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Research Article

Synthesis, Characterization, and Antibacterial Activity of Biocompatible Aluminium Complexes with N-(2-Hydroxy-5-methyl benzyl)phenylalanine

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New antibacterial ligand H_3hmbpa-5 (N-(2-hydroxy-5-methyl benzyl)phenylalanine) and its aluminium complexes Al(O-Pri)(Hhmbpa-5), Al(H_2hmbpa-5)(Hhmbpa-5), and Al(H_2hmbpa-5)$_3$ were synthesized in excellent yield in a stoichiometric ratio of 1 : 1, 1 : 2, and 1 : 3 (Al(OPri)3 : ligand), based on an idea of biological activity of the phenylalanine and its substituted N-aryl derivatives and Al (III) derivatives. The complexes were characterized by FT-IR, ^{1}H, ^{13}C NMR, UV-visible, and elemental analysis. An inhibition zone test was used for the evaluation of the antibacterial test. The results show that the ligand and aluminium complexes have superior antibacterial activity against *Pseudomonas aeruginosa* (MTCC-424), *Streptococcus aureus* (MTCC-103), *Klebsiella pneumoniae* (MTCC-3389), and *Klebsiella pneumoniae* (MTCC-109).

1. Introduction

The search for antibacterial agents has attracted very much attention in recent years due to the emergence of dangerous diseases caused by different pathogens [1–3]. Evolution of different antibacterial agents has become a dangerous problem worldwide. Hence, it is needful to synthesize effective antibacterial compounds that will help fulfill the needs of modern times. The common antibacterial agents include organic and inorganic compounds [4]. Metal-organic complexes have a lot of advantages in the wide antibacterial spectrum, high effectiveness, long-term persistence, thermal stability, and clinical approach as compared with traditional chemical disinfectants [5, 6]. As we know, the nature of ligand, metal, and structure of metal-organic complexes have large influence on their properties. It is proposed that the aluminium metal reacts with N, O, and S atoms of each electron donor group such as amino, phosphate, and imidazole [7]. Goyal et al. reported five and six coordinated several aluminium and gallium N-arylsalicylaldiminate derivatives [8]. Sharma et al. proposed the molecular structure of unsymmetrical dimer viz. bis(N-phenylsalicylideneiminato)aluminium-di(μ-isopropoxy)di(isopropoxo)aluminium(III) and its reactions with alkoxyalkanols, having four- and six-coordinated aluminium(III) [9]. Symmetrical five-coordinate homodinuclear aluminium compounds have also been reported [10, 11]. In recent years, phenylalanine and its derivatives with many metal ion have been reported as an antibacterial agent to oppose many harmful microorganisms [12, 13]. Hydroxyethylpiperazine derivatives of phenylalanine appear the antimalarial activity against *Plasmodium falciparum* [14]. Also, the antimalarial activity was showed by some short peptides of phenylalanine [15].

Some of the 4-amino-N acetylphenylalanine amide ATP phosphate ester derivatives are potent protein kinase inhibitors [16]. N-Tetrahydrofuroyl-Lphenylalanineand1-[(3,5-dichlorophenyl)sulfonyl]pyrrolidine-2-carboxamido-3-phenylpropanoic acid derivatives act as potent antigen-4

antagonists and studied for inflammatory autoimmune diseases [17]. Mahesh Bhatt's group reported antibacterial and antioxidant properties of new amide derivatives of 4-nitro-L-phenylalanine [18]. Vajpayee and Singh reported synthesis characterization and antibacterial activity of Schiff base ligand containing homo and heterodinuclear derivatives of aluminium [19]. Gecgel et al. prepared an aluminum-based MOF and its amine form as novel biologically active materials for antioxidant, DNA cleavage, antimicrobial, and biofilm inhibition activities [20]. Zhang et al. developed aluminum metal-organic frameworks with photocatalytic antibacterial activity for autonomous indoor humidity control [21].

In this article, we report the synthesis, characterization, and antibacterial activity of new ligand H_3hmbpa-5 and aluminium complexes. It is expected that the assemblage of the highly potent antibacterial activity of [N-(2-hydroxy-5-methyl benzyl)phenylalanine] with the aluminium complex may contribute to the treatment of many dangerous diseases caused by *Pseudomonas aeruginosa* (MTCC-424), *Streptococcus aureus* (MTCC-103), *Klebsiella pneumoniae* (MTCC-3389), and *Klebsiella pneumoniae* (MTCC-109).

2. Materials and Methods

2.1. Reagents and Instruments. All the chemicals viz-phenylalanine (Alfa), p-cresol (BDH), sodium acetate trihydrate (BDH), formaldehyde solution 37% w/v (BDH), glacial acetic acid (BDH), benzene (Fischer), isopropanol (BDH), potassium dichromate (BDH), con.c. sulphuric acid (Fischer), and aluminium isopropoxide (Alfa) were of AnalaR grade used in the formation of H_3hmbpa-5 and aluminium derivatives (**1a–1c**). Elemental Vario make EL-III model instrument was used for elemental analysis of the prepared complexes at 950–1200°C. The melting point of the synthesized compound was calculated by the MP70 model. The PerkinElmer spectrum-Two IR spectrometer was used for recording the IR data of prepared compound in the range of 4000 to 400 cm^{-1} using KBr disc. The Shimadzu UV-visible 1800 spectrophotometer was used for the analysis of UV-visible spectra of synthesized complexes. The ^1H and ^{13}C NMR spectra of prepared complexes were carried out by the Bruker AVANCE III model of the NMR spectrometer using DMSO-d6.

