



## Effect of cefotaxime concentrations on the surface roughness parameters and swelling rate of chitosan/Gum Arabic polymer blend

Alshaima M. Ali<sup>1</sup>, A.M. Abdelghany<sup>2</sup>, M. El-Henawy<sup>1</sup>, A.H. Oraby<sup>1</sup>

<sup>1</sup>Physics Department, Faculty of Science, Mansoura University, Mansoura 35516, Egypt

<sup>2</sup>Spectroscopy Department, Physics Research Institute, National Research Centre, 33 ElBehouth St., Dokki, 12311, Giza, Egypt

Received: 12/2/2022  
Accepted: 15/2/2022

**Abstract:** Flexible thin films of natural polymer blend containing 90wt% Chitosan (Cs), 10 wt% Gum Arabic GA) containing variable mass fractions of cefotaxime drug were successfully synthesized. The effect of dopant drug concentration was studied on correlation to the roughness parameters and swelling rate behavior of the studied nanocomposite. All doped samples were compared to their pristine undoped blend of 90Cs/10GA. Obtained data show that studied parameters are considered as concentration dependant. Studied samples characteristics suggest possible use in different medical, catalytic and optical applications.

**keywords:** Chitosan (Cs); Gum Arabic (GA); Cefotaxime (CF); Roughness parameters; Swelling rate.

### 1. Introduction

During the last century, polymeric material and their respective filled or doped samples or even their composite studied for possible uses in different fields especially in the medical field has grown with a better understanding of polymers backbones and possible modifications of their structure where it forms the most building blocks of our day's life [1-3]. Some living organisms are considered as an important source for natural polymers that starts synthesizing within the middle of the nineteenth century with lacking a proper understanding of their molecular structure [4]. One of the important applications during the last decades is to produce natural or synthetic polymers to produce nanoparticles to encapsulate insulin for oral delivery [5].

Chitosan, a straight-chain copolymer of  $\beta$ -(1-4)-D-glucosamine and acetyl-b-(1-4)-D-glucosamine, synthesized through alkaline N-deacetylation of naturally polysaccharide chitin, Chitosan ( $C_6H_{11}O_4N$ )<sub>n</sub> is the 2nd most abundant basic polysaccharide on the earth and is structurally similar to cellulose, which is composed of only one monomer of glucose. Chitosan and chitin have become materials of great interest as an under-utilized resource and

as a new functional biomaterial of high potential in various fields [6-8].

Gum Arabic (GA) or Acacia gum is an edible biopolymer obtained as exudates of mature trees of Acacia Senegal and Acacia Seyal, Acacia Karoo, Acacia polyacantha, Acacia sieberana [6]. GA is a branched-chain, complex hetero-polysaccharide, either neutral or slightly acidic, found as mixed calcium, magnesium, and potassium salt of a polysaccharide acid (Arab Acid). Gum Arabic is widely used in the food industry, it is non-toxic, odorless, and tasteless [9,10]. It interacts with water and has a wide range of applications such as emulsion, texture control, and aver encapsulation Gum Arabic at near-neutral pH, its carboxyl groups will be largely dissociated and lead the molecule to be open [11]. These properties enable both oppositely charged polymers with negatively charged polymers to interact with each other by intermolecular electrostatic interaction, forming polyelectrolyte complexation (PEC) [12]. Gum Arabic has many medicinal effects, including wound healing, producing a liquid solution with an acidic property (pH ~ 4.5). Known as an antioxidant biomolecule, the ability of several forms of amino acid residues increases [13].

The mechanical characteristics of chitosan have been established with the use of gum Arabic cross-linking. It has now been recorded to be used in folk medicine, such as used outside to mask inflamed surfaces [14].

Cefotaxime is a third - generation cephalosporin antibiotic agent, it is used for a wide-ranging of infectious processes activated by susceptible organisms, and can improve the antimicrobial effect [15,16].

Presented work aims to investigate the effect of cefotaxime concentrations on the surface roughness parameters and swelling rate of chitosan/Gum Arabic flexible thin polymer blend films synthesized using natural sources.

## 2. Experimental Work

### 2.1. Sample preparation

Chitosan (Cs) polymer of chemical formula  $[(C_6H_{11}NO_4)_n]$  and high weight with a deacetylation degree of 85% extra pure was obtained from exporter lab chemicals Co. (Alpha Aesar, USA). Gum Arabic (GA) polymer of chemical formula  $(C_{26}H_{34}N_2O_{13})$ , gotten from Bio Basic INC (CAS 9000-01-5). Gum Arabic of physical powdered appearance and Orange-brown colored shape material. Cefotaxime drug powder of chemical formula  $[(C_{16}H_{17}N_5O_7S_2)]$  and MW (455.47 g/mol) was purchased from Pharmaceutical Chemicals, Egypt.

