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Study the Effect of adding Polyacrylamide on Rheological and Mechanical Properties of Carboxymethyle cellulose Polymer as aqueous solutions

Prof. Dr. Abdul-Kareem J. Al-Bermany E-mail: dr.abdulkaream@yahoo.com
University of Babylon / Facility of Science / Physics department / Iraq
Dr. Fouad SH. Hashim E-mail: bur11111@yahoo.com
Haider H. jassim E-mail: haider.hassan389@yahoo.com
University of Babylon / College of Education for pure science/ Physics department/ Iraq

Abstract:
In this paper, we investigated the Rheological and mechanical properties of Carboxymethyle cellulose dissolves in distilled water at different concentrations (0.1%, 0.2%, 0.3%, 0.4% ,0.5 ,%,0.6 %,0.7% and 0.8%) (gm./ml) before and after adding (0.5, 1) gm. of PAAM for all concentrations, the Rheological properties such as shear viscosity, relative viscosity, specific viscosity, reduced viscosity and Viscosity Average Molecular weight are measured, all the viscosities depend on density and concentration, The results show that adding PAAM lead to increase the values of different types of viscosities then chose before.
Also the research included study of the Mechanical properties by ultrasonic waves velocity technique at frequency(25 KHz), these properties are absorption coefficient of ultrasonic waves, relaxation time, relaxation amplitude, specific acoustic impedance, compressibility, and bulk modules had been measured and all the results showed that all properties are increasing with the increase of the polymer concentration except compressibility is decreasing with the increase of the concentration before and after adding PAAM.
Keywords: Carboxymethyle cellulose, Polyacrylamide ,Rheological properties, Mechanical properties.

1-Introduction

In many cases water-soluble polymers give rise to substantial viscosity increases when dissolved in water. Therefore, a large number of water-based systems used in practical application contain polymers to control the viscosity (1). CMC is one of the most important cellulose derivatives, which have an immense importance to the industry and also in our everyday life. CMC is a linear, long chain, water soluble, anionic polysaccharide derived from cellulose (2). In addition, the purified cellulose is a white to cream colored as well as tasteless, odorless, and it is a free-flowing powder. Furthermore, due to its water-soluble heteropolysaccharides with high molecular weight properties, thus CMC is often blended with starch to provide desirable texture, enhanced product quality and stability, control moisture and also water mobility (3). CMC is an important industrial polymer due to its high viscosity, non-toxic, non-allergenic, biodegradability as well as production at lower cost. Furthermore, it is a most important water soluble derivative with various applications in paper, food, detergents, cosmetics, and textiles(4). CMC is generally prepared through the reaction of alkali cellulose with monochloroacetate or its sodium salt in an organic medium(5).
Although, due to hydrogen bonds that associated with structure of molecular cellulose, it neither melts nor dissolves readily in common solvents. The main step in carboxymethylation its water-soluble derivatives have found various applications. The purified product has contribution in food, pharmaceutical, detergents and cosmetics industries. It is used as a preservative for coating of fresh fruit and thickener for pharmaceutical products. Beside the wide use of CMC, it is used as drilling mud in oil industry (6). In addition, CMC is applied as a dye thickening in textile industry. CMC as detergent and surfactant are used as anti-dirt agent for protection of fibers surface. CMC has been synthesized from raw cellulose, wood, paper cotton-linter fibers, Lantana camara, banana plants and sugar beet pulp(7). Polyacrylamide, is a synthetic polymer derived from acrylamide monomer which was originally introduced for use as a support matrix for electrophoresis in 1959. Polyacrylamide is a polymer that is formed from units of acrylamide, a known neurotoxin. However, polyacrylamide itself is non-toxic, but is a controversial ingredient because of its potential ability to secrete acrylamide. Polyacrylamide is used in wide range of cosmetic products (moisturizers, lotions, creams, self-tanning products, etc.). Because polyacrylamide is chemically inert and stable over various conditions, polyacrylamide has been employed, whether clinically or under development, to serve as a useful matrix for several types of extracorporeal toxin removal . Polyacrylamides were first used as an implantable carrier for sustained delivery of insulin to lengthen the life of diabetic rats. Since then, various drug delivery systems based on polyacrylamide have been developed(8). It is also used as a carrier for other bioactive macromolecules and cells to produce the desired effects. Polyacrylamide-chitosan hydrogels are biocompatible and are used for sustained antibiotic release(9). PAAM is major applications flocculants in water treatment, paper manufacture, mining, and oil recovery; absorbents; gels for electrophoresis(10)(11).

ultrasonic technique is good method for studying the structural changes associated with the information of mixture assist in the study of molecular interaction between two species; some of mechanical properties of different polymers were carried by some workers using ultrasonic technique(12). The purpose of this research was to investigate the physical properties of carboxy methyl cellulose (CMC) with Polyacrylamide (PAAM) as aqueous solutions by ultrasound wave at fixed frequency (25 KHZ) and study the effect of adding PAAM on the physical properties of CMC to enhance its different applications.

2. Experimental:

2-1 Preparation of Solutions:

CMC (Panreac Spain) with assay (99.8%) and PAAM product by (BHD) with assay (99.9%) of high viscosity. The CMC solution was prepared by dissolving a known weights of CMC powder in affixed volume (500 ml) of distilled water under stirring at 70C^{o} for (60 min). The CMC concentrations were (0.1%, 0.2%, 0.3 %, 0.4%, 0.5%, 0.6%, 0.7% and 0.8%) gm./ ml ; then PAAM was added with different weights (0.5 and 1 gm.) to all CMC Concentrations. The resulting solution were stirred continuously for (60 min) until the solution mixture became a homogeneous.