2.2. Synthesis of Ligand H_3hmbpa-5 and Aluminium Derivatives Al(OPri)(Hhmbpa-5), Al(H_2hmbpa-5)(Hhmbpa-5), and Al(H_2hmbpa-5)$_3$. For the synthesis of H_3hmbpa-5 (Scheme 1), the procedure adopted is similar to that by Bradely et al. [22], and also in our previously published research work that is "Synthesis and characterization of boron derivatives with N-(o-hydroxy substituted benzyl) phenylalanines," in "Indian Journal of Advances in Chemical Sciences" [23]. The ligand H_3hmbpa-5 was obtained as solid powder in excellent yield (Table 1).

The synthesis of aluminium derivatives (**1a-1c**) was carried out under a fractionating column (30 cm long) closed with Raschig rings and attached to a condensation variable containing a still head [24]. The alcoholysis reaction is performed between aluminium triisopropoxide and H_3hmbpa-5 in the molar ratio of 1 : 1, 1 : 2, and 1 : 3 with ligand in benzene medium at 90–100°C about 8–12 hour depending upon how many no. of moles of isopropanol were liberated. The number of moles of isopropanol liberated in the binary azeotrope with benzene was estimated by iodometric titration.

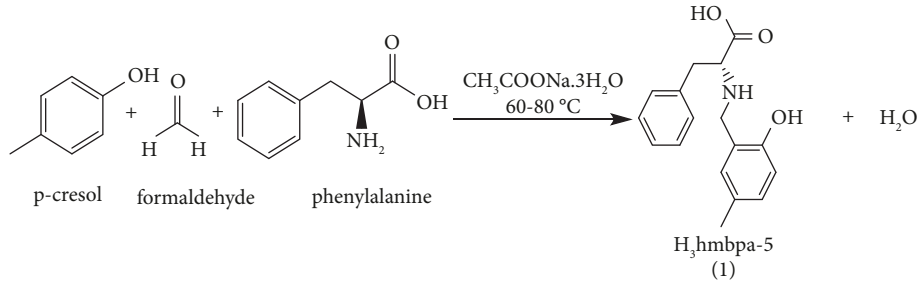
For the synthesis of Al(OPr^i)(Hhmbpa-5), a mixture of Al(OPr^i)$_3$ (0.6127 g, 3.00 m·mole) and H_3hmbpa-5 (0.8550 g; 3.00 m·mole) suspended in dry benzene (60 mL) was refluxed for cal. 8 hrs on a wax bath (90–100°C). The product was isolated in high yield: 0.924 g; m.p. 175°C, amount of isopropanol in azeotrope, calculated: 0.36 g, found: 0.34 g (2 mol).

For the synthesis of Al(H_2hmbpa-5)(Hhmbpa-5), a mixture of Al(OPr^i)$_3$ (0.5106 g, 2.50 m·mole) and H_3hmbpa-5 (1.4250 g, 5.00 m·mole) suspended in dry benzene (60 mL) was refluxed for cal. 9 hrs on a wax bath (90–100°C). The product was isolated in high yield: 1.44 g; m.p. 178°C, amount of isopropanol in azeotrope, calculated: 0.45 g, found: 0.41 g (3 mol).

Similarly, for the synthesis of Al(H_2hmbpa-5)$_3$, a mixture of Al(OPr^i)$_3$ (0.4085 g, 2.00 m·mole) and H_3hmba-5 (1.7100 g; 6.00 m·mole) suspended in dry benzene (60 mL) was refluxed for cal. 11 hrs on a wax bath (90–100°C). The product was isolated in high yield: 1.75 g; m.p. 185°C, amount of isopropanol in azeotrope, calculated: 0.36 g, found: 0.32 g (3 mol). The ligand and aluminium derivatives were obtained as solid powder and soluble in dimethylsulphoxide, dimethylformamide, and sodium hydroxide but insoluble in benzene and ether.

2.3. Antibacterial Activity. The bactericidal activity of the prepared ligand (**1**) and aluminium complexes (**1a–1c**) was tested against Gram-negative bacteria such as *Pseudomonas aeruginosa* (MTCC-424), *Klebsiella pneumoniae* (MTCC-3389), and *Klebsiella pneumoniae* (MTCC-109) and Gram-positive bacteria such as *Streptococcus aureus* (MTCC-103). All the previous strains were obtained from the Microbial Type Culture Collection and Gene Bank (MTCC), Institute of Microbial Technology, Chandigarh, India.

