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Nano tube structures by the intercalation of 5Flouro Cytosine with ZnO-layered hydroxide applied as anti fungal

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Abstract

y are indeed formed of positively charged brucite-like layer Layered metal hydroxides(LMH) materials are interesting host nano materials for controlled release type drug. They are formed of positively charged brucite –like layer, and exchangeable anions as well as water molecules are situated in the interlayer spaces(10), ZnO nanotube were synthesized by sol-gel reaction between zinc oxide and (5FlouroCytosine), Evolution of the anion was characterized by UV-VIS spectroscopy,), Fourier transform infrared (FT-IR) spectroscopy technique, X-ray diffraction (XRD), Scanning electron microscopy (SEM), Transmission Electronic Microscope (TEM) (AFM).shows a well crystalline nano tube structure was formed. Controlled release was fitted with different modules. Antimicrobial activity tests were done on the Candida fungal, the results showed that nano hybrid composite was more sensitive to the fungal inhibition than the uses of free drug in this study.

Keywords: ZnO; nano materials; antimicrobial activity; intercalations.

1.0 INTRODUCTION

Layered metal hydroxides (LMH) materials are interesting host nano materials for controlled release of anionic type drug. They are indeed formed of positively charged brucite-like layers, and exchangeable anions as well as water molecules are situated in the interlayer spaces. Therefore, organic-inorganic nanohybrids can be obtained when anionic pesticide species are introduced in the interlayer space of the LMH host structures directly during synthesis achieved by well known methods, such as .anionic exchange. ZnO has Zinc Oxide Layered hydroxides are a 2D material One of the versatile properties of layered hydroxide (ZLHs) has the ion exchange property. ZnO gust different anions such as in the intercalation of mixed solution from two anions (2,4-dichloro phenoxy acetate 4Chloro)and (1,3,5-tri chloro phenoxy acetate [2,3], used as a host octyle gallat (4) . ZnO intercalations are similar to layered double hydroxide (4,5). ZnO nanoparticles loaded coffee extract (6), of phenolic constituents(7,8), loaded active agents against food borne pathogens(9), many efforts are concentrated on the synthesis of ZnO as host materials in the synthesis of nano composites (10-16). Among those methods based on physical and chemical technologies to synthesize ZnO-organic nanoparticles complexion additives are always used to control the growth of the crystals (17-22). Addition of organic active molecules will give certain stability in the synthesized nanoparticles. So, it is necessary to find some effective methods to induce the conversion of the Zn complex into ZnO-organic nanoparticles. ZnO, fiber-like materials from the decomposition of bis(acetyl acetonat) zinc fibers at 110 °C in the presence of superheated steam was obtained (23), Octyl gallat is phenolic compound widely used in the food industries as antioxidant (24).

In this work we report the use of 5Flouro cytosine (5FC) as a template to intercalating with ZnO LHs to form stable complex" 5Flouro cytosine- ZnO (5FCZ))", nanoparticle via a simple hydrothermal synthesis (at 40 ^{o}C). The resulted composite was structural characterized and controlled release and bioactivities against some gram positive and negative bacteria's, to show the different activities between free (5FC) freely used as food preservative and 5FCZ nanoparticles without any organic materials. But, to the best of our knowledge, there is no report in the open online publications on the intercalation of 5FC in the anionic form with ZnO. The 5Flouro cytosine , one of the anti fugal molecules. This study are devoted to investigate the adsorption of the anion 5FC and its release from the interlayer of LMHs into the different aqueous media. Recently, several types of organic-inorganic nanohybrids using LMH as host structures, particularly $Zn^{2+}(OH)_{2-x}(A^{m-})_{x/m} . nH_2O$, have been described (25-26). This is due to the easy preparation, either by direct reaction with ZnO or by intercalation of zinc hydroxide salts. Indeed the intercalation of gallat, ellagic and 4-(2,4-dichlorophenoxybutyrate) anions in the interlayer space of zinc layered hydroxide (ZLH) has been achieved by hydration of zinc oxide in aqueous solutions (27-29). Several anionic UV absorbers have been also intercalated into zinc hydroxide by stirring in aqueous solution (30).

2.0 Results and discussion

2.1 Powder X-ray Diffraction

Powder X-ray diffraction patterns of the solids obtained that indicate after hydrothermal treatment at ZnO-LHs nanohybrids have been formed "Figure (1). For sample ZnO-LHs all samples have six characteristic peaks at 2 ɵ= 3.4 ; 7.4 and 17, which can be indexed to the (003), (006), and (009), planes corresponding to the basal reflections (d) the first reflection is recorded at 1.42nm, in this case the crystal parameter c = 3 (d003) = 4.3 nm approximately, the diffractions of the planes (006), (009) have d= 0.71 and 0.36nm respectively . On the other hand, the basal reflections of the ZnO fig(2) are recorded at d= 0.281, 0.259 and 0.247 nm respectively were in the same positions. The evidence for phase structure of the as-prepared sample was obtained by XRD pattern, as shown in "Fig. (2), all the diffraction peaks can be indexed to those of hexagonal ZnO. After refinement, the lattice constants, a=3.251 Å, c=5.210 Å, were obtained, which is very close to the reported value for ZnO (a=3.253 Å, c=5.209, JCPDS card, No.80-0075). Powder XRD patterns of the 5FCZ-LHs samples showed that the full width of half maximum (FWHM) value of (003)=0.43 deg., which indicated an increase of the crystallite size within the synthesis of 5FCZ-LHs. And all the sharp diffraction peaks indicate the good crystals of the obtained nanostructures.

