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Synthesis and Characterization of Polyacrylamide co acrylic acid Graphene Oxide Nanocomposite for Pharmaceutical Packaging Application

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Synthesis and Characterization of Polyacrylamide co acrylic acid Graphene Oxide Nanocomposite for Pharmaceutical Packaging Application

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Thesis Approval

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Dedication

At the final step of my research, I would like to dedicate it firstly to my special land "Palestine". As well as to Al-Quds University; The university of excellence and excellence and An Najah University which is my first incubator at the level of Bachelor's .

This thesis is dedicated to my supervisor Dr. Wadie Sultan and co-supervisor Dr. Sami Makharza who gave me a continuous support in the time I was in need. I will never forget to dedicate this achievement to Mrs. Maryam Faroun; the researcher at Nano lab who did not hesitate a moment to help me in anything.

To my husband, Eseed, who, whatever I said, I can not to thank him, my husband who was my first support to go in this way and did not hesitate for any time for giving me a moral and financial support.

To my child, Lelian, who is bearing my absence. When she will grow up ,she will be proud of her mother so much and forgive me for negligence towards her.

Dad,Mom and all my family who gave me the biggest support and always encouraging me to continue my education.

My second family, my husband's family and particularly my mother who supported me in different ways.

All my friends and people in my life who support, love and remember me.

I dedicate my research.

Declaration

I certify that this thesis submitted for the degree of master, is the result of my own research, except where otherwise acknowledged, and this thesis has not been submitted for the higher degree to any other university or institution.

Laila Motassem Abu Ayyash

Signed:

Date: 28/5/2022

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Laila Abuayyash

Abstract

Graphene oxide (GO) synthesized by oxidation reduction reaction (Hummers' method).It was used to be incorporated in polyacrylamide co acrylic acid (Poly(AAm-co-AA)) to study the thermal properties, mechanical properties, as well as the result of swelling in aqueous solution. Different concentrations of Poly(AAm-co-AA) (5,2.5,1.25, and 0.9%) with different concentrations of GO (0.05 and 0.1 %) were used to prepare Poly(AAm-co-AA) /Graphene oxide nanocomposite by using casting method.

Thermal properties were investigated by using Differential Scanning Calorimetery (DSC), the results described that the thermal properties improved and enhanced by incorporation of GO to Poly(AAm-co-AA) matrix. The glass transition temperature (Tg) was raised from 50 of neat Poly(AAm-co-AA) to 75 °C at 0.05% GO. The crystallinity was increased to a certain extent. The young's modulus was increased at 5% Poly(AAm-co-AA) as GO content increased to 0.1%. The swelling of Poly(AAm-co-AA) with different concentrations reduced by adding GO nanoparticles which explains the barrier properties of GO in the molecular level of polymer matrix. The FT-IR spectra of Poly(AAm-co-AA)/GO nanocomposite confirm the strong bonding between polymer and nanoparticles in different functional groups. Briefly, the incorporation of GO into Poly(AAm-co-AA) polymer matric improves the mechanical thermal and barrier properties of polymer because of strong interaction between GO particles and polymer.

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LIST OF ABBREVIATIONS

AFM	**Atomic Force Microscope**
CGMP	**Current Good Manufacturing Practice**
CMG	**Cystometrogram**
CVD	**Chemical Vapor Deposition**
EELS	**Electron Energy Loss Spectra**
FDA	**Food and Drug Administration**
FLG	**Few Layer Graphene**
GO	**Graphene Oxide**
GOFs	**Graphene Organic Frameworks**
IGO	**Improved Graphene Oxide**
LDA	**Local Density Approximation**
LEED	**Low Energy Electron Diffraction**
MCT	**Murcury Cadimum Telluride**
MC	**Methyl Cellulose**
NIR	**Intrinsic near-infrared**
Poly(AAm-co-AA)	**Poly acrylamide co acrylic acid**
PP	**Polypropylene**
PS	**Polystyrene**
PVA	**Poly Vinyl Alcohol**
RGO	**Reduced Graphene Oxide**
RT	**Room Tempereture**
SANCs	**Super adsorbent Nanocomposites**
STA	**Simultaneous Thermal Analysis Instrument**
TEP	**Thermoelectric power**
Tg	**Glass transition temperature**
TIMs	**Thermal Interface Materials**
Tm	**Melting temperature**
TGA	**Thermogravimetric Analysis**
TPa	**Tera Pascal**

Chapter One: Introduction:

Today,polyacrylamide polymer are one of the most famous used water soluble polymers. Technically the terminology of “polyacrylamide” is known as all products, which are produced by copolymerization with acrylic acid or its partial hydrolysis, and for which the name poly(acrylamide-co-acrylic acid) have to be used.[1]

Recently, chemically modified graphene (CMG) has been used in different studies in the scope of different applications, such as polymer nanocomposites, materials that are related to energy, different sensors, materials like ‘paper’, field-effect transistors (FET), and applications related to biomedical studies, due to its excellent properties like electrical, mechanical, and thermal ones.[1]

Packaging needs for pharmaceutical products are more complex than those of other non-edible. The pharmaceutical packaging materials are considered as protecting and preserving pharmaceutical products or drugs from damage by enclosing them. Packaging materials are any substance which enclose/surround the product after the time of production to till final usage [2].

Before any one starts to understand pharmaceutical packaging, many basic information is needed regarding chemistry, physiology, drug delivery pathways, drug properties, the food and drug administration (FDA),pharmaceutical current good manufacturing Practice (CGMP) and others. These complicated and large amount of information play a very important role in the design and required performance of pharmaceutical packaging [3].

The pharmaceutical packaging market is continuously improving and has faced annual growth of a minimum five percent per year in the last few years. Distribution of different products is now more worldwide than ever. Mass customization of packaging to allow its use in a variety of markets is an issue that needs concentration and discussion. Environmental properties, including sustainability, could always be an issue to think about to any packaging design [4].

A **Pharmaceutical Package container** is a substance or a device which has the Pharmaceutical or drug product and this package may or may not in direct contact with the drug[5].

1.1 Poly(acryl amide co acrylic acid)

1.1.1Polyacrylamide – co – acrylic acid structure and preparation

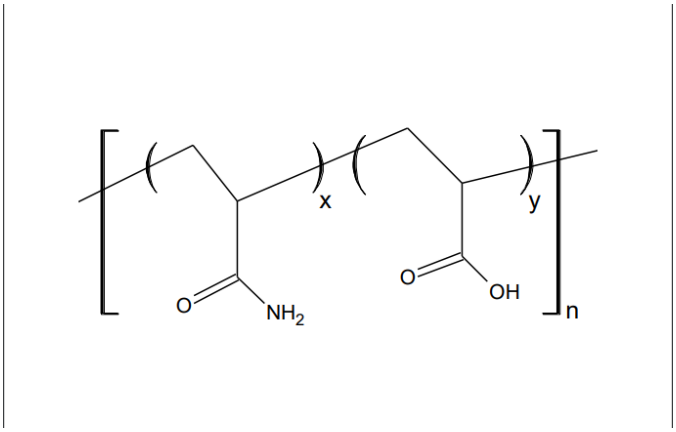


Figure 1.1:Chemical Structure Polyacrylamide co acrylic acid.

Synthesis of this copolymer can be done by copolymerization method of acrylic acid with acrylamide and by saponification (partial) of the functional group of amide in polyacrylamide (See figure1.1). The researchers followed two ways during the study with priority on the following properties: negligible degree of branching, controlled polymer's chain length (and chain length distribution), controlled chemical composition, and minimization of impurities [1].

In the Thakur's work, hydrogels that are based on acrylic acid and acrylamide which has a crosslink with N,N'-methylene bisacrylamide were synthesized by free radical polymerization in the solution without reducing or increasing the amount of acrylic acid as the ionic comonomer in water [6].

Poly(acrylamide-co-acrylic acid) hydrogels were prepared also by the free-radical crosslinking copolymerization of acrylamide monomer at accurate amounts, but at reducing or increasing the amount of acrylic acid as the ionic comonomer in water [7].

1.1.2 Swelling and mechanical properties of polyacrylamide co acrylic acid

In 2004, Poly(acrylamide-co-aclylic acid) was chosen as the basic material because of many properties like commercial availability and its high water affinity. The polymer in the swollen state did not have a very good mechanical strength [8].

Poly(acrylic acid) hydrogels are used in different applications because of their low cost and high water absorption capacity. In addition, this kind of super absorbent polymer has low salt retention capacity and very slow water absorption rate. Also, after it absorbs water, it shows dispersion elastic properties and inferior gel strength. These characteristics have highly reduced the product quality and its application fields [9].

Poly(acrylic acid) is one of the studied pH sensitive hydrogels .These hydrogels have ionic groups that detects pH changes due to its ionizable groups such as amino groups and carboxyl groups. This polymer shrinks and extends at pH below and above the pKa of the polymer (approximately pH 4.75).In the ionizable form of polymer it extends,so more amount of water is taken by the polymer.Because of their special deswelling and swelling attitudes, the pH-sensitive hydrogels are used for different potential applications that includes many fields such as chemical separation, chemical transducer, drug delivery and artificial organ [7].

1.2GO Structure and Synthesis

The graphene's study is the most attractive topics in the science of materials and physics(condensed matters) [10]. Graphene has very good properties for applications to be used in different fields.

1.2.1Chemical Structure:

See figure 1.2 for chemical structure of GO. Graphene is the main building block of the all graphitic structures of carbon.GO contains a single atomic layer of sp^2 hybridized carbon atoms organized in a shape of honeycomb. Research through this substance has been considering that graphene may have different exciting and interesting exciting physical characteristics, for instance, high degree of stiffness ,strength, thermal conductivity and an impermeability to the gases. One of the most clear application of graphene is in field of nanocomposites [11].