These antibacterial activities were evaluated qualitatively by the agar disc diffusion method. This method was used to find the toxicity of bacterial pathogens which multiply largely to detect growth or inhibition within 24–48 hrs of incubation. Bacterial broth cultures were prepared to the density of 10^5 cells/mL. Aliquots of 100 μl were spread evenly into individual nutrient agar plates. On each plate, six equidistant disc plates were put on agar with 0.5 mm diameter sterilized forceps, 2 mm from the edge of the plate. Three quantities (1 μg, 5 μg, and 15 μg/disc) of each sample of ligand and aluminium complexes were transferred to respective agar disc, and plates were incubated at 37°C for 24–48 h. Sterilized DMSO was included as the negative control, and 50 μg/disc oxytetracycline (OTC) was used as the positive control. All the experiments were performed in triplicate. The appearance of clear inhibition zones around the wells seemed as positive results and was measured in mm.



Scheme 1: Synthesis of H_3hmbpa-5 from p-cresol, formaldehyde, and phenylalanine.

Table 1: Analytical data of synthesized ligand (**1**) and aluminium complexes (**1a–1c**).

Ligand/aluminium complexes	Colour	Yield (%)	Elemental analysis (%) found (calcd.)				
			C	H	O	N	Al
H_3hmbpa-5	Light brown	85	71.52 (71.57)	6.64 (6.67)	16.83 (16.84)	4.89 (4.91)	—
Al(OPr^i)(Hhmbpa-5)	Brownish white	95	65.93 (66.03)	5.18 (5.21)	15.42 (15.51)	4.43 (4.52)	8.63 (8.72)
Al(H_2hmbpa-5)(Hhmbpa-5)	Brownish white	93	68.58 (68.68)	5.84 (5.92)	16.05 (16.13)	4.62 (4.70)	5.43 (5.53)
Al(H_2hmbpa-5)$_3$	Brownish white	91	69.55 (69.62)	6.03 (6.10)	16.23 (16.35)	4.68 (4.77)	3.02 (3.06)

3. Results and Discussion

*3.1. Structure of Ligand (**1**) and Aluminium Complexes (**1a–1c**).* The ligand was synthesized using p-cresol, formaldehyde, and phenylalanine according to Scheme 1. Aluminium complexes were synthesized using Al(OPr^i)$_3$ and H_3hmbpa-5 according to Scheme 2. All the analytical data obtained for ligand and aluminium complexes are in good compatibility with 1 : 1, 1 : 2, and 1 : 3 stoichiometric ratios of metal to ligand. The UV-visible spectra of ligands and complexes were studied in the range of concentration (10^{-4}-10^{-5}). The molecular formula and structure of ligand (**1**) and aluminium complexes (**1a–1c**) were deduced according to the molar ratio method that is obtained using the iodometric titration [25] and spectroscopic analysis. In the synthesized complexes **1a**, **1b**, and **1c**, aluminium metal is four, five, and six coordinated, respectively.

3.1.1. FT-IR Spectra. The FT-IR spectra (Figure 1) of ligand (**1**) and aluminium complexes (**1a–1c**) were recorded using KBr pellets in the region of (4000–400) cm^{-1}. The comparison of IR displacements between the ligand and the obtained aluminium complexes are presented in Table 2.

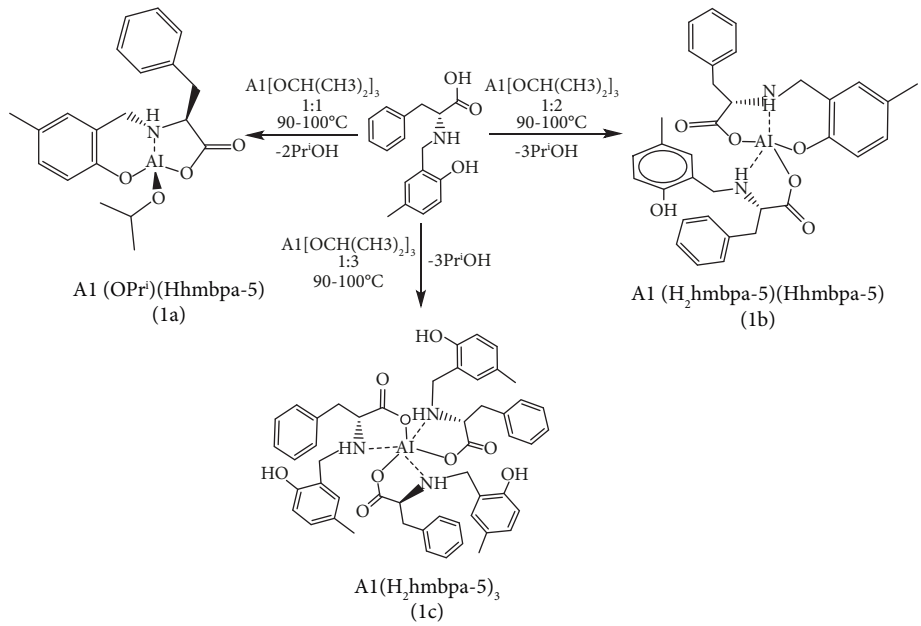
The appearance of a broad band in region 3440–3200 cm^{-1} in the case of H_3hmbpa-5 (**1**) shows the overlapping of the phenolic O-H and aromatic C-H stretching. The medium and weak bands at 2930 cm^{-1} and 2850 cm^{-1}, respectively, may be assigned to C-H stretching of both -CH_2- and -CH_3 groups of the cresol ring. A weak broad band at 2350 cm^{-1} shows the N-H stretching of the >NH_2^+ group. The overlapping of the asymmetric O-C=O and aromatic C=C stretching identified here as a shouldered band at 1625 cm^{-1}. The overlapping of aromatic C=C stretching and C-H bending of both -CH_2- and -CH_3 groups of the cresol ring is identified here by the appearance of strong bands at 1500 cm^{-1} and 1460 cm^{-1}, while a sharp band of medium intensity at 1340 cm^{-1}may be ascribed to the symmetric O-C=O stretching. The occurrence of a sharp band at 1280 cm^{-1} corresponds to the interaction of O-H bending and C-O stretching of the phenolic group. While medium bands at 1230 cm^{-1} and 1135 cm^{-1} may be attributed to the C-N stretching and phenolic C-C-O stretching, respectively. Here too, very weak absorptions below 600 cm^{-1} occur because of both aromatic ring skeletal vibrations.

