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Experimentally Designed of PVC/$NiAl_2O_3$/AlF_3 Nanocomposite by Sol-Gel Method

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Abstract: In this study, we have synthesized nano Aluminum Fluoride (nAF) nanoparticles by the sol-gel method and studied the nano-sized morphology of crystals. In the other section, the PVC/$NiAl_2O_3$/AlF_3 (nPNA) nanocomposite was successfully prepared and characterized by FT-IR, and HRTEM techniques. FTIR peaks of the PVC and nPNA have been shown spherical shape of PVC and also spherical shapes nanoparticles of nPNA loaded on PVC. A solvothermal method has been successfully introduced and applied for catalyst efficiency. This nanocomposite was used for the removal of Congo red dye. For this purpose, the morphology and the structure of crystals have been changed by modification on precursor gel. Meanwhile, precursor gel preparation and the interaction on the nano-sized area have been studied. This study exhibited that PVC/$NiAl_2O_3$/AlF_3 (nPNA) nanocomposite is an effective catalyst for the synthesis of some organic derivatives. The results show that the as-prepared nanocomposite is an efficient catalyst and that PVC/$NiAl_2O_4$/AlF_3 nanocomposite can be used in the next-generation of some organic reactions and faster production of various materials.

Keywords: Nano Catalytic activity, Nano-Size, Morphology, Sol-gel, TEM

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Research paper

1 INTRODUCTION

Synthesis of the sol-gel method has been so widely developed that nano-metal oxides, either pure or organic-inorganic hybrid materials, are applied and available now [1-8]. One of the most common applications of Al_2O_3 and $nAlF_3$ (and their derivatives) is as a catalyst support. Many physical properties of the catalyst support, such as temperature, pH, aging time, raw materials and preparation technique, have a major impact on the performance of the catalyst in sol-gel processes [9-11]. Ono et al. [12] were the first to report this procedure and they managed to study the effectiveness of the pH and number of pH-swing frequencies on the Aluminium Oxide formation. Maity et al. [13] prepared the catalyst supported by alumina via various techniques, such as pH-swing, and determined that this method improved the pore size distribution (PSD). [14].

Aluminum fluoride has a number of current and potential applications as an additive to the molten electrolyte of the aluminum production cell in order to lower the melting point and increase the electrical conductivity. Moreover, among metal fluoride catalysts and catalyst supports for halogen exchange reactions, AlF_3 is one of the most important catalysts [15-18]. We have recently developed a reduction method of converting Ag nanospheres into nanorods [19], nanoplates [20], their antibacterial activity [21-22], an improved and easy synthetic route for silver nanoparticles in poly (diallyldimethylammonium chloride) (PDDA) [23], synthesis of Gold/HPC hybrid nanocomposite [24], preparation of Ag/ZnO nanocomposite [25-26] and comparison nanosilver particles and nanosilver plates for the oxidation of ascorbic acid [27].

As part of our efforts to investigate the usefulness of nanocomposite catalysts for the synthesis of organic and heterocyclic compounds [28-30], we report an efficient process for the synthesis of arylidende barbituric acid derivatives from the cyclo-condensation reaction between barbituric acid, various aldehydes by using 1 mol% ammonia mediated at room temperature.

In the present paper, nPNA composite was successfully prepared (from nAF) and then, catalytic activity of nPNA has been considered in the synthesis of two organic derivatives.

2 EXPERIMENTAL

2.1. Characterization

Transmission electron microscopy (TEM, Model Hitachi 4500-M) was used to study morphology and particle size. It was performed using Bruker AXS diffractometer D8 ADVANCE with Cu-Kα radiation in the range $2\theta=10^{o}–80^{o}$. FT-IR spectra of the samples were done using a spectrometer (Model Perkin–Elmer) in the wavelength range 400-4000 cm^{-1}.

2.2. Materials and Methods

Baribituric acid (BBA) and various aldehydes were purchased from Aldrich company and used as received without further purification. Solution pH ranged between 7 and 10. The resulting gel nAF was kept at 50 ^{o}C for 20 h. Then, obtained gel were filtered and rinsed with a NH_3.

2.3. Catalytic Activity

The catalytic activity of the $PVC/NiAl_2O_4/AlF_3$ (nPNA) was shown as below optimization procedure. BBA, nPNA, and methanolic solution of various benzaldehydes were added to hot water and stirred until the color precipitate of the products has been prepared. The precipitate of solid was filtered. A solvothermal method has been successfully introduced and applied for catalyst efficiency. The ABBA derivatives have been characterized by FT-IR spectroscopy.

