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Original Research Article

Microwave synthesis of AlO(OH)) and $Mg(OH)_2$ nanoparticles and evaluation of their antifungal activity

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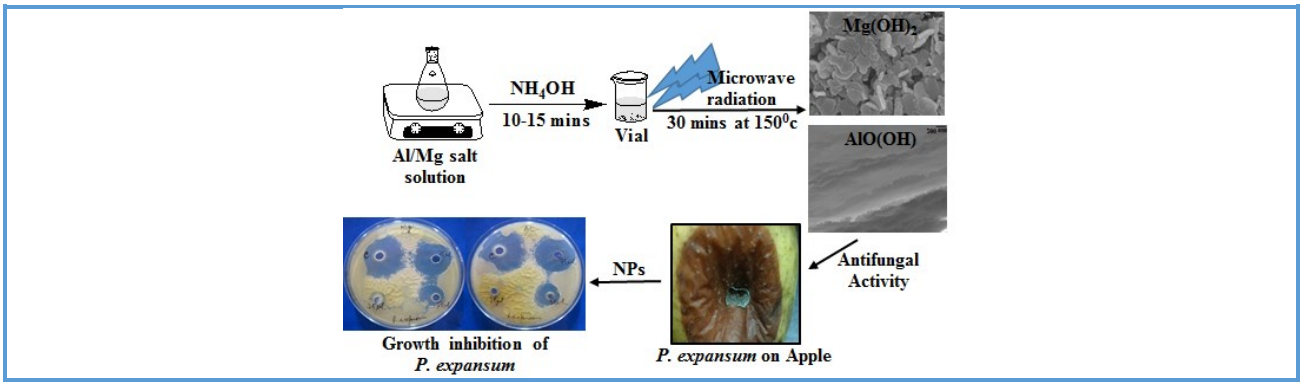
ABSTRACT

Microwave heating has been adopted as green approach for the synthesis of bohmite (AlO(OH)) and brucite ($Mg(OH)_2$) nanoparticles (NPs) for antifungal activity. The synthesis of AlO(OH) and $Mg(OH)_2$ NPs were carried out at 150 °C and the resulting NPs have an average diameter of 10-20 nm. The $Mg(OH)_2$ and AlO(OH) have trigonal and orthorhombic crystal structure, respectively. The antifungal activity of the synthesized NPs was assessed using the *Penicillium Expansum* (*P. expansum)* through agar well diffusion method. The $Mg(OH)_2$ and AlO(OH) revealed comparable significant antifungal activities towards *P. expansum*. About 79% and 74% reduction in the growth of the fungus was obtained respectively of AlO(OH) and $Mg(OH)_2$ as compared to the standard control haxahit. Nanomaterials bind on the surface of the fungi thereby preventing the normal activity of fungi and inhibit their growth, ultimately kill them. Reported NPs have significant potential to replace expensive nanomaterials in the field of antimicrobial studies.