Flexible thin films of 90wt% Chitosan (Cs), 10 wt% Gum Arabic (GA) containing variable mass fractions of cefotaxime drug were successfully synthesized through the traditional solution casting route adopting distilled water as a common solvent. The concentration of the drug was determined and calculated before the preparation of the doped samples. Sample nomination and composition were listed in table (1).

**Table (1)** sample nomination and composition

Sample	Nominal Composition		
	CS	GA	CF × 47.6 mg
	Wt%		
Blend	90	10	0
B1	90	10	1
B2	90	10	2
B3	90	10	3
B4	90	10	4
B5	90	10	5

The surface morphology of the synthesized thin films was investigated using a scanning electron microscope (SEM) (Quanta Field emission FEG 250 Netherlands) operated at 30 kV voltage. A (2 cm×2 cm) piece of the dry sample was immersed in a buffer solution of constant pH ranging between 4 and 9 and incubated at 37 °C for 24 h until the samples reached the equilibrium state of swelling. The swollen films were weighted before ( $W_d$ ) and after immersion ( $W_s$ ). The swelling ratio was calculated using [13]:

$$\text{Swelling ratio}(\%) = \frac{W_s - W_d}{W_d} \times 100$$

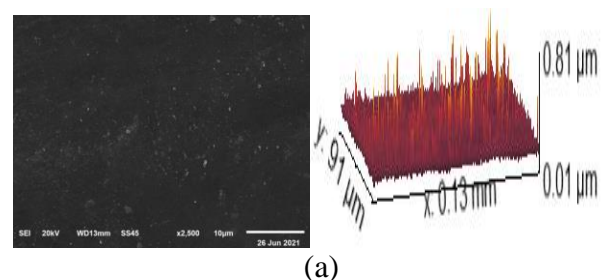
## 3. Results and Discussion

### 3.1. SEM analysis

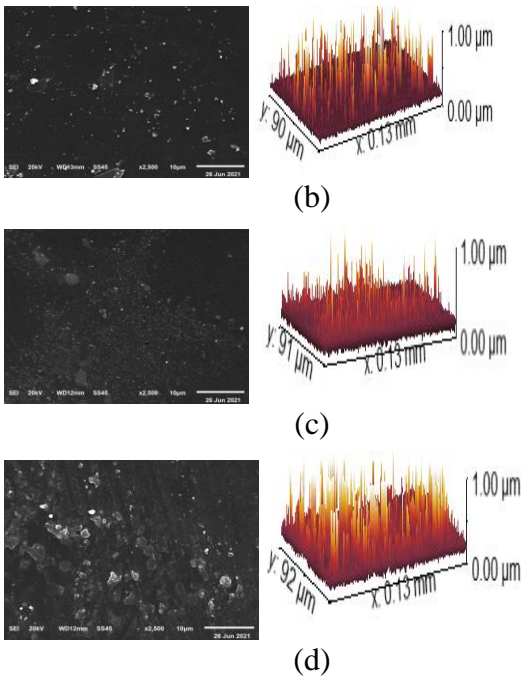
SEM permits to form of a quantitative analysis of different morphological aspects of a corroded surface, including estimating surface roughness, characterizing the corrosion pits (their dimension and shape), locating the corrosion, and determining the morphology of corrosion products (such as oxides).

The surface morphology indicated via SEM images with a constant magnification of the studied samples synthesized via traditional solution casting evaporation route result from mixing of different polymers can give information about the interaction and distribution of filling materials throughout the polymeric matrix before and after incorporate with cefotaxime drug.

Figure (1.a-d) reveals exemplified SEM micrographs of the synthesized samples. The analyzed sample micrographs were used to calculate surface parameters including average roughness ( $R_a$ ), Root means square roughness ( $R_q$ ), the maximum height of the roughness ( $R_t$ ), maximum roughness valley depth ( $R_v$ ), maximum roughness peak height ( $R_p$ ), and Average maximum height of the roughness ( $R_{tm}$ ).