The band due to the phenolic group as observed in H_3hmbpa-5 is found to be absent in Al(OPr^i)(Hhmbpa-5), suggesting the bonding of the phenolate oxygen with aluminium. A shift in $\nu_{sCOO,}$ as compared to H_3hmbpa-5, suggests the bonding of the carboxylate oxygen with aluminium in **1a–1c**. In IR spectra (Figure 1) of complexes **1a–1c,** the absorptions at 610–550 and 480–470 confirmed the presence of Al-O and Al-N bonds, respectively [26].

3.1.2. UV-Visible Spectra. Electronic spectra of ligand and aluminium complexes were taken in the (200–900) nm range in DMSO (Figure 2). In the electronic spectrum of H_3hmbpa-5, a high-intensity absorption band appears at 245–275 nm due to $\pi \longrightarrow \pi^*$ transition of the aromatic ring. On the other hand, the spectra of Al(OPr^i)(Hhmbpa-5) show $\pi \longrightarrow \pi^*$ transition at 310–335 nm and $n \longrightarrow \pi^*$ transition at 505–555 nm. The spectra of Al(H_2hmbpa-5)(Hhmbpa-5) show $\pi \longrightarrow \pi^*$ transition at 305–330 nm and $n \longrightarrow \pi^*$ transition at 490–530 nm, while Al(H_2hmbpa-5)$_3$ shows a high-intensity$\pi \longrightarrow \pi^*$ transition at 300–325 nm and $n \longrightarrow \pi^*$ transition at 480–560 nm [27].

3.1.3. ^{1}H and ^{13}C NMR Spectrum. The proton NMR spectrum of H_3hmbpa-5 and the aluminium complex were recorded in dimethyl sulfoxide (DMSO-*d*6) solution using the Bruker AVANCE III NMR spectrometer.

In ^{1}H NMR spectra of prepared compound H_3hmbpa-5 resonate at 4.53 (dt, J = 8.1, 5.6 Hz 1H, NH) confirmed the C-N bonding between formaldehyde and



Scheme 2: Synthesis of aluminium derivatives Al(OPr^i)(Hhmbpa-5), Al(H_2hmbpa-5)(Hhmbpa-5), Al(H_2hmbpa-5)$_3$ from H_3hmbpa-5, and Al(OPr^i)$_3$ in 1 : 1, 1 : 2, and 1 : 3 molar ratio.

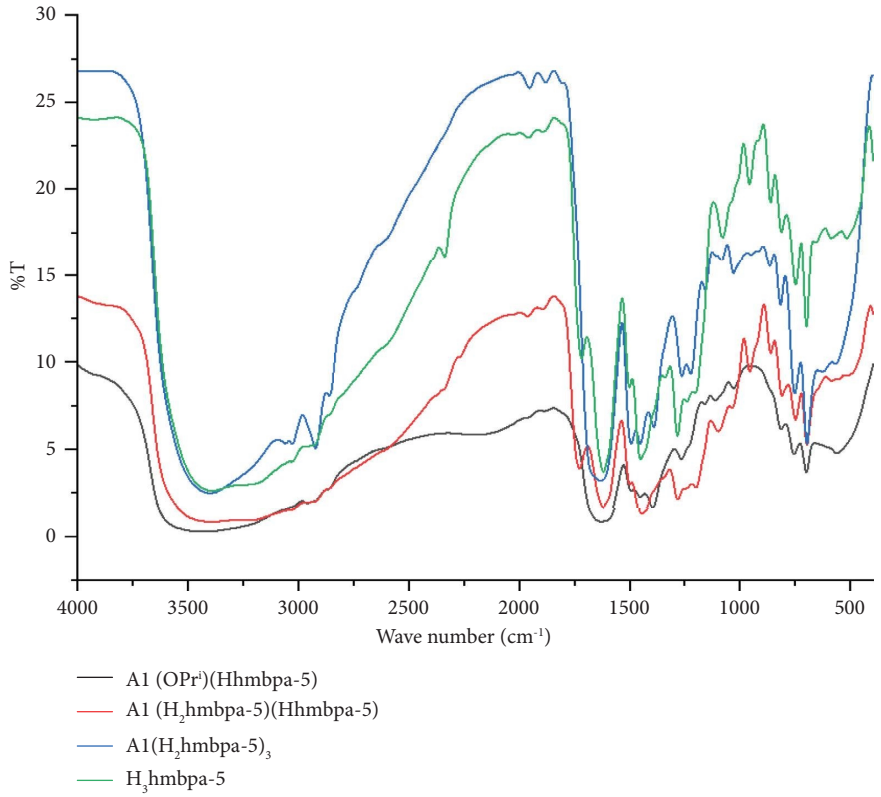


Figure 1: FT-IR spectrum of H_3hmbpa-5 and the aluminium complexes (**1a–1c**) (KBr disk).