Graphical Abstract



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Introduction

Decrease in productivity of food and transfer of infections from food to humans due to chronic infections caused by microbes like fungus, bacteria etc. needs imediate attention. Antibiotics have been the preferred treatment for such infections however these microbes often developed resistance against the antibiotics [1]. About 70% of bacterial infections no longer respond to the antibacterial medications that are designed to cure them and some few were reported for fungus too [2]. Fungus like *Candida* and *Aspergillus* have been reported to develop resistant to fluconazole [3, 4]. As an alternative to antibiotics, many scientist and reasearchers are exploring nanoparticles (NPs) to prevent the growth of pathogenic microbes or kill these microbes. Use of NPs has less chance of promoting resistance in microbes than the antibiotics as the action of NPs against growth and development of microbes is surface dependent [5, 6]. Therefore, attention has been focused on new and exciting NPs based materials with antimicrobial activity [7]. NPs of Ag, Au, Pt, Ti, Si, Cu their alloy and oxides have been extensively used as antimicrobial agent [8, 9]. Their potencies against microbes have been well known and applied in the field of nanobiotechnology for past few years. However, from economical point of view, used of these expensive nanomaterials for treatment of infections is not feasible. On the other hand, oxides of Zn NPs have been reported to have effective action against *P. expansum* [10]. Sol–gel synthesized $Mg(OH)_2$, $Ca(OH)_2$, and mixtures of $Mg(OH)_2$ and $Ca(OH)_2$ NPs have showed potential inhibition efficiency against *A. niger* and *P. oxalicum* [11]. *Cymbopogon citrates* leaf extract synthesis of Al_2O_3 NPs was reported by Mohammad and his group in 2016 [12]. Minimum inhibitory concentrations (MIC) of Al_2O_3 NPs against *Candida spp* is 283.3±84.9 to 1200±282.8 mg mL^{-1}, minimum fungicidal concentration (MFC) in the range of 766.6±205.4 to 1866.6±188.5 mg mL^{-1} against the tested *Candida spp* [12]. Dong et.al has reported used of $Mg(OH)_2$ synthesized by electrolytic method as antibacterial activity against *Escherichia coli* (E. coli) and *Burkholderia phytofirmans* in liquid culture and in paper sheets [13]. *Carica papaya* L. leaf extract synthesis of MgO was reported and used for studying soil borne anti-fungal phythogen activity. MgO (0.5 gm/mL) nano particle suppressed fungal invasion through root irrigation with control efficiency of 50.20 and 62.10%, respectively for tobacco black shank and black root rot disease [14]. All these studies suggested that, oxides and hydroxides NPs play significant role in controlling the growth of microbes. We are looking forward for cost-effective and benign NPs for controlling growth and infection of microbes.

In this direction, the present work proposed the used of green synthesized $Mg(OH)_2$ and AlO(OH) NPs for controlling growth of the fungus *Penicillium expansum (P. expansum)* which is one of the oldest and commom *Penicillium* that causes spoilage of pome fruit/blue mold [15]. The $Mg(OH)_2$ and AlO(OH)NPs were fabricated through a simple and environment friendly microwave assisted route at 150 °C for 30 mins in aqueous medium. The microwave (MW) synthesis is considered to be a green approach which enable homogeneous heating using electromagnetic wave (with frequencies 300 MHz–300 GHz) and easy nucleation of NPs as compared to conventional method [16, 17]. Short thermal induction period, selective formation of morphology, superheating of solvents, and localized high temperatures make MW synthesis suitable method for production of metal/metal/alloy particles in nano range [18]. Moreover, it minimize the cost of the process by

and the waste disposal problems [19]. The *P.expansum* usually causes infections on apples and pears by producing mycotoxin patulin, a neurotoxin that can enter the food supply via apples and products of apple like such as juice and cider [20]. In India about 75% of country's apple production comes from Jammu & Kashmir, where 48% of the area is covered under apple as per the horticulture census 2016-17 [21]. In most of the farms, apples are hand-picked increasing the chance of exposure to this fungus. Considering such possible exposure, control of *P. expansum* is vitally important.

Experimental

Materials and methods

Aluminum nitrate nonahydrate, magnesium nitrate hexahydrate and ammonium hydroxide were obtained from Sigma Aldrich pvt. Bangalore, India. Throughout the synthesis process double distilled water was used.

Synthesis of AlO(OH) and $Mg(OH)_2$

1 gr of aluminium and magnesium nitrate were dissolved in 10 mL of distilled water and ammonium hydroxide solution (500 µL) was introduced under constant stirring for 10-15 min. This ammoniated solution was transferred into the vial of Anton Paar Microwave Synthesis Reactor-300 for 30 min at 150 °C. Allowed to carry out the reaction for 30 min.White color powder precipitate out from the solution indicating the formation of AlO(OH) and $Mg(OH)_2$. These powders were collected by centrifuging for 10-15 mins at 5000 rpm and washed thrice with ethanol followed by distilled water. To study the antifungal activity, these powders were redispered in 10 mL of distilled water by sonicating for 30 min. For characterization purposed, powder collected were dried at 80 °C for 6 h.

