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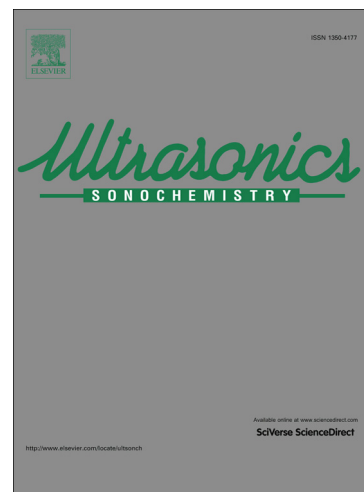
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Synthesis and Characterization of a New Nano Lead(II) 0-D Coordination Supramolecular Compound: A Precursor to Produce Pure Phase Nano-sized Lead(II) Oxide

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Abstract

Nano-structure of a new 0D Pb(II) coordination supramolecular compound, $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2$ (**1**), L=8-HQuin= 8-hydroxyquinolin ligand has been synthesized by use of a sonochemical process and characterized by scanning electron microscopy (SEM), X-ray powder diffraction (XRPD), Fourier transform infrared spectroscopy (FTIR) and elemental analyses. The structure of compound **1** was determined by single-crystal X-ray diffraction. The single crystal X-ray data of compound **1** implies that the Pb^{+2} ions are five coordinated. Each lead atom is coordinated to nitrogen and oxygen atoms of 8-hydroxyquinolin ligand. Topological analysis shows that the compound **1** is 1,2,3,4,4M12-1net. Nanoparticles of lead (II) oxide have been prepared by calcination of lead (II) coordination polymer at 500 °C that were characterized by scanning electron microscopy (SEM), X-ray powder diffraction (XRPD) and IR spectroscopy.

Keywords: Nanoparticles; Lead (II) oxide; Coordination supramolecular; Ultrasonic.

Introduction

The design and preparation of coordination supramolecular compound are becoming an exciting field during the past decade and have been widely studied as they represent an important

interface between synthetic chemistry and materials science. The synthesis of coordination supramolecular compound with various metal ions and ligands have led to a wide range of potential applications as e.g. molecular wires, electrical conductors, molecular magnets, in host-guest chemistry as well as in catalysis[1-11]. Pb(II) cations form a range of coordination supramolecular compound and polynuclear complexes which display interesting structural features as a consequence of the large radius, adoption of different coordination numbers and the possible occurrence of a stereochemically active lone pair of electrons [12]. The absence of crystal field stabilization energy effects also allows the Pb(II) cations to adopt a range of different coordination geometries not restricted to octahedral, tetrahedral or squareplanar. 8-Hydroxyquinoline (8-HQ), as a very useful ligand, has often been incorporated into conjugated coordination supramolecular compound. These coordination supramolecular compound may find potential applications in OLED devices, optical sensors, and dye-sensitized solar cells due to their exceptional optical properties and thermal stability. However, these coordination supramolecular compound usually possess very poor process ability due to lack of thermoplasticity and solubility in common solvents [13-24]. A number of different synthetic approaches have been offered for the preparation of coordination supramolecular compound. Some of them are slow diffusion of the reactants into a polymeric matrix, diffusion in the gas phase, evaporation of the solvent at ambient or reduced temperatures, precipitation or recrystallization from a mixture of solvents, temperature controlled cooling, hydrothermal synthesis and sonochemical syntheses [25-27]. Sonochemical methods can cause homogeneous nucleation and a considerable reduction in crystallization time compared with conventional oven heating when nanomaterials are prepared [28]. Many researchers have investigated the effect of ultrasound on chemical reactions, and most theories imply that the chemical or physical effects of ultrasound

originate from acoustic cavitation within collapsing bubbles, which generates extremely localized hot spots having temperatures of roughly 5000 K, pressures of about 500 atm, and a life time of a few microseconds. Between the microbubble and the bulk solution, the interfacial region around the bubble has very large gradients of temperature, pressure, and the rapid motion of molecules leading to the production of excited states, bond breakage, the formation of free radicals, mechanical shocks, and high shear gradients [29]. The use of high-intensity ultrasound to enhance the reactivity of metals as stoichiometric reagent has become a synthetic technique for many heterogeneous organic and organometallic reactions [30-31].

Recently, nano sized coordination supramolecular compound with finite repeating units, have aroused a growing interest due to their special properties distinctive from conventional bulk coordination supramolecular compound[32]. Up to now, various coordination supramolecular compound nanocrystals including nanoparticles, nano-cubes, nano-rods, and nanotubes have been successfully synthesized [38-41]. Nano-particles of coordination polymers are fascinating to explore, because they are interesting candidates for applications in gas storage, conductivity, molecular recognition and separations, catalysis, chirality, photonics and magnetic materials [42-44]. The results of ultrasound radiation on chemical reactions were reported in the recent works [45-47]. Also, PbO is a polar inorganic crystalline material with many applications, for example; leaden plug, glass ceramic and dye industry. PbO is very fascinating because of its numerous phases such as Pb_2O , Pb_2O_3 and Pb_3O_4 . Till now, many methods have been developed to synthesis PbO nanocrystals, including vapor phase growth [48], vapor-liquid-solid process [49], soft chemical method [50], electrophoretic deposition [51], sol-gel process [52,53], homogenous precipitation [54], etc. The use of metal organic supramolecular compounds as precursors for the

preparation of inorganic nanomaterials such as lead(II) oxide has not yet been investigated thoroughly [55,56].