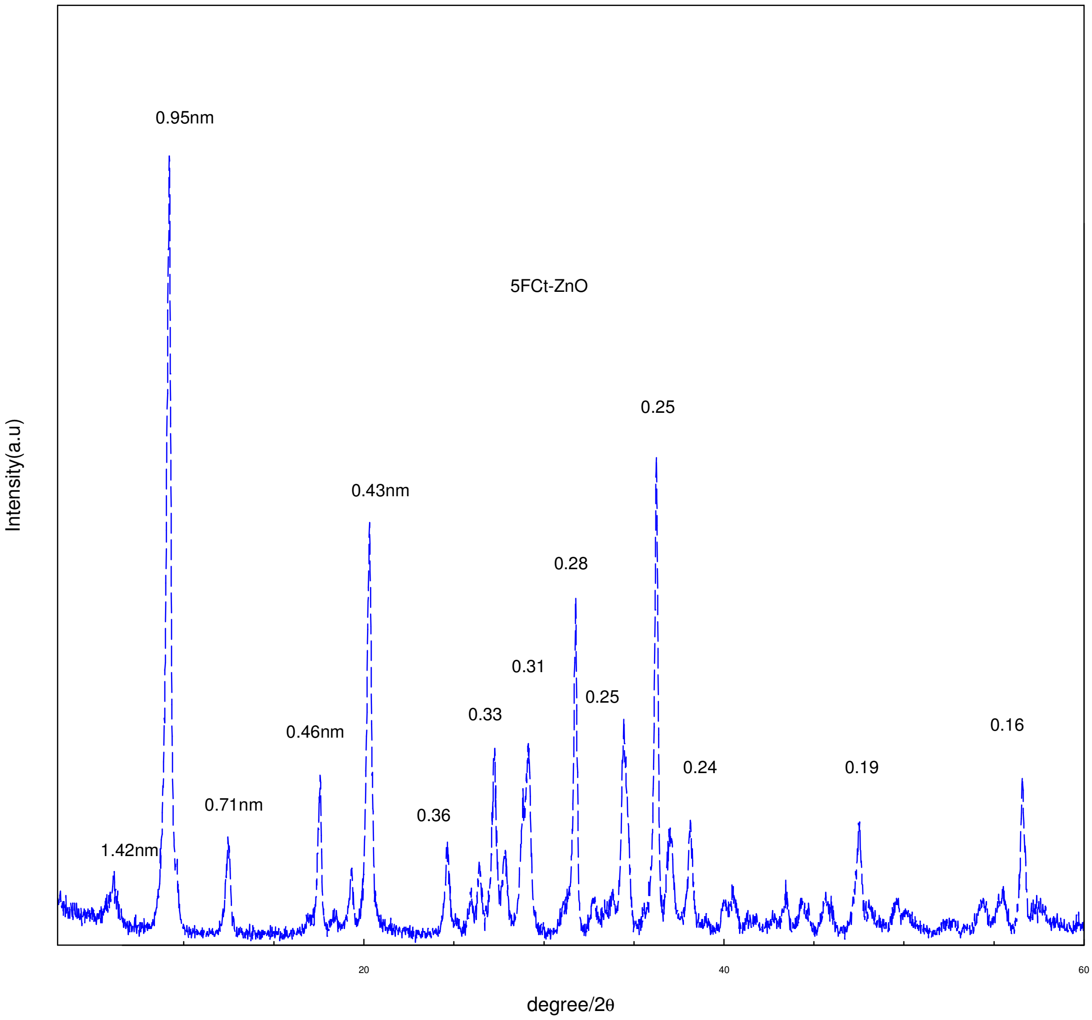
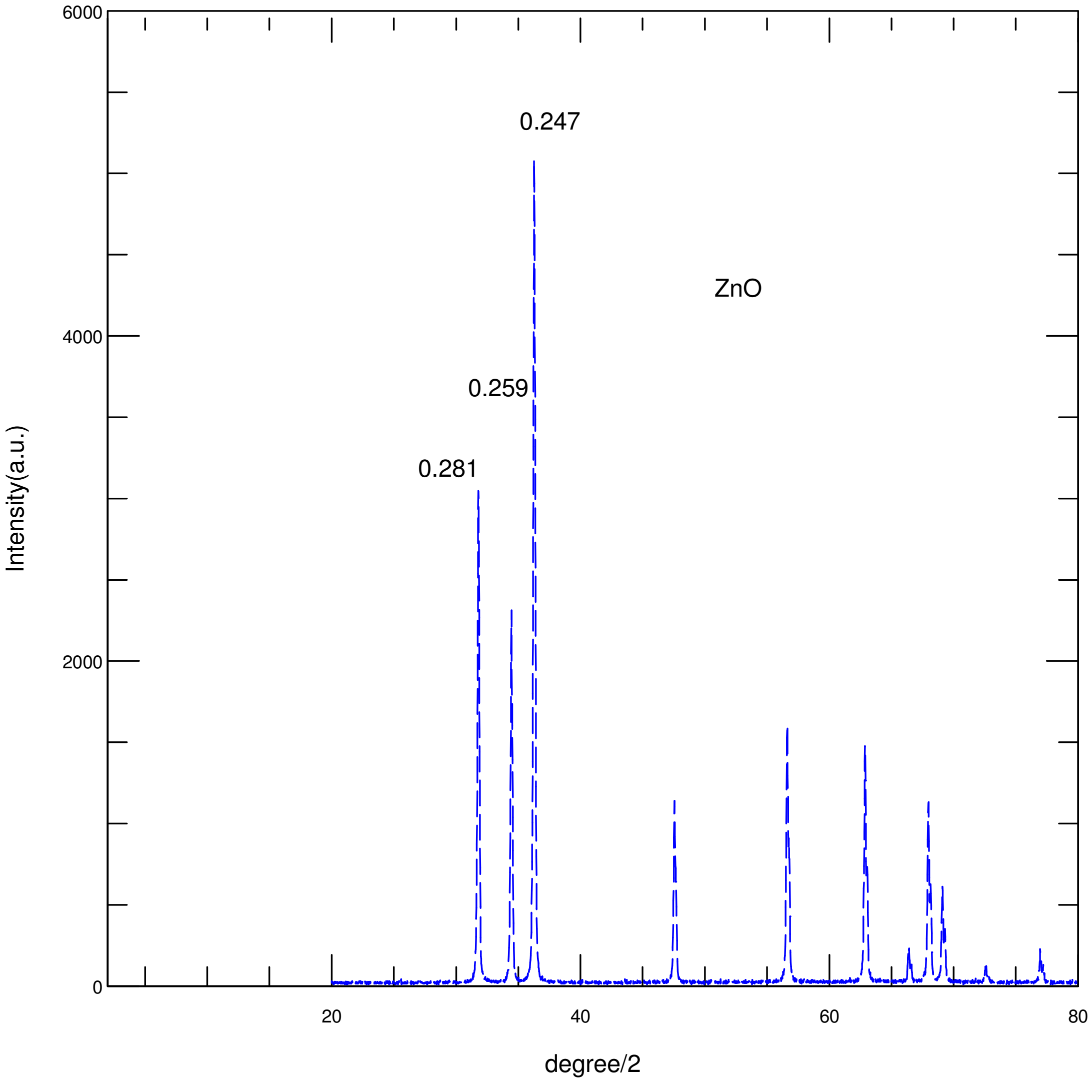