2.2 Rheological measurements

2.2.1 Density and viscosity measurements:

The density of the CMC solution (ρ) was determined by the density bottle method and the shear viscosity measured before and after adding PAAM for all concentrations using Ostwald viscometer with accuracy of ± 1.05% (13,14), elsewhere different types of viscosity were determined before and after the adding PAAM by the equations (1, 2, 3 and 4), The shear viscosity had been calculated by the following equation (14)(10):

$$\eta_s / \eta_o = (\rho_s\, t_s) / (\rho_o\, t_o) \ldots\ldots (1)$$

Where (ρ_s) and (η_s) are density and shear viscosity of solute respectively, (ρ_o) and (η_o) are density and viscosity of distilled water respectively, (t_s) and (t_o) are the flow time for solution and distilled water respectively Relative viscosity ($\eta_{rel.}$) is simply the ratio of the viscosity of the polymer solution to the viscosity of the pure solvent at the same temperature. or the ratio of the two efflux times, and is given by the Jones-Dole equation(15) (16):

$$\eta_{rel.} = (\eta_s / \eta_o) = 1+ \eta_{red.} \ldots\ldots.(2)$$

Where (c) is the concentration of solutions and($\eta_{red.}$) is the reduced viscosity, The specific viscosity (η_{sp}) and the reduced viscosity($\eta_{red.}$) was calculated by the equations(15)(10) (16):

$$\eta_{sp.} = (\eta_{rel.} - 1) = \eta_{red.}\, c \ldots\ldots\ldots (3)$$
$$\eta_{red.} = \eta_{sp.} /c \ldots\ldots\ldots\ldots (4)$$

The intrinsic viscosity has been obtained practically its value represent the intersection to y-axis as C goes to zero of graph between reduced viscosity and concentration, which represented the piratical value of intrinsic viscosity before and after adding PAAM. The intercept values of these curves are shown in table (1). The intrinsic viscosity can be calculated theoretically by using philippoff equation as follow:

$$\eta_{rel} = [\, 1 \; + [\, \eta] \frac{c}{g}]^{8} \ldots\ldots (5) \quad \text{Philipp off Equation}$$

The relation between [η] and relative viscosity was determined by Arrhenius, so it's called Arrhenius equation as follows(13):

$$\ln \eta_{rel} = [\eta\,]\, C \ldots\ldots (6) \quad \text{Arrhenius Equation}$$

Viscosity Average Molecular weight had been calculated by the following equation(15) (17):

$$[\eta\,] = K M^{a}_{v} \ldots\ldots\ldots\ldots (7)$$

Where k, a are constant depends on the type of the polymer, for CMC the values of constants are (a=0.91, K=1.23*10^{-4}) (15) (18) .

The effective molecular radius(r) was calculated by the following equations(10)(19):

$$\eta_{rel} = 1+ 6.3*10^{24}\, r^3\, C_m \ldots\ldots\ldots\ldots\ldots\ldots (8)$$

$$\text{slope} = 6.3*10^{24}\, r^3 \ldots\ldots\ldots\ldots\ldots\ldots (9)$$

$$r = \sqrt[3]{slope/6.3 * 10^{24}} \quad \text{..... (10)}$$

Where slope equal to the value of slope plotted between relative viscosities against concentration.

2.3 Mechanical measurements

2.3.1 Ultrasonic :

Ultrasonic measurements were made by pulse technique of sender-receiver type (SV-DH-7A/SVX-7 velocity of sound instrument) with constant frequency (25 KHz),as shown in Fig. below the receiver quartz crystal mounted on a digital venire scale of slow motion, the receiver crystal could be displaced parallel to the sender and the samples were put between sender and receiver. The sender and receiver pulses (waves) were displaced as two traces of cathode ray oscilloscope, and the digital delay time (t) of receiver pulses were recorded with respect to the thickness of the samples (x). The pulses height on oscilloscope (CH1) represents incident ultrasonic wave's amplitude (A_0) and the pulses height on oscilloscope (CH2) represents the receiver ultrasonic wave's amplitude (A) after passing the solution.

Generator and Receiver of Ultrasonic Waves

The ultrasonic wave velocity (v) was calculated using the following equation :

$$v = x / t \quad \text{..... (11)}$$

The relaxation time (τ) was calculated from the equation(20) :

$$\tau = 4 \eta_s / 3\rho v^2 \text{..... (12)}$$

Where (t) is time that the waves need to cross the samples (digital obtained from the instrument).
The acoustic impedance of a medium (Z), it was calculated by equation(21):

$$Z = \rho v \text{..... (13)}$$

Bulk modulus (K) is the substance's resistance to uniform compression, it is defined as the pressure increase needed to decrease the volume; it was calculated by Laplace equation(22):

$$K = \rho v^2 \text{..... (14)}$$

Compressibility (β) is a measure of the relative volume change of a fluid or solid as a response to a pressure (or mean stress) change, it was calculated by the following equation(23) :

$$\beta = (\rho v^2)^{-1} \text{..... (15)}$$

The absorption coefficient (α) was calculated from Lambert – Beer law(24) :