Results and Discussion

Formation of $Mg(OH)_2$ and AlO(OH) was established from powder X-ray diffraction pattern (PXRD) recorded using D8-Eco Advance Bruker. PXRD pattern of fabricated particles is given in Figure 1. $Mg(OH)_2$ is in trigonal phase having prominent peaks (Figure1**a**) at 18.4^0,32.8^0, 37.8^0, 50.8^0, 58.5^0, 62^0, 68.1^0 and 71.9^0 highlighting the (001), (100), (101), (102), (110), (111), (103) and (201) planes (powder diffraction file number 96-100-0055)[22]. While AlO(OH) have diffraction peaks (Figure 1**b**) at 14.5^0, 28.23^0, 38.5^0, 45.7^0, 49.4^0, 51.68^0, 55.22^0, 60.34^0, 64.8^0, 67.19^0 and 72.19^0 which can be attributed to (020), (120), (031), (131), (200), (220), (151), (080), (231), (002) and (251) plane of orthorhombic crystal structure as per powder diffraction file number 96-901-5089 [23]. The Scanning electron microscope (SEM) images of the fabricated NPs were recorded using Hitachi 3600 SEM instrument, and image highlights the uneven or irregular surface of the NPs exposing active surfaces. Elemental composition of the nanosystem was study using energy dispersive X-ray analysis (EDAX) intrument Thermo Scientific Ultradry 5225 Verona Rd Madison. Nanomaterials of $Mg(OH)_2$ and AlO(OH) strictly consist of Mg, O and Al, O as given in Figure 2**c** and 2**d**.

Field emission scanning electron microscope (FESEM) images of synthesized nanomaterials are displayed in Figure 3**a**, **b**, **c** and **d**. Figure 3**a**, **b**, **c** and **d** highlight the nanoscopic nature of $Mg(OH)_2$ and AlO(OH) respectively. Due to small size and high surface energy, particles seem to agglomerate to give flake like structure, as given in Figure 3.

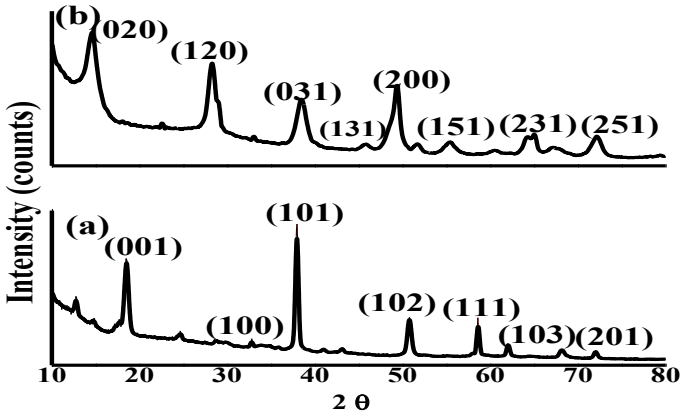


Figure 1. PXRD of (a) $Mg(OH)_2$ and (b) AlO(OH)