In most of the coordination supramolecular compound the bridging ligands are solely coordinated to a single metal ion. In this work, we choose a O- and N- donor ligand, 8-hydroxyquinolin and ClO_4^- anion with the aim to construct a new mixed-ligand nano coordination compound. Also, we report on the preparation of $[\text{Pb}_4(8\text{-HQin})_6](\text{ClO}_4)_2$ (**1**), however this compound was used for preparation of PbO nanoparticles.

2. Experimental

2.1. Materials and physical techniques

All reagents and solvents for the synthesis and analysis were commercially available from Merck Company and used as received. Methanol was used to prepare solutions. A multiwave ultrasonic generator (Sonicator_3000; Misonix, Inc. Farmingdale, NY, USA), equipped with a converter/transducer and titanium oscillator (horn), 12.5 mm in diameter, operating at 20 kHz with a maximum power output of 600W at room temperature for 1 hr, was used for the ultrasonic irradiation. IR spectra were recorded using Perkin-Elmer 597 and Nicolet 510P spectrophotometers. Microanalyses were carried out using a Heraeus CHN-O- Rapid analyzer. Melting points were measured on an Electrothermal 9100 apparatus. X-ray powder diffraction (XRD) measurements were performed using an X'pert diffractometer of Philips company with monochromated $\text{Cu}\alpha$ radiation ($\lambda = 1.54056 \text{ \AA}$). Simulated XRD powder pattern based on single crystal data were prepared using Mercury software [57]. The crystallite sizes of selected samples were estimated using the Scherrer method. The samples were characterized with a scanning

electron microscope (SEM) (Philips XL 30) with gold coating. The X-ray single crystal data for compound **1** was collected at 296(1) K on STOE IPDS II diffractometers (Mo $K\alpha = 0.71073\text{\AA}$). Cell parameters were retrieved using X-AREA [58] software and refined using X-AREA on all observed reflections. Data reduction and correction for Lp (Lorentz-polarization) and decay were performed using X-AREA software. Absorption corrections were applied using MULABS [59] in PLATON [60]. All structures were solved by direct methods and refined by full-matrix least squares on F^2 for all data using SHELXTL [61] software. All calculations were performed by PLATON. All non-hydrogen atoms were refined anisotropically. Hydrogen atoms were positioned geometrically and refined with a riding model approximation with their parameters constrained to the parent atom with $U_{iso}(\text{H}) = 1.2 U_{eq}(\text{C})$.

2.2. Synthesis of $[\text{Pb}_4(\text{8-HQuin})_6](\text{ClO}_4)_2$ (**1**) as single crystal

Lead (II) acetate (0.325 g, 1 mmol), 8-hydroxyquinolin (0.14g, 1 mmol) and sodium perchlorate (2 mmol, 0.244g) were loaded into one arm of a branch tube and both of the arms were filled slowly by methanol. The chemical bearing arm was immersed in an oil bath kept at 60 °C. Crystals were formed on the inside surface of the arm kept at ambient temperature, After 3 days, colorless crystals were deposited in the cooler arm were filtered off, washed with water and air dried.

Compound 1: White crystals, m.p. 320°C. Analysis: found (C: 34.21, H: 1.89, N: 4.38%; calculated for $\text{C}_{54}\text{H}_{36}\text{Cl}_2\text{N}_6\text{O}_{14}\text{Pb}_4$; C: 34.27, H: 1.92, N: 4.44%).

IR (cm^{-1}) selected bands: $\nu = 725(\text{s}), 818(\text{vs}), 1101(\text{s}), 1227(\text{vs}), 1316(\text{s}), 1380(\text{s}), 1493(\text{s}), 1561(\text{s}), 3040(\text{w}), 3423(\text{w})$

2.3. Synthesis of $[\text{Pb}_4(8\text{-HQin})_6](\text{ClO}_4)_2$ (**1**) nano-structures under ultrasonic irradiation

High-density ultrasonic probe immersed directly into the solution of lead (II) acetate (10 ml, 0.1 M) in methanol, then into this solution, a proper volume of 8-hydroxyquinolin ligand and sodium perchlorate in methanol solvent (10 ml, 0.1 M) was added in a drop wise manner. The solution was irradiated by sonochemical with the power of 60W and temperature 45 °C for 1 h. The obtained precipitates were filtered, subsequently washed with water and then dried.

Compound 1: White crystals, m.p. 332°C. Analysis: found (C: 34.25, H: 1.90, N: 4.41%; calculated for $\text{C}_{54}\text{H}_{36}\text{Cl}_2\text{N}_6\text{O}_{14}\text{Pb}_4$; C: 34.27, H: 1.92, N: 4.44%).

IR (cm^{-1}) selected bands: $\nu = 725(\text{s}), 818(\text{vs}), 1102(\text{s}), 1227(\text{vs}), 1316(\text{s}), 1380(\text{s}), 1493(\text{s}), 1561(\text{s}), 3040(\text{w}), 3423(\text{w})$

2.4. Synthesis of PbO as nanoparticles

For preparation of PbO nano-particles calcinations of nano powder compound **1** was done at 500 °C in static atmosphere of air for 4h. IR spectrum and powder XRD diffraction show that calcination was completed and the entire compounds were decomposed.