$$A/A_0 = e(-\alpha x) \ldots\ldots (16)$$

Where (A_0) is the initially amplitude of the ultrasonic waves,(A) is the wave amplitude after absorption and (x) is the thickness of the sample. Attenuation is generally proportional to the square of sound frequency so the relaxation amplitude (D) was calculated from the following equation where (f) is the ultrasonic frequency(25):

$$D = \alpha / f^2 \ldots\ldots\ldots (17)$$

3. Results and Discussion:

3.1 Rheological properties:

The values of density with concentration are shown in (Fig.1) this Fig shows the density is increasing with increase of the concentration since the density defined as mass per unit volume and we adding different weight of polymer to fixed volume of solvent so there are linear increment for density. Shear Viscosity is increasing with concentration as shown in (Fig.2) this attributed to the mechanism that hydrogen bonding of water attached to oxygen sites, this leads to solvation sheaths and increase in the size of the molecules so its viscosity (13), furthermore water act as plasticizer that reduce tensile strength and increase its chains. Relative viscosity, specific viscosity and reduced viscosity as show in figure (3), figure (4) and figure (5) respectively possess the same behaviors of shear viscosity because they derived from it as shown in equations (2,3,4) adding PAAM made enhancement for these viscosities because the viscosity describes a fluid's internal resistance between molecules so when we add polyacrylamide there will be more molecules, the additional forces between molecules leads to an additional contribution to the shear stress(14). the theoretical intrinsic viscosities were calculated by using philippoff equation and arrhenius equation. The comparison between our experimental values and the theoretical values obtained by these two equations are shown in table (1). The exponential behavior of viscosity with concentration was attributed to the structural change associated with liquid polymer solution and probably indicating entanglement interaction(26).

Intrinsic Viscosity $[\eta]$ (dl/gm.)			
polymers	Theoretical		Experimental
	Arrhenius equation	Philippoff equation	
CMC	13.2	13.4	13.9
CMC+ 0.5 gm. PAAM	18.2	18.4	18.8
CMC + 1gm. PAAM	23.2	23.4	24

Table(1) shows comparison between the theoretical and experimental values of Intrinsic Viscosity

Viscosity Average Molecular weights before and after adding PAAM were calculated by using equation(7). The values of [η] were taken experimentally from table (1) and constants (K) and(a) depending on the polymer type and shown in the following table (2). The calculated values of the viscosity-average molecular are shown in table (2). High represent comparison between the theoretical values of viscosity average molecular weights obtained by philippoff and arrhenius equations and experimental values obtained by using intrinsic viscosity.

Viscosity Average Molecular Weight (M_v)					
Polymers	a	The constant (K) x 10^{-4}	Theoretical		Experimental
			arrhenius equation	Philippoff equation	
CMC	0.91	1.23	337448.5	343071	357164
CMC+ 0.5 gm. PAAM	0.8	1.8	1803011.9	1827812.5	1877615.6
CMC + 1 gm. PAAM	0.8	2.19	1911194	1931811.2	1993925.4

Table(2) comparison between the theoretical and experimental values of Viscosity Average Molecular weight (Mv)

Effective molecular radius for High and Low concentration were increased after adding PAAM as show in table (3).

Effective radius (r) (cm)		
polymers	Low Con. x 10^{-7}	High Con. x 10^{-7}
CMC	5	6.5
CMC+ 0.5 gm. PAAM	9.2	12
CMC +1 gm. PAAM	10.5	13

Table (3) Comparison between effective radius for High and Low concentration

3.2 Mechanical properties:

A useful method of studying mechanical properties of liquids is based on ultrasound. Figure (6) shows the relationship between the speed of ultrasound with a concentration. This figure shows that the velocities of ultrasonic wave remain constant with the increase of the CMC polymer concentration. Because of the lack of interaction or interlock between the solute and solvent molecules where the molecules dissolved in solution without the presence of the bonds to binds it, which makes the free movement of molecules within the solution, which in turn allows the passage of ultrasound and this helps to make the speed of waves fixed with concentration. and Adding PAAM increase the velocity, this attributed that ultrasonic waves interact with polymers causing association between the two types of molecules that lead to increase the velocity (27). (Fig.7) Shows that Ultrasonic relaxation time are increasing with concentration, also (Fig.7) Shows that relaxation time increased when adding PAAM this attributed to the fact that ultrasonic energy depends on viscosity thermal conductivity, scattering and intermolecular processes , thermal conductivity and scattering effects are known to be negligible (14), (Fig.8) shows that The specific acoustic impedance increasing at increasing concentration, since adding PAAM increased acoustic impedance because PAAM polymer chains fills the valances by swallowing water molecules and be closer to CMC macromolecules(27). the bulk modulus is increasing with concentration (Fig.9). The compressibility is decreasing with the increase of concentration (Fig.10) and attributed to the fact that in Laplace equation no(15). There are inverse proportionality between compressibility and ultrasonic velocity(27). (Fig.11) shows that absorption coefficient is increasing, this is because the attenuation of the ultrasonic wave in liquids is determined mainly by the size, shape, and distribution of particles dispersed in the carrier liquids, this is attributed to the fact that when polymer concentration increase there will be more molecules in solution this lead to more attenuation against wave propagation and the changes in the particle size distribution function rather than the increase in the shear viscosity , the attenuation can be attributed to the friction and heat exchange between the particles and the surrounding medium as well as to the decay of the acoustic wave in the forward direction due to scattering by the Particles , increasing of solution concentration always results in increase of solution viscosity, and vice versa(28). The relaxation amplitude is increasing with increasing concentration as shown in figure (12) this attributed that the displacement of excited molecules became small because moment of inertia for polymer macro molecules reduce (25).

