



SYNTHESIS, CHARACTERIZATION AND APPLICATION OF BARIUM TUNGSTATE: A COMPREHENSIVE REVIEW

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Abstract: BaWO₄ crystallizes into a space group I41/a tetragonal scheelite structure. Nanocrystalline BaWO₄ can be produced from a variety of salts, and the synthesis process, reaction temperature, precursor concentration, solution pH, and reaction duration all affect the shape of the particles. The synthesis process and the level of crystal lattice distortion determine the optical characteristics of BaWO₄. This is a wide band gap semiconductor with blue luminescence and is an excellent host material for doping. Long luminescence life periods, large Stokes/anti-Stokes shifts, crisp band emissions, and exceptional photo-stabilities are characteristics of rare earth activated tungstate luminous materials.

Keywords: BaWO₄; structural properties; optical properties; doping, photoluminescence

1. Introduction

One of the 21st century's emerging technologies is nanotechnology. Studying and interacting with matter at incredibly small scales is the focus of nanoscience and nanotechnology. A novel approach to creating intricate materials and gadgets by precise manipulation of matter's properties and assembly at the nanoscale length scale is called nanoscience. The capacity to regulate or work with particle size at the atomic scale is strengthened by it. The physics and interactions between atoms exhibit unique characteristics at the nanoscale that are absent at bigger dimensions. Every attribute of matter changes to allow for specific applications as the grain or particle size in solid matter is reduced to nanoscale ranges. For example, inert materials can become catalysts, opaque substances can become transparent, stable materials can become flammable solids, and insulators can become conductors.

A nanocrystal's composition, structure, phase, shape, size, and size distribution all affect its properties. Furthermore, the success of "bottom-up" approaches towards future nanodevices depends on the architectural control of nanosized materials with well-defined geometries. [1-3]. Because of their exceptional size and shape-dependent characteristics and wide range of possible applications, inorganic nanoparticles with controllable and uniform size and shape have garnered a lot of attention over the past ten years [4–13]. To put it briefly, one of the main goals of the development of nanotechnologies is the control of the shape of the nanoparticles [10,11]. For the controlled synthesis of polyhedral, other synthetic techniques have already been devised [14–20].

Because of their unique optical, electrical, magnetic, and thermal characteristics as well as their potential use in gas sensors, photoelectronic devices, and catalysts, metal oxide nanoparticles have been the subject of extensive research. The structure and luminescent characteristics of metal tungstate nanoparticles are highly intriguing. The heaviest tungstate in the family of alkaline earth tungstates is barium tungstate (BaWO₄). BaWO₄ crystallizes in the tetragonal scheelite type structure at ambient circumstances, just as a lot of other compounds of the ABX₄ type. Due of its superior magnetic, photoluminescence, and electrical conductivity qualities, BaWO₄ has to be thoroughly studied [21, 22]. BaWO₄ based materials are among the most reactive

alkaline earth tungstates and can be used in a wide range of technical applications, including optical filters, humidity sensors, light-emitting diodes, and scintillators.

2. Nanocrystalline BaWO₄

Due to their intriguing structural and photoluminescence characteristics, metal tungstates with the formula MWO₄ have garnered a lot of interest [25–29]. Scintillation counters, lasers, and optical fibers have all found use for these materials [30, 31]. A number of the divalent transition metal tungstates have found commercial use in fluorescent lights and lasers, while others are particularly significant because of their magnetic and electrical conductivity. Furthermore, these materials find use as humidity sensors and catalysts [32, 33]. In MWO₄ compounds, M²⁺ will be in the wolframite-type monoclinic structure, where the tungsten atom takes on an overall six-fold coordination, if its ionic radius is less than 0.77 Å (Ni = 0.69) [34]. Nonetheless, in larger bivalent cations with an ionic radius greater than 0.99 Å (Ba=1.35), the tungsten atom takes on tetrahedral coordination and the structure is referred to as the scheelite-type tetragonal structure [35].