3. Results and discussion

Reaction between the organic nitrogen and oxygen -donor ligand (8-hydroxyquinolin) (L), sodium per chlorate and lead (II) acetate yielded crystalline material formulated as a new 0D CSC $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2(\mathbf{1})$. Single crystal X-ray diffraction analysis (Tables 1–2) of compound **1** was carried out and reveals that $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2(\mathbf{1})$ complex crystallizes in triclinic space group *P*-1. Also, shows compounds **1** is 0-D coordination supramolecular. $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2(\mathbf{1})$ is air-stable and high-melting solid that is soluble in DMSO.

In compound **1**, Pb atoms are coordinated by two N atoms, three O atoms and have square pyramid coordination sphere as O_3N_2 . In the crystal molecules lie on crystallographic binary axes, so the asymmetric unit contains half of a molecule (Fig 1). Pb–N and Pb–O distances are about 2.39–2.50 Å (Table-2 and Fig. 2). In compound **1** strong bond construct the monomer complex $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2$, which is expanded in supramolecular monomer complex $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2$ by weak interactions. At least compound **1** is a supramolecular compound with molecular structure (Fig. 3). The simplification and classification of the monomer molecules by Topos Pro in compound **1** gives the underlying topology 1,2,3,4,4M12-1, which is abundant for 0D coordination compounds (more than 36 examples in TTO collection of Topos Pro (Fig.4) [62]. Every $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2$ monomer is connected to other monomers by van-der-Waals bonds H–H, C–H, O–H, C–N, N–O, but the area of these contacts between central molecule and 15 surrounding molecules is different. Thus, the largest area of interactions are provided π - π stacking of L ligands (Fig. 3). There are only 2 molecules having π - π contacts with the central one, they form molecule packing which additionally have O–C, C–H, N–C and H–H interactions. Obviously π - π interactions play major role in formation of the molecular packaging by surrounding each molecule in geometry of connection(Fig 5).

The elemental analysis and IR spectra of the nanoparticles and the single crystalline material of compound **1** are indistinguishable and they exhibit the expected absorption bands in the IR (Fig. 6). The IR spectra of compound **1** show absorption bands with a variable intensity in the frequency range 1227–1493 cm^{-1} correspond to vibration of the pyridine rings of ligand 8-HQuin. The relatively weak absorption bands at 3040 cm^{-1} are due to the C–H modes involving the aromatic ring hydrogen atoms.

Figure 7 shows the simulated XRD pattern from single crystal X-ray data of compound **1** in comparison with the XRD pattern of nano typical sample of compound **1** prepared by sonochemical process. There is an acceptable match with some minor differences between the simulated (Fig. 7a) and experimental powder X-ray diffraction patterns (Fig. 7b). This indicates that the compound obtained sonochemically is essentially identical to that obtained by conventional single crystal evaporation. Estimated from the Sherrer formula for the calculation of particle sizes from the broadening of the XRD peaks $D=0.891\lambda/\beta\cos\theta$, where D is the average grain size, λ is the X-ray wavelength (0.15405 nm), and θ and β are the diffraction angle and full-width at half maximum of an observed peak, respectively, the average size of the particles was found to be around 115 for compound **1** which is in agreement with the value obtained from the SEM image (Fig. 8). For further investigation we used other solvents such as acetonitrile by the sonochemical process; however, particles size of compound were not in nanoscales.

PbO nanoparticles can be generated by thermal decomposition of nano powder of compound **1** at 500°C for 3 h. The final product upon calcination of the compound **1**, based on XRD pattern (Fig. 9), is PbO. These particles show a high degree of crystallinity, with sharp and well defined peaks in the XRD pattern. PbO particles are the only crystalline phases present and all diffraction

peaks were indexed with PbO with $a = 5.464 \text{ \AA}$ and S.G= Pnam which is in ICSD card file No.189978

Fig.10 shows SEM image and the corresponding particle size distribution histogram of PbO particles, produced by calcinations of compound **1** at 500 °C.

4. Conclusion

One new Pb(II) coordination supramolecular compound $[\text{Pb}_4(8\text{-Quin})_6](\text{ClO}_4)_2(\mathbf{1})$ (L=8-HQuin=8-hydroxyquinolin ligand), has been synthesized utilizing a thermal gradient approach and also by sonochemical irradiation. Compound **1** was structurally characterized by single crystal X-ray diffraction. The crystal structure of compound **1** is made up of 0D supramolecular compound and shows the coordination number in the Pb(II) ions in compound **1** is five. The direction of bonds in **1** shows that coordination around the Pb(II) ion in this complex is hemidirected geometry with a stereochemically active lone pair as expected. Additionally, the X–Pb–Y angle in this complex suggests that there is a hole in the coordination sphere due to lone pair-bond pair repulsion. Nano-particles of compound **1** has been characterized by scanning electron microscopy (SEM), X-ray powder diffraction (XRD), elemental analyses and IR spectroscopy. Nano-particles of Pb(II) oxide has been prepared by calcination of Pb(II) CSC. Pure phase PbO nanoparticles were obtained by calcinations of compound **1** at 500 °C for 3 h. The nano oxide was characterized by scanning electron microscopy (SEM) images, X-ray powder diffraction (XRD) and IR spectroscopy.

Supplementary material

Crystallographic data for the structures reported in the paper has been deposited with the Cambridge Crystallographic Data Centre as supplementary publication No: CCDC-1517108. Copies of the data can be obtained on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK [fax: +44 1223/336033; e-mail: deposit@ccdc.cam.ac.uk].