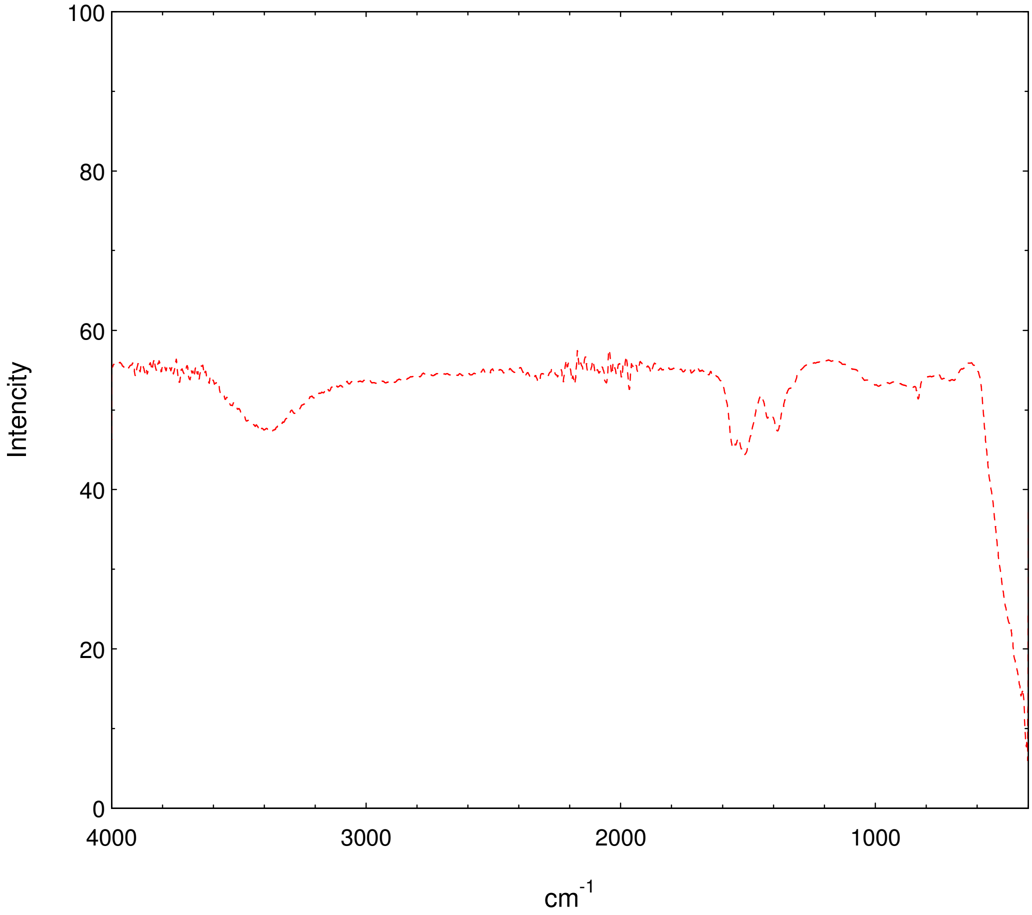
Fig (1) PXRD diffractions of 5FCZ-LHs shows the evaluations of (003),(006), and (009) planes diffractions and the characteristic peaks related to ZnO..