BaWO₄ crystallizes into a space group I4₁/a tetragonal scheelite structure. The Ba and W atoms in the BaWO₄ unit cell have an S₄ point symmetry, which coordinates the tungsten atoms to four oxygen atoms in a tetrahedral arrangement. The bonding angles between O–W–O in the somewhat deformed oxygen coordination polyhedra are 108.56° and 111.30°. The site symmetry is D_{2d} because the barium atoms are coordinated to form BaO₈ polyhedrons, which are eight oxygen atoms located in the corners of the tetragonal unit cell [36, 37]. The O atoms occupy the C1 sites, while the site symmetries of the Ba and W atoms are S⁴ and Td symmetry [38]. For the tetrahedral WO₄^{2−} anions and Ba²⁺ cations, the crystal structure is strongly ionic [39]. Figure 1.1 depicts the tetragonal BaWO₄ crystal structure.

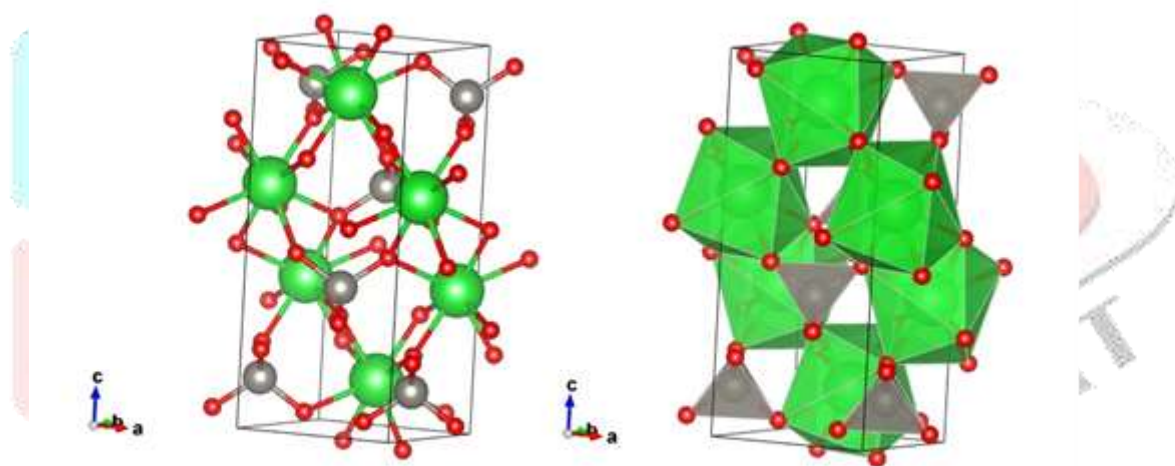


Figure 1.1 Crystal structure of tetragonal BaWO₄

3. Literature Overview

It has been reported that MWO₄ type oxides with scheelite structure, where M=Ca, Sr, Ba, Pb, and Cd, are helpful as humidity sensors [44], scintillators [41], oxide ion conductors [42, 43], and laser host materials [40]. Solid-state method [45,46], co-precipitation [47], chemical precipitation [48][49], solvothermal [50], sol-gel, reverse micellar reactions [51], microwave hydrothermal [39,52,53], cyclic microwave irradiation [54], sucrose template method [35], sonochemical method [55,56], combustion [57], hydrothermal [58], and reverse microemulsion [59] were some of the techniques used to synthesize BaWO₄ nanoparticles. The solution-based chemical synthetic methods are highly significant in the design and manufacture of fine materials. They have been effective in surmounting numerous constraints associated with the conventional solid-state, high-temperature approaches. Major issues including agglomeration, extensive diffusion routes, and contaminants can also be removed, producing goods with better homogeneity. In this work, the chemical precipitation approach is used since it is relatively easy, less expensive, and requires no complex equipment.

Several salts, including BaCl₂, Ba(NO₃)₂, Na₂WO₄, BaCO₃, WO₃, and others, have been used in the literature as precursors for the synthesis of nanocrystalline BaWO₄[60–63]. The primary variables affecting the size and shape of the nanoparticles are the synthesis process, reaction temperature, precursor concentration, solution pH, and reaction duration [61, 64–66]. The surface shape of BaWO₄ microstructures