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Table 1. Crystal data and structures refinement for [Pb₄(8-Quin)₆](ClO₄)₂ (1).

Empirical formula	C ₅₄ H ₃₆ Cl ₁₂ N ₆ O ₁₄ Pb ₄
Formula weight	1892.55g/mol
Temperature	291(2) K
Wavelength	0.71069 Å
Crystal system	Triclinic
Space group	P -1
Unit cell dimensions	a = 10.248(5) Å, α = 65.520(5)° b = 11.768(5) Å, β = 75.865(5)° c = 12.668(5) Å, γ = 82.445(5)°
Volume	1347.5(10) Å ³
Z	1
Density(calculated)	2.332 Mg/m ³
Absorption coefficient	12.629 mm ⁻¹
F(0 0 0)	876
Crystal size	0.18 × 0.13 × 0.11 mm ³
Theta range for data collection	1.81 to 26.00°
Reflections collected	10814
μ	12.63mm ⁻¹
Index ranges	-10 ≤ h ≤ 12 -14 ≤ k ≤ 14 -15 ≤ l ≤ 15
Max. and min. transmission	0.3372 and 0.2096
(sin θ/λ) _{max}	0.617 Å ⁻¹
Theta(max)	26.0°
Radiation type	Mo Kα
Refinement method	Full-matrix least-squares on F ²
Data/restraints/parameters	5213 / 0 / 355
Goodness-of-fit-on F ²	0.991
Refinement	R[F ² > 2σ(F ²)] = 0.050 wR(F ²) = 0.111 S = 0.99
R1 [I > 2σ(I)]	10814, 5213, 3606
Rint	0.0590
T _{min} , T _{max}	0.210, 0.337
Largest diff. peak and hole	1.564 and -2.744 eÅ ⁻³
CCDC no.	1517108

Table 2. Selected bond lengths/ Å° for compound $[\{\text{Pb}_4(8\text{-Quin})_6\}(\text{ClO}_4)_2]$ (1).

Pb(1)—O(1)	2.269(8)	C(10)—C(11)	1.42(3)
Pb(1)—O(3)	2.391(7)	O(1)—Pb(1)i	2.640(7)
Pb(1)—N(1)	2.506(11)	C(23)—C(22)	1.435(17)
Pb(1)—O(1)i	2.640(7)	C(8)—C(9)	1.38(2)
Pb(1)—O(2)	2.675(8)	C(8)—C(7)	1.398(19)
Pb(2)—O(2)	2.254(8)	C(8)—H(8)A	0.9300
Pb(2)—O(3)	2.419(8)	C(16)—C(17)	1.39(2)
Pb(2)—N(3)	2.502(11)	C(5)—C(6)	1.435(16)
Pb(2)—N(2)	2.536(12)	C(9)—C(4)	1.40(2)
Cl(1)—O(5)	1.329(18)	C(9)—H(9)A	0.9300

(i) 1-x, 1-y, -z.