phenylalanine. In ^{13}C NMR spectra of H_3hmbpa-5, both -CH2- group resonate between 48.40–45.40 and 37.80 ppm [28]. In ^{1}H NMR spectra of H_3hmbpa-5 -CH2- protons are diastereotopic in nature and resonate between 3.98 and 2.84 ppm.

In ^{1}H NMR spectra of **1a,** the disappearance of multiplet at (7.14–7.10 m 1H) confirmed the absence of phenolic proton and confirmed the linkage of phenolic oxygen with aluminium. The appearance of a singlet at 7.20 (s, 1H) confirm the presence of phenolic proton in **1b,** while the

Table 2: Characteristics infrared frequency (cm^{-1}) of aluminium derivatives (**1a–1c**).

1/1a–1c	$\nu_{O\text{-}H}$ [a]	$\nu_{N\text{-}H}$, $\nu_{C\text{-}H}$ [b]	$\nu_{C\text{-}H}$ [c]	$\nu_{as\ COO}$	$\Delta\nu_{COO}$	ν_{sCOO}	$\nu_{C\text{-}N}$ [d]	$\nu_{Al\text{-}O}$	$\nu_{Al\text{-}N}$
1	3440–3200 (mb)	3240–3000 (mb)	2930 (m) 2850 (w)	1625 (s)	285	1340 (m)	1230 (mb)	—	—
1a	—	3280–3000 (b)	2960 (mb) 2860 (m)	1630 (mb)	240	1390 (s)	1265 (b)	610 (s) 570 (m)	480 (m) 460 (m)
1b	3600–3300 (b)	3200–3000 (mb)	2935 (w) 2910 (mb) 2850 (w)	1643 (s)	257	1386 (mb)	1263 (m)	595 (m) 550 (s)	485 (m) 475 (m)
1c	3550–3300 (vb)	3150–3000 (mb)	2990 (m) 2840 (w)	1642 (vs)	262	1380 (s)	1250 (s)	600 (s) 560 (m)	490 (m) 470 (w)

[a]Absorption frequency (cm^{-1}) of the phenolic (O-H) group; [b]Absorption frequency (cm^{-1}) of N-H and C-H of both aromatic rings; [c]Absorption frequency (cm^{-1}) of C-H of -CH3 and both -CH2- groups; [d]Absorption frequency (cm^{-1}) of C-N bond. Vb = very broad, b = broad, m = medium, mb = medium broad, w = weak, wb = weak broad, s = sharp, vs = very sharp, sb = sharp broad, and sh = shoulder hump.

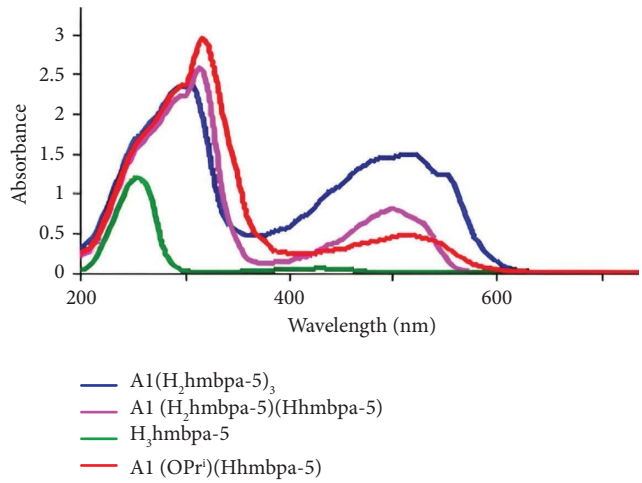


Figure 2: UV-visible spectrum of ligand (**1**) and aluminium complexes (**1a–1c**).

multiplet at (7.14–7.10 m 1H) in **1c** also confirmed the presence of Ar-OH proton.

[N-(2-hydroxy-5-methylbenzyl)phenylalanine](H3hmbpa-5): ^{1}H NMR (500 MHz, DMSO) ((δ 7.31–7.18 (m, 5H Ar-H) Ring proton of phenylalanine part), 7.14–7.10 (m, 1H cresol part and Ar-OH), 6.90–6.84 (m, 1H cresol part), 6.75 (d, J = 8.5 Hz, 1H cresol part), 4.53 (dt, J = 8.1, 5.7 Hz, 1H, NH), 3.98 (ddd, J = 15.4, 5.7, 0.9 Hz, 1H, CH_2), 3.89–3.79 (m, 2H, CH,CH_2), 3.06 (ddt, J = 13.9, 6.3, 0.9 Hz, 1H, CH_2), 2.84 (ddt, J = 13.9, 6.3, 1.0 Hz, 1H, CH_2), 2.22 (d, J = 0.9 Hz, 3H, CH_3). ^{13}C NMR: (125 MHz, DMSO) δ 175.33(COOH), (154.92(Ar-C-OH), 126.00(C-1),131.02(C-6),129.78(C-5),129.14(C-4),115.99(C-3) Ring carbon of cresol part) (137.54(C-1), 128.94(C-2,C-6), 127.21(C-3,C-5), 127.07(C-4) Ring carbon of phenylalanine part), 61.62(CH), 48.40(CH_2), 37.80(CH_2), and 20.75(CH_3).