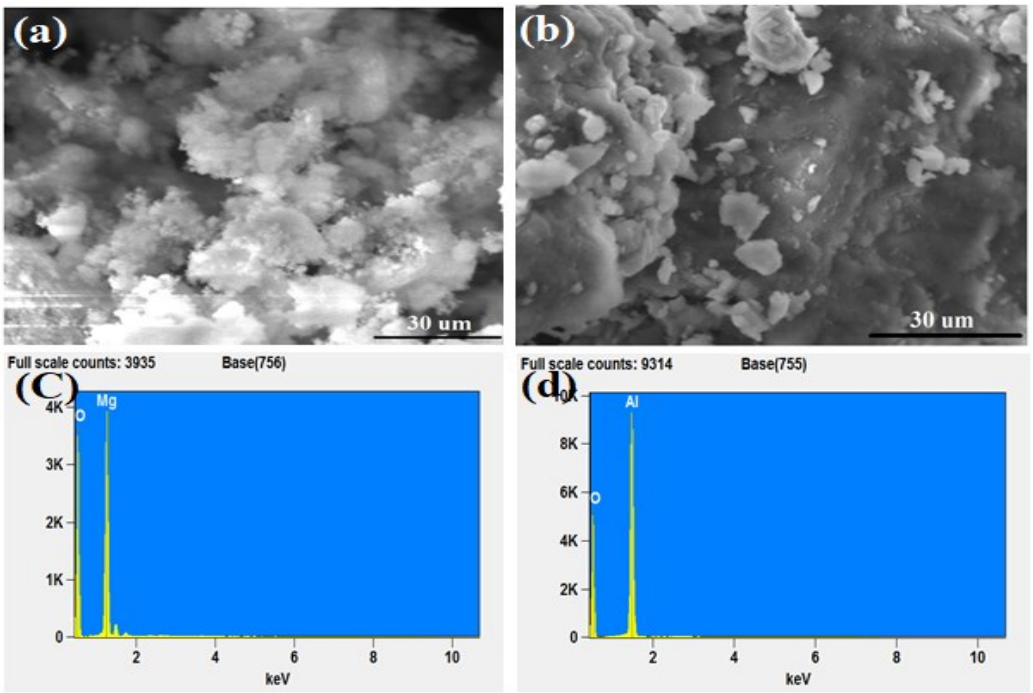
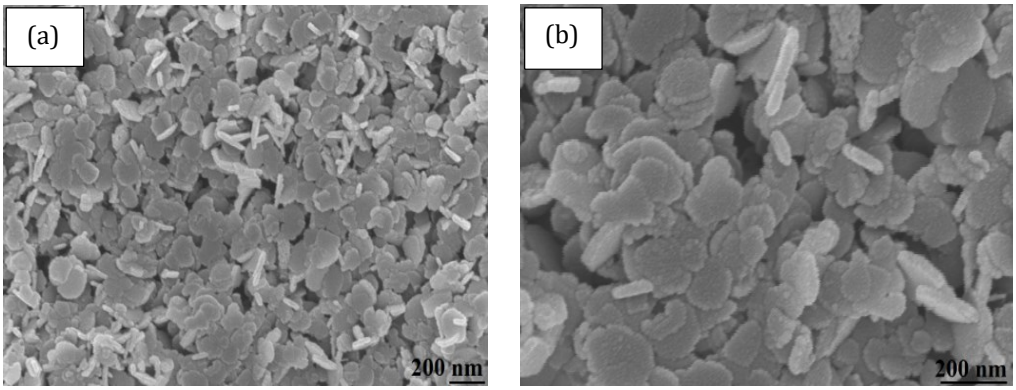


Figure 2. SEM image of (a) $Mg(OH)_2$,(b) AlO(OH) and EDAX of (c) $Mg(OH)_2$ and (d) AlO(OH)



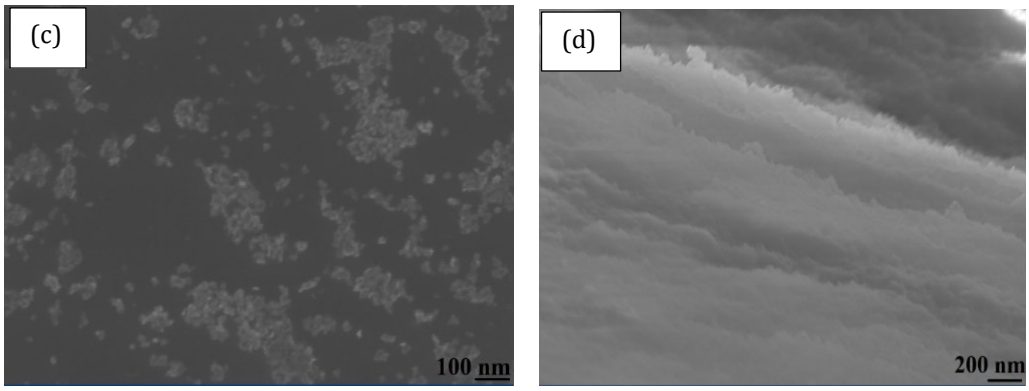


Figure 3. FESEM image of $Mg(OH)_2$ (a and b) and AlO(OH) (c and d)

The average diameter of both the NPs is in range of 10-20 nm (determined using Image J).