2.4. Result and Discussion

During the preparation, the chemical reactions (reaction 1) that occurred are highlighted as changes in the various crystalline phases observed in the XRD pattern. It can therefore be said that Nano Aluminum fluoride (nAF) was utilized in this sample as precipitant [15-16].

$$\underline{\textit{alkaline regions}}$$

$$Al(CH_3COO)_3 + Al_2O_2F_4 + 5HF \xrightarrow{\underline{\textit{In glass reactor}}} 3nanoAlF_3 + 3\,CH_3COOH + H_2 + O_2 \quad (NH_3\ Solvent) \quad (1)$$

(nAF)

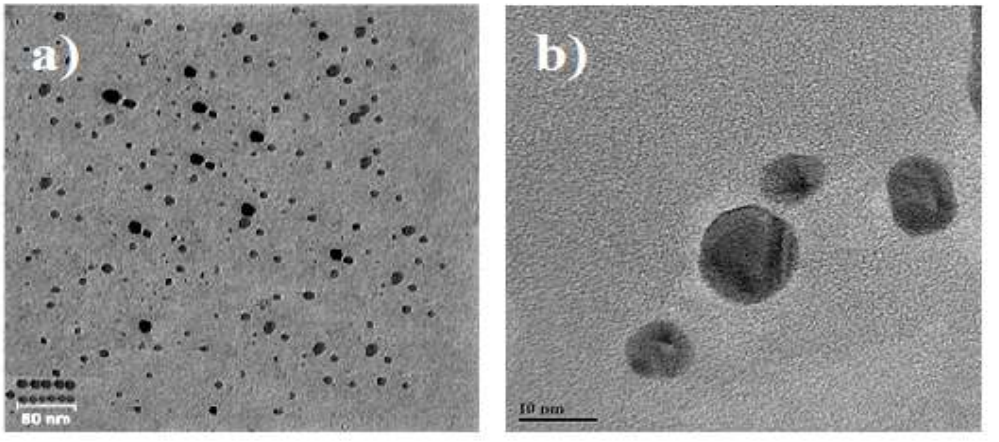

Fig. 2 HR TEM images of the $nAlF_3$ sample.

Figure 1 (a, b) represents the HRTEM direct image of $nAlF_3$ sample. The size of the particle is spherical in shape and has a size in the range of 10-18 nm which was a good similarity to the size obtained in the optical model. TEM images were prepared to determine the produced nanoparticles' size, morphology, and uniformity. The results showed the production of hexagonal $nAlF_3$ nanoparticles, and it matches the XRD patterns.

The FTIR spectrum of $nAlF_3$ is shown in "Fig. 2". In this sample, there is not broad absorption bands at 3300-3500 cm^{-1} associated with O-H stretching of adsorbed water. The sharp peak at 1645 cm^{-1} belongs to the bending vibration of H-OH in (a small amount of water) H_2O molecules. The band at 1075 cm^{-1} is related to symmetrical Al-F bending modes. The adsorption peak at 1635 cm^{-1} belongs to the bending vibration of O-H group in the adsorbed water [31-35].