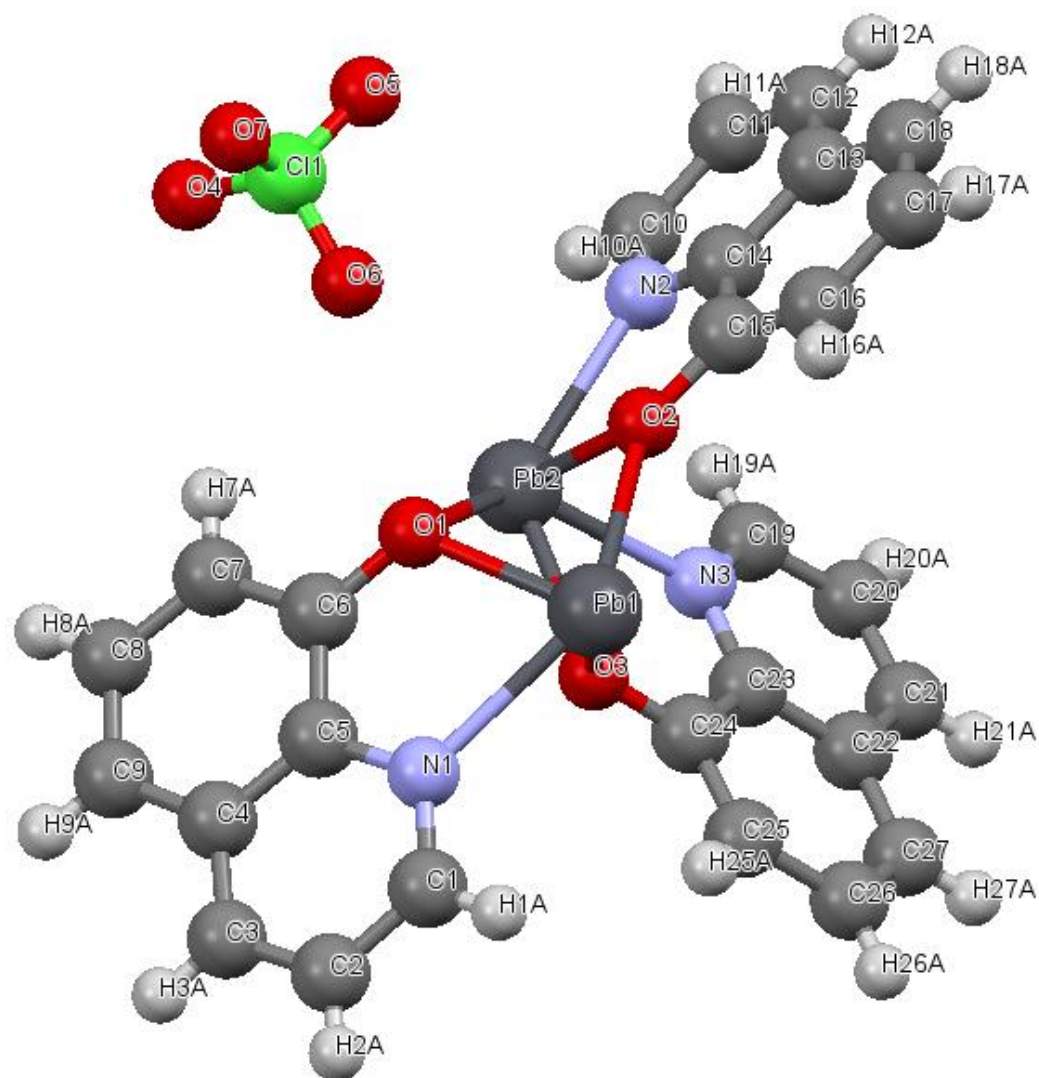


Figure 1. Asymmetric unit cell of compound **1** (which is as part of the crystallographic unit cell)

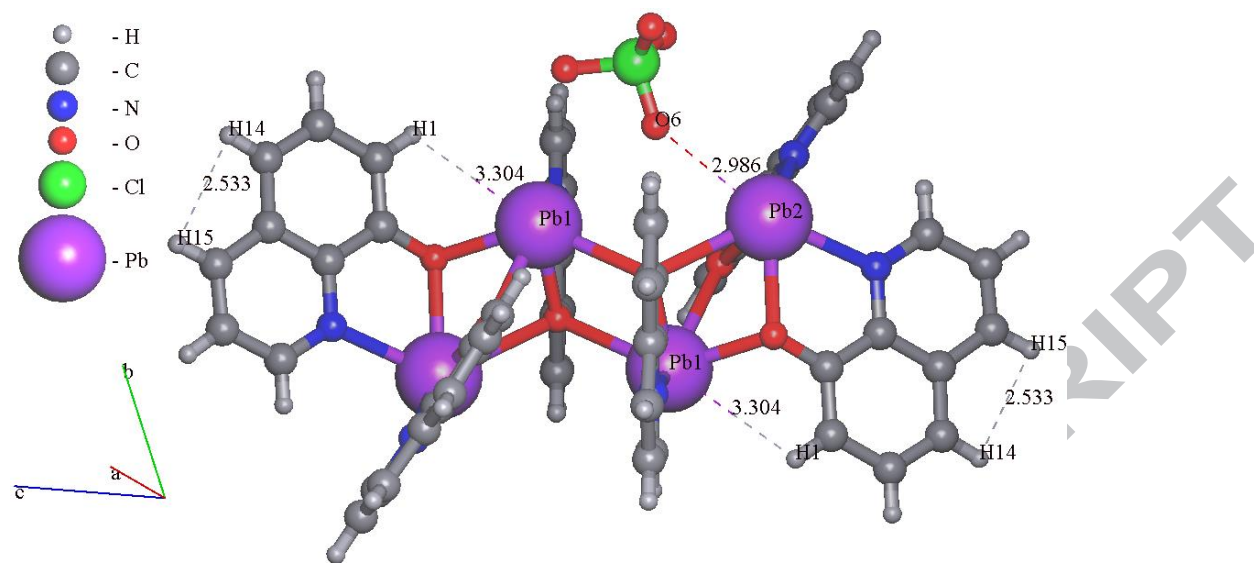


Figure 2. Dashed lines represent weak interaction and distance of bounds in compound **1**.

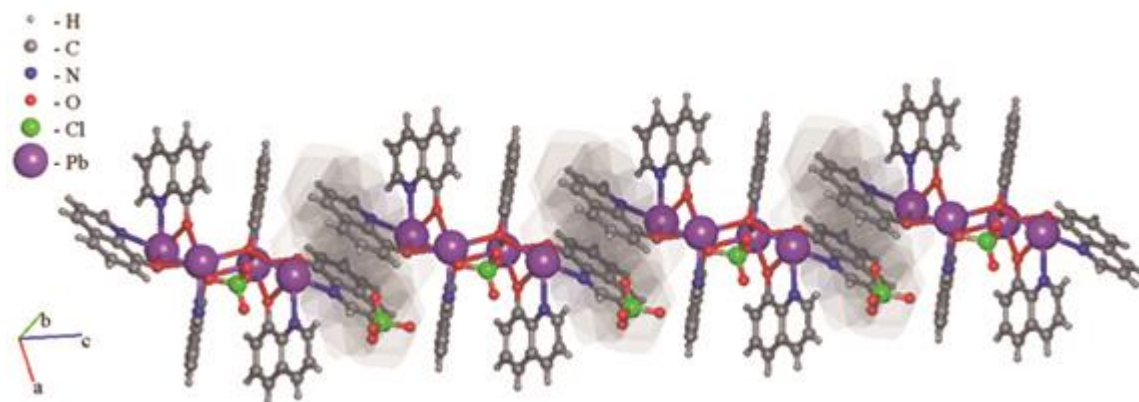


Figure 3. Four monomers in compound 1.