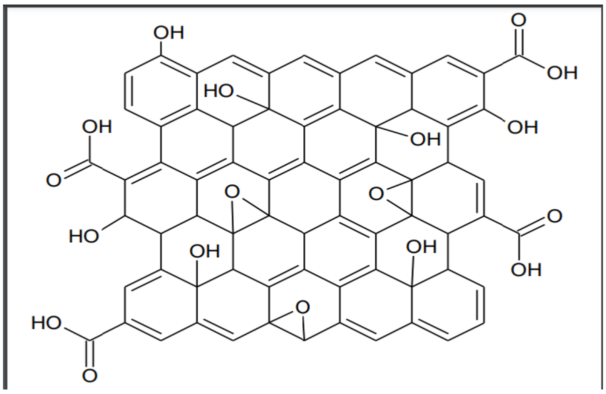


Figure 1.2: Chemical Structure of GO

1.2.2Method of preparation:

Comprehensive reviews in the synthesis for dispersions of GO sheets and RGO sheets, synthesized from GO, have newly appeared. Mainly GO is prepared by either the Staudenmaier, Brodie, Hummers method, or some combinations of that methods. The above methods include oxidation of the graphite at different stages [12].

Researchers mentioned that one method for synthesis of graphene is to breakdown graphite into graphene through methods like a liquid phase exfoliation or mechanical breakage (top-down approach) [13]. They mentioned other methods to prepare graphene is by using techniques such as epitaxial growth on silicon carbide or epitaxy of molecular beam, chemical vapor deposition (CVD) (known as bottom-up approach). The easiest way of synthesis of few or single layer graphene is through the mechanical breakdown. This method is done from either good-quality natural graphite or highly-oriented pyrolytic graphite [13].

Blake with his team mentioned that one of the first useful and successful methods was the dispersion and exfoliation of graphite in solvents specially organic solvents like N-methylpyrrolidone or dimethylformamide. More than 50% of graphene monolayers could be obtained when we control the agitation levels and the purification of the suspention [14].

In 2009. The researchers focused on the production of graphene from graphite by bottom up method. Many reports concerning on this point since 40 years ago[15].

The high quality of graphite crystals can be obtained by scotch tape, but in microscale level. This method is not applicable in large-scale preparations for practical applications. Alternative ways mentioned for large scale synthesis such as gas phase synthesis by CVD on Ni or Cu, chemical exfoliation of bulk graphite, organic synthesis of graphene nanoparticles, and epitaxial graphene growth on SiC wafers. Commenting on the above methods, the CVD synthesis is most useful in preparation of larger scale with a good quality of graphene films reaches up to meter scale [16].

The epitaxial graphene synthesis includes CVD growth on epitaxial matched metal surfaces. A researcher in an article published in 1969 talked about surface science unassigned low-energy electron diffraction (LEED) patterns can be rationalized in terms of a “monolayer of graphite [17].

According to the history of micromechanical exfoliation, in 1999, Ruoff with coauthors put the basics for micromechanical exfoliation and also studied outlines for the importance of graphene for a wide variety of basic applications [18].

The synthesis of graphene oxide particles through colloidal dispersions of graphite in solvents is known as exfoliation and oxidation of graphite. This is mainly done by mixing of graphite powders in different organic solvents for example NMP or DMF, then transfer it to high intensity ultrasound. This process is a significant process in synthesis of few layers of graphene oxide but the yield is not in large quantities[19].

Other methods mentioned that graphene platelets synthesized through substrate-free gas-phase by using a microwave plasma reactor. Muli-layer graphene synthesis through arc-discharging have recently been published. According to physical vapor deposition, scientists believe that preparation of graphene powders will increase in the future following this method and researchers concentrate on this method and suggest it as a scope of research. Also many articles published about the importance of CVD method in the production of large scale of graphene powder useful for different application in science [20].

Recently, Marcano, et. al,2010 reported the large scale synthesis of GO nanoribbons from multiwalled carbon nanotubes through using different reagents such as concentrated H_2SO_4 and $KMnO_4$.They explored that the addition of H_3PO_4 to the above reaction produced graphene oxide nanoribbons with the advantage of obtaining more intact graphitic basic planes. After the reduction of graphene oxide nanoribbons they had a very good conductivity. The researchers concluded that the above oxidation procedure which is ($KMnO_4$ and mixture in 9:1 of concentrated H_2SO_4/H_3PO_4 and $KMnO_4$, called the improved method) can be used for synthesis of improved GO (IGO) with less defects in the basic plane as compared to GO synthesized following the special Hummers' method [21].

1.3 Properties of graphene oxide:

1.3.1 Mechanical Properties of GO

It was reported that the Young's modulus of defect-free graphene is equal to 1.0 TPa.The fracture strength approximately about 130 GPa. Cystometrogram (CMG) was also explored by similar atomic force microscope (AFM) indentation method [22].

The mechanical properties of monolayer graphene oxide exhibited a lower effectual Young's modulus (about 23.4 GPa when the thickness of the layer is 0.7 nm) when compared to result reported for graphene alone. It was found that the value of graphene oxide is approximately 39.776.8 MPa concerning the pre-stressed membranes obtained through the method of solution-based deposition. It is 1 order of the magnitude less than that achieved by other for mechanically breaked graphene. The novel (FEM)-based mapping methods,AFM imaging and finite element method are of general ways for achieving the prestress of different thin membranes and elastic modulus for them [23].

A search was made in 2012 concluded that entering polyvinyl alcohol (PVA) polymer into the GO space increases the hydrogen-bonding network between the composite and the nanoparticles, in a pattern similar for that is found in the natural biomaterials, creating increase strength of the composite [24].

According to the stress strain of GO-Cement, it shows a wider stress–strain curve through the post-peak area, this leads to a less probability of sudden failure. The researchers mentioned that the addition of GO to GO-cement composite leads to an enhancement in the surface area. [25]

Medhekar et. al., explained that the high stiffness and the mechanical properties of GO platelets is due to the presence of functional groups with high density and hydrogen bonding as well. [26].

A research was made on GO and epoxy composite to compare the mechanical strength between the epoxy free and the GO epoxy composite. The results showed an increase in mechanical strength such as hardness, tensile test and impact test [2].

In another research,the mechanical charachters of graphene oxide was made based on intrinsic Young's modulus and strength, of graphene oxides, they made an amorphous and ordered graphene oxide for comparison. The Young's modulus for ordered graphene oxide is found in a range from (380 to 470) GPa as the coverage of oxygen groups through the sheet varies. According to amorphous graphene oxide the Young's modulus range is smaller in a range between 290–430 GPa. As oxygen coverage raises, both the intrinsic strength and young's modulus decrease monotonically because of the breakage of the sp^2 carbon network and decreasing the energetic stability for the amorphous and ordered graphene oxides. Adding to the above, the band gap of the GO becomes narrower under uniaxial tensile strain, giving an effective way to tune the electronic features of graphene oxide-based substances.[27]

1.3.2 Electronic Properties of Graphene Oxide

Graphene oxide has a good electrical properties, the energy gap of the graphene oxide is around 0.1 - 4.0eV. Researchers agree that oxidation of graphene will enhance the electronic properties of graphene. [28]

According to electrons that are found in graphene, it seems to be almost insensitive to electron-electron interactions and disorders. It has a very long mean free paths. The scientists concluded that its features is not as the properties of semiconductors or metals. Graphene is special in its structure, because it is very robust with a special phonon modes that we do not found in ordinary 3D solids.[29]

The researchers focus on graphene in their researches and ignore the other structures because of its special optical and electrical properties. The electrons are different in graphene because they have long mean free paths without facing the electron-electron disorder and interactions . Because of that , the graphene's properties is different from those of other common semiconductors and metals associated with the electronic and physical structures.[30]

1.3.3Thermal Properties of GO

A search published in 2019, the thermal properties of graphene mentioned that the specific heat of graphene has not been measured directly, the calculations for the specific heat was made by referencing the information and data for graphite. The specific heat of a material depends by two components, electrons and phonons (or lattice vibrations). The Micro-Raman spectroscopy is a method to introduce electrons and to measure the thermal conductivity of graphene [30].

Experimental studies were made in 2022 concluded that graphene (with its electron mobility with the range from 1,000 to 7,000 cm^2 Vs^{-1})has an amount of peak value for thermoelectric power (TEP) at about 80 μV K^{-1} at room temperature. The TEP sig, which shows the major charge carriers, changed from the positive to the negative as the gate bias crossed the charge neutrality point. The same results were obtained with a TEP of ~50–100 μV K^{-1} were obtained in different studies [31].

Table 1.1: Thermal conductivity enhancement in nanocarbon composites (Rasheed, Shihab et al. 2021.)

Filler	Enhancement %	Volume Fraction	Base Material
MWCNT	160%	1.0%	Oil
SWCNT	125%	1.0%	Epoxy
SWCNT	200%	5.0%	Epoxy
Graphite nanoparticles	3000%	25%	Epoxy
GO nanoparticles	30%-80%	5.0%	Glycol
GO	400%	5.0%	Epoxy resin
Graphene	500%	5.0%	Silver epoxy
Graphene	1000%	5.0%	Epoxy

Bauld et. al,2018 mentioned that Graphene, with a structure of a (2D) crystal of sp2 hybridized carbon atoms, has an excellent thermal properties. According to the thermal conductivity of individual graphene flakes have excess of 5000 W m1 K1 .[32]. It is mentioned that for thermal management applications graphene materials have been usually defined only because to their bulk properties [32].

1.3.4 Optical Properties

It is reported that the scientists used spectroscopic ellipsometry to estmate the optical constants of reduced graphene oxide. The changes in thickness and optical characteristics of graphene oxide sheet in reduction through long-term exposure, broad band white light is monitored. They determined the averaged optical constants of the reduced graphene oxide layer and the optical constants of the graphene oxide from a multiple-time-step analysis and a multiple-location analysis.[33].

In a research published in 2021,it was concluded that graphene oxide was analyzed based on the absorbance spectrum, X-ray diffraction, and the optical energy gap value. For some samples that the scientists prepared, the absorption coefficient value of test samples raises in the UV region but reduces in the visible region. According to the optical energy gap amounts of the test samples were very near each other, the value is about (2.35-2.50) eV[31].

Some studies following the optical properties of GO in suspension form were investigated by photothermal reduction in infrared irradiation. The blue emission which has a wavelength around 500 nm was because of to the carbonyl (C=O) groups. According to the yellow-red emission which has a wavelength about 610 nm emitted due to epoxy/hydroxyl (C-O/-OH) functional groups. As the IR power density increases and the reduction time , the reduction in the intensity of the yellow-red emission was observed but blue emission is prominent [34].