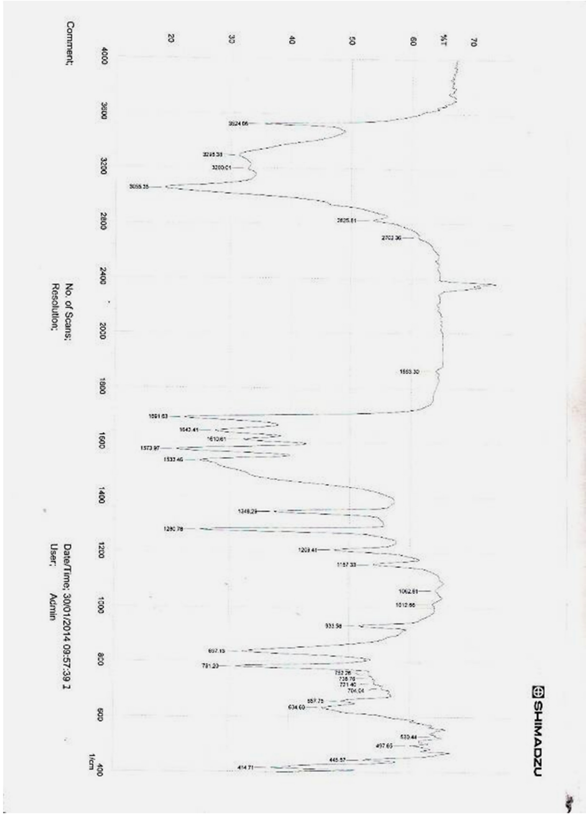
Fig(2) PXRD diffractions of ZnO shows the characteristic planes diffractions of (101).(011) and (002).

2.2 FTIR technique:

The FTIR spectrum of the hybrid is a complement of the xrd results and provide further evidence for the intercalation in addition, some absorption bands are slightly shifted due to the interaction of the anion (Fig4) and the host layer (Fig3). The typical broad absorptions bands of zinc layer hydroxide superposing with the anion hydroxide observed and the water molecules at 3524 cm^{-1} and the band due to the (C–O-H) stretching vibration of C-H stretching in the organic chain at 2825 cm^{-1}, the asymmetric and symmetric stretching of C=O appears at 1691 cm^{-1}. The bands at 1643, 1610 and 1280 cm^{-1} attributed to characteristic of the vibrations of 5FC present in the nanohybrids that is indicating the presence of 5FC molecule in the nanohybrids. In the 5FCZ nanocomposites (fig4) shows a combination spectrum of both the host ZnO and the guest anions, the bands located at 1384 cm^{-1} due to the stretching vibration of C-N in the aromatic ring. The presence of carboxylate group, COO can be deduced by the observation of bands at 1691 are due to C=O stretching , the band at 1573 are due to C-O-H bending , 783 and the band 893 cm^{-1} are symmetric and anti symmetric vibrations of the COO- group in the 5FC intercalated in the inter layers of ZnO. [23] . which are attributed to asymmetric and symmetric vibration, respectively [24 -29] whereas a band at around 1012, 1062 cm^{-1} is corresponded to (–C-0-)stretching vibration.



Fig(3) FTIR of pure Zno shows the characteristic vibrations



Fig(4) FTIR spectrum of 5FCZ-LHs shows the chractertic vibrations of5FCZ nanohybrids.

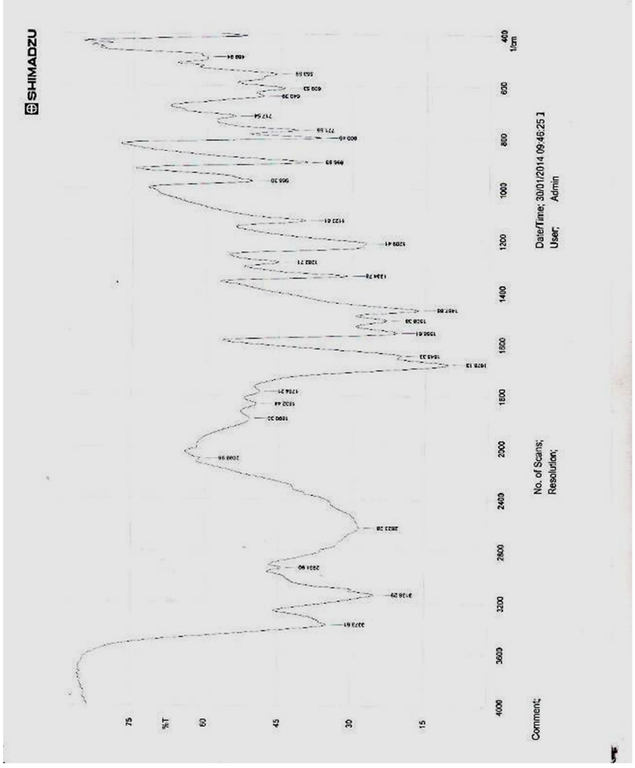


Fig.(5)FTIR spectrum of 5FC shows the chractertic vibrations

2.3 Atomic forcing microscope(AFM) study of the surface morphology:

Surface morphology studies: Atomic Force Microscopy image was recorded with scanning probe Microscopy (SPM-AA3000) instrument employing soft wear WSXM(nanotech.) the glass slides were cut to1X2cm and clean by putting them in(1:1)ethanol:deionized water solution and treated by ultrasonic(Ultrasonic FALC) instrument.

Fig. (6) Shows AFM images of the 5FC-ZnO LHs nano particles synthesized with 5FC-ZnO-LHs, with a mean size of (120.8nm). The AFM observation is in good agreement with the data obtained by PXRD technique. Fig(6A in two dimensional) and in the three dimensional (6B) shows a nano tube structures in the case of (5FCZ) average diameter of 120.8nm. This was a good indication to the intercalation done.

