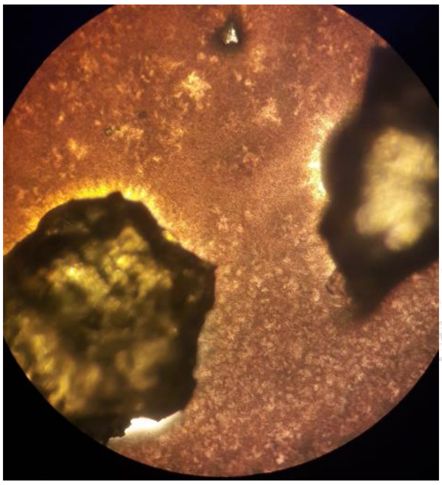


Figure No. 17: 0.1 g SDS E. coli sample

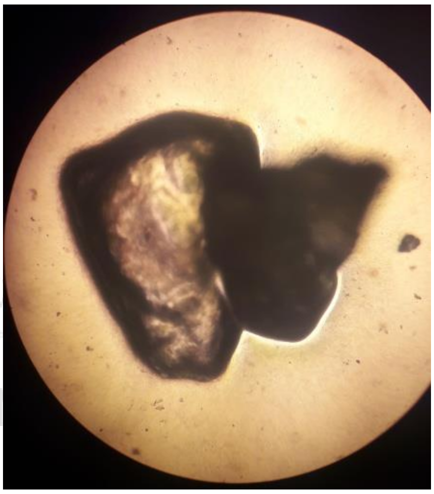


Figure No. 18: 0.25 g SDS Bacillus sample

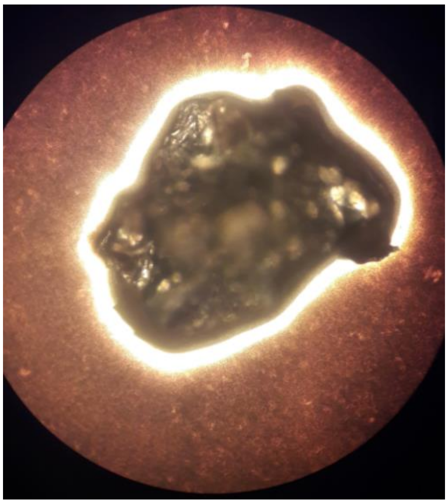


Figure No. 19: 0.5 g SDS Bacillus sample

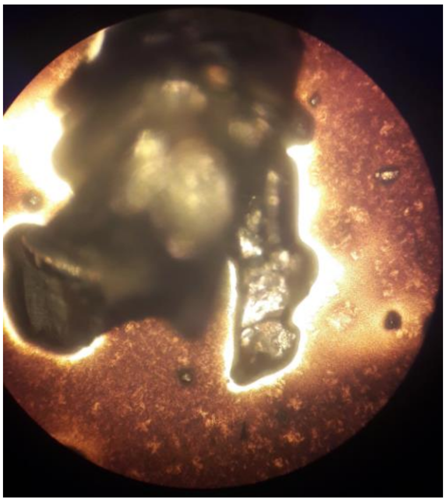
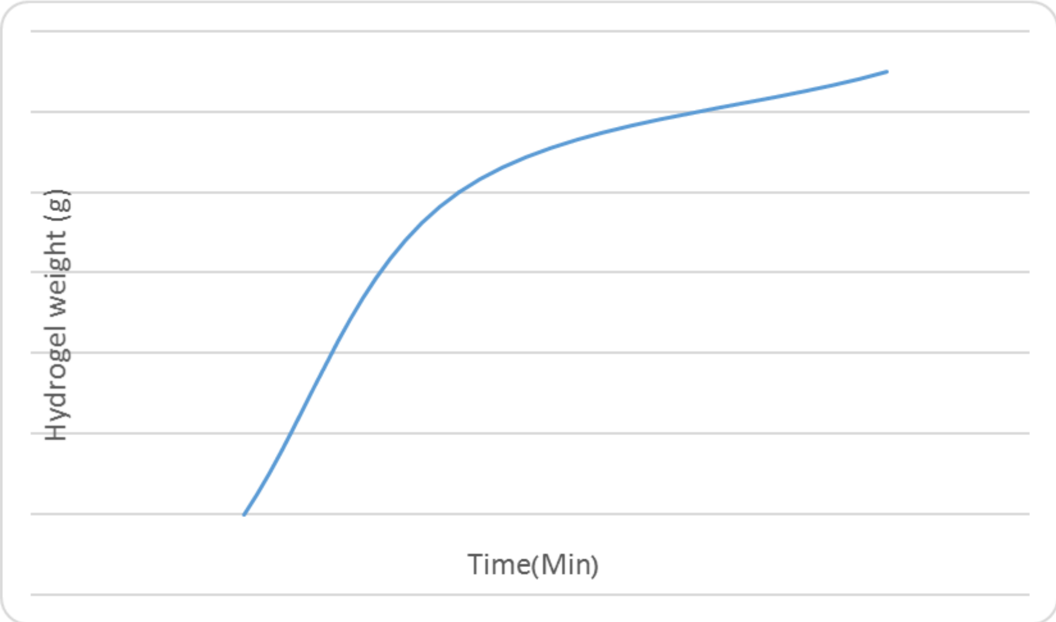