1.4GO/Polymer Nanocomposite

Sadrolhosseini et. al,2018 were mentioned that graphene oxide and other graphite derivatives were used as fillers in nanocomposite to be used in different applications. Polymers that may used in the nanocomposites are polyvinyl alcohol (PVA), polypropylene (PP), polystyrene (PS), PMMA, epoxy, silicon, polyester, foam, polyurethane, polycarbonate, poly(vinyldiene fluoride) [20].

Several methods have been utilized to prepare GO nanocomposite. the GO/epoxy composite was prepared by using the casting method at room temperature[2]

In 2019, the nanocomposite from Platinum nanoparticles and graphene oxide were prepared in aqueous solution using a technique called laser ablation .An investigation for the effect of optical nonlinear, optical linear, nonlinear and thermal properties of the above nanocomposite solution was made [30].

In order to use the graphene nanomaterials in heat dissipation, scientists have worked hard on assembling graphene into a host matrix, and introducing composite layers. The previous materials could be ideally good to be used as thermal interface materials (TIMs) [32].

1.5 The importance of packaging processes

Packaging is mentioned as one of the biggest industry worldwide, worth about $280 billion. Purchaser healthcare for packaging express 4% ($11.2 billion) from the total of packaging industry. As drug manufacturers proceed towards the 21st century, they challenged many problems that packaging could help them meet. Earlier, packaging was not an important issue for many pharmaceutical industries, showed as the final step in pharmaceutical manufacturing. Recently, the first stage of drug development process the manufacturer have to consider the type and shape of packaging materials. The packaging of the drugs rapidly become a very important part in the pharmaceutical delivery systems as a basic element of the sales issue, through which producers can differentiate their products from others. According to that the need of pharmaceutical packaging is raising, and would continue to be greater as companies depends more on labeling and packaging as media to promote and protect their products, increasing patient compliance, also meeting new regulations [35].

1.6 Purposes of packaging:

1.Transport and storage purpose:

The package shape and properties should be a thing that can be easily handed. They have to be prepared so that they can be protected efficiently [5].

2. Product identification:

Packaging is a very important tool that helps in product identification.

3.Convenience and marketing:

Packages should have properties that add convenience in different ways, for instance, handling distribution, display, stacking, sale, re-closing, opening, use, dispensing ease of disposal, reuse, recycling. It also can be used by marketing to encourage the buyers to purchase the drug specially food supplements [37]

4. Physical and biological protection:

Packaging materials are important to protect the product from biological or any physical hazard.It protects from light ,oxygen,moisture,and any mechanical damage.It also should provide a suitable environment for the product and protects it against any undesired microbial growth. [5].

1.7 Category of packaging container:

Primary packaging	Materials that are in direct contact with the drug	

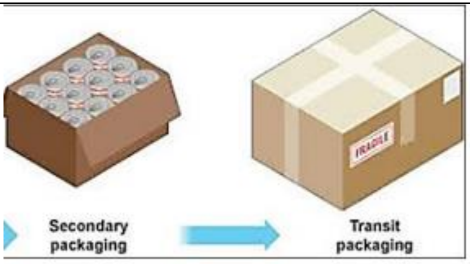
Secondary Packaging:	Packaging that locates outside the primary packaging and it is used to group primary packages together such grouping blisters in a cartoon	
Tertiary Packaging	It is used for bulk handling, transport shipping and warehouse storage.	

Table 1.2:Category of packaging container[38]

1.8 Importance of nanotechnology in pharmacy:

Nowadays, the technology of the future is Nanotechnology. According to the molecular level it is responsible for manufacturing of functional groups. These systems are specialized by a different electronic, physical and optical, structures which are captivating for disciplines from the substances to pharmaceutical sciences. The main point of nanotechnology that scientists can manipulate and appreciate different materials at the scale of molecules and atoms . The compatibility of biological molecules with nanoparticles is very attractive and has obtained special attention in the preparation of new materials for use in medicine , leads for gene therapy, imaging, drug delivery, novel drug preparations in addition to applications in the prevention, diagnosis and treatment of diseases. [39]

The revolution of Nanomaterials systems design has started because of nanotechnology application in the biomaterials which affects the drug safety and efficacy. In the last years, the growth of nanotechnology and nanoscience have increased rapidly following its applications. There is an increasing agreement that nanotechnology, specifically as a drug delivery system, would bring important advantages in the treatment and diagnosis of diseases. The advantage of nanoparticles that they have very large (functional) surfaces that makes them to entrap or attach other compounds and molecules, such as peptides and drugs therapeutic purposes [40].

1.9 Carbon-based nanomaterials:

Graphene is sheet that has two-dimension nanomaterial composed of a layer of sp^2 hybridized of carbon atoms, it has obtained very important position in the medical field because of its special mechanical, thermal and electrical properties [41]. After the discovery of graphene, researchers have concentrated on discovering different properties, different types of nanomaterials and different applications in electronic devices , photoconductive materials and optoelectronic devices solar cells, drug delivery, tissue engineering and medical imaging [42]. Graphene has a property of the capacity of enzyme loading, so it is a good nanomaterial in the electrochemical sensors.Recently, all of (GO), graphene, and RGO have also achieved great attention in the field of biomedicine [43]. Nano-GO displayed as the best in drug adsorption when compared with other different carbon-based substances used for drug adsorption due to stacking [44]. According to the intrinsic near-infrared (NIR) optical absorption, researchers have explored graphene-based photothermal therapy, followed by obtaining special antitumor therapeutic uses. Furthermore, different materials, including proteins, conducting polymers, quantum dots, cyclodextrin and noble metals bounded onto the top of graphene to explore enhancement to the magnetic and optical characters for cancer therapy and multimodal imaging [45]. The in vivo and in vitro toxicity of graphene was interested several groups [46]. It was concluded that excretion affects the toxicity of the graphene particles, adding to the above parameter the chemistry condition and particle size, all of them play a very special role in the biodistribution. Without functionalization, grapheme oxide or graphene was very toxic but after the adding of biocompatible coating on the surface, GO did not have any cytotoxicity at doses that was given in the trials [47]. The application of graphene oxide in biomedical field is concentrated on drug delivery and gene because graphene can go through the plasma membrane and enhance the cellular uptake of active ingredients [48]. In addition, the suitable biocompatible and hydrophilic polymers and excipients are still being explored to enhance the properties of GO [49].

1.10Background of study:

Recently, carbon nanostructures are attractive for different researchers in worldwide and different modified structures of carbon reported in various scientific reports.

Nano-sheet of GO has an atomic thick layer which has a poly-aromatic sp^2 hybridization carbon structure that is characterized with very high surface area.

The property of having very high surface area of graphene components make them ideal forms for drug delivery and drug loading purposes. Much attraction has been concerned on searching about polymer films to improve their mechanical features and room temperature conductivity by mixing different polymers.

Now a day's, the super-absorbents, which Poly acryl amide co acrylic acid (Poly(AAm-co-AA)) is one of them, are moderately crosslinked polar polymer networks which can take large amount of water or other polar fluids and they having a great interest in recent researches in different applications.

According to Graphene oxide -based nanocomposites, They have been very important focused on and demonstrated to have special performances in many ways. Recently, there are only a small number of investigations devoted to GO-based nanocomposite hydrogels.

It has been little investigations about GO-based super-absorbent hydrogels (Like Poly(AAm-co-AA)-GO nanocomposite) are available until now. In fact, GO is a good idea for fabricating novel superabsorbent hydrogels, the reason behind that is having large numbers of hydrophilic groups on its surface.

1.12Problem statement:

The large gap in the pharmaceutical research fields and a need of a film which has a good mechanical, thermal and swelling properties for pharmaceutical manufacturing led to think about preparing a nanocomposite of Poly(AAm-co-AA) with GO.

GO which has two-dimensional sheet of sp2-bonded atoms of carbon, it has very attractive and the attention increased because of its good thermal, electrical and conductivities and high specific surface area and principle low manufacturing cost**.**

Polyacrylamides are one of the most widely used water soluble polymers. As hydrogels are suitable materials not only for imitating the precise structure and function of living systems but for controlling the drug releasing pattern inside the living body**.** As Poly(AAm-co-AA) itself did not have enough mechanical strength in the swollen state, the combination between GO and Poly(AAm-co-AA) will enhance the properties of the polymer and expected to obtain better features.

1.12 Significance of study:

Many researchers in pharmaceutical industry keep on studying, developing, and investigating on new drug delivery systems and new films to cover the pharmaceutical products, as well as consumer market drug in order to fill in the gap of present drugs limitation. They aim to improve the efficacy, safety and stability of drug which directly move along with enhance patient compliance and improve pharmaceutical industries. As for present research study on Poly(AAm-co-AA)-GO nanocomposite, it is to improve the stability for drugs which was detected as limitations. These limitations are an opening door towards better formulation in pharmaceutical applications.

This research was conducting to study the Poly(AAm-co-AA)-*GO nanocomposite* to enhance the mechanical ,physical ,structural properties of the film.

1.13 Objectives:

The present study will focus on the objectives as follow:

1. To prepare Poly(AAm-co-AA) films with different concentrations.

2. Preparation of GO

3. To prepare Poly(AAm-co-AA)-GO nanocomposite with different concentrations.

4. To Measure the physical and structural characteristics of the nanocompopsites and films.

1.14 Research hypothesis:

The Poly(AAm-co-AA)-GO nanocomposite can be prepared and measured for its physical and structural properties and have characteristics more interesting compering with the neat Poly(AAm-co-AA) films.

Chapter Two: Literature Review

2.1Introduction:

Recently, carbon nanostructures are attractive for different researchers worldwide and different modified structures of carbon reported in various scientific reports. Nano-sheet of graphene oxide has a single atom thick layer which has a poly-aromatic sp2 hybridization carbon structure that is characterized with very high surface area.

The property of having very high surface area of graphene components make them ideal forms for drug loading and drug delivery purposes.

Much attraction has been concerned on searching about polymer films to improve their mechanical features and room temperature conductivity by mixing different polymers.