Infrared (IR) spectra of fabricated $Mg(OH)_2$ and AlO(OH) is given in Figure 4. $Mg(OH)_2$ show stretching at 3702 cm^{-1}, 1365 cm^{-1}, 821 cm^{-1} which can be attributed to O-H, CO_3^{2-} and Mg-O-Mg stretching [24]. Vibrations at 465 cm^{-1}, 481 cm^{-1} and 738 cm^{-1} can be attributed to AlO_6, bending of H-bonds in the octahedral structure of boehmite OH-Al=O was observed at 1075 cm^{-1} [25], peak at 1317 cm^{-1} can be attributed to nitrate peak which may have interfered from the precursor.

Peaks at 3080 cm^{-1} and 3286 cm^{-1} are of interlayer OH symmetric and anti-symmetric stretching vibrarions in boehmite [26].

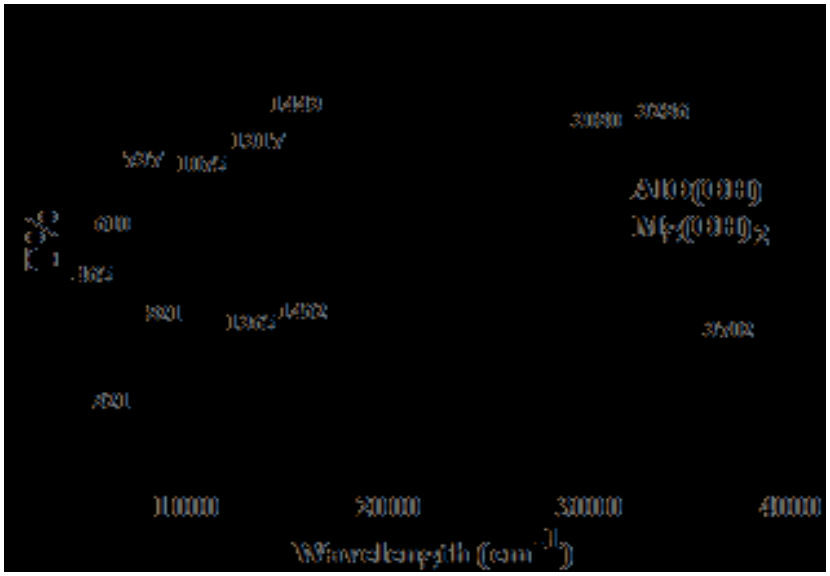


Figure 4. IR spectra of $Mg(OH)_2$ and AlO(OH)

Figure 5**a** and **b** displays the antibacterial activity of the fabricated NPs. Effect of synthesized materials on *P. expansum* was studied using agar well diffusion method. The 7 days old fungal cultures grown on potato dextrose medium were used to monitor the antifungal activity of selected NPs. A sample of 0.03 mL of inoculate fungal pathogen were inoculated in 30 mL of molten saboured dextrose agar (SDA) medium in culture tubes.

The homogenous SDA with fungal was poured into 90 mm Petri plates. After solidifying the culture tubes in laminar airflow chamber, wells were made on the agar plate using 5 mm standard cork borer. The Hexahit 0.1 mg/mL (20 µL/disc) was used as standard and 0.10 gm/mL nanomaterial of same concentration were added to respective wells. Incubation was allowed to take place for 2 days at 25±2 °C. The effect of the synthesized NPs against the *P. expansum* pathogen were evaluated and compared with the standard well. The antifungal activity was calculated by measuring the zone of inhibition by using the standard scale (Table 1).