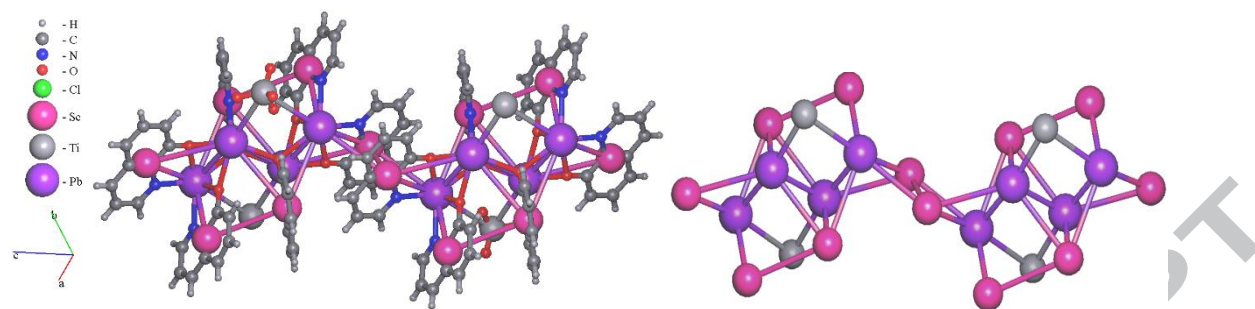


Figure 4. Topological representation of coordination networks in compound 1.

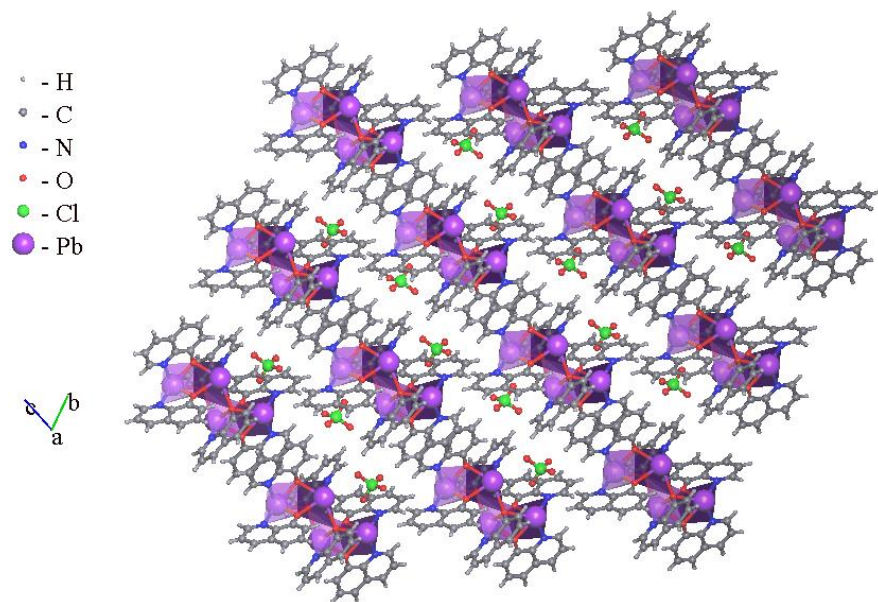


Figure 5. A fragment of the 3D framework in compound 1.

