Influence of Sodium Carboxymethyl cellulose on Sedimentation of Aluminium Phosphate Precipitate at Various Temperatures

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ABSTRACT
*Sedimentation of aluminium phosphate formed in presence of sodium caboxymethyl cellulose biopolymer was studied at various temperatures. The main objective is to present the influence of sodium carboxymethyl cellulose on sedimentation behavior of aluminium phosphate precipitate. Known volume (10ml) of sodium carboxymethyl cellulose at different concentrations was mixed with constant volume (30ml) of disodium hydrogen phosphate solution in a sedimentation measurement apparatus-pressed meter. Resulting solution was mixed separately with constant volume (30ml) of aluminium chloride solution. The settling of the precipitate formed was monitored and recorded at different temperatures (30, 40 and 50^{o}C). Blank sedimentation run (with 10ml of deionized water instead of 10ml of sodium carboxymethyl cellulose) was inconclusive. For every measurement conducted, precipitate sedimentation profile curve was described and On-set Sedimentation Rate (OSR) and Sedimentation Volume Ratio (SVR) computed. It was observed that without sodium carboxymethyl cellulose, aluminium phosphate precipitate readily dissolved into the medium shortly after mixing or during stirring. Precipitate remained un-dissolved in presence of sodium carboxymethyl cellulose biopolymer. Poor correlations in OSR ($0.50 \leq R^2 \geq 0.05$) and SVR ($0.49 \leq R^2 \geq 0.005$), indicating non-*dependence *with concentration of sodium carboxymethyl cellulose, implying no chemical interaction of aluminium phosphate precipitate with sodium carboxymethyl cellulose, were described at all temperatures investigated. These observations suggest that sodium carboxymethyl cellulose biopolymer enhances precipitation of aluminium phosphate but poorly influenced sedimentation of the precipitate formed. The observations also imply that sodium carboxymethyl cellulose would be good stabilizing agent for drug formulations containing aluminium phosphate.*

Keywords: *sedimentation, aluminium phosphate, precipitate, sodium carboxymethyl cellulose, temperature*

1.0 INTRODUCTION

Sedimentation, also known as settling, describes the motion of particles in suspension or molecules in solution in response to external force (Atkins & Paula, 2006; Fatimi *et al.*, 2010; Obunwo *et al.*, 2017). It is a physical phenomenon that happens when materials in suspension are denser than the suspending medium.The phenomenon is an advantage in water purification technology where suspensions are further sedimented to enhance proper separation of flocculated or precipitated particles. However, in formulation of suspension drugs, sedimentation phenomenon is a disadvantage because it causes compaction of the active ingredients at the bottom of the container. Two important parameters used in describing and quantifying sedimentation characteristics of pharmaceutical formulations include sedimentation volume ratio and sedimentation rate (Patel, 2010; Santosh and Naga, 2016). Sedimentation volume ratio is a measure of the degree of sediment compaction. Sedimentation rate is a measure of particle-size distribution and is usually evaluated from settling onset time or supernatant clarity (Mekhamer *et al.*, 2009; Kang *et al.*, 2013).

Aluminium Phosphate is a chemical compound with the formula $AlPO_4$. It is an inorganic salt found in minerals and pharmaceutical industries where they are used as a catalyst to obtain chemotherapeutics drugs commercially available in gel. It is a crystalline powder that does not have any odour, slightly soluble in hydrochloric and nitric acid and soluble in water. It is the most used antacids, in industries; they have it in ceramics dental, cement, cosmetics, paints, paper and like calcium and iron, the phosphate of aluminium has been in use as adjuvant (enhancer of vaccine potency) in the formulation of injectable suspension drugs against bacterial and viral diseases (Paneque-Quevedo, 2013; Chawla *et al.*, 2016; Angelova and Yordanov, 2018).

Sodium carboxymethyl cellulose is a non- toxic and tasteless white flocculant powder. It is stable and soluble in water. It is an aqueous solution in neutral and alkaline transparent viscous liquid. It is insoluble in organic solvents such as ethanol. In pharmaceutical industry, sodium carboxymethyl cellulose is widely used in oral and topical application or oral and parental administration. It may also be used as tablet binder and disintegrant and can also be used to stabilized emulsions (Devish and Rakesh, 2020). Sodium carboxymethyl cellulose can form highly viscous colloidal solution with adhesive, emulsifying, flowing, thickening water protective colloid, film forming acid and it is physiologically harmless, so it is widely used in food, pharmaceutical, cosmetic, oil paper, textiles construction and other area of production (Ergun and Huebner, 2016). It is also applied in formulation of suspension drugs to enhance physical stability of active pharmaceutical ingredients. (Okorie *et a*l., 2011; Kadaji *et al.*, 2011; Mastropietro *et al*., 2013 and Lee *et al*., 2014).