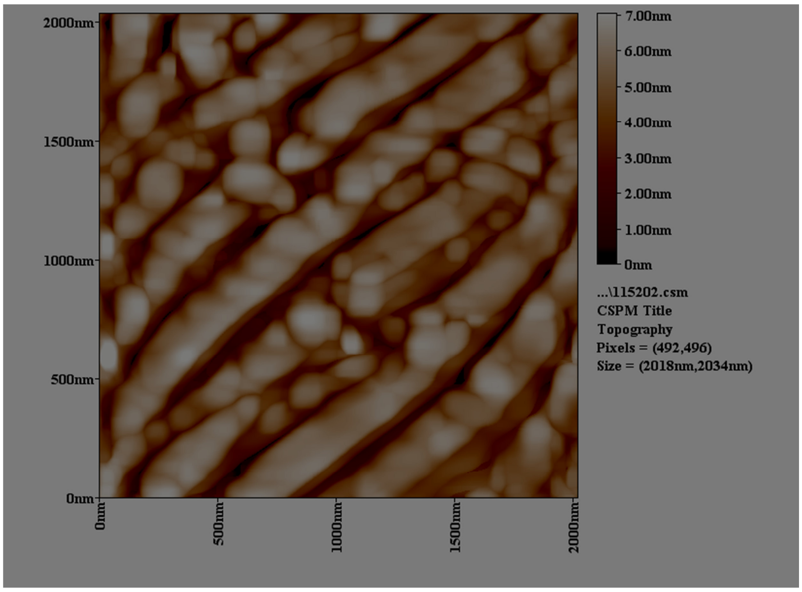


Fig .(6)A- image of AFM shows the two dimentional coordinations of the 5FCZ-LHs

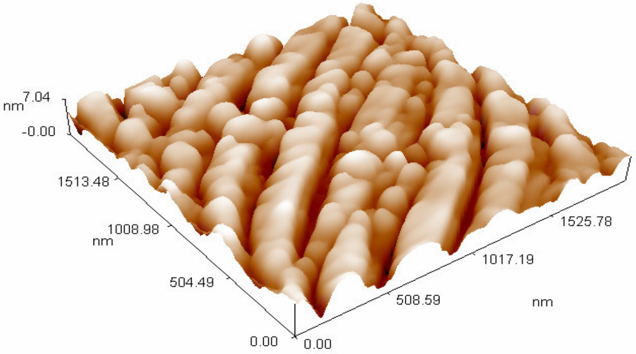
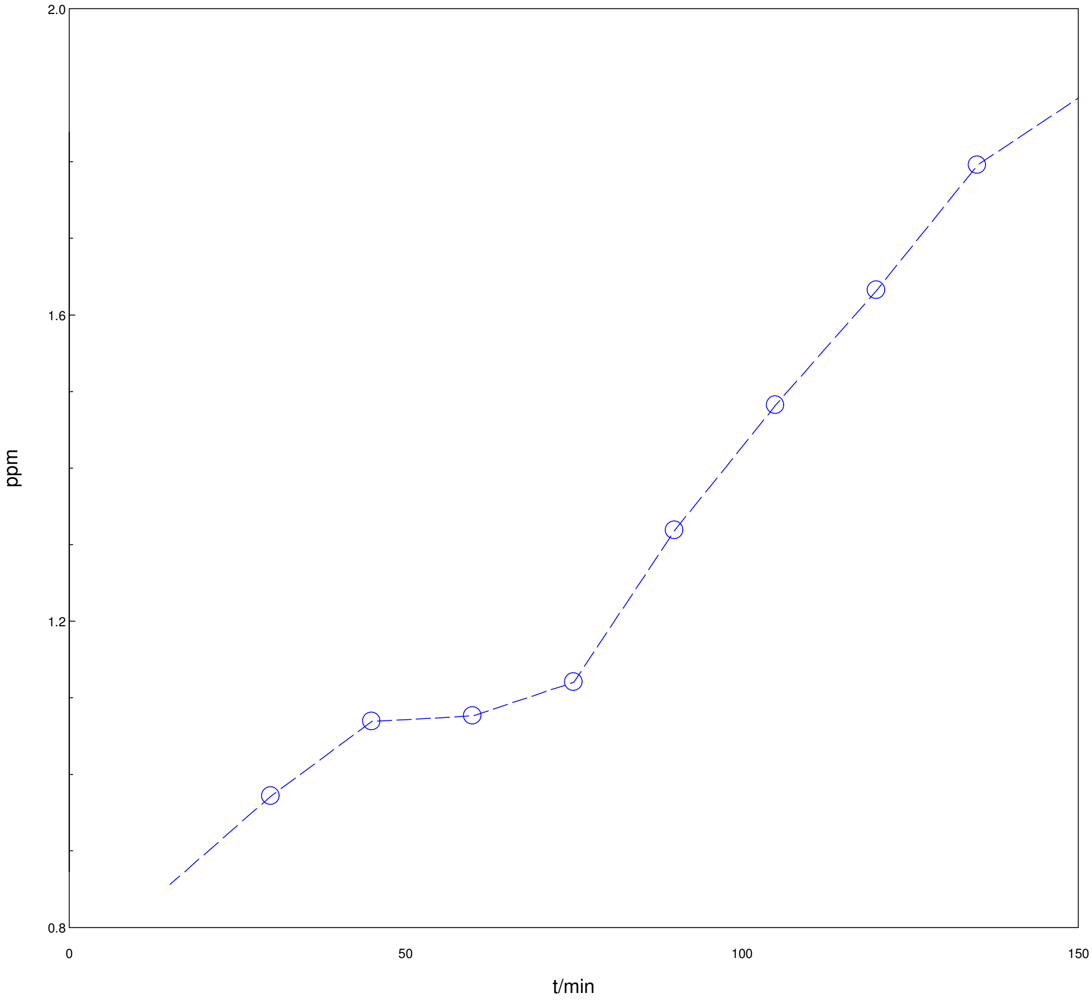


Fig.(6B) image of AFM shows the three dimentional coordinations of the 5FC-ZnO-LHs.