produced via microemulsion mediated by the hydrothermal method is influenced by the concentration of surfactant and the length of the hydrothermal treatment, according to Liu et al.'s findings [67]. Zhang et al. used the solvothermal approach in 2005 to create bundle-like nanorod arrays that are 1-2 μm long and 200-300 nm wide, with a diameter of 15 nm for each nanorod [50]. Shi et al. [60] described the fabrication of penniform BaWO_4 nanostructures consisting of nanowires or nanobelts guided by a block copolymer in cationic reverse micelles. With the use of polymethacrylic acid (PMAA) and a precipitation process between BaCl_2 and Na_2WO_4 solutions, Zhao et al. created a variety of BaWO_4 hollow structures, such as spheres, peanuts, and ellipsoids [61]. Hernandez-Sanchez et al. produced BaWO_4 nanodots by regulating the temperature during the solution precipitation process [64]. BaWO_4 powder with an octahedral-like morphology was synthesized by Cavalcante et al. using the co-precipitation method and processed in a home microwave-hydrothermal (MH) at 413 K for various periods [47]. Kwan et al. used a reversed micelle templating technique to create unique BaWO_4 nanorod superstructures [51]. Fishbone-like nanostructures were created by Zhang et al. using the solution growth technique in w/o microemulsions [68]. They discovered that the product's morphology is greatly influenced by the molar ratio of cations to anions and that the water content has a significant impact on the product's size. The literature has reported on the straightforward microwave-assisted method of synthesizing BaWO_4 crystals with various morphologies, including nanosheets, nanobelts, flowerlike, quadrangled plates, and sheaves of dendrites [23]. By changing the reaction parameters, such as the molar ratio of $[\text{Ba}^{2+}]/[\text{WO}_4^{2-}]$, the pH value of the starting solution, and the concentration of PVP aqueous solutions, the morphologies of the samples were adjusted.

Using various surfactants allows for control over the samples' crystallite size and surface shape. By using a hydrothermal process and several surfactants, Xie et al. were able to successfully manufacture BaWO_4 crystals with a variety of morphologies, including olive-like, flake-like, and whisker-like structures [58]. Zhang et al. used glycerol as a capping agent and cosolvent with water in a surfactant-free hydrothermal process to generate dumb bell-like BaWO_4 microstructures [58]. The use of various surfactants and capping agents to regulate the size and shape of BaWO_4 nanoparticles was investigated by Talebi et al. [69]. The BaWO_4 nanocrystals produced by a reverse microemulsion process have different surface morphologies and sizes depending on the molar ratio of water to surfactant and reactant concentration [59]. Zawawi et al. [70] investigated the porosity of the BaWO_4 powder produced by the sucrose solution evaporation method.

By adjusting the surface chemistry of the crystals, which is connected to the relative stability of the faceted crystals, Oliveira et al. presented a theoretical model based on the Wulff architecture that describes the potential crystal morphologies. The BaWO_4 crystals were found to include (112), (001), and (100) facets with low surface energy values, according to both theoretical and experimental evidence [49].

Numerous studies have examined the impact of calcination temperature on the characteristics of BaWO_4 nanoparticles produced using various techniques. When the calcination temperature was raised, Lim et al. examined the alterations in surface morphology and PL emission intensity of the BaWO_4 powders produced via the solid-state metathetic method [71]. Following calcinations, Anicete-Santos et al. discovered structural alterations and fluctuations in PL emission intensity of the BaWO_4 powder [53]. Sadiq et al. [48] showed that the BaWO_4 nanoparticles produced by the chemical precipitation approach had altered surface shape, increased particle size, and increased PL intensity.

BaWO_4 has a tetragonal scheelite-type structure at room temperature and pressure; at high pressure and temperature, however, it becomes monoclinic. There is no clear similarity between the scheelite and wolframite types and the monoclinic BaWO_4 that is generated at high pressure and temperature [72]. According to Kawada et al., barium atoms are found in the spaces between the closely spaced, two-dimensional WO_6 octahedra that are joined by an edge and corner-sharing in monoclinic BaWO_4 . When comparing the barium atoms' coordination number to the structure under normal circumstances, it has risen. At a pressure of 5 GPa and a temperature of 621 K, Lacomba-Perales et al. detected the transition from the scheelite phase to the monoclinic BaWO_4 -II phase [73]. Tan Da-Yong et al. stated [74] that above 7 GPa, the structural alteration of BaWO_4 crystals generated by the solid-state technique from tetragonal scheelite type to monoclinic fergusonite type was observed. They noticed that the pressure and axial parameter were constantly changing. They also demonstrated how the tiny displacive distortion of rigid WO_4 tetrahedrons and the substantial compression of soft BaO_8 polyhedrons couple to produce the first-order feature of this phase transition. Understanding the scheelite to fergusonite transition in additional scheelite-structured compounds, such as molybdates, germanates, and silicates, will be aided by such coupling. Gomis et al. also examined the