The effect of sodium carboxymethyl cellulose on sedimentation of precipitate is not well known. However, studies by Khaled & Abdelbaki (2012); Mastropietro *et al*., (2013) showed that polyvalent metal ions such as calcium, aluminium and zinc have decremental effect on flow properties of sodium carboxymethyl cellulose biopolymer. Also, Iboroma *et al.* (2018) asserted that interaction of suspending medium with materials (ions, molecules, precipitates, etc.) in suspension could influence the physical stability behaviour of suspension. In this work, the sedimentation of aluminium phosphate precipitate formed without and with solution of sodium carboxymethyl cellulose was studied, by (1) precipitating known amount of aluminium phosphate precipitate in presence and in absence of sodium carboxymethyl cellulose at 30, 40 and 50^{o}C and (2) by sedimenting of the precipitate so formed. The purpose was to present the influence of sodium carboxymethyl cellulose biopolymer on sedimentation of aluminium phosphate precipitate.

2.0 MATERIALS AND METHODS

2.1 Materials and Reagents

Analytical grade JHD chemicals including Aluminium chloride, disodium hydrogen phosphate and Sodium carboxymethyl cellulose made by Guangdong Guanghua Sci-Tech Co. Limited were used for the study. De-ionized water obtained from Central Chemistry laboratory was used to prepare solutions. A precipitate sedimentation measurement apparatus (Plate 1) consisting of hot-plate, magnetic stirrer, cylindrical tube, water-bath, thermometer and a stirring rod was set up. Digital stopwatch (XL-5853) was used for taking timely readings of the sedimentation process and a LED light (Dp 90828) was used to provide back light for proper illumination.

2.2 Preparation of Standard and Other Working Solutions

0.5M solutions of aluminium chloride (molecular weight [$AlCl_3.6H_2O$] = 241.43g) and disodium hydrogen phosphate (molecular weight [$Na_2HPO_4.12H_2O$] = 358.14g) were prepared by weighing 120.73g and 179.07 using Denver Instrument Company (TP-512A) weighing balance. Each salt was dissolved in de-ionized water and made up to volume in one litre volumetric flask. Also, 2000ppm solution of sodium carboxymethyl cellulose (Na-CMC) was prepared by dissolving 2g of Na-CMC in 1 litre of water. Other working solutions (200ppm, 400ppm, 600ppm, 800ppm, 1000ppm, 1200ppm, 1400ppm, 1600ppm and 1800ppm) of Na-CMC were prepared by dilution of 25ml, 50ml, 75ml, 100ml, 125ml, 150ml, 175ml, 200ml, 225ml and 250ml of the stock in 250ml measuring cylinder.

Plate 1: Precipitate Sedimentation Measurement Apparatus- The PreSed Meter.

3.3 Batch-wise Precipitation and Sedimentation Tests

30ml of 0.5M solution of Na_2HPO_4 (as phosphate ion precursor) was mixed in a gradulated measuring cylinder with 30ml of 0.5M solution of $AlCl_3$ (as aluminium ion precursor) to produce aluminium phosphate precipitate suspension. The precipitate formed was stirred for 1min and was allowed to settle under gravity at different temperature settings (30, 40 and 50°C) of the sedimentation measurement apparatus. The effect of sodium carboxymethyl cellulose concentration on sedimentation of the precipitate formed was carried out by adding fixed volume (10ml) of different concentrations (200ppm-2000ppm) of sodium carboxymethyl cellulose, first to the phosphates ion solution before adding the aluminium ion solution. To zero the effect of sodium carboxymethyl cellulose and obtain sedimentation data in absence of sodium carboxymethyl cellulose, blank sedimentation tests, without sodium carboxymethyl cellulose, were tried, by adding 10ml of deionized water instead of 10ml of sodium carboxymethyl cellulose solution. Attempt to measure sedimentation of aluminium phosphate precipitate in absence of sodium carboxymethyl cellulose failed because precipitate dissolved into the reaction medium shortly after mixing and thus no sedimentation data was recorded in absence of sodium carboxymethyl cellulose. Formation of insoluble aluminium precipitates is illustrated using equations 3.1.