Al(OPri)(Hhmbpa-5) **(1a)**: ^{1}H NMR (500 MHz, DMSO) (δ 7.31–7.19 (m, 3H Ar-H) Ring proton of phenylalanine part), (7.13 (dd, J = 1.9, 0.9 Hz, 1H cresol part), 7.02–6.96 (m, 1H cresol part), 6.81 (d, J = 8.0 Hz, 1H cresol part), (4.22 (hept, J = 6.0 Hz, 1H, CH (OPr^i)), 4.02–3.94 (m, 1H, CH_2), 3.98–3.91 (m, 1H CH), 3.86 (ddt, J = 15.4, 5.6, 0.9 Hz, 1H, CH_2), 3.12 (dt, J = 7.5, 5.5 Hz, 1H NH), 3.04 (ddt, J = 14.0, 6.1, 0.8 Hz, 1H, CH_2), 2.85 (ddt, J = 13.9, 6.0, 0.9 Hz, 1H, CH_2), 2.21 (d, J = 0.8 Hz, 3H, CH_3), and 1.30 (dd, J = 6.1, 1.5 Hz, 6H, CH_3). ^{13}C NMR (125 MHz, DMSO) δ (173.12, COOH), (155.21, Ar-C-O), 137.16, 131.64, 129.03, 128.99, 128.92, 127.77, 127.21, 126.25, 114.92, (Ar-C of both ring), (70.36, CH), (62.61, CH (OPr^i)), (46.77, 37.78, CH_2), and (22.72, 20.72, CH_3).

Al(H_2hmbpa-5)(Hhmbpa-5) **(1b)**: ^{1}H NMR (500 MHz, DMSO) δ 7.31–7.21 (m, 8H, Ar-H) Ring proton of phenylalanine part,7.20 (s, 1H, Ar-OH), 7.18–7.10 (m, 2H cresol part), 7.03–6.97 (m, 1H cresol part), 6.90–6.83 (m, 2H cresol part), 6.75 (d, J = 8.5 Hz, 1H cresol part), 4.05–3.80 (m, 6H CH), 3.64 (dt, J = 7.7, 5.9 Hz, 1H NH), 3.10–3.01 (m, 2H CH_2), 3.04–2.97 (m, 1H CH_2), 2.83 (ddt, J = 13.9, 6.1, 0.9 Hz, 2H CH_2), and 2.21 (d, J = 0.9 Hz, 3H, CH_3). ^{13}C NMR (125 MHz, DMSO) δ (172.05, 172.02, COOH), (154.95, 153.75Ar-C-O), (137.20, 137.12, 132.09, 131.02, 129.78, 129.25, 129.19, 129.17, 129.15, 129.03, 128.98, 128.92, 128.20, 127.46, 127.27, 127.21, 127.07, 125.96, 116.39, 115.99(Ar-C of both ring)), (63.11, 62.86 CH), (48.27, 46.16, 37.50, 37.38 CH_2), and (20.76, 20.75CH_3).

Al(H_2hmbpa-5)$_3$ **(1c)**: ^{1}H NMR (500 MHz, DMSO) δ 7.31–7.18 (m, 5H Ar-H) Ring proton of phenylalanine part, 7.14–7.10 (m, 1H cresol part and Ar-OH), 6.87 (dd, J = 8.6, 2.1 Hz, 1H cresol part), 6.75 (d, J = 8.5 Hz, 1H cresol part), 3.96 (ddt, J = 15.2, 5.7, 0.9 Hz, 1H CH_2), 3.92–3.86 (m, 1H CH_2), 3.89–3.81 (m, 1H CH), 3.64 (dt, J = 7.6, 5.7 Hz, 1H NH), 3.01 (ddt, J = 13.9, 6.0, 0.9 Hz, 1H CH_2), 2.81 (ddt, J = 14.0, 6.0, 0.9 Hz, 1H CH_2), and 2.21 (d, J = 0.9 Hz, 3H, CH_3) ^{13}C NMR (125 MHz, DMSO) δ (172.03, 171.99,171.96, COOH), (155.00, 154.97, 154.95,Ar-C-O), (137.24, 137.20, 137.16, 131.17, 131.09, 131.02, 129.86, 129.82, 129.78, 129.36, 129.30, 129.25, 129.19, 129.14, 129.08, 129.03, 128.98, 128.92, 127.34, 127.27, 127.21, 127.20, 127.13, 127.07, 126.05, 126.01, 125.96, 116.05, 116.02, 115.99, (Ar-C of both ring)), (62.96, 62.92, 62.88, CH), (48.31, 48.29, 48.27,CH_2), (37.55, 37.50, 37.46, CH_2), and (20.84, 20.80, 20.75, CH_3).