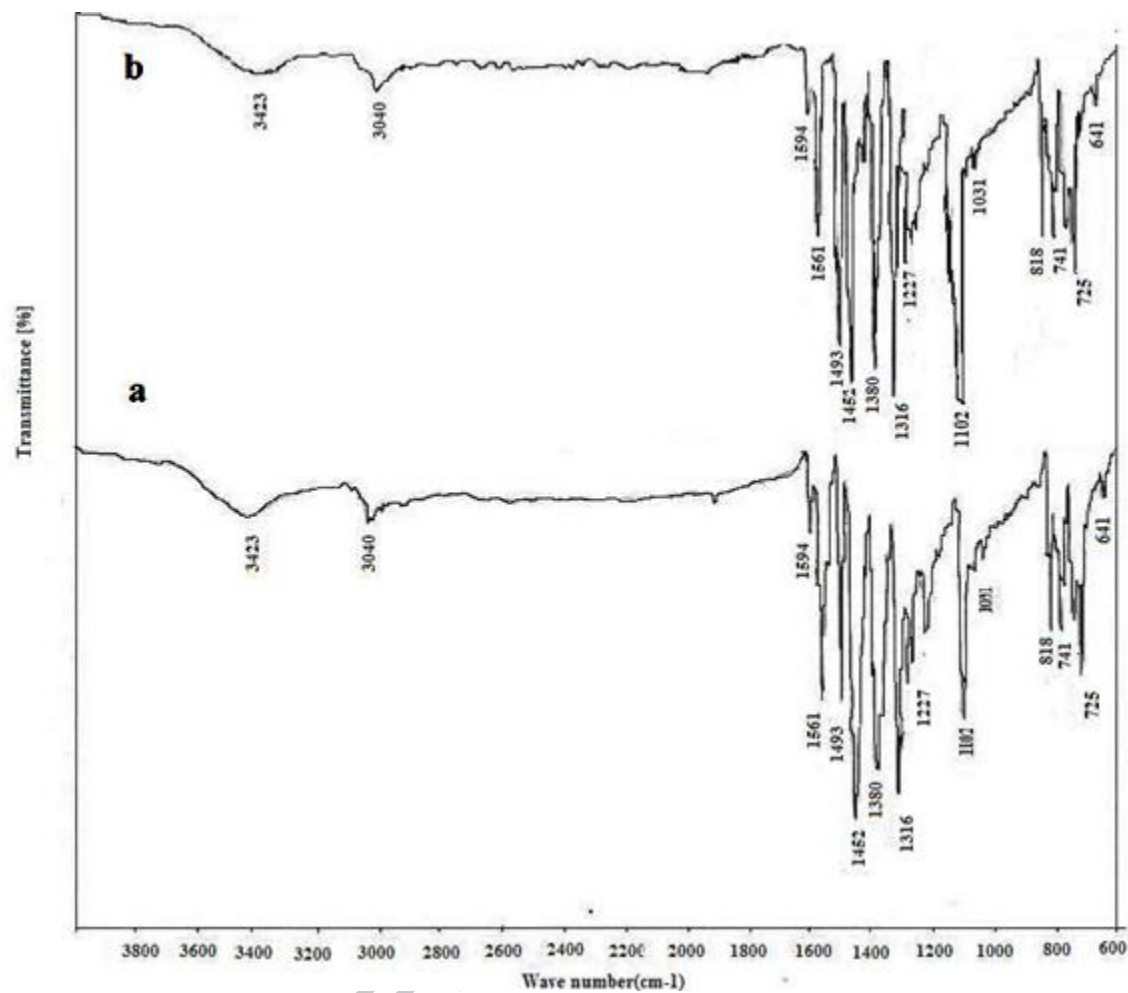


Figure 6. The IR spectra of (a) bulk materials as synthesized of compound 1, (b) nano-sized compound 1 prepared by sonochemical method

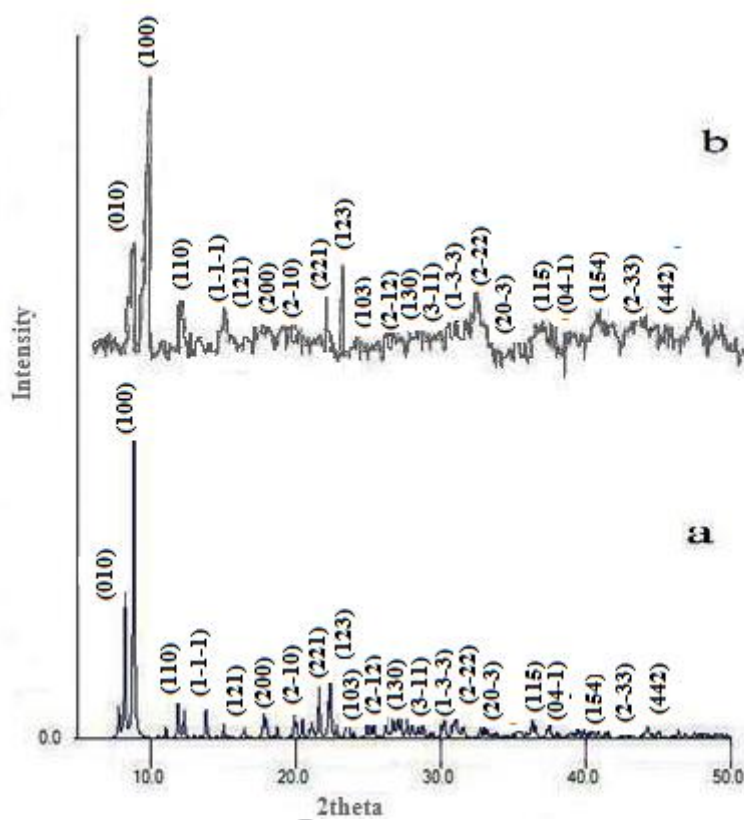


Figure 7. XRD patterns: (a) simulated pattern based on single crystal data of compound 1, (b) nano particles of compound 1.

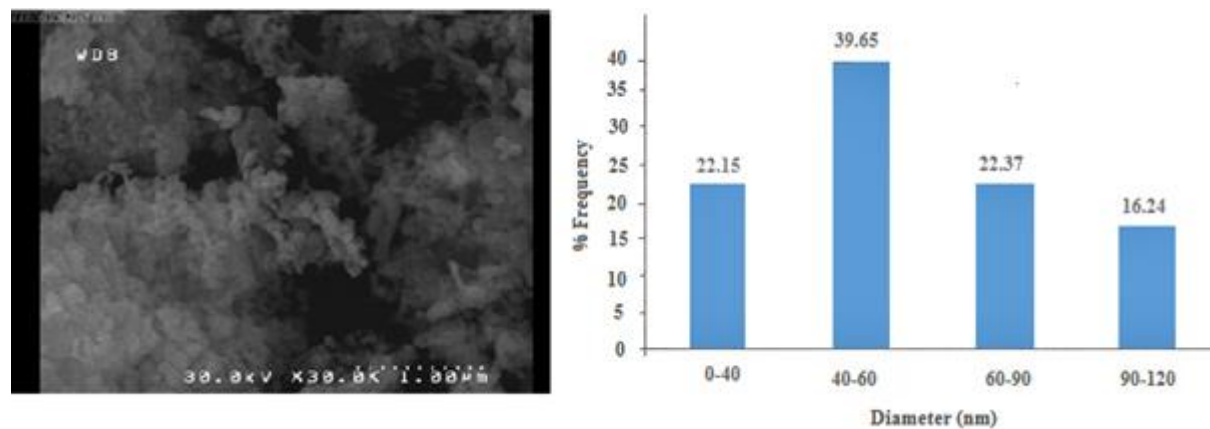


Figure 8. SEM image and the corresponding particle size distribution histogram of compound 1 nanoparticles.