$$Al^{3+}_{(aq)} + PO_4^{3-}_{(aq)} \rightarrow AlPO_{4(s)} \qquad (3.1)$$

The displacement of the precipitate suspension (hp) was monitored and recorded every minute, for 10 minutes, when the sedimentation was observed to be complete. This gave the precipitate height at equilibrium sedimentation (hp_{ES}). For every sedimentation test, data were recorded and precipitate displacement plotted as $[-(hp_i - hp_0)]$ against $(t_i - t_0)$, where hp_i was the height of the precipitate suspension at a given time, t_i, and hp_0 was the initial height of the suspension at time zero, t_0. Precipitate height at equilibrium sedimentation, (hp_{ES}), which is the height of the suspension zone or interface when the sedimentation was observed to be near complete, was also recorded. In these tests, the On-set Sedimentation Rate (OSR) and Sedimentation Volume Ratio (SVR) of precipitate were determined in a single measurement. The rate at which the suspension moved (or displaced) at early stages was used to measure the OSR and the ratio of the height of the suspension at equilibrium sedimentation to the height of the sample gave the SVR (Your recent Publication). The experiments were carried out at temperatures ranging from 30 to 50°C.

3.0 RESULTS AND DISCUSSION

Precipitation and sedimentation tests performed in absence (0ppm) and in presence (200ppm - 2000ppm) of sodium carboxymethyl cellulose suggest that sodium carboxymethyl cellulose enhances precipitation of aluminium phosphate but poorly influenced sedimentation of the precipitate formed. It was observed that in absence of sodium carboxymethyl cellulose, aluminium phosphate precipitate readily dissolved into the medium shortly after mixing. Thus, no sedimentation data was recorded for aluminium phosphate precipitate in absence of sodium carboxymethyl cellulose. However, in presence of sodium carboxymethyl cellulose (200ppm - 2000ppm), precipitate was stable without dissolution at the various temperatures and the sedimentation profiles are given in Figure 1. As may be seen in Figures 1, the sedimentation profiles described by aluminium phosphate precipitate are irregular and also showed similar sedimentation pattern. For example, at 30°C, profile corresponding to 2000ppm exhibited shortest settling time but exhibited longest settling time at 40°C. At 50°C, profile for 200ppm gave shortest settling time. Also, profiles corresponding to 200ppm and 2000ppm exhibited similar slow settling times at 40°C.