Avg. Diameter:120.82 nm

Diameter(nm)<	Volume(%)	Cumulation (%)	Diameter(nm)<	Volume(%)	Cumulation (%)	Diameter(nm)<	Volume(%)	Cumulation (%)
40.00	1.94	1.94	100.00	4.85	29.13	150.00	10.68	76.70
60.00	1.94	3.88	110.00	8.74	37.86	160.00	7.77	84.47
70.00	3.88	7.77	120.00	10.68	48.54	170.00	6.80	91.26
80.00	10.68	18.45	130.00	6.80	55.34	180.00	6.80	98.06
90.00	5.83	24.27	140.00	10.68	66.02	190.00	1.94	100.00

Table(1) shows the the pores distributions of granuals in the 5FC-ZnO-LHs nanohybrids.



Fig(7)The controlled release in carbonate media of 0.5M

2.4 Controlled Release of 5F-Cytosine(5FC) into Aqueous Media.

The drug release properties of 5FC from the nanohybrid interlamellae
into various aqueous media using (0.5 M of Na_2CO_3 have been conducted. "Figure (7) shows the release profiles in different aqueous solutions. The effects of various aqueous systems on the release of 5FC were evaluated according to the maximum accumulated release. It was observed that carbonate dominated the accumulated release percentage due to its affinity toward the interlayer of the layered hydroxides. As can be seen in "Figure (8), a rapid release of 5FC occurs at the initial stage, which is followed by a slower release of 5FC, 5FC is almost (97%) % replaced by CO_3^{2-}, resulting in the highest accumulated release among the media studied. The maximum release time shows that 5FC is exchange with CO_3^{2-} at (180) min. All the previous discussions were

about the 0.5M concentrations. The effect of concentration of aqueous systems on the release of 5FC was investigated according to the maximum accumulated release it was clear that the 0.5M aqueous system dominates the accumulated release percentage for all aqueous systems as shown in fig(1). It should be mentioned that the initial release rate of 5FC during the first (20) min in carbonate aqueous solution is much faster than that in the other aqueous systems. In the basic medium initial release is too slow but during the 180min the accumulation of 5FC is 60%, in the case of acidic the initial release is more than in the basic, with accumulation of 50% during 1440min. fig(8), (9),(10).