Poly(acrylamide-co-acrylic acid) (Poly(AAm-co-AA))has a very wide range of applications for example a stabilizer and a flocculent in different ecological and technological processes. Poly(AAm-co-AA)is polar and hydrophilic polymer that is used in many biological and medical applications in form of active hydrogels.

Now a day's, the super-absorbents, which Poly(AAm-co-AA)is one of them, are moderately cross linked polar polymer networks which can take large amount of water or other polar fluids and they having a great interest in recent researches in different applications.

According to Graphene oxide -based nanocomposite, They have been very important focused on and demonstrated to have special performances in many ways .Recently, there are only a small number of investigations devoted to GO-based nanocomposite hydrogels.

It has been little investigations about GO-based super-absorbent hydrogels(Like Poly(AAm-co-AA)-GO nanocomposite) until now. In fact, GO is a good idea for fabricating novel superabsorbent hydrogels the reason behind that is having large numbers of hydrophilic groups on the surface.

2.1 Preparation of polymer/GO Nanocomposite:

In a study of Jing and his coworkers (2020), they have synthesized poly(acrylamide-*co*-acrylic acid)–Al^{3+}/poly(vinyl alcohol)/graphene oxide double network nanocomposite hydrogels by a freeze–thaw cycle and free-radical polymerization method in the presence of aluminum ions and graphene oxide. The pH-sensitivity, mechanical properties and surface morphology and can be tuned by adjusting the contents of Al ,GO and PVA [50].

The following study prepared electro conductive hydrogels that contains Acrylamide which is copolymerized with acrylic acid , carboxymethyl cellulose and GO by a free radical polymerization method. They explored the RGO effect as a nanomaterial on enhancing the strength abilities of the synthesized hydrogel [51].

Dia,et al.(2019), a research studied the hydrogels which were synthesized by grafting copolymerization of acrylamide and acrylic acid onto pineapple peel carboxymethyl cellulose with addition of graphene oxide (GO). The swelling , structure, and multiple responses to pH, salt, and organic solvents were studied. They concluded that addition of GO resulted in a more cross-linking density of the network and thus decreased the swelling ability. They also prepared nanocomposites from starch that has superabsorbent capability by a strategy of facile one-pot. It was synthesized by the simultaneous formation of magnetic iron oxide nanoparticles and in situ radical solution polymerization of superabsorbents based on poly(acrylic acid-*co*-acryl amide) grafted onto starch backbones in the presence of graphene oxide (GO) nano sheets. The morphology, chemical structure, and phase composition were characterized by using scanning electron microscopy (SEM), Fourier transform infrared FTIR, transmission electron microscopy (TEM), UV-Vis spectroscopy, X-Ray diffraction (XRD), vibrating sample magnetometer (VSM) , thermal gravimetric analysis (TGA), and). In this study, they concluded that swelling behaviors showed that the mixture of GO nanoplates in the polymeric network leads to a remarkable improvement in the swelling capacity of the SANCs.[52]

Huang, et al.(2012), researchers prepared GO/Poly acryl amide co acrylic acid super-absorbent hydrogels by a method called in situ radical solution polymerization successfully. The result that they got a hydrogen bonds and possible different covalent bonds between GO and poly acryl amide co acrylic acid chains. [53]

2.2Mechanical properties of Polymer in composite:

A composite mixture of a poly(acrylic acid-co-acrylamide)/bentonite/kaolin composite was prepared and the mechanical features of this composite was studied. They used Acrylic acid and acrylamide as a function of water absorbent monomers. The repeated water absorption, water preserving capability, mechanical properties, salt resistance of the composite hydrogel were analyzed and discussed.[9]

2.3 Mechanical Properties of Polymer/GO Nanocomposite

Mónica Cobos, et al. (2018) prepared RGO and studied results for loading it on PVA on the mechanical properties. According to the elastic modulus it has no change after adding GO , they clarified this result to an experimental error. By adding GO it was found that the elongation reduced and the tensile strength raised from 47 to 56 MPa.[54]

Yibo Zhao et al. (2018) were interested in PVA/GO nanocomposite hydrogel as an artificial cartilage material. The Young's modulus for poly vinyl alcohol/GO has a higher value than PVA, it was improved with increasing GO concentration. [55]

Other research was investigated the GO/ methyl cellulose mechanical characters. That mixture was synthesized by simple assisted solvent evaporation assembly. 1,3,5%wt. of graphene oxide was used with methyl cellulose. The results shows increasing in the young's modulus and stress with addition of graphene oxide .The young's modulus reached to (104± 3) MPa and the stress (3.8± 0.2) GPa , but neat methylcellulose only has a value of 56±4 MPa for young's modulus and 1.27± 0.2 GPa for stress at break. [56]

In 2017,Meryem Goumri et al. studied mechanical characteristics of poly vinyl alcohol/GO and poly vinyl alcohol/RGO. By adding graphene oxide, the yield stress and Young's modulus and have increased, When you compare neat polymer with nanocomposite the yield strain for graphene oxide nanocomposites was enhanced when compared to pure polymer.However, using 2%wt. of reduced graphene oxide ,the Young's modulus and yield decreased.[57]

Y. Zhang et al made a search about the mechanical characters in 2018.They investigated graphene oxide/carbon nanotube carried on carboxymethyl cellulose by simple mixing and evaporation technique for solution. Adding the GO in to the carboxymethyl cellulose leads to raising in the tensile strength value from 37.28 MPa to 39.72 MPa. According to Young's modulus,it was also enhanced ,however the yield strain was reduced .The addition of carbon nanotube to form the nanocomposite sheets was resulted in the same results as in GO.[58]

2.4 Thermal Properties of polymer/GO Nanocomposite:

Graphene has special thermal characters, the thermal conductivity of graphene is about 5000 Wm-1 k -1 (Cui et al., 2016). The thermal characters is measured by using different instruments, Pyrisl Thermoanalyzer DSC, STA and TGA.

The thermal characters for MC/GO nanocomposite were examined by Rama Kanta Layek, et al.in 2018.They used STA in their research .The thermograph results show a small increase in the thermal properties for graphene oxide /methylcellulose nanocomposite compared with neat polymer.[56]

Mónica Cobos , et al.(2018) were prepared RGO .After that they e loaded the graphene oxide on poly vinyl alcohol polymer and examined the thermal. The Tg of the nanocomposite increases as amount of GO increases.However the crystallization and melting temperature reduced with raising the amount of GO.

In 2018,Kumar enhanced the thermal characters of Poly ethylene terephthalate by addition of RGO,his results showed that the polymer has a higher degra[54] dation rate than the nanocomposite. That means adding RGO enhances the thermal properties of the neat polymer.[59]

At the same pattern, a search in 2018 was interested in the scope of preparing a combination of cellulose nanocrystals and graphene in different concentrations.Then,they studied its effect after adding the mixture on poly (lactic acid) layers through melt blending method. The enhancement in thermal properties were noticed by increasing the concentration of GO. [60]

According to Meryem Goumri with her coworkers research in 2017.They interested in two Nanofillers; graphene oxide and reduced graphene oxide fillers on poly vinyl alcohol. They used DSC and TGA.They noticed enhancement in thermal stability. The temperature of degradation was increased compared with the neat polymer. They used DSC for measuring Tg and the result was increasing the Tg by addition of GO more than RGO to the pure polymer. Tm showed no effect by adding the graphene oxide. When adding reduced graphene oxide or graphene oxide the crystallization degree is reduced.[57]

Chapter Three: Materials and Methods

3.1 Materials and equipment:

Materials that were used in the research are: Polyacryl amide co acrylic acid Poly(AAm-co-AA) powder, 80+%, M.wt = 150000-520000g/mol and D=0.75g/ml which was purchased from Sigma Aldrich. graphite flakes from Hebron university, reagent $KMnO_4$, $NaCl$,H_2SO_4 concentrated . The below equipment were used at Nano laboratory: Analytical Balance, Bath Sonicator (type T 490DH), model 37600 Mixer ,Spin coater that has timer reaches up to 30sec and a speed reaches up to 1000rpm, ,Digital Caliper (STAINLESS HARDENED) 0-150mm. DSC that was used (DSC Perkin Elmer 4000) which is heated up to 4500 C and has a heating rate equals to 10 C/min and used under the nitrogen atmosphere,Mechanical Test Machine.(FTIR) spectrometer of Bruker IFS 66/S, that surrounded with a liquid nitrogen and has cooled MCT detector and has a KBr beam splitter, the last equipment belongs to physics lab at Al-Quds university. The tip Sonicator, MODEL VCX750 37 with 750 watts was used at Hebron University to produce nanographene oxide particles .Figure (3.1) show the photograph of equipments used**.**

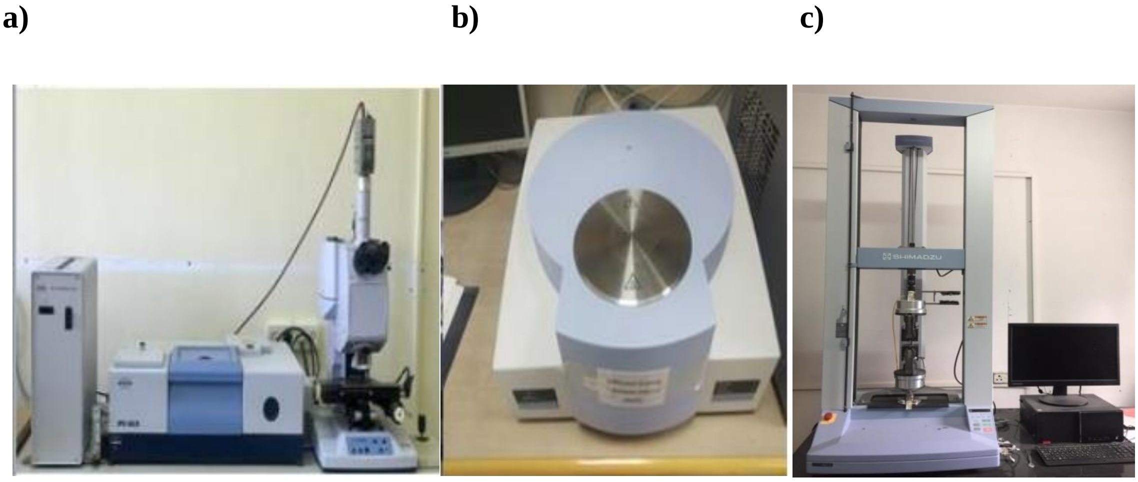


Figure 3.1: photograph of equipment. a) FT-IR, b) DSC, c) Mechanical Tester

3.2 Methods

This approach includes methods of preparation: thin films, Poly(AAm-co-AA) film /GO-Poly(AAm-co-AA) nanocomposite,and their analytical methods.