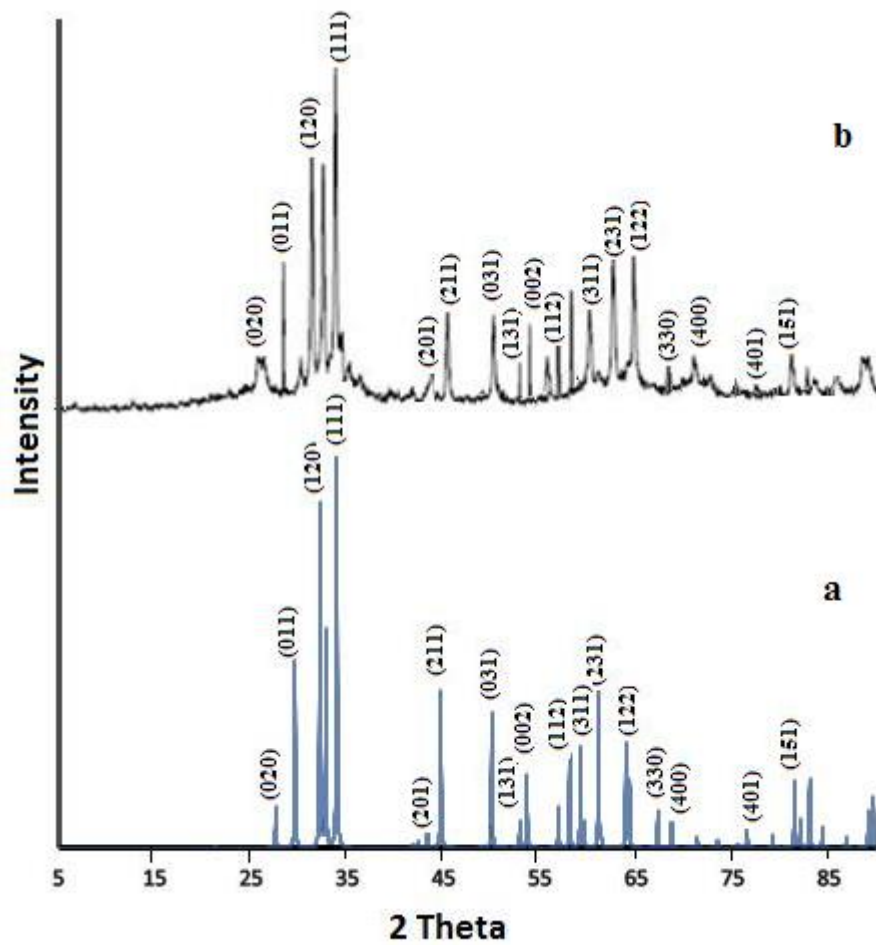


Figure 9. XRD pattern of PbO (a) simulated pattern based on single crystal data

(b) nanoparticles prepared by calcination of nano powder compound 1.

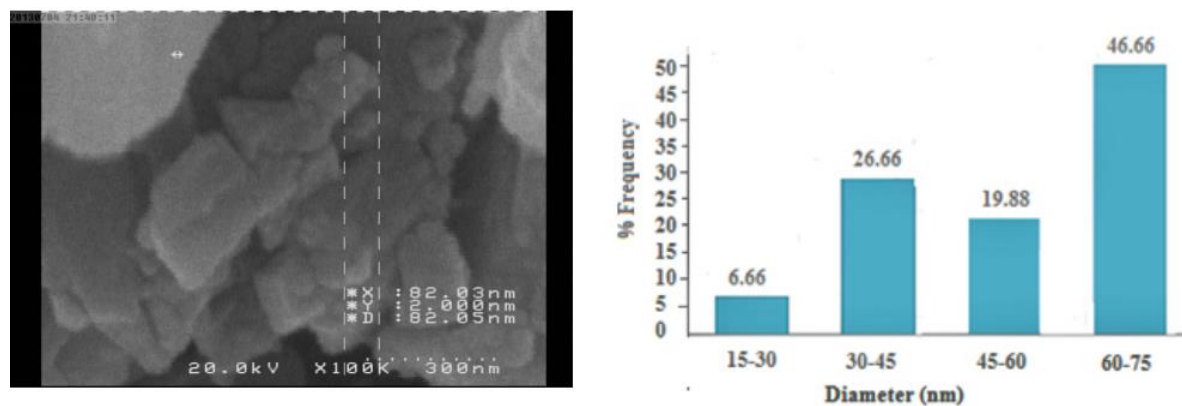


Figure 10. SEM image and the corresponding particle size distribution histogram of PbO nanoparticles prepared by calcination of nano powder of compound 1 at 500 °C.

> Nanoparticles of a lead(II) coordination supramolecular compound (CSC), have been synthesized by a sonochemical process.

> The nanoparticles were characterized by scanning electron microscopy, X-ray powder diffraction, Fourier transform infrared spectroscopy and elemental analyses.

> The topological analysis of the compound was studied

> lead (II) oxide nanoparticles have been prepared by calcination of lead (II) CSC.