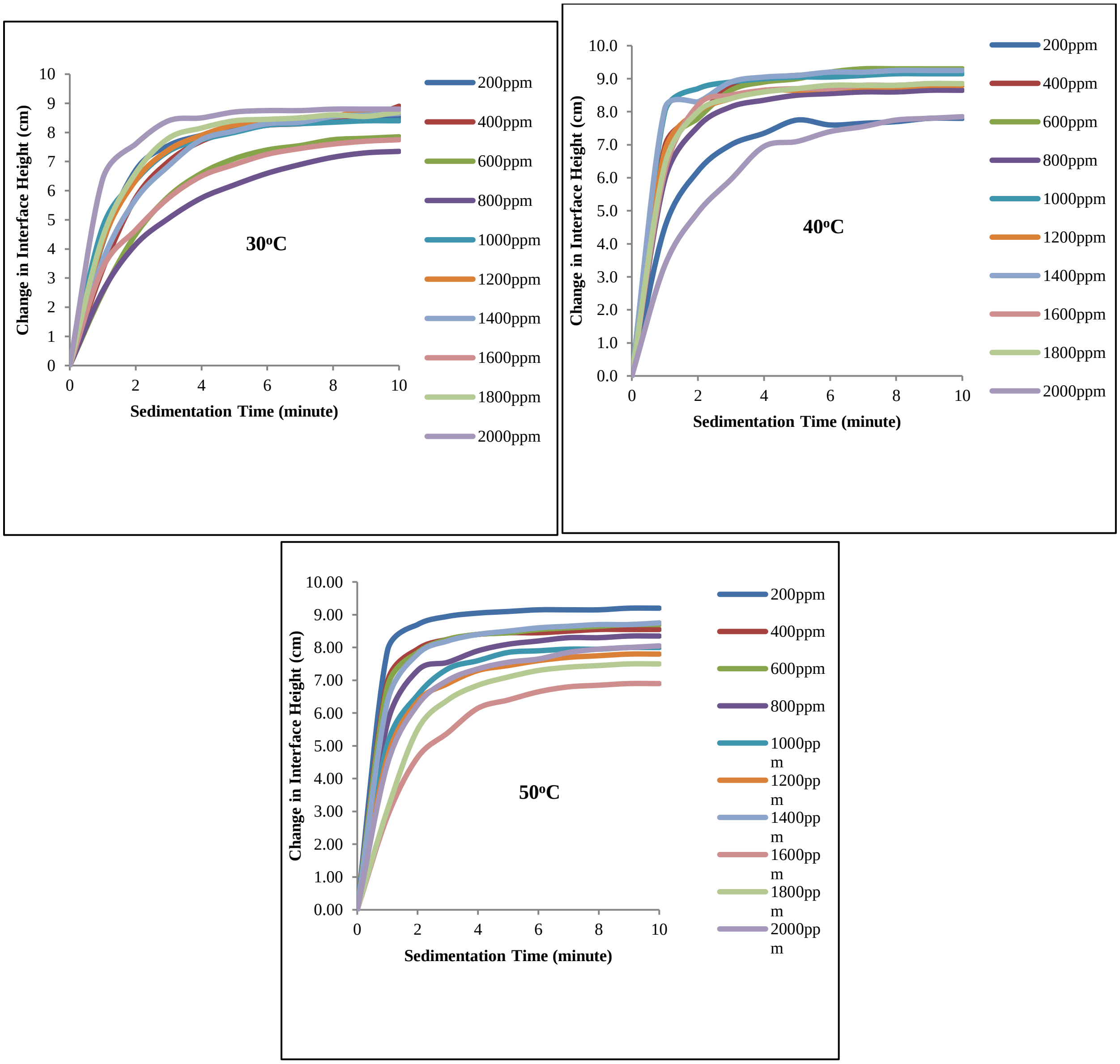
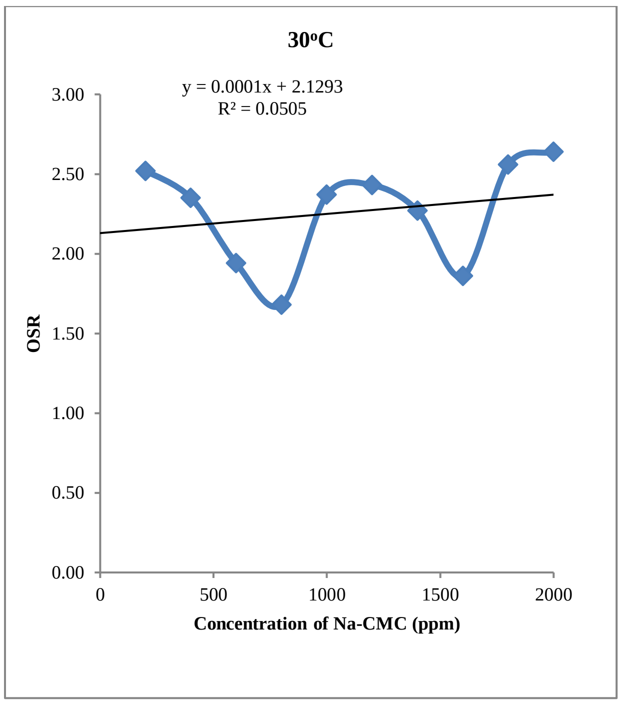
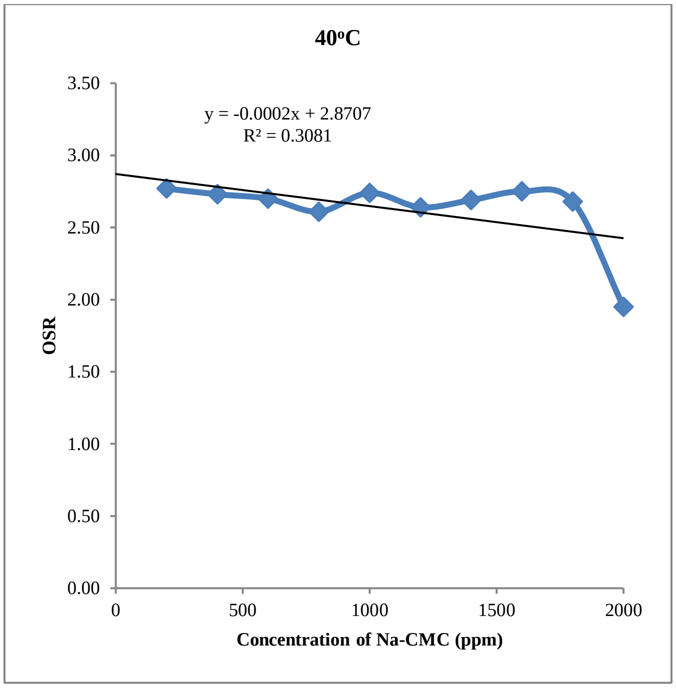