change in BaWO₄ single crystal from its low-pressure tetragonal form to a significantly denser monoclinic structure, which was created using the Czochralski process. Using GSAS software, Rietveld refinements of the low- and high-pressure phases' crystal structures from the quasi-hydrostatic tests were carried out [75].

Zhang et al. used XPS spectra to investigate the chemical and bonding environment of the BaWO₄. Ba is present in the Ba²⁺ state, as shown by the peaks seen in the region corresponding to the Ba 3d binding energy at 780.2 eV (Ba 3d_{5/2}) and 795.6 eV (Ba 3d_{3/2}). W is in its [WO₄]²⁻ (W⁶⁺) state, as indicated by the peaks at 35.6 and 37.8 eV, which correspond to the W 4f state. There was also a strong oxygen 1s peak at 531.0 eV, which is indicative of oxygen in its O²⁻ oxidation state [58].

The production technique and the degree of crystal lattice distortion determine the optical characteristics of BaWO₄ [76]. With a band maximum occurring at 220-250 nm, BaWO₄ has a single absorption band that corresponds to the ligand to metal charge transfer in the region 218-274 nm. The degree of crystal lattice distortion determines the precise location of the maximum [77]. In the (WO₄)²⁻ group, excitation from O_{2p} to W_{6s} occurs through absorption of UV radiation [35]. According to published research, BaWO₄ is a direct bandgap semiconductor and that barium has no discernible impact on the sample's bandgap [78]. Because of the radiative transition of self-trapped excitons within the (WO₄)²⁻ tetrahedral group, the BaWO₄ emits a wide blue luminescence [79]. A wide range of temperatures was observed by Nikl et al. when they observed the excitonic luminescence of BaWO₄. They concluded that: (i) emission originates from the lower-lying triplet state split by Jahn-Teller (JT) interaction; (ii) both triplet states affected by JT interaction contribute to the observed emission; and (iii) emission from one or both triplet states is additionally affected by the presence of the low-symmetry local fields because the symmetry of the emitting center is not perfectly tetrahedral [80]. BaWO₄ thin films made using the polymeric precursor technique had an optical bandgap of 5.78 eV, according to measurements made by Pontes et al. [81]. Blistanov et al. [82] observed the presence of two broad bands in the photoluminescence (PL) emission of BaWO₄, one in the green area (520-530 nm) and another in the blue region (about 450 nm). Zhang et al. claim that the size of the nanoparticles and the film-forming impact determine the precise location of the PL emission peak in BaWO₄ film [21]. According to Lima et al., depending on the milling period, the optical bandgap of BaWO₄ powders physically disordered by high-energy mechanical milling varied between 5.76 and 5.23 eV. The strength of the broad PL emission they saw, which ranges from 500 to 800 nm, is dependent on the milling duration. Thus, they deduced that a crucial requirement for producing a strong and widespread PL emission is structural disorder in the lattice [83]. For BaWO₄ single crystal, Lacomba-Perales et al. measured the optical bandgap and found 5.26 eV [84]. The BaWO₄ sample produced by Parhi et al. using the solid-state metathetic method with microwave energy assistance has a bandgap of 4.8±0.2 eV [85]. Sadiq et al. [48] found that for the BaWO₄ powder prepared by the chemical precipitation process, there was an increase in bandgap with calcination temperature (5.77-5.88 eV). However, BaWO₄ nanoparticles made by sonochemical means yielded a bandgap of 3.2 eV according to Khademolhoseini et al. [56].