3.2.1 Preparation of poly(AAm-co-AA) thin films:

A 0.9, 1.25 2.5, 5, wt. % of Poly(AAm-co-AA) (See figure 3.2) Samples of Poly(AAm-co-AA) with different concentrations in solution.The solutions were prepared by weighing appropriate amount of polymer and adding purified water under continual stirring for about 30 min at RT. Afterwards, the polymer solutions were poured on the Petri dishes after that spin coated in different rates (200- 550 rpm) by using different times (5, 15, 30, 60s), with a 4ml volume. For low concentration of poly(AAm-co-AA) 0.9%, spin coater shows hard process of preparation due to loss of material during spinning at any speed and any time interval. other samples were not distributed uniformly due to high viscosity. The opaque physical appearance was produced from 5%wt. polymer. Finally, samples allowed to dry in an incubator at RT for about 3 days to be used for characterization see figure 3.3.

Figure 3.2:Samples of Poly(AAm-co-AA) with different concentrations in solution.

Figure 3.3.Samples of Poly(AAm-co-AA)films.

3.2.2Synthesis of GO solution:

Figure3.4 shows the GO was synthesized through an oxidation process as that begun by addition of graphite flakes into NaCl . Grinding for both components were done. A concentrated H_2SO_4 was then added to the mixture above for intercalation and stirring for 24 hrs. After that time, $KMnO_4$ was added carefully to the resultant mixture at a slow rate and stirring for 3 hrs. The mixture was further diluted by addition of 200 ml of distilled water with continuous stirring.Ultrasonication of the mixture was made for 2 hours. for particle size reduction. A sample of GO solution with a lateral size of 200nm was obtained.

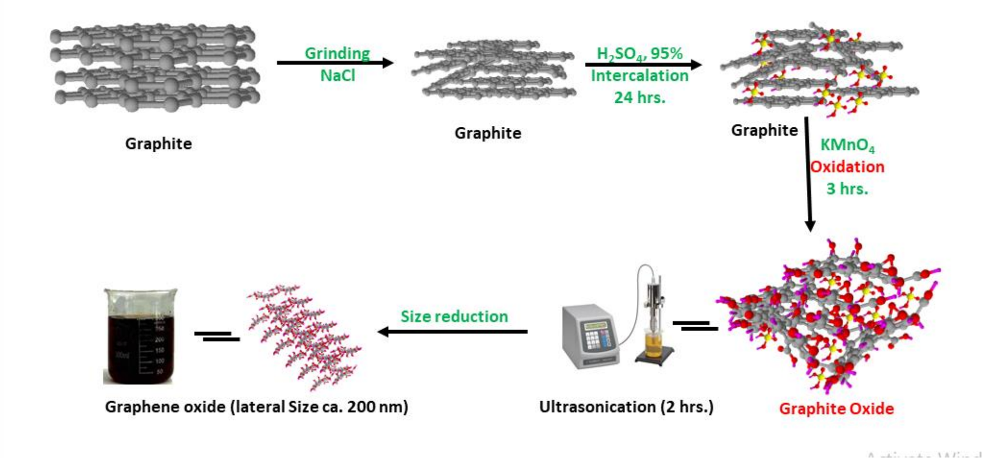


Figure 3.4:Schematic illustration of GO preparation.

3.2.3 Preparation of Poly(AAm-co-AA)/GO nanocomposite films:

See figure 3.5 for polymer/GO synthesis.Different concentrations of GO were synthesized by dilution from the concentrated stock solution of GO (5g/l). After the dilution,The solution was separated into 5 parts to add poly(AAm-co-AA) and prepare polymer/GO nanocomposite solutions that has a concentration of 0.9, 1.25 2.5, 5, wt. % of polymer. The above four concentrations of polymer/GO nanocomposite were put for sonication in bath sonicater at room temperature for 30 minutes.

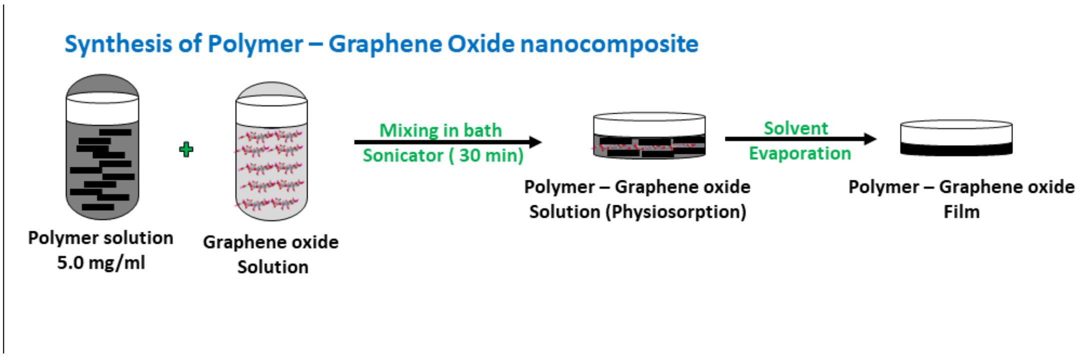


Figure 3.5: Synthesis of polymer-graphene oxide nanocomposite

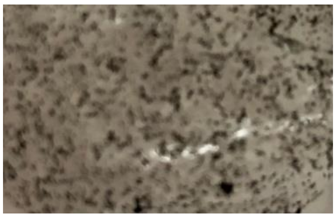


Figure 3.6:Poly(AAm-co-AA)/GO nanocomposite film

As shown in figure 3.6,the Solutions were adopted to casting method with 4 ml volume on petri dishes (plastic ones) and set to dry at room temperature and humidity for 3 days to get the samples ready for characterization.

Matrix of Poly(AAm-co-AA)/GO nanocomposite is shown in the (Table 3.1) and the flow chart for Polymer /graphene oxide nanocomposite films synthesis show in the flow chart (Figure 3.7)

No.	**Code**	**Conc. of GO %**		**Conc. of Polymer %**			
		0.05	**0.1**	**5**	**2.5**	**1.25**	**0.9**
1	$\mathbf{A_{5}G_{0.05}}$	×		×			
2	$\mathbf{A_{2.5}G_{0.05}}$	×			×		
3	$\mathbf{A_{1.25}G_{0.05}}$	×				×	
4	$\mathbf{A_{0.9}G_{0.05}}$	×					×
5	$\mathbf{A_{0.6}G_{0.05}}$	×					
6	$\mathbf{A_{5}G_{0.1}}$		×	×			
7	$\mathbf{A_{2.5}G_{0.1}}$		×		×		
8	$\mathbf{A_{1.25}G_{0.1}}$		×			×	
9	$\mathbf{A_{0.9}G_{0.1}}$		×				×
10	$\mathbf{A_{0.6}G_{0.1}}$		×				

Table 3.1 :Matrix of Poly(AAm-co-AA)/GO nanocomposite preparation.

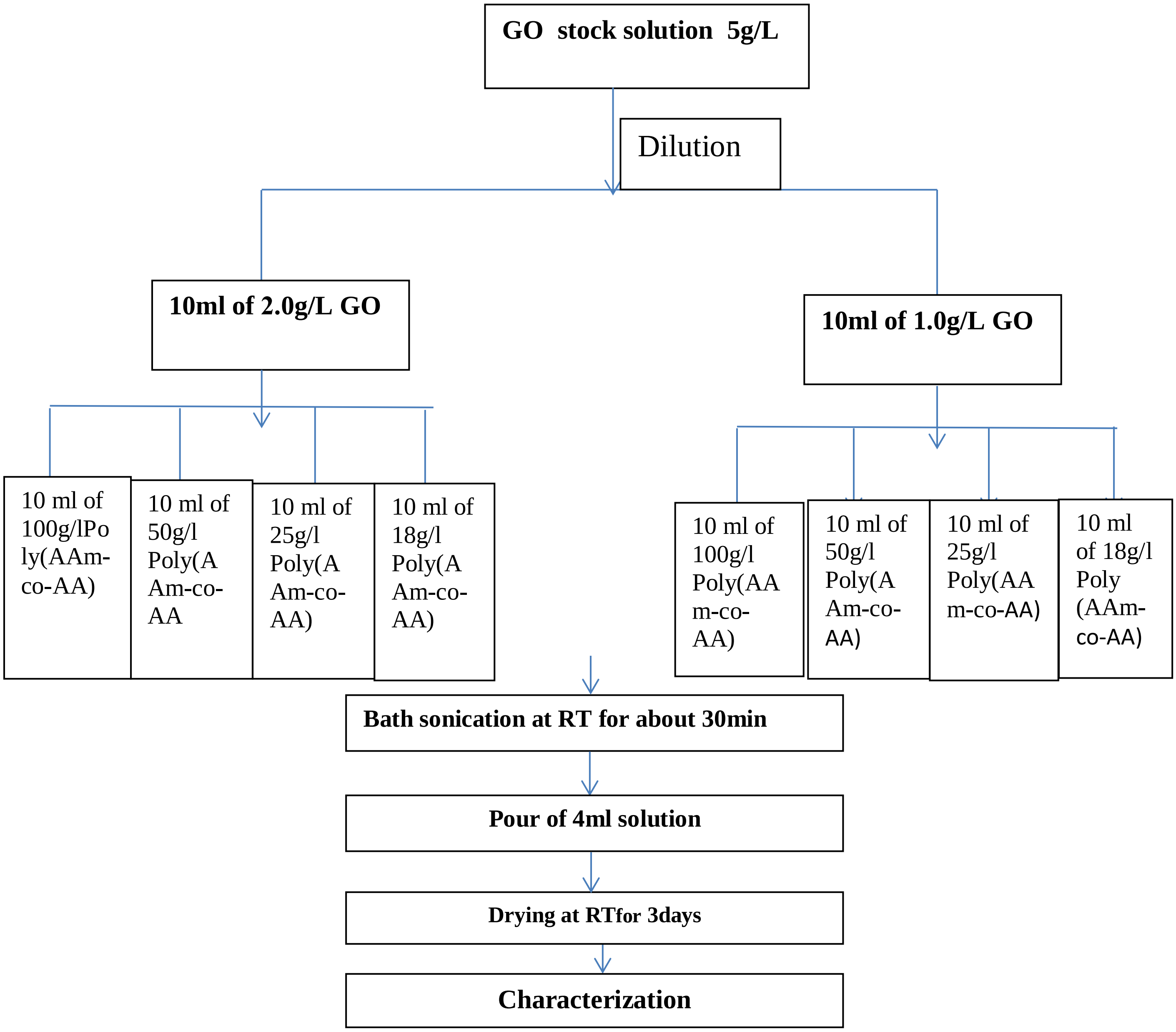


Figure 3.7: Flow chart for Poly(AAm-co-AA)/GO nanocomposite films preparation

3.2.4 Analysis and characterization of Poly(AAm-co-AA)/GO films.

a) Measurement of thickness:

The thickness measurment of Poly(AAm-co-AA) films were measured using both calculation and digital caliber .Calculations depend on taking the weight of the test sample that was cut into square shape, after that calculate the volume following equation: Volume equals mass divided by density. The volume that was calculated is used for calculation of thickness following this equation: Volume of cube= length* width* height which is the thickness). The thickness of Poly acryl amide co acrylic acid/GO films was estimated by using digital caliber, for accuracy reasons the thickness of the test sample was measured at 4 different places and average result is used for result presentation.

b) Thermal properties:

DSC was prepared for pure 5,2.5,1.25,0.9% Poly(AAm-co-AA) and for samples with different concentration of GO with different concentration of the polymer. About 3.0 mg of the test sample was cut by a scissors then take the weight on the digital balance, after that the test samples were sealed in the inner part of DSC pans, the heat capacity changes are tracked by changes in heat flow.

The process above gives thermal information about Tg, crystallization/melting manners, solid-solid transitions, degree of the crystallinity , specific heat, decomposition behavior purity determination, oxidative stability cross-linking reactions.

According to heating program, it was put in the range of 25 ˚C to 200˚C after that cooled to 25 ˚C at cooling rate of 10˚C to get crystallization and melting temperature. The tested sample was heated again to 400 ˚C at the same cooling rate to get the Tg and the degradation temperature.

c) Mechanical properties

The elastic modulus and tensile properties for films were prepared using Mechanical Test Machine from Shimadzu. Mechanical features of the material provides data about mechanical resistance and if it meets with the requirements of our application. Rectangular strips of the test films were obtained with dimensions of (2×4cm) and the thickness kept at (0.01_0.05) mm.Main idea of this test is putting test sample between the two clamps, that pull the sample until it breaks. Elongation and force are measured then plot of the stress against strain can be obtained. Mechanical test was prepared only for high concentration of polymer.

d) Fourier-Transferred Infrared Spectrometer (FT-IR)

The FTIR is a technique known as a fingerprint for any sample.It depends on infrared radiation which goes through test sample, according to the fate of radiation some of it will be absorbed and other will be transmitted. The obtained spectrum shows the molecular transmission and absorption . Fourier-Transferred Infrared Spectrometer can provide qualitative analytical data about each type of sample and guess the unknown substance also determine the number of components inside the mixture. In this research the absorption spectra in the region of (4000-400 cm-1) were taken .

e) Measurement of swelling :

The swelling test were calculated by putting the weighted samples in phosphate buffer (KH_2PO_4, pH=7.2) for 20 minutes , After that, the film was removed, and weighted to calculate the amount of solution adsorbed by sample.:

Swelling ratio= ((m2-m1)/m1)*100%

m1: Initial weight of film.

m2: After immersing in phosphate buffer for 20 min .

Chapter Four: Results and Discussion

4.1 Superior properties of graphene oxide

4.1.1 Thermal properties of Poly(AAm-co-AA) with graphene oxide:

Figure 4.1,4.2 and 4.3 shows the thermograms of neat Poly(AAm-co-AA) , Poly(AAm-co-AA):GO (5.0mg/ml:0.5mg/ml). The thermal properties of Poly(AAm-co-AA) /graphene oxide were estimated by using DSC. The Tg of neat Poly(AAm-co-AA) is about 50˚C. After adding graphene oxide to the Poly(AAm-co-AA), it was increased to reach about 75˚C at 0.5% concentration of GO. This attitude was explained due to the strong physical interaction between graphene oxide and the matrixes of the polymer. That means the thermal properties were improved for the prepared nanocomposite compared to the neat polymer. The Tm for the poly acrylamide co acrylic acid is about 290˚C. By adding graphene oxide to the polymer in concentrations 5% and 2.5% , the Tm is 286 ˚C and 280˚C respectively. The small difference in the melting temperature through the 2 tested concentrations of polymer is because of the volume or spaces between the polymer chains are more in the lower polymer concentration.

According to the percent of crystallinity, the depth of the Tm peak was increased in all nanocomposites thermograms compared with the polymer see table 4.1.This means that part of the amorphous regime is transformed into a crystalline part which means that the presence of graphen oxide enhances the closed packing. The above results go with results that was obtained in a search about nanocomposite with epoxy and GO[61]. The Tg of GO epoxy nanocomposite raises with increasing GO concentration An increasing in Tg of GO-epoxy nanocomposite was explained from a high specific surface area and wrinkled GO, which can bind with epoxy chain and restricts the mobility of matrix. The homogeneous dispersion of graphne oxide increases the surface area and increase the contact.[61]

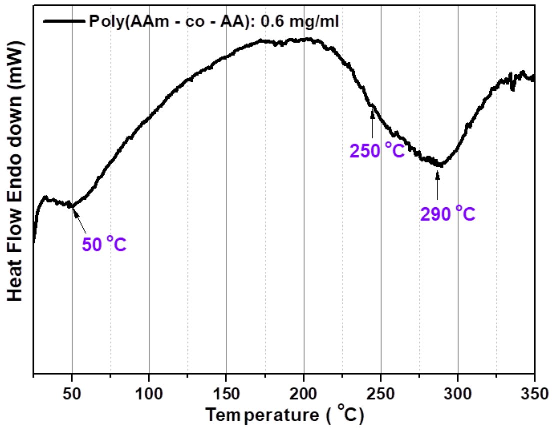


Figure 4.1:The DSC thermogram of neat Poly(AAm-co-AA).

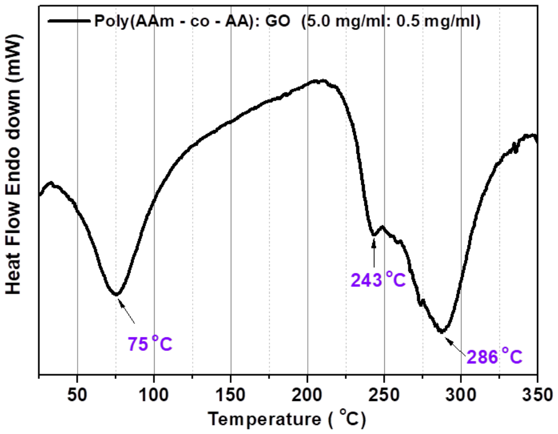


Figure 4.2: The DSC thermogram of of Poly(AAm-co-AA):GO (5.0mg/ml:0.5mg/ml).

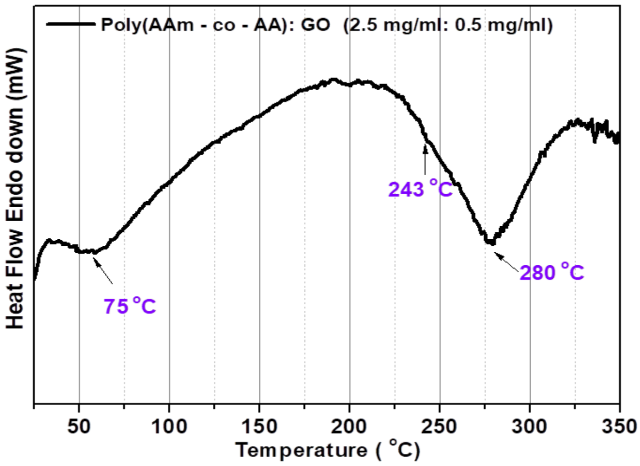


Figure 4.3:The DSC thermogram of Poly(AAm-co-AA):GO (2.5mg/ml:0.5mg/ml).

Sample	**Tg (˚C)**	**Tm (˚C)**	**Depth of Tm peak**
Poly(AAm-co-AA) neat film	**50**	**290**	**Not deep**
Poly(AAm-co-AA):graphene oxide (2.5:0.5)mg/ml	**75**	**280**	**More than the polymer alone**
Poly(AAm-co-AA):Graphene oxide (5:0.5)mg/ml	**75**	**286**	**More than the polymer alone**

Table 4.1:DSC results

4.1.2 Mechanical properties of poly(AAm-co-AA) with graphene oxide:

Figure 4.4.a shows the stress-strain curviture of poly acrylamide co acrylic acid. figure 4.4.b reveals the stress–strin curve for poly acrylamide co acrylic acid: GO(5.0:0.5)mg/ml,and figure 4.4c shows the stress – strain curve of poly acrylamide co acrylic acid:GO(5.0:1.0)mg/ml. The elongation of Poly(AAm-co-AA) film was around 97%.