Figure 1: Sedimentation Profiles of aluminium phosphate Precipitate in Presence of sodium carboxymethylcellulose for Various Temperatures

Temperature affects sedimentation via change in medium viscosity (Obunwo*et al*., 2014). Sodium carboxymethyl cellulose biopolymer, a suspending agent, also affects sedimentation through viscosity change. The influence of sodium carboxymethyl cellulose biopolymer and temperature on sedimentation of precipitate is based on the assumption that chemical interaction between suspending medium and precipitate may occur and more so at high temperature. The poor correlations in the computed OSR ($0.50 \leq R^2 \geq 0.05$) and SUR ($0.49 \leq R^2 \geq 0.005$) described are indicative of low chemical interaction between aluminium ions and sodium carboxymethyl cellulose molecule at the temperatures investigated. The results of study also described the suitability of sodium carboxymethyl cellulose biopolymer as suspending medium for aluminiumphosphate precipitate suspension. OSR and SVR of aluminium phosphate increased only by (4.8% and 14.1%) and decreased by only (3.3% and 2.7%) at 30^{o}C and 40^{o}C. The 17.7% decrease in OSR and 40% increase in SVR observed at 50^{o}C are abnormal and unexpected. The observations are indicative of low interaction of sodium carboxymethyl cellulose with aluminium phosphate precipitate at the various temperatures.

This observation also implies that sodium carboxymethyl cellulose solution would be good suspending agent for drug formulations containing aluminium phosphate.





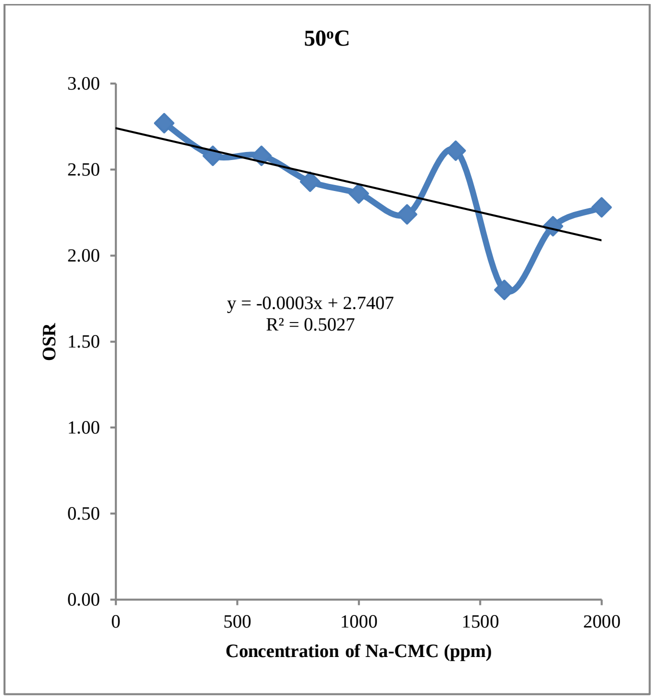
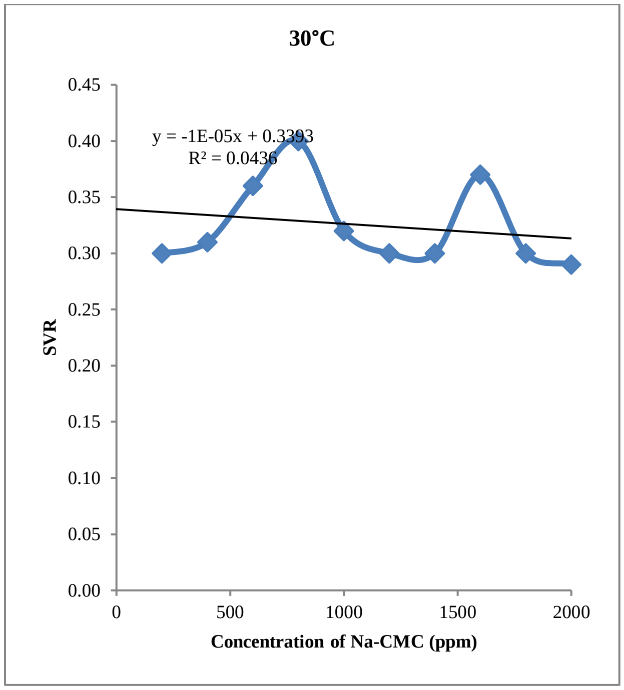
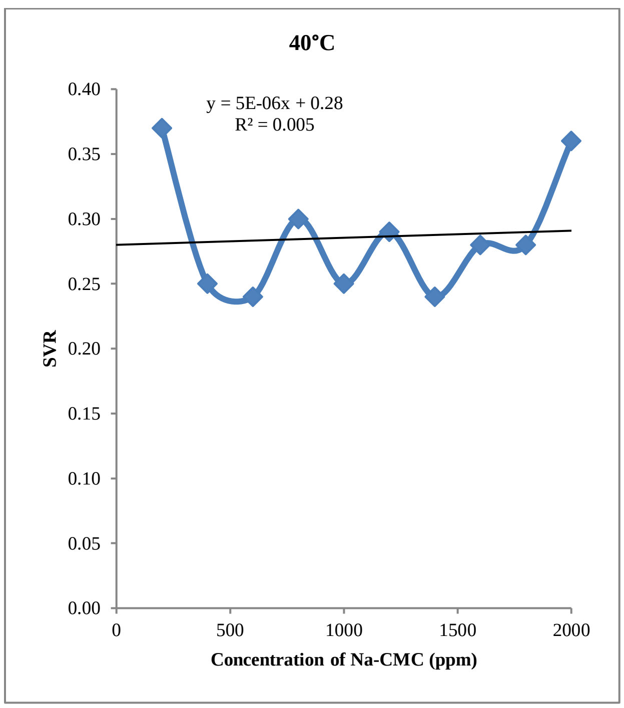