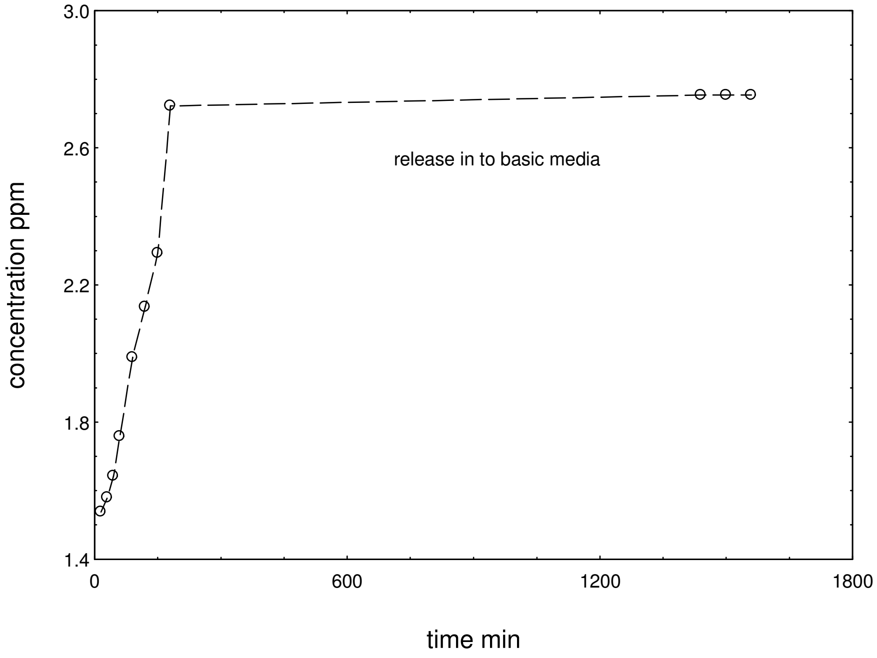


Fig (8) release of 5FC into a basic media PH=13.5

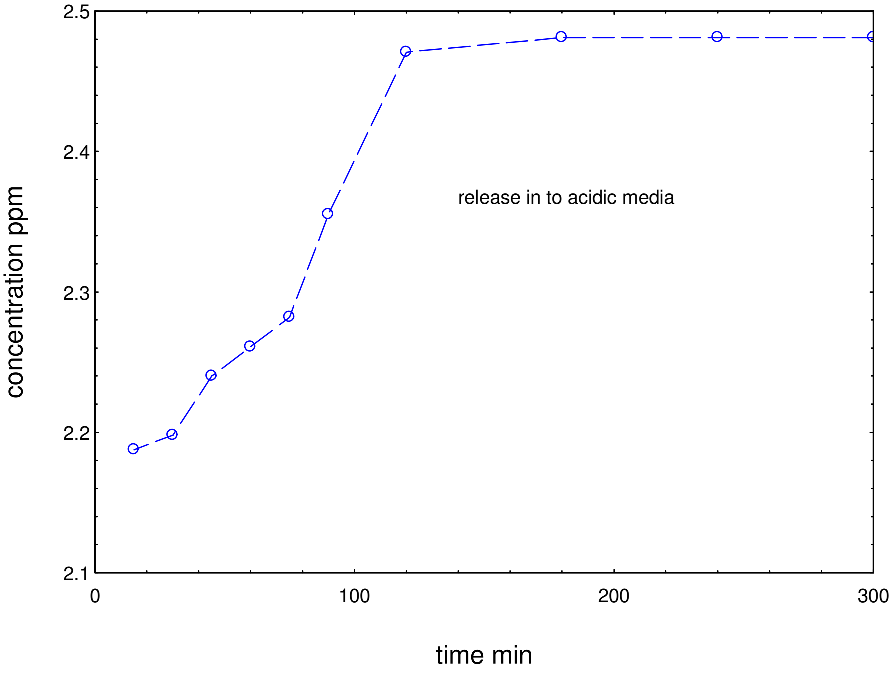


Fig (9) evolution of the release of 5FC in acidic media at PH 2

2.5 Release Kinetics

Release kinetics of 5FC has been evaluated with various models such as zeroth order, first order and pseudo-second order "Figure (7). The corresponding rate constants together with the r^2 values obtained from the fitting with zeroth , 1st, and 2nd order model. By comparing the correlation coefficient, r^2 values obtained from the fitting with those modules, it is clear that the release profile of 5FC from the nano hybrid was governed by the pseudo-second order kinetics modules in initial time till 80min then the release control change during the coming time as fig.(8) shows. The rate constant k obtained from the pseudo-second order kinetic model was more pronounced in case of 0.5 M carbonate solution, the nature of aqueous solution and the concentration of aqueous solution. Additionally, it can be seen from "Figure (9)" that the aqueous solution of 0.5M concentration can be considered more convenient to present the release process due to it could be released much more amount of 5FC intercalated .

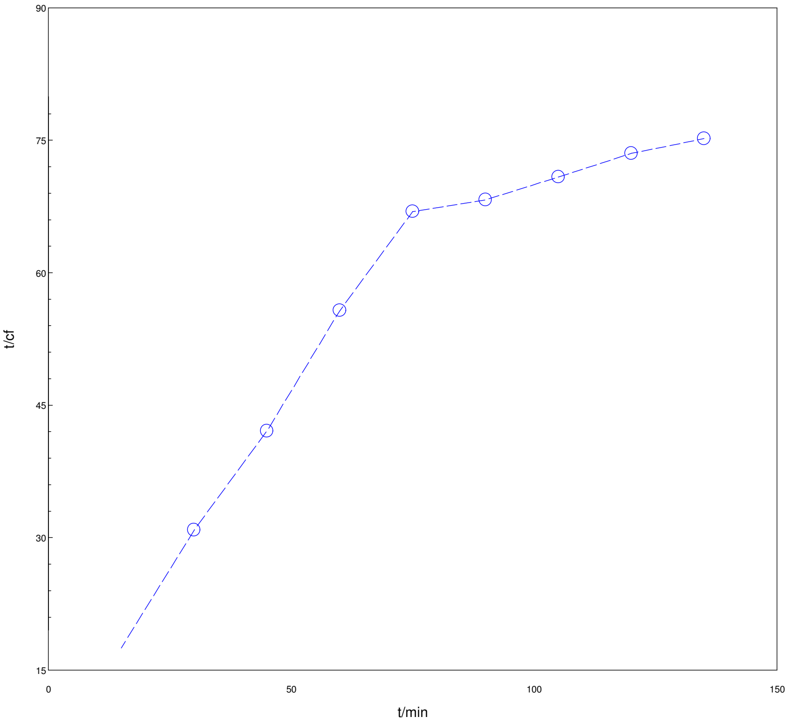
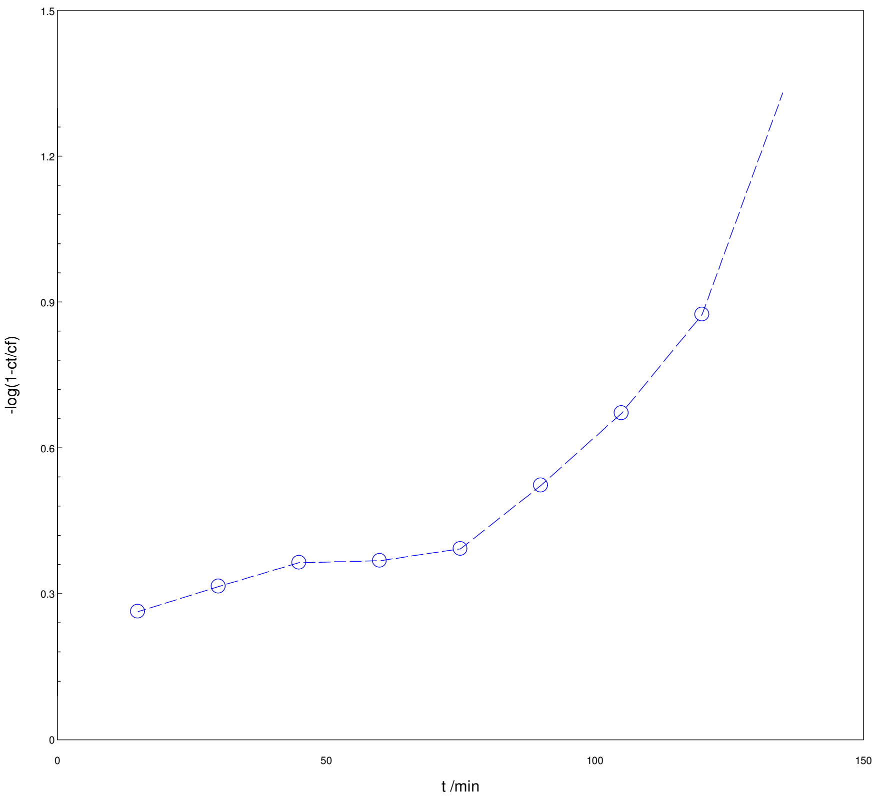


Fig (10) fitting 2nd order model to the release of 5FC in to carbonate media(0.5M).