4. Conclusion:

1-Adding PAAM to CMC made enhancement for the viscosity, Increasing concentration leads to increasing viscosity because of the mechanism that hydrogen bonding of water attached to oxygen sites, this leads to salvation sheaths and increase in the size of the molecules so its viscosity so it can be used as thicker colloid blend in coating process.

2-ultrasonic absorption coefficient increases with increasing concentration before and after adding PAAM so it can be used as coated materials for moving bodies in order not to be detect by ultrasonic technique .

3- also can be used as coated materials for teaching room and different factories to absorb sound noises of instruments.

4- When concentration increases the velocity increases there will be complexes molecules were formed in the solution by the effect of peroxide and roots that rebounded to Network formations between polymer chains when adding PAAM, so the blend can be used as good medium for transferring ultrasonic waves in such medical instruments.

5- Adding PAAM reduced compressibility this lead to increase interaction between polymer molecules this cause enhancement for mechanical properties against environments .

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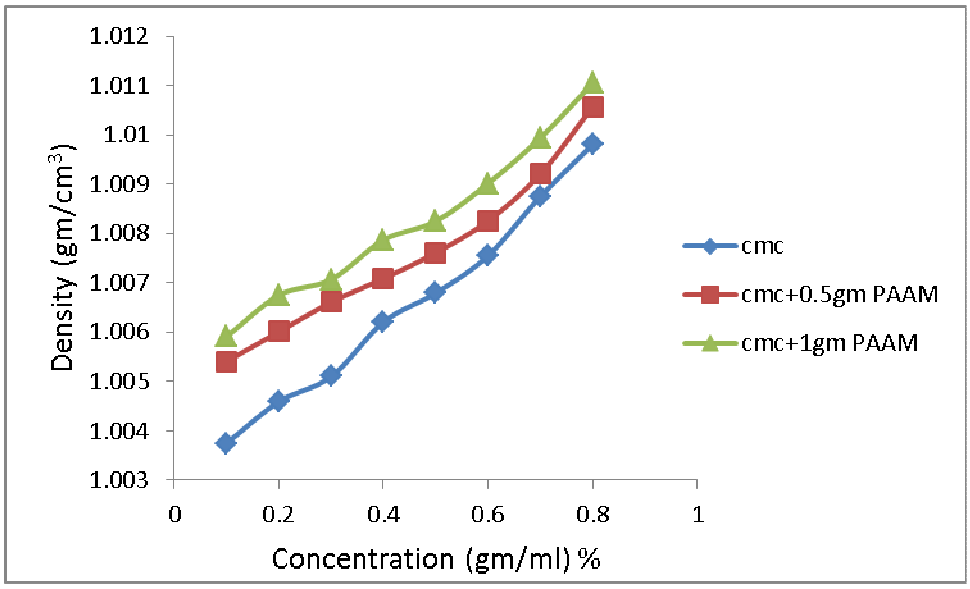
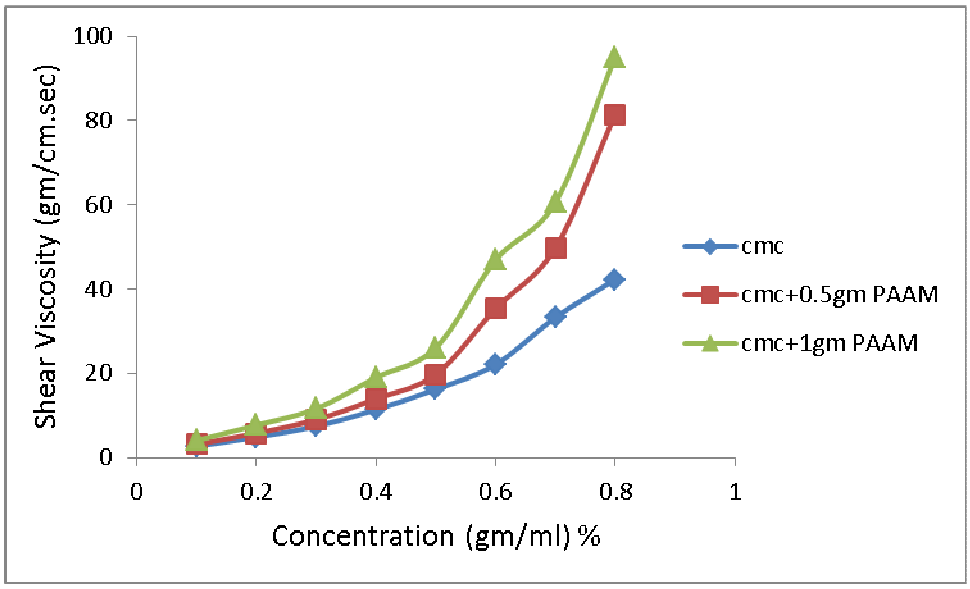
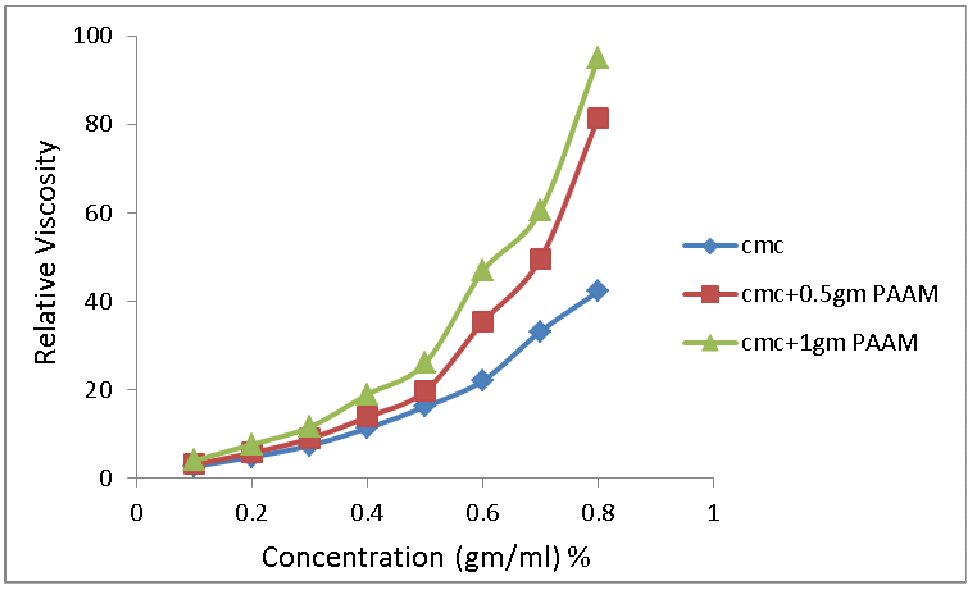