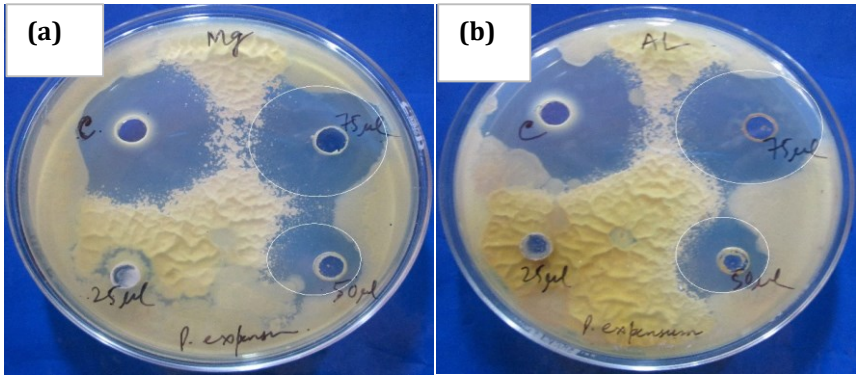


Figure 5. a) and b) displays the antifungal activity of $Mg(OH)_2$ and AlO(OH) on *P. expansum*

Table 1. Antifungal activity of $Mg(OH)_2$ and AlO(OH) nanoparticles

Sl.NO	$Mg(OH)_2$ (mm)			AlO(OH) (mm)			Control (mm)
	25 (µL)	50 (µL)	75 (µL)	25 (µL)	50 (µL)	75 (µL)	20 (µL)
1	-	13	19	-	13	20	20
2	-	12	19	1	11	19	22
3	-	13	18	2	12	21	21.3

Value are presented as mean ±SD

The $Mg(OH)_2$ and AlO(OH) seem to exhibit comparable significant antifungal activities. 25 µL of the selected NPs seems to be non effective in preventing the growth of fungus, often growth of fungus show concentration dependent inhibition**.** On increasing the concentration of NPs to 50 (µL) and 75 (µL), inhibition in the growth of *P. expansum was* observed. The AlO(OH) was able to reduce the growth of the fungus by 79% while $Mg(OH)_2$ reduces the growth by about 74% as compared to standard control hexahit. The oxides and hydroxides of metal NPs exhibit antimicrobial activity by generating reactive oxygen species like hydroxyl radicals ($OH^{\bullet}$), superoxide anion (O^{2-}) and perhydroxyl radicals ($HO_2^{\bullet}$), which disrupts the integrity of microbial membranes and cell wall through decomposition and destruction [27]. And these metal oxide and hydroxides in nanoscale bind themselves to intracellular proteins of the microbes and inactivate their biological activities there by killing the microbes [28]. Till date, ZnO and TiO_2 have been explored to control the growth of *P. expansum* [29, 30]. Automated turbidimetric analysis was used to assess the influence of the ZnO NPs on *P. expansum,* however different inoculums sizes have no significant effect for the use of spore concentrations between 10^4 and 10^6 spores/mL of *P. expansum* was observed [29]. Retardation in the development of *Penicillium* rot in apple significantly (P=0.05)

was reported on used of TiO_2 powder and TiO_2 coated plastic film due to its photocatalytic reaction [30]. Dimkpa *et al.* [31] has reported inhibitory activity of ZnO of 70 (±15) nm at minimum of 3 mmol/L (&0.244 mg/mL) against *Penicillium expansum*. Apart from metal oxides, silver nanoparticles synthesized using culture filtrate of lactic acid bacteria (13.84±4.56 nm) show strong growth inhibition against *Penicillium expansum* (15.87±1.01 mm) [32]. Silver chitosan nanocomposite with size less than 10 nm also reported to show significant antifungal activity against *P. expansum* [33]. Activity of fabricated NPs reported in present work seems to be more significant (or comparable) than those reported in literature.

Conclusions

Microwave heating has been adopted as an environmental benign approach to synthesize NPs of AlO(OH) and Mg $(OH)_2$. Superheating of solvents and localized high temperatures of microwave resulted in the formation of desired nanoparticles in 10-20 nm range. Nanoparticles such obtained show potential activities against the growth of fungus *P. expansum*. AlO(OH) reduce the growth of *P. expansum* by 79% while $Mg(OH)_2$ reduces about 74%, can be attributed to their small size and ability to generate reactive oxygen species. This work highlighted the significant role of green synthesized hydroxide nanoparticles as antimicrobial agent.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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