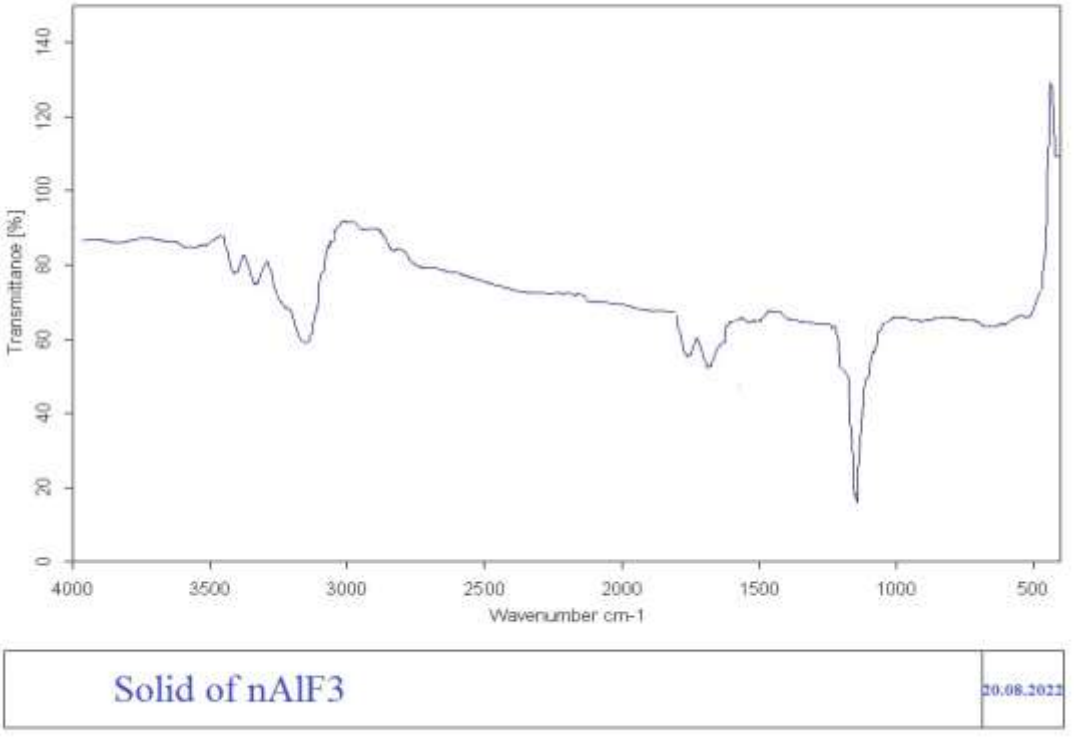

Fig. 2 FTIR spectrum of the nano AlF_3.

PVC/$NiAl_2O_4$/AlF_3 (nPNA) nanocomposite was prepared using the reaction of PVC, $AlCl_3$, and $NiCl_2$ in the presence of NaOH. The chemical structure of PVC and PVC/$NiAl_2O_4$/AlF_3 was studied by FT-IR spectroscopy and the results are presented in "Fig. 3".

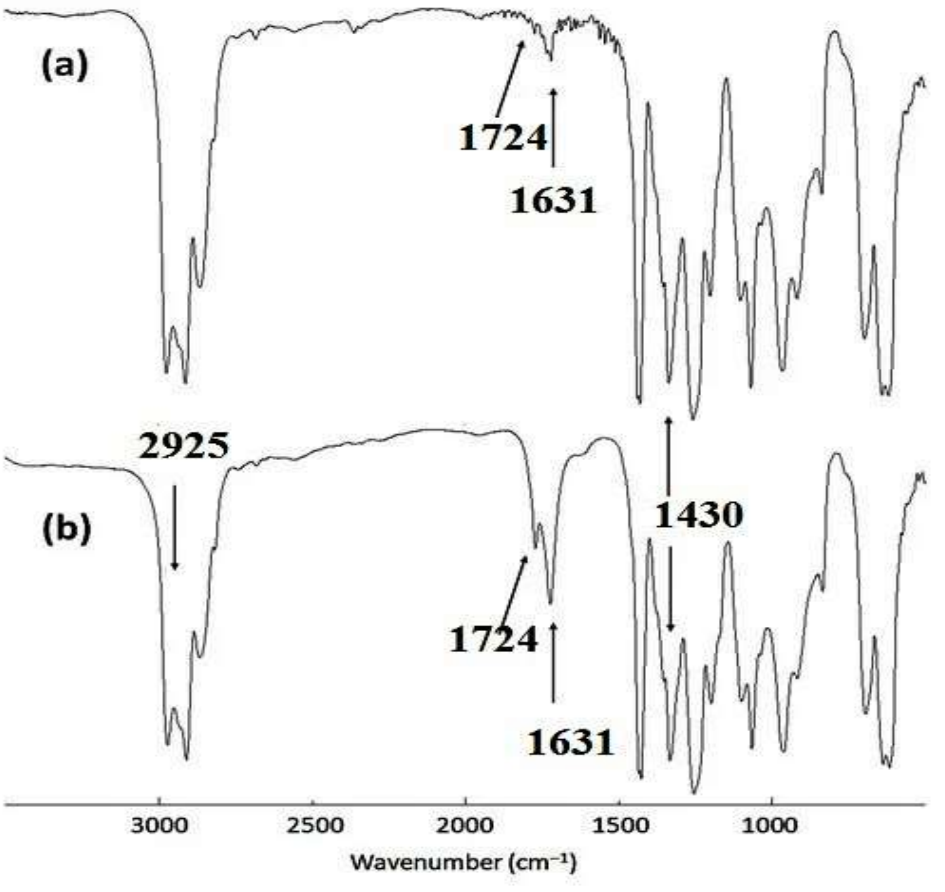

Fig. 3 FT-IR spectra of: (a): PVC, and (b): PVC/$NiAl_2O_4$/AlF_3 composite.