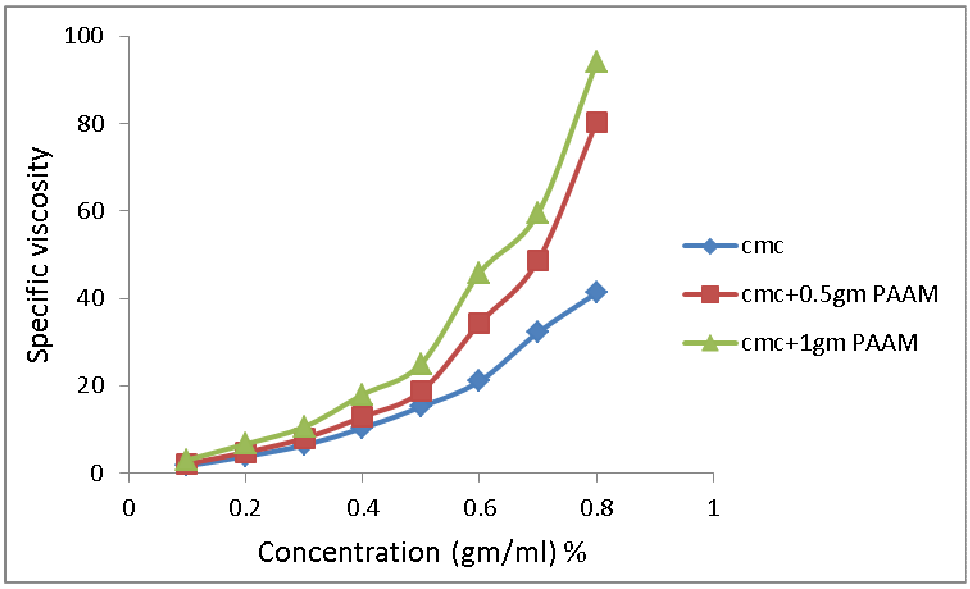
Figure(1) Density due to concentration



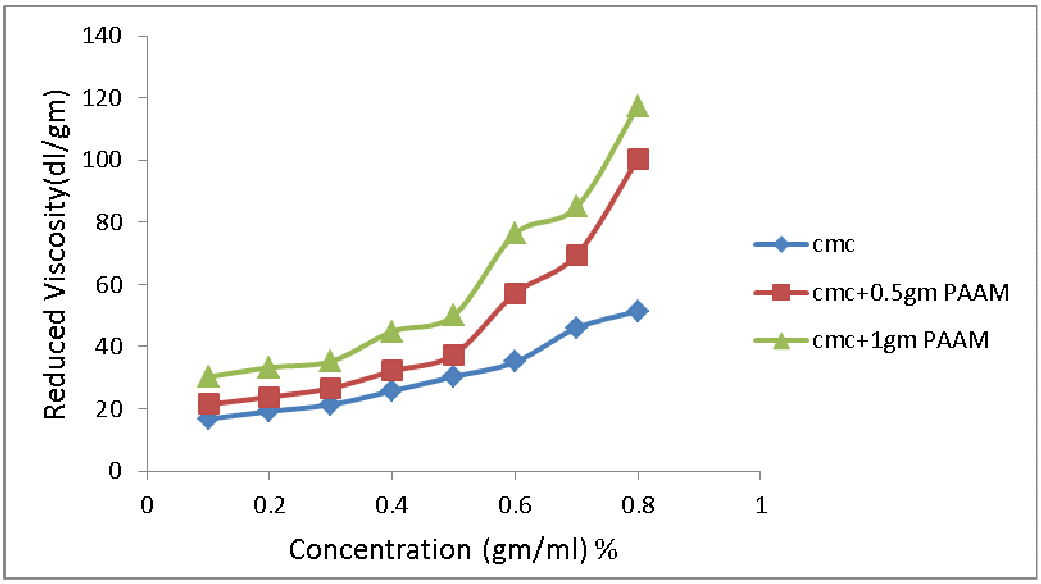
Figure(2) Shear viscosity due to concentration