Fig(11) fitting first order model applying on the release of 5FC from the inter layer of 5FC-ZnO-LHs into carbonate media.

2.6 Surface morphology of 5FCZ

The surface morphology was done using Hitachi S-4800 Field emission scanning Microscope in Fig. 12(a) and transmission electronic microscope TEM image of nano tube structure (Fig. 13) show that the nano tube fabricated by the sol-gel method are hallow tube and uniform in size, with tube inner diameter of 62nm, the outer diameter of twin tube about 135nm, morphology of the 5FCZ hallow tube is easily observable in Figure 12-13, Homogeneous dispersion of 5FCZ in the 5FC/ZnO matrix is seen, and indicates good compatibility between the filler and the intercalated blend matrix. The individual 5FCZ vis-ible in the image have diameters of approximately 62 nm and lengths of up to about 112mm, which are in agreement with values usually obtained by AFM and XRD studies

In Figure 15, the nano tube 5FCZ. The presence of 5FCZ was confirmed further by using D XR The results for crystal shape and size are in agreement with the TEM observations for tube structure of 5FCZ. Some aggregates could be seen at lower magnification, and may have formed during evaporation of water or may have been present in the original suspension.

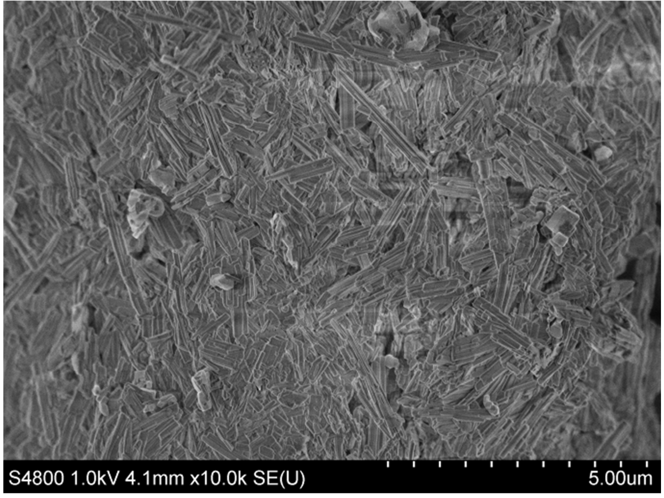


Fig. 11 SEM image of hallow tube structure of 5FCZ

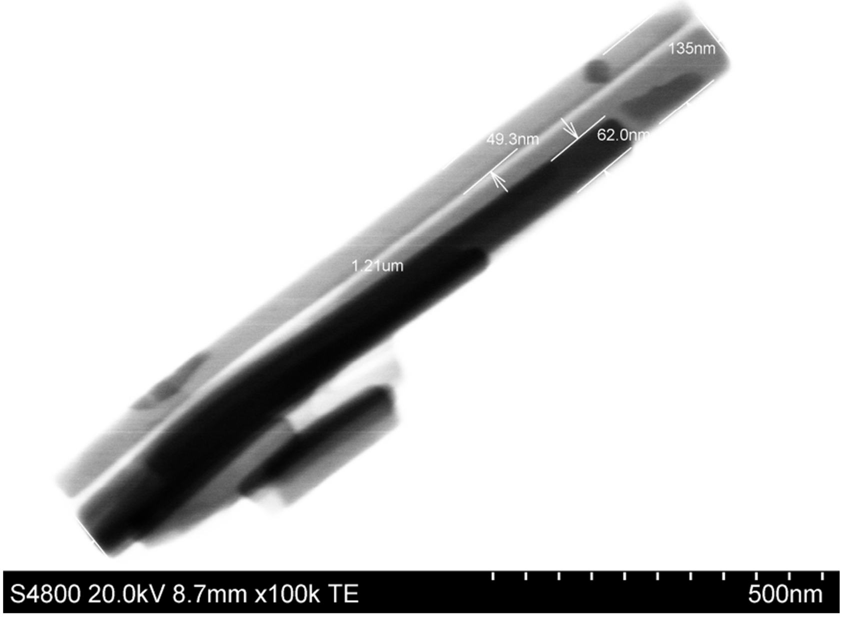


Fig.12 shows the TEM image of hallow tube structure

3.0 Conclusion

5FCZ nano tube were prepared with ZnO by sol-gel methods, and characterized using FTIR spectroscopy, X-ray diffraction, SEM,TEM spectroscopy, 5FC was intercalated into the nano tube structures. Controlled release studies showed the system of release was governed by the pseudo second order model. The 60 %. Drug release was in acidic media, and 50% in to basic media, the total release equilibrium of 5FCfrom 5FCZ was 60 % in 100 minutes. The drug release profiles showed initially rapid release, which became slower as time progressed. The best fitting of mathematical model was the pseudo-second order model.

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