The youngs modulus for poly acrylamide co acrylic acid is 125%, for poly acrylamide co acrylic acid:GO(5.0:0.5)mg/ml is 200%, and for poly acrylamide co acrylic acid :GO(5.0:1.0)mg/ml is 300%. It is well known that when modulus is higher, more stress is required to obtain the same amount of strain; an idealized rigid body (Close to poly(poly acrylamide co acrylic acid):GO nanocomposite attitude) will have an infinite Young's modulus.On the other hand, the soft material (for example fluid) will deform without force, and would have zero Young's modulus.Our results exhibit clear enhancement in mechanical features and it was increased from a good dispersion of GO on Poly(AAm-co-AA) matrix, and a strong hydrogen bonding between Poly(AAm-co-AA) and GO. Also increasing the concentration of GO in the polymer leads to raise the youngs modulus and so the mechanical featurs of the nanocomposite.According to plastic strain region,it was clear that the reagon disappered in the nanocomposite.This is due to the nanocomposite attitude that was similar to the fiber materials mechanical behavior and loss of polymer behavior.These results agrees with a search was made in 2010 about chitosan and GO.The researchers obtained an improvement in youngs modulus results for chitosan with GO 1%Wt.[62].The above results also goes with [63] for nanocomposite of GO 3% with poly (acrylic acid) grafted onto amylose. The tensile strength and elastic modulus of the nanocomposite hydrogel increased by 124% and 26%, respectively with 3 wt% GO loading.

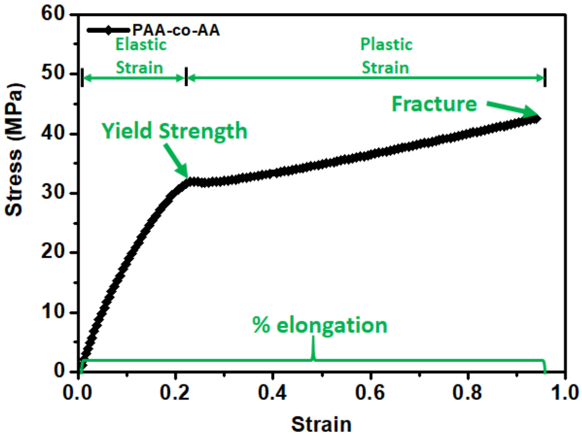


Figure 4.4.a:Stress strain curve of poly(AA-co-AA)

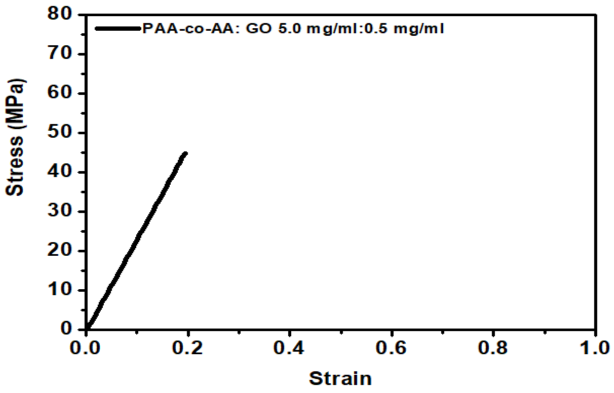


Figure 4.4.b:Stress strain curve of poly(AA-co-AA):GO sample

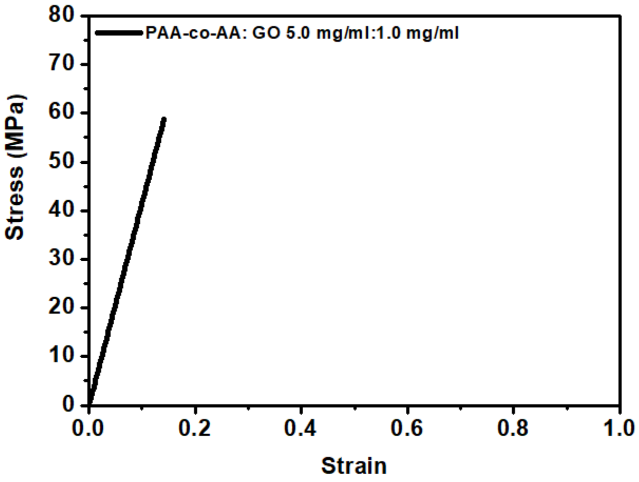


Figure 4.4.c:Stress strain curve of poly(AA-co-AA)GO sample

Figure 4.4.a shows the stress-strain curve of Poly(AAm-co-AA) alone.Figure 4.4.b shows the stress –strin curve for Poly(AAm-co-AA):GO(5.0:0.5)mg/ml.Figure4.4.c shows the stress –strain curve of Poly(AAm-co-AA):GO(5.0:1.0)mg/ml

4.1.3 Swelling ratio of Polymer (Poly(AAm-co-AA)) graphene oxide nanocomposite.

As shown in Table 4.2 the swelling ratio test results for poly acrylamide co acrylic acid in the prepared concentrations.Swelling ratio for neat polymer that has highest concentration shows the least swelling ration .Thats due to the presence of the pores in the polymer chain with lowest concentratio.Because of that more volume of solution goes through the polymer chains.According to the swelling ratio of nanocomposite with different concentrations of GO%. The higher the GO content in the nanocomposite the lower swelling ration obtained.These resultes could be explained because of the effect of GO particles. Polymer chains have a pores in its structure, which can carry large number of solution, but an addition of graphene oxide decreases the pores through the chaines of the polymer, and produce more rigidity which cannot expanded to hold more water molecules. In conclusion, the graphene oxide acts as a barrier for penetration of water molecules.In a search forErceg,et al.(2021),they explained increase in swelling ratio at pH around 7 compared with pH around 3 for poly (AAm-co-AA)because of Carboxylic groups of acrylic acid units are in ionized (anionic) form at high pH . Negatively charged groups which are carboxylic groups repel each other which makes expansion of polymer network and that leads to the greater water uptake**.[64]**

%Poly(AA-co-AA)	**%GO**	**%Swelling ratio**
5%	0%	0.21%
2.5%	0%	0.3%
1.25%	0%	0.91%
0.9%	0%	1.67%
5%	0.05%	0.19%
2.5%	0.05%	0.25%
1.25%	0.05%	0.8%
0.9%	0.05%	1.32%
5%	0.1%	0.1%
2.5%	0.1%	0.18%
1.25%	0.1%	0.68%
0.9%	0.1%	1.17%

Table 4.2:Swelling ratio results

4.1.4Thickness test of pure Poly (AAm-co-AA), Poly (AAm-co-AA)/GO nanocomposite.

The thickness for poly acrylamide co acrylic acid polymer films and poly acrylamide co acrylic acid /GO nanocomposite was measured by using caliber for both and by calculations and caliber for neat polymer:Each of the films was weight individualy in to get the mass.The below aquation was used for calculations.

$V=l\times w\times h$

v:volume

l:length

w:width

h:hight

$d=m/v$

d:density

m:mass

v:Volume

The thickness was from 0.39mm to 0.045mm depending to the polymer concentration , that means as the concentration increased the thickness will increase.See the results in table 4.3

Polymer %	GO%	Average thickness by caliber (mm)	Average thickness by calculations (mm)
5%	0%	0.325	0.295
2.5%	0%	0.133	0.095
1.25%	0%	0.060	0.055
0.9%	0%	0.045	0.04
5%	0.05%	0.332	--
2.5%	0.05%	0.171	--
1.25%	0.05%	0.08	--
0.9%	0.05%	0.051	--
5%	0.1%	0.390	--
2.5%	0.1%	0.210	--
1.25%	0.1%	0.116	--
0.9%	0.1%	0.089	--

Table 4.3:Thickness results for polymer alone and polymer/GO by calculation and caliber

4.1.5 Quality Assurance (IR of Graphene Oxide and Polymer)

4.1.5.1 FT-IR of Poly (Acryl amide co acrylic acid) with graphene oxide

The spectra of FT-IR were used inorder to estimate the interaction between Poly(AAm-co-AA) and graphne oxide. FT-IR spectrum for graphene oxide is shown in the Figure 4.5 The functional group for oxygen were seen as the following: C-O and C=O can be assigned at the bands of 1062 cm^{-1} and 1730 cm^{-1}, respectively. The (O-H) groupe can be noticed at 3364 cm^{-1}. C=C group was seen at 1614 cm^{-1}.

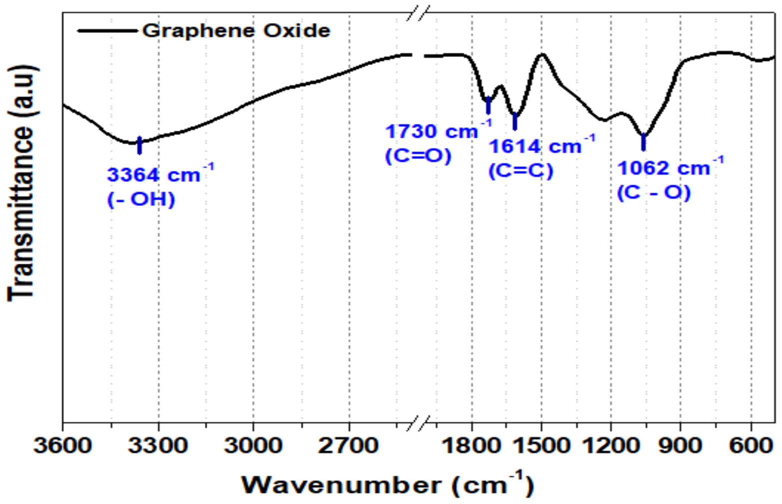


Figure 4.5: FTIR Spectrum of graphene oxide particles.

The FT-IR spectrum for poly acrylamide co acrylic acid is shown in Figure 4.6 for its spectrum, the functional groups for the polymer were seen as following:,CH_2, NH_2, C–N C=O, can be observed at the bands of 3190 cm^{-1} ,3337 cm-^{1}, 1428 cm^{-1} ,1636 cm^{-1}respectively.At 1118 cm^{-1} there is also a peak related to amide group .The peak at 1322cm^{-1} is the characteristics (C–O) stretching peak of –COOH.