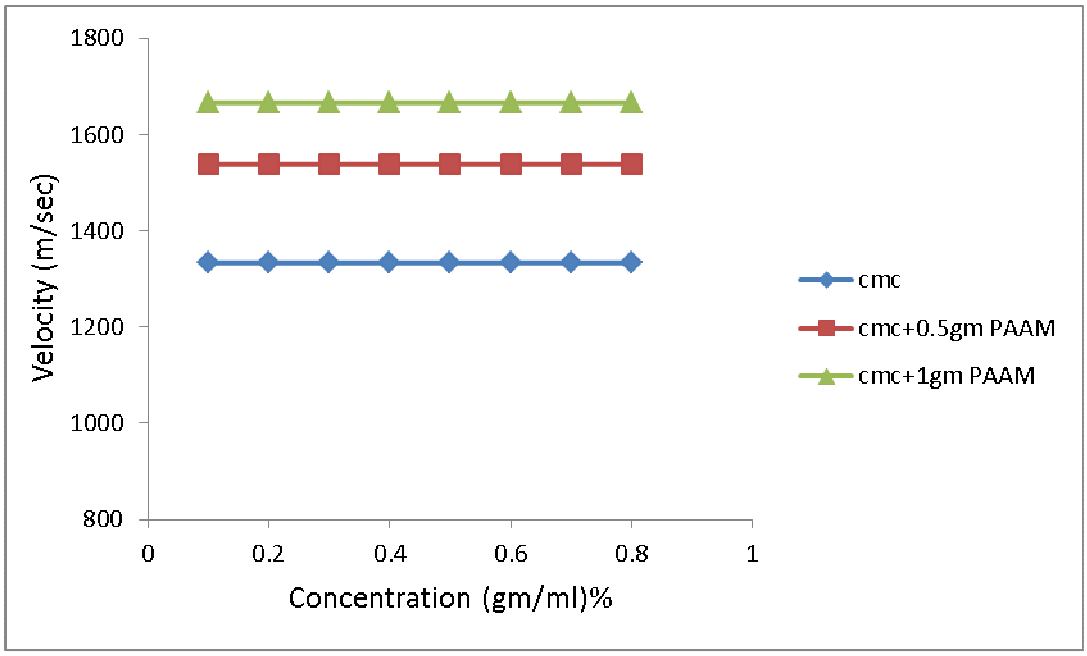
Figure(3) Relative viscosity due to concentration