Tyagi et al. examined the photoluminescence of BaWO₄ single crystals that were cultivated using the Czochralski method within a temperature range of 77–300 K. An ultraviolet emission band was detected at normal room temperature, however it was completely extinguished after undergoing vacuum annealing. The blue emission resulting from radiative transitions within the (WO₄)²⁻ was present in all samples [86]. In the study conducted by Zhang et al., it was observed that the BaWO₄ microstructures exhibited photoluminescence (PL) emission peaks at wavelengths of 387 and 426 nm [58]. Cavalcante et al. observed that the optical bandgap of BaWO₄ powder, which was produced using the microwave hydrothermal method, exhibited a range of values (4.4-3.84 eV) depending on the duration of the processing. The literature [39] reported a green photoluminescence caused by aberrations in the WO₄ tetrahedral group. Anicete-Santos et al. discovered that the presence of a disorder in the arrangement of Ba atoms at a small distance is the cause of the green photoluminescence emission observed in BaWO₄ powders made using the polymeric precursor approach [53]. Phuruangrat et al., Shen et al., and Lim et al. reported the presence of blue luminescence in BaWO₄ powders that were produced by cyclic microwave irradiation [54, 65, 71]. The emission peak and intensity of photoluminescence (PL) emission exhibited variations in response to changes in reaction time and preparation temperature [65,71]. According to Lim et al., the shoulder peaks observed in the PL spectrum can be attributed to the Jahn-Teller splitting effect and the Frenkel defect [87]. Wu et al. discovered that the photoluminescence intensity of BaWO₄, which was produced using microwave synthesis, may be altered by employing various surfactants [88].

Cerny et al. [89] investigated the stimulated Raman scattering (SRS) of 1.06- μm , 50-ps pulses in BaWO_4 and KGW ($\text{KGd}(\text{WO}_4)_2$) crystals. A Raman shift of 925 cm^{-1} was detected in BaWO_4 , a highly promising material characterized by its significant spectroscopic peak intensity and integral intensity. They suggested BaWO_4 , which has an efficiency of 50%, as a potential candidate for use in solid-state Raman Laser systems. It can be utilized across a broad range of pumping radiation wavelengths and with pulses lasting from picoseconds to nanoseconds. Ge et al. [90] conducted a study on the thermal and mechanical properties of the BaWO_4 crystal produced using the Czochralski process. The crystal's density exhibited a linear decline as temperature increased, indicating favorable thermal conductivity properties in the produced crystal. The crystal's microhardness measurement indicated that it possessed a low level of hardness, making it susceptible to easy processing and polishing.

According to Basiev et al. [91], BaWO_4 crystals are capable of serving as a very effective source for Raman lasing on both picosecond and nanosecond time scales. Raman lasers utilizing BaWO_4 and $\text{Ba}(\text{NO}_3)_2$ Raman crystals were produced and studied. Various stocks were used to obtain components that had a maximum wavelength of around 2.2 μm , achieving an efficiency of up to 10%. The researchers conducted a study on stimulated Raman scattering within the mid IR region (2.31-2.75-3.7 μm) using a BaWO_4 crystal. The crystal was subjected to pumping at wavelengths of 1.9 and 1.56 μm . In a BaWO_4 crystal, the first Stokes component at a wavelength of 2.31 μm and the fourth Stokes components at wavelengths up to 2.75 μm and 3.7 μm were generated using 1.9 μm and 1.56 μm pumping, respectively [92]. Lasers within this specific range of wavelengths are utilized in several fields such as medical, air communication lines, and lidar technology. Zverev et al. conducted an analysis on the UV and IR absorption in BaWO_4 crystals and found that these crystals have the potential to be highly effective in Raman lasers operating in the visible and near IR spectral bands (264 – 5300 nm) [93].

Zhang et al. conducted a study on the electronic structures, dielectric function, and absorption spectra of both the pristine BaWO_4 crystal and the BaWO_4 crystal with a barium vacancy ($\text{V}^{\text{Ba}^{2-}}$) using density functional theory. The findings revealed that the BaWO_4 crystal has anisotropic optical properties, and its optical symmetry aligns with the lattice structure geometry of the crystal. The 370 nm absorption band may be attributed to the presence of a barium vacancy [94].

There is a lack of comprehensive reports regarding the magnetic characteristics of the BaWO_4 samples. According to VSM tests conducted by Khademolhoseini et al. [56] and Talebi et al. [69], BaWO_4 samples exhibited ferromagnetic properties, with a saturation magnetization of 0.007 emu g^{-1} .