3.2. Antibacterial Activity. The *in vitro* antibacterial activity of the ligand and aluminium complexes was evaluated qualitatively by the agar disc diffusion method [29], tested against Gram-negative bacteria such as *Pseudomonas aeruginosa* (MTCC-424), *Klebsiella pneumoniae* (MTCC-3389), and *Klebsiella pneumoniae* (MTCC-109) and Gram-positive bacteria such as *Streptococcus aureus* (MTCC-103). These antibacterial activities were evaluated qualitatively by the agar disc diffusion method. The radius of zone of inhibition values (mm) of the ligand and aluminium complexes in different concentrations (1 mg, 5 mg, and 15 mg) with positive control of 50 mg oxytetracycline are present in Table 3 and Figures 3–6.

From these results, it is clear that the free ligand and aluminium complexes have an excellent antibacterial effect against all the tested bacteria. Also, it is clear from Table 3 and Figures 3–6, the ligand (**1**) and aluminium complexes (**1a–1c**) show larger antibacterial effect against *P. aeruginosa*

Table 3: Results of antibacterial activity of the synthesized compounds.

1/1a–1c	Con.c. (mg/mL)[a]	The radius of the zone of inhibition (mm)			
		P. aeruginosa (MTCC-424)	*S. aureus* (MTCC-103)	*K. pneumoniae* (MTCC-3389)	*K. pneumoniae* (MTCC-109)
1	1/5/15/50	0.85/0.85/1.1/1.25	0/0/0.35/0.85	0/0.25/0.45/0.70	0.45/0.40/0.90/1.25
1a	1/5/15/50	0.05/0.70/0.75/1.25	0/0.0/0.35/0.75	0/0/0.30/0.70	1.25/0.45/0.75/1.25
1b	1/5/15/50	0.45/0.75/0.80/1.25	0/0.45/0.20/0.80	0/0/0/0.55	0.0/0.75/0.95/1.30
1c	1/5/15/50	0.55/0.60/0.75/1.25	0/0.25/0.25/0.75	0/0/0/0.75	0.45/0.70/0.95/1.45
DMSO	—	0/0/0/0	0/0/0/0	0/0/0/0	0/0/0/0

[a]1/5/15 mg/mL con.c were taken for each ligand and aluminium complexes with positive control of 50 mg/mL oxytetracycline.

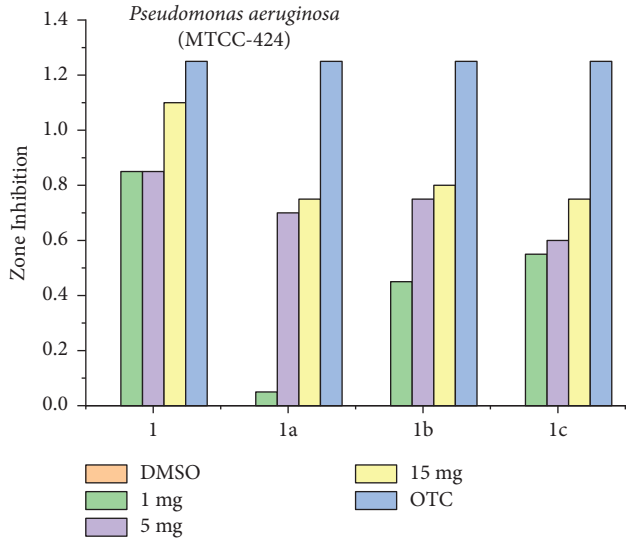


Figure 3: Radius of the zone of inhibition (mm) against *P. aeruginosa* (MTCC-424) in the range of 1–15 mg/mL sample (**1**, **1a**, **1b**, and **1c**) with positive control of 50 mg/mL oxytetracycline (OTC).

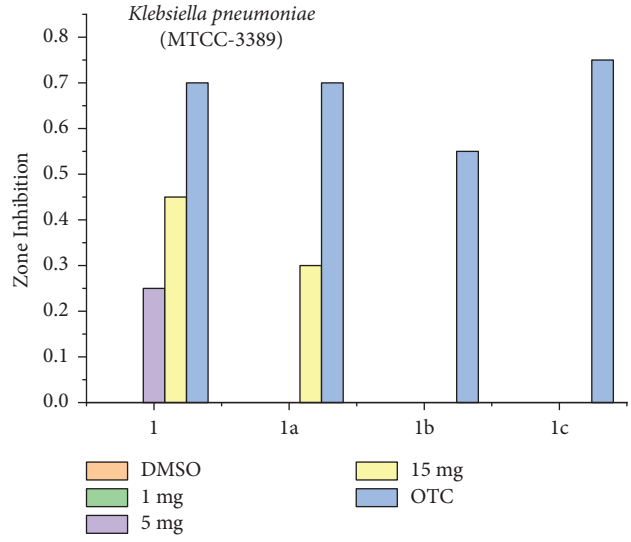


Figure 5: Radius of the zone of inhibition (mm) against *K. pneumoniae* (MTCC-3389) in the range of 1–15 mg/mL sample (**1**, **1a**, **1b**, and **1c**) with positive control of 50 mg/mL oxytetracycline.