Figure No. 20: 0.1 g SDS Bacillus sample

The effect of time

At 30, 60, and 120 minutes, the hydrogel production was evaluated and the optimum time was 60 minutes, as shown in the diagram.



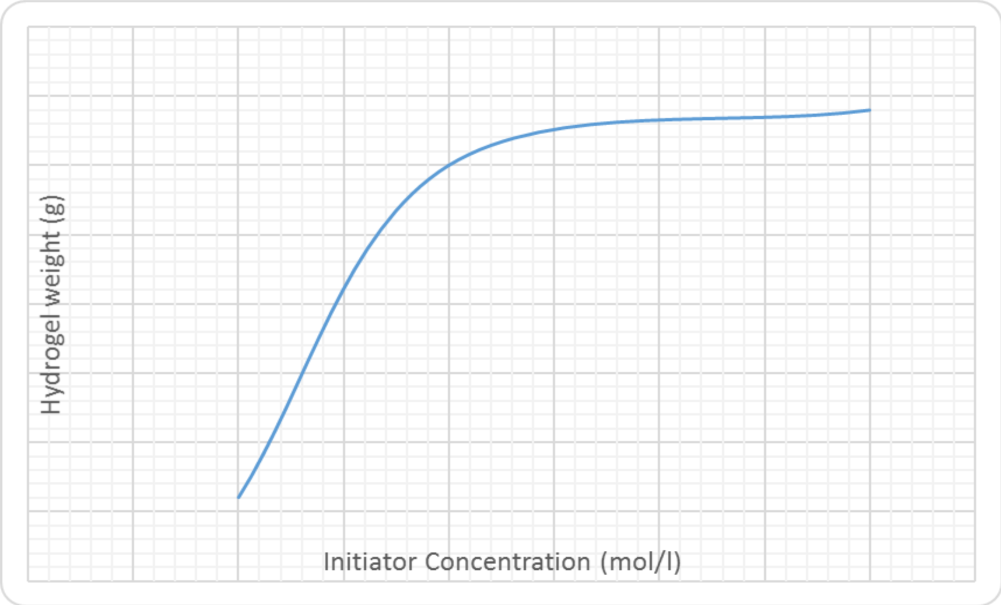
Graph 3: The effect of time

The Effect of Monomers Concentration Percentage

The acrylic acid (AA) polymer solvent is water and the methyl methacrylate (MMA) polymer solvent is acetone and toluene. For this reason, as the percentage of methyl methacrylate (MMA) increases, the produced hydrogel amount increases.

Initiator Concentration Effect

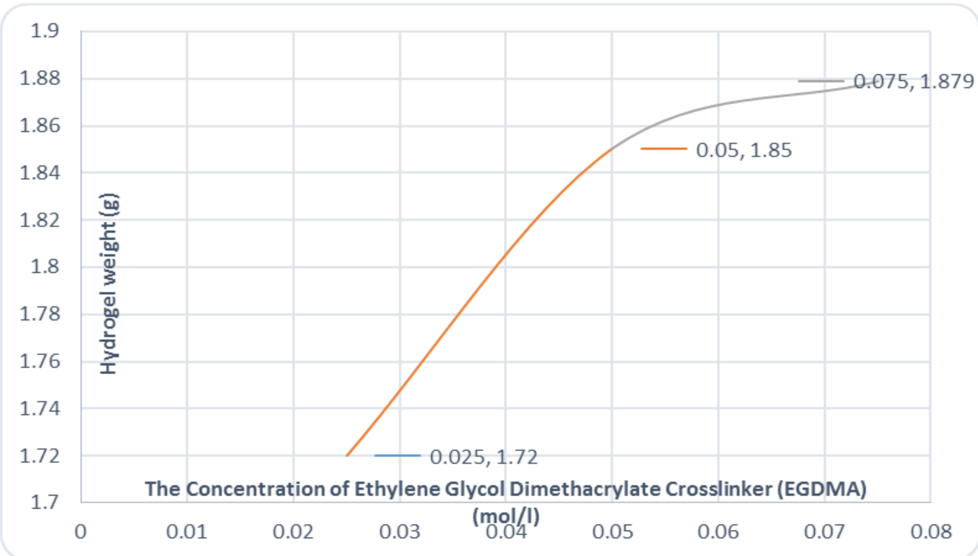
As the amount of initiator decreases, the possibility of forming active centers in radicals is reduced and the amount of hydrogel is reduced. And with increasing initiator amount, the possibility of forming active centers in radicals is increased and the amount of hydrogel increases; like the catalyst which partially increases the reaction rate and then the increase of the catalyst has no effect on the reaction rate. And by increasing the initiator concentration from 0.04 M or (0.24 g) to 0.08 M or (0.48 g), it is possible to form homopolymers and is not suitable for hydrogel formation.