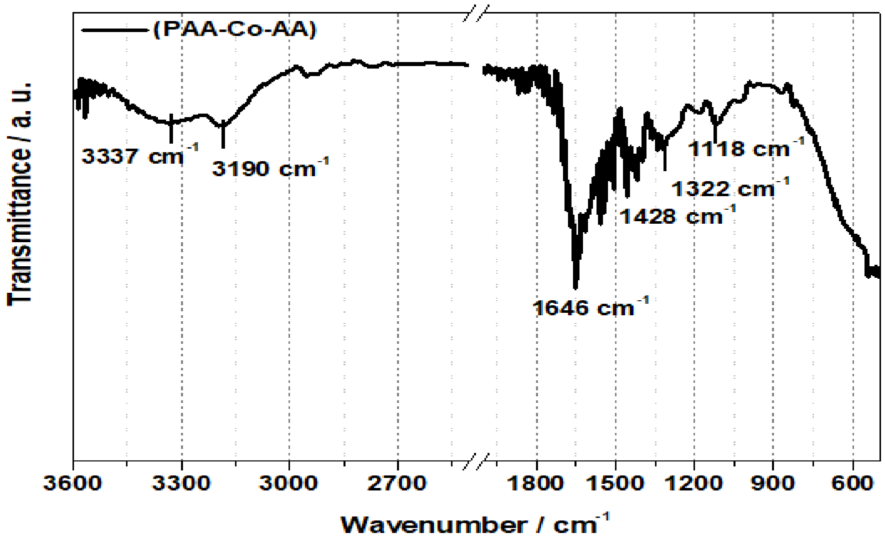


Figure 4.6: FTIR Spectrum of Poly(AAm-co-AA)

The shift of COOH CH2 ,CN,C=O groupes is a marker for the interaction occurred between Poly(AA-co-AA) and GO. At all concentration of the polymer the shift is obvious in lower or higher frequency as shown in (Figure 4.7 a, b and c.)

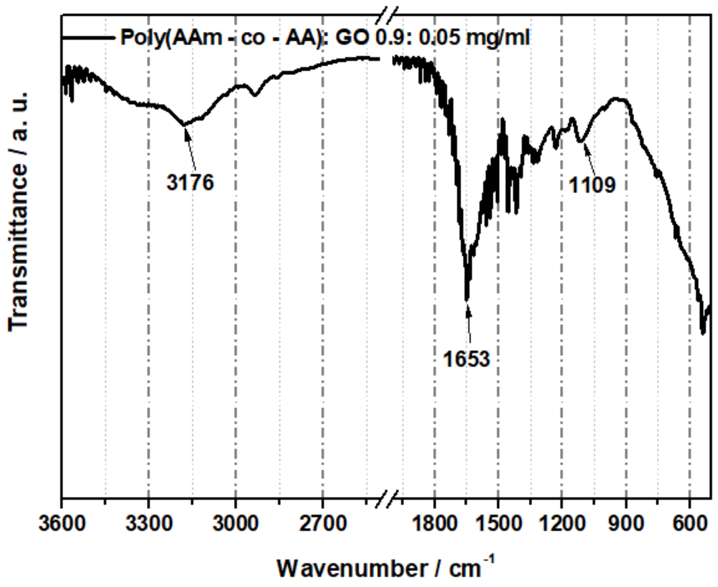


Figure4.7 a) FT-IR spectrum of Poly(AAm-o-AA):GO(0.9:0.5)mg/ml

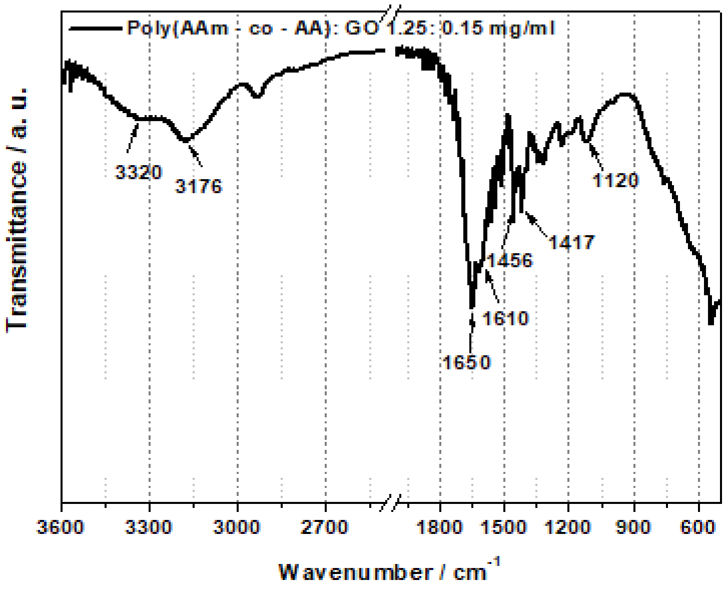


Figure4.7 b) FT-IR spectra of Poly(AAm-o-AA):GO(1.25:1.0)mg/ml

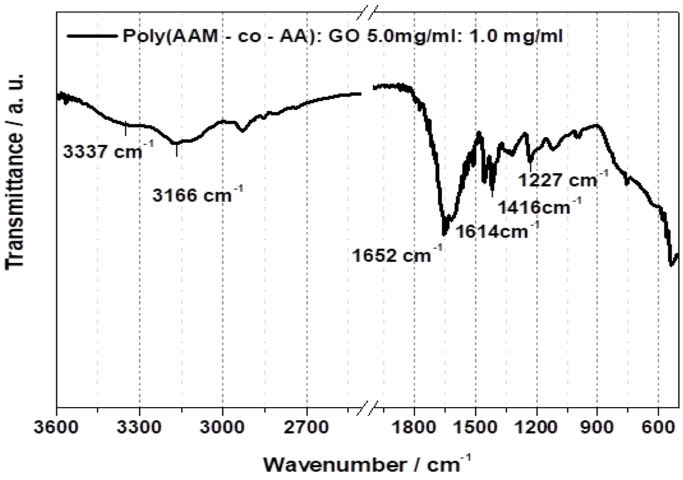


Figure4.7 c) FT-IR spectrum of Poly(AAm-o-AA):GO(5.0:1.0)mg/ml

Table 4.4 shows the degree of wavenumber shift to left or to right for the peaks in nanocomposite . This clear shift for different functional groupes attributed to the interaction between GO and polymer chains.

Functional groupe name	Wavelength of GO (cm^{-1})	Wavelength of Poly(AA-co-AA) (cm^{-1})	Wavelength of Poly(AA-co-AA) with GO nanocomposite (cm^{-1})	Shiftting
O-H	3364			
NH_2		3337	3337	
CH_2		3190	3166	24 to right
C=O		1646	1652	6 to left
C=C	1614		1614	
C–N		1428	1416	12 to left
COOH		1322	1227	95 to left

Table 4.4 FT-IR results for each of GO,Poly(AAm-co-AA) film,Poly(AAm-co-AA)/GO nanocomposite.

5. Conclusion and Future Work

Poly(AA-co-AA)/graphene oxide nanocomposite were synthesized by using simple casting technique.The interaction leads to improvement in the thermal properties. It leads to raise in Tg, melting, degradation, and tempereture of crystallization.Barrier properties of graphene oxide was noticed by decreasing in the swelling as increasing the concentration of GO. FT-IR of Poly(AA-co-AA)/GO nanocomposite indicate the strong interaction between polymer and GO particles .

Based on these results, the development of the polymer charactes can be refered to a homogenous dispersion of geaphene oxide in polymer chains and effictive interaction between the two components. In the future, it should be applied to specific drugs as a capsule shell that include hydrophobic and hydrophilic drugs .

6.Referances

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تحضير و وصف مركب اكسيد النانوجرافين مع حمض البولي اكريل اميد كو اكريليك لتطبيقات تغليف المستحضرات الصيدلانية .

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ملخص :

تم انتاج أكسيد الجرافين باستخدام تفاعل االكسدة واالختزال (طريقة هامر). تم استخدام أكسيد الجرافين لتضمينها في بوليمر حمض بولي اكريل امايد كو اكريليك من أجل دراسة الخواص الحرارية والخواص الميكانيكية, وكذلك اختبار التورم.

تم استخدام تراكيز مختلفة من بوليمر حمض بولي اكريل امايد كو اكريليك(5 ,2.5 ,1.25 ,0.9)%مع تراكيز مختلفة من اكسيد الجرافين(1.0 و 0.5)%لتحضير مركب النانو حمض بولي اكريل امايد كو اكريليك / اكسيد الجرافين باستخدام طريقة الصب البسيطة.

تمت دراسة الخواص الحرارية باستخدام مسعر المسح التبايني (DSC) وأظهرت النتائج أن الخواص الحرارية تحسنت من خالل دمج أكسيد الجرافين في بوليمر حمض بولي اكريل امايد كو اكريليك .

درجة االنتقال الحراري (Tg)زادت من 50 لبوليمر حمض بولي اكريل امايد كو اكريليك النقي إلى 75درجة مئوية عند اضافة 0.5 %من أكسيد الجرافين. المساحة فوق منحنى منحدر درجة حرارة التحلل زادت أيضا عند اضافة اكسيد الجرافين وهذا يدل على أن جزء من مادة بوليمر حمض بولي اكريل امايد كو اكريليك تحول من الشكل الابلوري الى الشكل البلوري مما يؤدي الى التقليل في الماسافات بين سلاسل بوليمر حمض بولي اكريل امايد كو اكريليك , وهذا دليل على زيادة صلابتها .

قوة الشد في بوليمر حمض بولي اكريل امايد كو اكريليك زادت بزيادة نسبة أكسيد الجرافين إلى200% عندما كان تركيز أكسيد الجرافين 0.5% و 300% في عندما كان تركيز أكسيد الجرافين1.0% .

نسبة التورم عند لبوليمر حمض بولي اكريل امايد كو اكريليك قلت بزيادة نسبة أكسيد الجرافين والتي تفسر خصائص حاجز اكسيد الجرافين في المستوى الجزيئي لمصفوفة البوليمر.

تؤكد أطياف تحويل فورييه الطيفي باألشعة تحت الحمراء (FT-IR)من مركبات النانو بوليمر حمض بولي اكريل امايد كو اكريليك / أكسيد الجرافين الترابط القوي بين البوليمر والجسيمات النانونية في عدة مجموعات وظيفية.

باختصار, دمج أكسيد الجرافين في مصفوفة بوليمر كحول حمض بولي اكريل امايد كو اكريليك يحسن الخواص الحرارية والميكانيكية والحاجز للبوليمر, بسببب تفاعل القوي بين جزيئات البوليمر وأكسيد الجرافين.