Figure 4: Graph of OSR of Aluminium Phosphate against Concentration of sodium carboxymethylcellulose for Various Temperatures


30°C
y = -1E-05x + 0.3393
R² = 0.0436
SVR
0.45
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
0
500
1000
1500
2000
Concentration of Na-CMC (ppm)


40°C
y = 5E-06x + 0.28
R² = 0.005
SVR
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
0
500
1000
1500
2000
Concentration of Na-CMC (ppm)

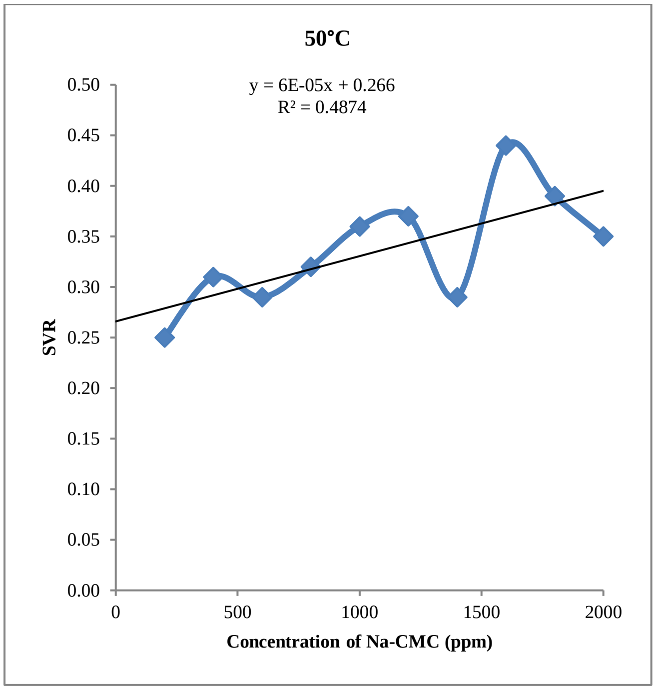


Figure 5: Graph of SVR of Aluminium Phosphate against Concentration of sodium carboxymethylcellulose for Various Temperatures

6.0 CONCLUSION

Sedimentation of aluminium phosphate precipitate formed without and with various concentrations of sodium carboxymethyl cellulose solutions was studied at different temperature settings of a sedimentation-measurement apparatus (The PreSed Meter). It was found that sedimentation of aluminium phosphate precipitate is poorly affected by concentration of sodium carboxymethyl cellulose biopolymer. The results also suggest that sodium carboxymethyl cellulose enhances precipitation of aluminium phosphate. The observations imply that sodium carboxymethyl cellulose would be good stabilizing agent for drug formulations containing aluminium phosphate.

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