(a)



**Fig (1.a:d)** SEM of Cs/GA (90:10) with CF drug (a) Blend, (b) B1, (c) B3, and (d) B5.

It was clear that the roughness parameters values that were normally categorized as amplitude, spacing, and hybrid parameters [17] were increased with increasing cefotaxime concentration as introduced in Table (2). The sudden change in roughness parameters of the sample containing maximum concentration of the drug may be attributed to the formation of saturated solution resulting in the appearance of clusters observed within the SEM micrographs figure (1.d).

**Table (2)** roughness parameters of the studied samples

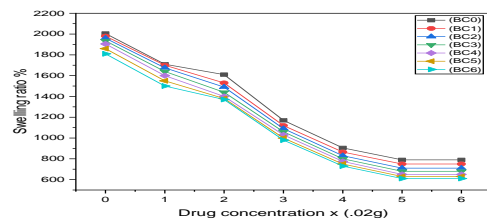
Sample	$R_a$	$R_q$	$R_t$	$R_v$	$R_p$	$R_{im}$
Blend	12.1	17.5	260.2	120.1	130.2	210.4
B1	15.2	21.4	308.6	150.3	150.6	230.8
B2	22.4	28.6	360.9	160.5	180.7	260.7
B3	32.7	35.4	415.6	180.7	195.8	295.8
B4	35.5	40.4	510.8	210.4	220.4	340.5
B5	39.9	45.3	650.8	240.4	305.4	370.7

The data collected can be utilized to determine the best use for the samples tested. An increase in roughness parameters coupled with an increase in surface area recommends that the studied samples could be used as a high-surface-area catalyst throughout industrial applications or for drug delivery and wound healing in medical applications, particularly with CF antibiotic drugs that have superior antibacterial properties [18].

### 3.1. Swelling rate test

Homeostasis, intercellular lipid, permeability barrier, cohesion, and other structural properties of the skin's uppermost layer are physiologically characterized as stratum corneum. Many factors influence these characteristics, including the skin's pH. The stratum corneum is known as the acid mantle because the pH of healthy and normal skin is in the range of 5.0–6.0. The pH of healthy and normal skin is defined as 5.0–6.0, and the stratum corneum is known as the acid mantle. Acid mantle pH is influenced by sebaceous glands, eccrine glands, apocrine glands, epidermal cells, gender, and age. If the skin pH is unbalanced, it leads to many disorders like skin illness (irritation and inflammation) as well as reduced cell cohesion and permeability barrier in the stratum corneum [13,19]. The swelling behavior is essential for polymer films used for wound healing because fluid uptake is a key agent during tissue regeneration. Swelling is a usual phenomenon of the interaction of a Polymer with solvent. Insertion of such liquid within polymer matrix tends to swell it. This could be either reversible or irreversible [20-23]. Many times, some polymers tend to absorb moisture which tends to change the associated properties. The total moisture insertion is not always reversible.

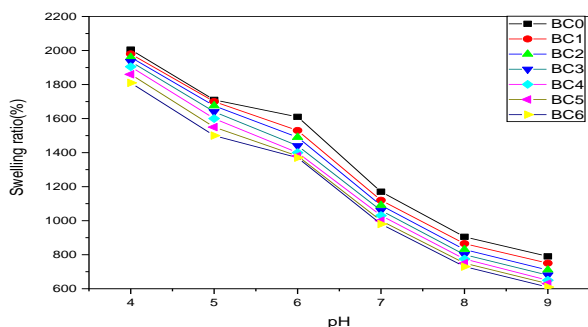
Figure (2) explains the swelling behavior of the studied blend sample and other samples that contain different mass fractions of the CF drug as a function of drug concentration and variable pH. This data revealed that blend samples have a good adsorption capacity and with increasing the pH values, the swelling rate of blend decreased. Such behavior is consistent with the fact that chitosan polymer dissolved in slightly acidic media and with increasing alkalinity, it's difficult to dissolve unless it is combined with other polymers.



**Figure (2)** Swelling ratios of blend samples and samples containing variable concentrations of CF drug with various pH values.

CS is recognized for its water-absorbing, its swelling ability, and its biocompatibility. So their blend curves shows the good swelling behavior. With increasing the pH, the swelling behavior of CS was noticed to be decreased. So these results encourage using CS/GA for the treatment of severe wounds and can be used for the preparation of hydrogel in the treatment of chronic wounds like diabetic foot ulcers [24-26]. By adding cefotaxime antibiotic, the swelling behavior of CS/GA also decreases with increasing drug concentration and it is proved by the blend curve in figure (2). These types of films can be utilized for antibiotic drug delivery or as injectable drug delivery carriers.