Vidya et al. conducted a study on the temperature dependency of the dielectric characteristics of BaWO_4 powders that were sintered at 810°C. They discovered that the dielectric constant exhibits minimal variation with temperature within the tested range. At room temperature, the loss factor is 1.56×10^{-3} and it reduces as the temperature rises. The dielectric constant has a value of 9 at ambient temperature and 8.5 at 250°C when the frequency is 5 MHz [57]. In addition, they suggested employing the BaWO_4 material for LTCC applications [57]. Krzmanc et al. discovered that BaWO_4 ceramics, produced via the solid-state process, exhibit a consistent dielectric constant value of 9 within the temperature range of 800-1100°C, when measured at a frequency of 11 GHz [95].

BaWO_4 has been extensively studied as a highly promising material for the photocatalytic degradation of organic dyes in water when exposed to UV light. The photocatalytic activity of the samples is influenced by the pH of the solution, the concentration of BaWO_4 nanoparticles, and the starting concentration of the dye. The photocatalytic efficacy of BaWO_4 particles, produced using the sonochemical technique, was evaluated by examining their ability to adsorb methylene blue and rhodamine B. The results indicated that the dyes were completely absorbed within a time frame of 10-15 minutes [55]. Sadiq et al., Khademolhoseini et al., and Talebi et al. have also documented the use of BaWO_4 catalyst for the decomposition of rhodamine B [48] and methyl orange [56,69]. Vijay et al. documented the process of photodegradation of Azure B by employing BaWO_4 as a catalyst. The deterioration rate reached its peak at a pH of 7.3 [96].

Doping is a successful technique for enhancing the surface structure and optical characteristics of BaWO_4 samples [97]. Multiple publications exist regarding the investigations of BaWO_4 nanoparticles doped with rare earth elements. Tungstate luminous materials activated by rare earth elements possess exceptional photostability, significant Stokes/anti-Stokes shifts, prolonged luminescence life spans, and distinct band emissions

[26]. Below is a comprehensive literature study that provides complete information about that specific property.

The BaWO₄ material can exhibit different luminescent emissions when doped with Eu³⁺, Tb³⁺, and Tm³⁺ ions [98–103]. Jinsheng et al. discovered that the addition of Pr³⁺ ions to BaWO₄ would result in a deep red emission [104]. Cavalcante et al. [105] examined the peaks resulting from f-f transition in the blue region of Pr³⁺ in Pr³⁺ doped BaWO₄. Research conducted on the BaWO₄ sample doped with Dy³⁺ reveals that many emission lines can be observed, which correspond to the f-f transition in Dy³⁺ [106–108]. Doping with rare earth elements Nd³⁺, Er³⁺, and Yb³⁺ enables the achievement of photoluminescence emission in the NIR range. Hou et al. reported a significant emission band at 977 nm in BaWO₄:Yb samples, which have potential uses in telecommunication and laser emission [109,110].

According to Chao et al., BaWO₄:Yb³⁺/Tm³⁺ nanocrystals exhibited four distinct photoluminescence emission lines with wavelengths of 454, 475, 647, and 790 nm when excited at a wavelength of 980 nm [111]. The emission intensity reached its peak when the concentration of Tb³⁺ was 1 mol%, but it decreased when the concentration exceeded this value. Yb³⁺ functions as a sensibilization center. Multiple observations indicate that the addition of Ce³⁺ and Ce⁴⁺ can enhance the strength of blue luminescence emission in BaWO₄ samples [112–115].

4. Conclusion

The morphology and size of the BaWO₄ nanoparticles can be altered by modifying the synthesis technique, reaction temperature, precursor concentration, solution pH, reaction time, and through the process of doping. BaWO₄ has a tetragonal scheelite-type crystal structure, but under the influence of high pressure and temperature, it undergoes a transformation to a monoclinic structure. This is a semiconductor with a large energy gap that emits blue light and is very suitable for introducing impurities. Tungstate luminous materials activated by rare earth elements exhibit exceptional photo-stability, significant Stokes/anti-Stokes shifts, extended luminescence lifetimes, and distinct band emissions.

5. References

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