The absorption bands in the infrared spectrum of polyvinyl chloride at 503, and 650 cm^{-1} are assigned to the amorphous absorption band of C–Cl stretching, and C–Cl crystalline absorption band, respectively. The υ(C–C) stretching vibrations band has been showed around 845. The bands at 1100 and 1175 cm-1 correspond to perpendicular chain stretch and parallel chain stretch [36]. The absorption band at 1359 cm^{-1} corresponds to CH_2 wag and the other band at 1430 cm^{-1} is due to the bending mode of CH_2. The absorption band at 1631 cm^{-1} is assigned to the carbon=carbon double bond stretching vibration for conjugated bonds, or either aromatic or aliphatic or both. The band at 1724 cm^{-1} is probably from the carbonyl stretching vibration. The wideband at 2925 cm^{-1} corresponds to the CH_2 asymmetric stretching mode, the peak broadening is due to the intermolecular and intra-molecular hydrogen bonds [37]. However, the small shift in 650, 845, and 1724 cm^{-1} band positions confirmed the formation of PVC/$NiAl_2O_4$/AlF_3 nanocomposite in the PVC polymer matrix. The peaks at 689, 614, and 427 cm-1 concerning Metal-O (Al-O and Ni-O) stretching [38-40].

2.5. Catalytic Activity of Arylidene Barbituric Acid Derivatives

The color of barbituric acid is white, while the color of FCG-1 (R=COOH) is pale-brown and FCG-2 (R=NH_2) is orange. The melting point of FCG-1 is about 293 ºC and is more than the melting point of FCG-2 (223 ºC), due to the COOH group that causes increasing hydrogen bond strength in FCG-2.

"Table 1" shows the conditions for the synthesis of ABBA, under various derivatives. The yield of the products is very low in acetonitrile, dichloromethane and is a medium in methanol and water, while the yield increased to ≥ 97% using a water-methanol (l: 1 v/v) mixture. Preparation of FCG-2 (40 sec) is faster than FCG-1 (210 sec) because NH_2 is an electron donor while COOH is an electron acceptor. The results

presented in this article are a continuation of previous works [41]. For example, the yield and time for the preparation of FCG-2 (entry 2 in "Table 1") are higher than this compound prepared by Rajput and Kaur [41]. By increasing the catalyst, the yield of FCG-1 and FCG-2 increased from 82 to 92 and 90 to 97%. Also, by changing the solvent from methanol to methanol-water, the yield of FCG-1 and FCG-2 increased. In the absence of the PVC/$NiAl_2O_4$/AlF_3, only a trace (< 50%) of FCG-1 and FCG-2 was obtained which is in agreement with previous reports [42]. This reaction has been monitored by the formation of colored precipitate product and is simple, and clean. The purified compounds have been recrystallized and characterized by FT-IR and UV-vis spectroscopies. Until now, there have been many reports on the preparation of arylidene barbituric acid by various catalysts [41], [43-46].

Table 1 Optimization of the preparation of arylidene barbituric acid derivatives

Entry	R	Time (min)	Yield (%)	Catalyst amount (g)	Solvent
		Arylidene	Barbituric	Acid Derivatives	
1	COOH	4	92	0.03	Methanol-Water (1: 1)
2	NH_2	1	97	0.03	Methanol-Water (1: 1)
3	COOH	6	82	0.02	Methanol-Water (1: 1)
4	NH_2	2	90	0.02	Methanol-Water (1: 1)
5	COOH	17	80	0.03	Methanol
6	NH_2	6	89	0.03	Methanol
7	COOH	27	77	0.02	Methanol
8	NH_2	9	80	0.02	Methanol

3 CONCLUSIONS

In conclusion, a multi-step precipitation method with the sol-gel technique was implemented. The TEM images and FT-IR spectroscopy indicated that the prepared nAlF_3 had low crystallinity and images showed that particle size is nanometer scale. Using the TEM images, the crystal size of the sample was 19.5 nm. The FT-IR spectroscopy confirmed the formation of nAlF_3 and PVC/$NiAl_2O_4$/AlF_3 nanocomposite in PVC polymer matrix. nPNA has been prepared and used as a new catalyst for the preparation of ABBA. The result confirmed that the nPNA composite is a good catalyst. This catalyst produces the FCG-1 and FCG-2 compounds in a very short time and with high yields. FTIR peaks of the PVC and nPNA have been shown spherical shape of PVC and also spherical shapes of nanoparticles of nPNA loaded on PVC. The main information from this study can be fixed or altered to optimize responses at more levels for future studies. These results show that PVC/$NiAl_2O_4$/AlF_3 nanocomposite can be used in next-generation of some organic reactions and faster production of various materials.

4 ACKNOWLEDGMENTS

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5 CONFLICTS OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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