**Figure (3)** shows the various swelling rates of the blend samples at different values of pH (4.0, 5.0, 6.0, 7.0, 8.0, and 9.0) containing different concentrations of CF drug. There is a change in the swelling ratio of the blend has been noticed when placed in various pH of the aqueous solution. At (pH 4.0- 6.0), the swelling ratios of the system decrease by increasing the CF concentration. When the pH values increased the swelling behavior decreased due to the increase of cross-linked, rigidity of network, and compact structure of the blend due to intra/inters molecular interaction which decreased swelling and solubility of drug.



**Figure (3)** Swelling ratios of (Cs/GA) blend with different concentrations of CF drug against various pH values.

#### 4. Conclusions

90 weight percent CS/10 weight percent GA an ordinary casting procedure was used to synthesis a mix flexible thin film sample with other samples of the same composition having a little dopant of cefotaxime medication. Both blending and adding CF to the examined mix resulted in an increase in surface roughness metrics and a decrease in the swelling rate of

the nanocomposites under investigation. Both events can occur and be explained in the context of a crosslinking process that causes a delay in the swelling process, resulting in a reduction in the swelling rate. The obtained data demonstrates the concentration dependency on the physical properties of the sample under consideration. It was also discovered that samples with greater CF concentrations had essentially same behaviour. Such concentration was recommended for possible use in different medical, catalytic and optical applications.

#### 5. References

1. Saheb, D. N., & Jog, J. P. (1999). Natural fiber polymer composites: a review. *Advances in Polymer Technology: Journal of the Polymer Processing Institute*, **18**(4), 351-363.
2. Thomas, S., Joseph, K., Malhotra, S. K., Goda, K., & Sreekala, M. S. (Eds.). (2012). *Polymer composites, macro-and microcomposites (Vol. 1)*. John Wiley & Sons.
3. Nurazzi, N. M., Sabaruddin, F. A., Harussani, M. M., Kamarudin, S. H., Rayung, M., Asyraf, M. R. M., ... & Khalina, A. (2021). Mechanical Performance and Applications of CNTs Reinforced Polymer Composites—A Review. *Nanomaterials*, **11**(9), 2186.
4. Toh, H. W., Toong, D. W. Y., Ng, J. C. K., Ow, V., Lu, S., Tan, L. P., ... & Ang, H. Y. (2021). Polymer blends and polymer composites for cardiovascular implants. *European Polymer Journal*, **146**, 110249.
5. Sabbagh, F., Muhamad, I. I., Niazmand, R., Dikshit, P. K., & Kim, B. S. (2022). Recent progress in polymeric non-invasive insulin delivery. *International Journal of Biological Macromolecules*, **203**, Pages 222-243.
6. Abdelghany, A. M., Meikhail, M. S., & Awad, W. M. (2022). Electrical conductivity of chitosan/PCL hosting network for CdSe quantum dots. *Polymer Bulletin*, **79**, 4381–4393.
7. Aboelwafa, M. A., Abdelghany, A. M., & Meikhail, M. S. (2021). Preparation, Characterization, and Antibacterial Activity of ZnS-NP's Filled Polyvinylpyrrolidone / Chitosan Thin Films. *Biointerf. Res. Appl. Chem.*, **11**(6),