Figure(4) Specific viscosity due concentration



Figure(5) Reduce viscosity due to concentration



Figure(6) Velocity due to concentration

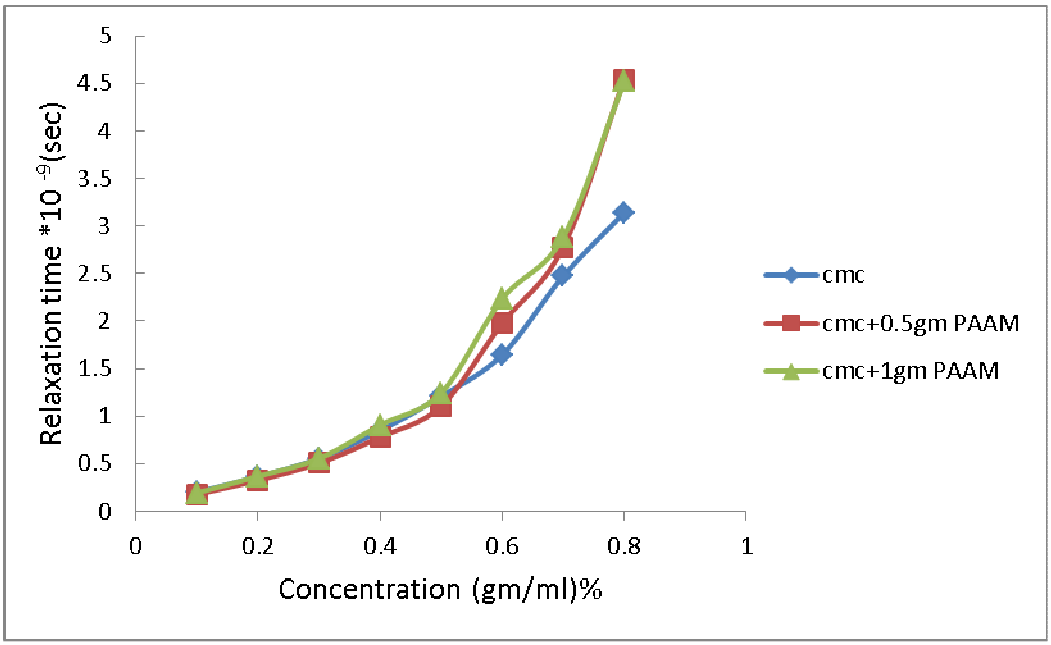


Figure (7) Relaxation time due to concentration

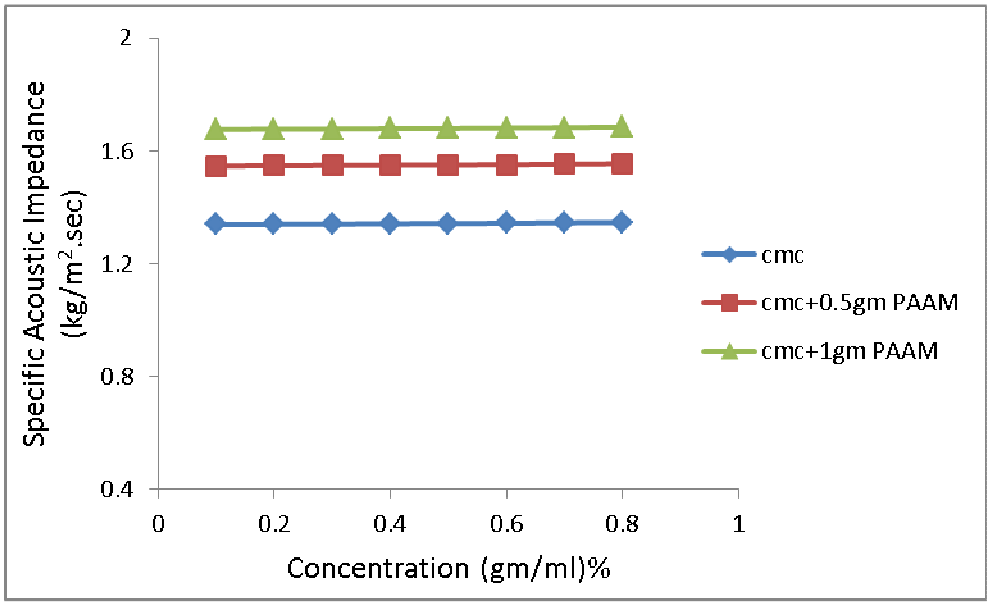


Figure (8)Acoustic impedance due to concentration

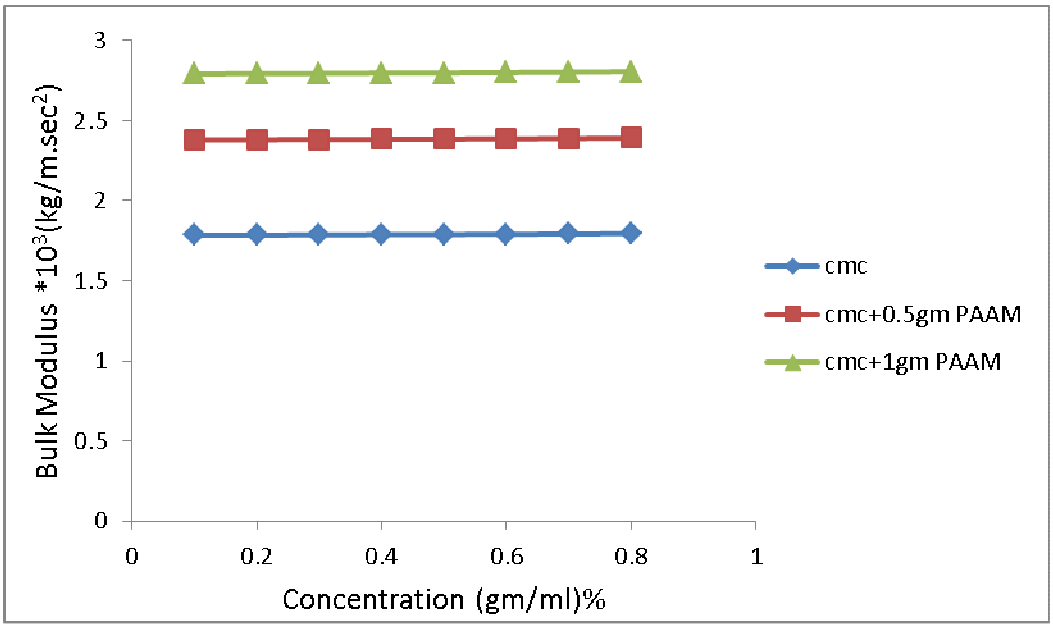