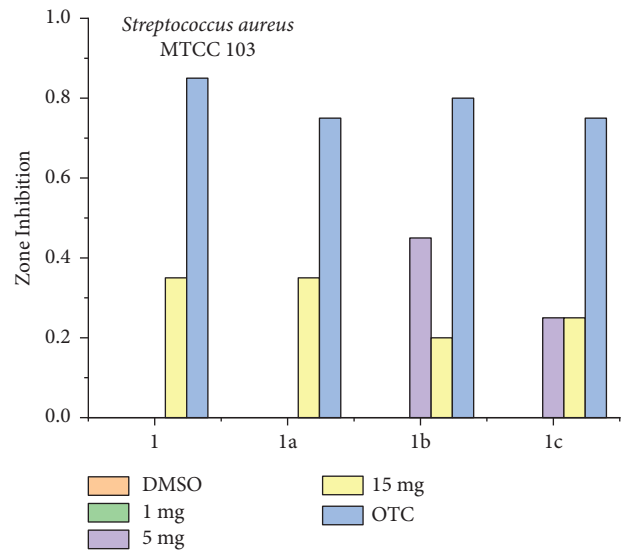


Figure 4: Radius of the zone of inhibition (mm) against *S. aureus* (MTCC-103) in the range of 1–15 mg/mL sample (**1**, **1a**, **1b**, and **1c**) with positive control of 50 mg/mL oxytetracycline.

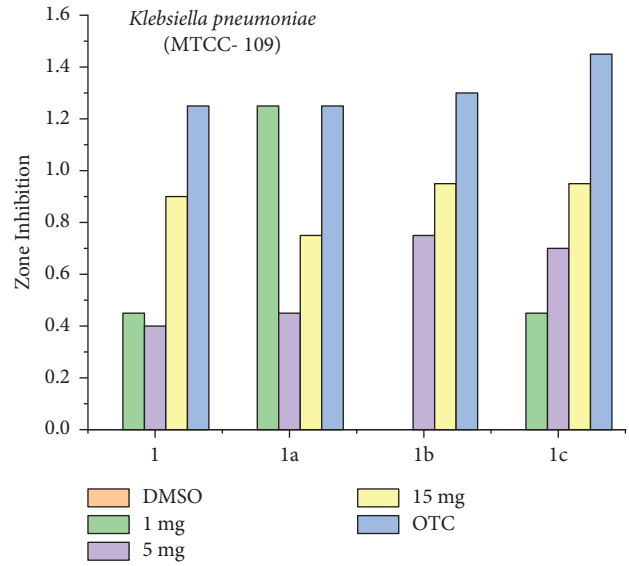


Figure 6: Radius of the zone of inhibition (mm) against *K. pneumoniae* (MTCC-109) in the range of 1–15 mg/mL sample (**1**, **1a**, **1b**, and **1c**) with positive control of 50 mg/mL oxytetracycline.

(MTCC-424) and *K. pneumoniae* (MTCC-109) but show smaller effects against *S. aureus* (MTCC-103) and *K. pneumoniae* (MTCC-3389).

Furthermore, the ligand H_3hmbpa-5 and Al(O-Pr^i)(Hhmbpa-5) in all concentrations (1 mg, 5 mg, and 15 mg) show excellent antibacterial effect against all the tested bacteria as compared to Al(H_2hmbpa-5)(Hhmbpa-5) and Al(H_2hmbpa-5)$_3$. We can conclude that these ligands and complexes have excellent growth inhibiting effects on tested Gram-negative and Gram-positive bacteria.

The mechanism of antibacterial activity of the prepared complexes is yet not still reported. The antimicrobial mechanism of the ligand and aluminium complex will be further investigated in the ongoing test of our lab.

4. Conclusion

As a result of this work, new antibacterial ligand H_3hmbpa-5 and aluminium complexes **1a–1c** were synthesized and characterized by elemental analysis, FT-IR, ^{1}H, ^{13}C NMR, and UV-visible spectroscopy. On the basis of these spectroscopic data, it is suggested that the aluminium derivatives

are conformable with the ratio 1 : 1, 1 : 2, and 1 : 3 for aluminium to the ligand. The obtained aluminium complexes are conformable with a mononuclear structure and suggested that the ligand H_3hmbpa-5 act as a bidentate and tridentate ligand. *In vitro* studies show that the ligand and aluminium complexes has excellent antibacterial effect towards both Gram-negative bacteria *Pseudomonas aeruginosa* (MTCC-424), *Klebsiella pneumoniae* (MTCC-3389), and *Klebsiella pneumoniae* (MTCC-109) and Gram-positive bacteria *Streptococcus aureus* (MTCC-103). Therefore, this study provides a new idea to synthesize a metal-organic complex having antibacterial ligand for a potential antibacterial agent in the medicinal field.

Data Availability

The supplementary data associated with the structures and antibacterial activity of prepared ligand and aluminium complexes are available as a separate file.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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Supplementary Materials

The structure of compounds **1**, **1a**, **1b**, and **1c** was established through FT-IR, ^{1}H, ^{13}C NMR, and UV-visible spectroscopy. The supplementary file contains ^{1}H, ^{13}C NMR spectrum, and also images of experimental slides of antibacterial activity of the prepared complexes. (*Supplementary Materials*)

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