- 14336-14343.
8. Bussiere, P. O., Gardette, J. L., Rapp, G., Masson, C., & Therias, S. (2021). New insights into the mechanism of photodegradation of chitosan. *Carbohydrate Polymers*, **259**, 117715.
  9. Vuillemin, M. E., Muniglia, L., Linder, M., Bouguet-Bonnet, S., Poinsignon, S., Morais, R. D. S., ... & Jasniewski, J. (2021). Polymer functionalization through an enzymatic process: Intermediate products characterization and their grafting onto gum Arabic. *International Journal of Biological Macromolecules*, **169**, 480-491.
  10. Zahran, M., Khalifa, Z., Zahran, M. A., & Azzem, M. A. (2021). Gum Arabic-capped silver nanoparticles for electrochemical amplification sensing of methylene blue in river water. *Electrochimica Acta*, **394**, 139152.
  11. Digby, Z. A., Yang, M., Lteif, S., & Schlenoff, J. B. (2022). Salt Resistance as a Measure of the Strength of Polyelectrolyte Complexation. *Macromolecules*, **55(3)**, 978–988.
  12. Putro, J. N., Lunardi, V. B., Soetaredjo, F. E., Yuliana, M., Santoso, S. P., Wenten, I. G., & Ismadji, S. (2021). A Review of Gum Hydrocolloid Polyelectrolyte Complexes (PEC) for Biomedical Applications: Their Properties and Drug Delivery Studies. *Processes*, **9(10)**, 1796.
  13. Pereira, A. G., Nunes, C. S., Rubira, A. F., Muniz, E. C., & Fajardo, A. R. (2021). Effect of chitin nanowhiskers on mechanical and swelling properties of Gum Arabic hydrogels nanocomposites. *Carbohydrate Polymers*, **266**, 118116. DOI: 10.1016/j.carbpol.2021.118116
  14. Shavisi, N., & Shahbazi, Y. (2022). Chitosan-gum Arabic nanofiber mats encapsulated with pH-sensitive *Rosa damascena* anthocyanins for freshness monitoring of chicken fillets. *Food Packaging and Shelf Life*, **32**, 100827.
  15. Javaid, S., Ahmad, N. M., Mahmood, A., Nasir, H., Iqbal, M., Ahmad, N., & Irshad, S. (2021). Cefotaxime Loaded Polycaprolactone Based Polymeric Nanoparticles with Antifouling Properties for In-Vitro Drug Release Applications. *Polymers*, **13(13)**, 2180.
  16. Kryuk, T. V., Tyurina, T. G., & Kudryavtseva, T. A. (2021). Sodium Cefotaxime–Potato Starch Conjugate as a Potential System for Antibacterial Drug Delivery. *Pharmaceutical Chemistry Journal*, **55(8)**, 803-807.
  17. Menazea, A. A., & Awwad, N. S. (2020). Pulsed Nd: YAG laser deposition-assisted synthesis of silver/copper oxide nanocomposite thin film for 4-nitrophenol reduction. *Radiation Physics and Chemistry*, **177**, 109112.
  18. Fecske, S. K., Gkagkas, K., Gachot, C., & Vernes, A. (2020). Interdependence of amplitude roughness parameters on rough gaussian surfaces. *Tribology Letters*, **68(1)**, 1-15.
  19. Jayaramudu, T., Ko, H. U., Kim, H. C., Kim, J. W., & Kim, J. (2019). Swelling behavior of polyacrylamide–cellulose nanocrystal hydrogels: swelling kinetics, temperature, and pH effects. *Materials*, **12(13)**, 2080.
  20. Ramzan, M., Obodo, R. M., Mukhtar, S., Ilyas, S. Z., Aziz, F., & Thovhogi, N. (2021). Green synthesis of copper oxide nanoparticles using *Cedrus deodara* aqueous extract for antibacterial activity. *Materials Today: Proceedings*, **36**, 576-581.
  21. Choe, C., Schleusener, J., Choe, S., Ri, J., Lademann, J., & Darvin, M. E. (2020). Stratum corneum occlusion induces water transformation towards lower bonding state: A molecular level in vivo study by confocal Raman microspectroscopy. *International Journal of Cosmetic Science*, **42(5)**, 482-493.
  22. Montaser, A. S., Rehan, M., El-Senousy, W. M., & Zaghoul, S. (2020). Designing strategy for coating cotton gauze fabrics and its application in wound healing. *Carbohydrate Polymers*, **244**, 116479.
  23. Abou El-Reash, Y. G., Abdelghany, A. M., & Abd Elrazak, A. (2016). Removal and separation of Cu (II) from aqueous solutions using nano-silver chitosan / polyacrylamide membranes. *International journal of biological macromolecules*, **86**, 789-798.
  24. Lee, Y. H., Hong, Y. L., & Wu, T. L. (2021). Novel silver and nanoparticle-

- encapsulated growth factor co-loaded chitosan composite hydrogel with sustained antimicrobially and promoted biological properties for diabetic wound healing. *Materials Science and Engineering: C*, **118**, 111385.
25. Elmorsy, E. E., Abdelghany, A. M., Ayad, D. M., & El, O. A. (2021). Synthesis and Physicochemical Studies of Polyvinyl Alcohol Polymer Modified with Copper Thiosemicarbazide Complex. *Lett. Appl. NanoBioScience*, **10**, 2624-2636.
26. Wang, M., Xu, L., Hu, H., Zhai, M., Peng, J., Nho, Y., ... & Wei, G. (2007). Radiation synthesis of PVP/CMC hydrogels as wound dressing. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, **265(1)**, 385-389.