Figure (9) Bulk Modulus due to concentration

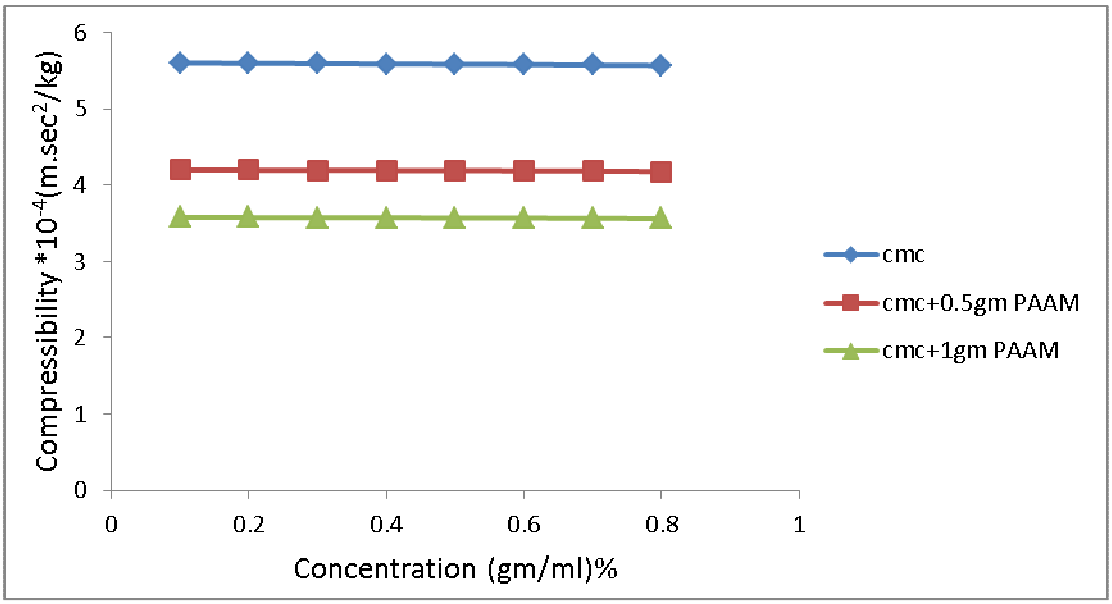


Figure (10) Compressibility due to concentration

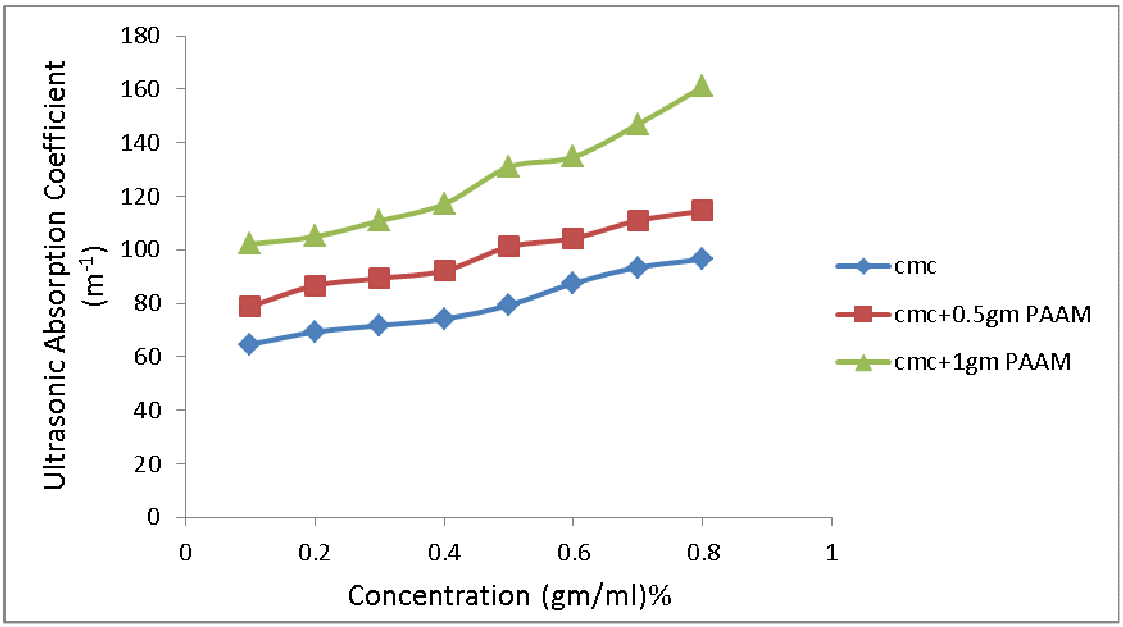


Figure (11) Absorption coefficient due to concentration

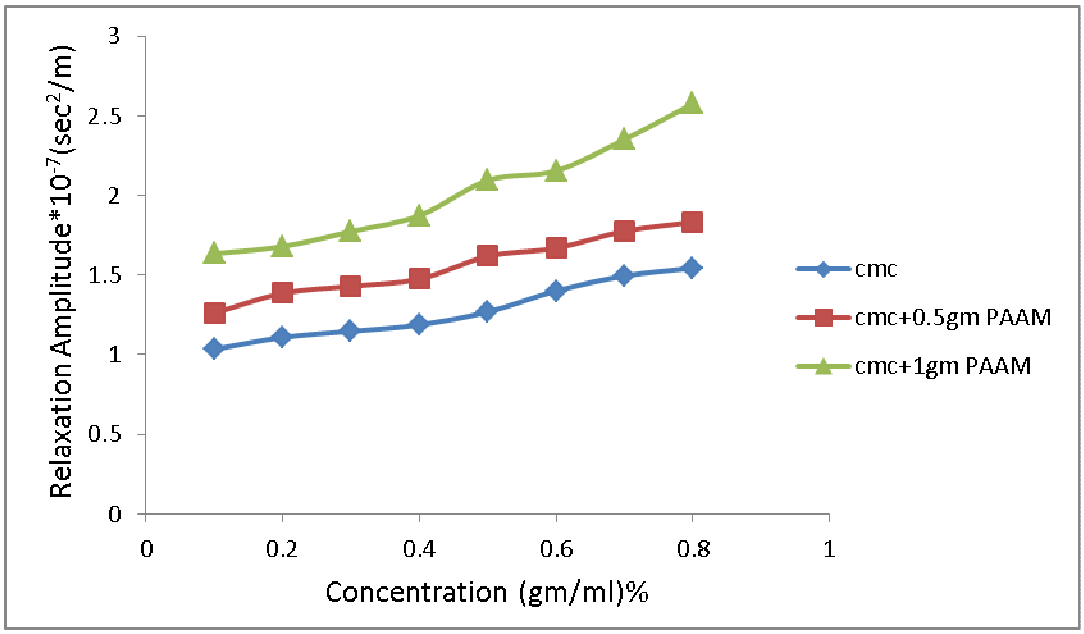


Figure (12) Relaxation amplitude due to concentration

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