Graph 4: Initiator Concentration Effect

The Concentration Effect of Ethylene Glycol Dimethacrylate Crosslinker (EGDMA)

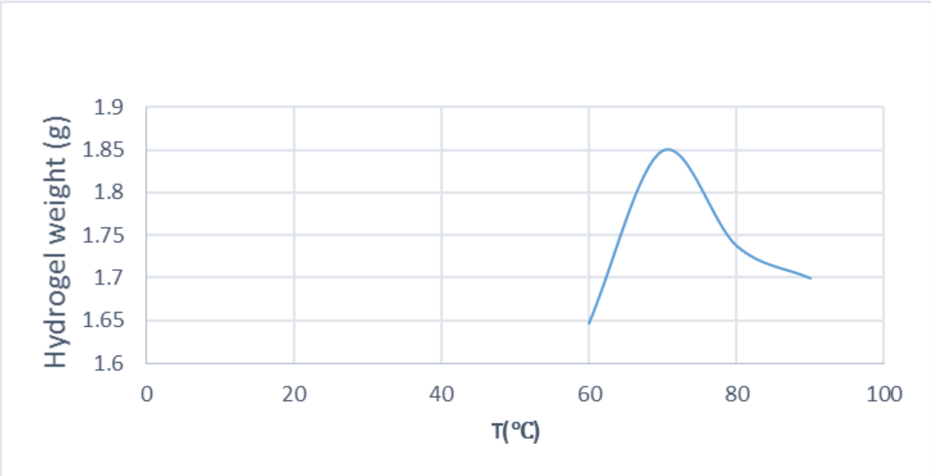
Effect of ethylene glycol dimethacrylate (EGDMA) as crosslinker on the hydrogel production in the amounts of (0.025 M) 0.11 ml, (0.05 M) 0.22 ml, (0.075 M) 0.33 ml was investigated. The optimum crosslinking concentration of ethylene glycol dimethacrylate (EGDMA) (0.075 M) is considered (0.075 M) 0.33 ml, as shown in the diagram.



Graph 5: The Concentration Effect of Ethylene Glycol Dimethacrylate Crosslinker (EGDMA)

The Effect of Temperature

The results show that 70 ° C is the optimum temperature for production of maximum weight of hydrogel.



Graph 6: The effect of temperature

DISCUSSION

In this study, acrylic acid-methyl methacrylate hydrogel was synthesized using a radical initiator in the presence of ethylene glycol dimethacrylate cross linker and the role of different factors such as optimum concentration of monomers, optimum initiator concentration, optimum percentage of monomers, optimum temperature, reaction time and optimum concentration of crosslinker was determined. After synthesis in addition to the water adsorption property, the antibacterial properties of hydrogel in the presence and absence of anionic and cationic surfactants were investigated by agar test. Hydrogel structure was confirmed by scanning electron microscope (SEM), thermal gravimetric analysis (TGA) and FT-IR infrared spectroscopy.

The total volume in the experiment was chosen to be 25 ml.

REFERENCES

[1] Nagam SP, Jyothi AN, Poojitha J, Aruna S, Nadendla R. A comprehensive review on hydrogels. Int J Curr Pharm Res. 2016;8(1).
[2] Ganji F, VASHEGHANI FE. Hydrogels in controlled drug delivery systems. 2009.
[3] Ullah F, Othman MBH, Javed F, Ahmad Z, Akil HM. Classification, processing and application of hydrogels: A review. Materials Science and Engineering: C. 2015;57:414-33.

[4] Laftah WA, Hashim S, Ibrahim AN. Polymer hydrogels: A review. Polymer-Plastics Technology and Engineering. 2011;50(14):1475-86.
[5] Satarkar NS, Hawkins AM, Hilt JZ. Hydrogel nanocomposites in biology and medicine: Applications and interactions. Biological Interactions on Materials Surfaces: Springer; 2009. p. 319-42.
[6] Shin J, Braun PV, Lee W. Fast response photonic crystal pH sensor based on templated photo-polymerized hydrogel inverse opal. Sensors and Actuators B: Chemical. 2010;150(1):183-90.