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Advanced Functional Thin Films via Nebulizer Spray Pyrolysis: Synthesis, Structure, and Applications

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Systematic Review

Keywords: nebulizer spray pyrolysis, aerosol deposition, thin films, ZnO, Al-doped ZnO, CZTS, ZnTe, magnesium phthalocyanine, solvent effects, structure–property relationships

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Advanced Functional Thin Films via Nebulizer Spray Pyrolysis: Synthesis, Structure, and Applications

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Abstract

Nebulizer-assisted spray pyrolysis (NSP) is a solution-mediated route for producing functional thin films through controlled aerosol generation, droplet transport, solvent evaporation and thermally activated precursor conversion at a heated substrate. Its appeal lies in the relative simplicity of the apparatus, flexibility in precursor chemistry and potential for coating extended substrate areas. Nevertheless, film formation is governed by coupled phenomena—including droplet-size distribution, solvent volatility, precursor speciation, substrate temperature, aerosol residence time and post-deposition treatment—that determine phase constitution, morphology, defect populations and functional response. This review examines NSP as a materials-engineering platform, with particular attention to oxide semiconductors such as ZnO and Al-doped ZnO (AZO), and to spray-pyrolysed chalcogenide absorbers such as Cu_2ZnSnS_4 (CZTS). Published work on solvent-dependent ZnO deposition and AZO coatings is considered alongside the author's contributions to ZnO, CZTS, ZnTe and magnesium phthalocyanine (MgPc) thin-film research. A necessary distinction is maintained between materials demonstrated by nebulizer/spray pyrolysis and comparative systems prepared by physical vapour deposition: the cited ZnTe and MgPc studies used vacuum or thermal evaporation and are therefore discussed as complementary examples of structure–property engineering, not as NSP demonstrations. The review develops a unified process–composition–structure–defect–property framework, evaluates characterization strategies and identifies research priorities in precursor design, reproducibility, phase verification, scale-up and application-specific optimization.

Keywords: nebulizer spray pyrolysis; aerosol deposition; thin films; ZnO; Al-doped ZnO; CZTS; ZnTe; magnesium phthalocyanine; solvent effects; structure–property relationships.

1. Introduction

Functional thin films underpin a broad spectrum of technologies, including transparent conducting electrodes, gas sensors, photovoltaic absorbers, photodetectors, optical coatings and emerging hybrid electronic devices. Their performance is determined not only by chemical composition but also by crystallographic order, grain connectivity, surface topography, defect chemistry, interfaces and thickness uniformity. The choice of deposition route is consequently a central part of materials design.

Spray pyrolysis converts a precursor-containing liquid into an aerosol that is transported towards a heated substrate. In nebulizer-assisted systems, the liquid is dispersed into fine droplets, which undergo solvent evaporation and precursor decomposition before or during contact with the hot surface. The deposited material subsequently nucleates and develops into a film. The technique is attractive because it can avoid high-vacuum infrastructure and accommodate a range of inorganic precursor chemistries. Reviews of ultrasonic spray pyrolysis emphasize the importance of solution flow, pyrolysis temperature, nozzle-to-substrate distance, carrier-gas flow, deposition time and precursor concentration [1].

The present article extends the discussion beyond a single compound. It considers common processing principles relevant to ZnO, AZO and CZTS, while examining the extent to which lessons from ZnTe and MgPc thin-film studies can inform a broader understanding of annealing, optical absorption and photoconductivity. The distinction between deposition routes is essential: ZnTe and MgPc in the cited Raj–Xavier studies were prepared by vacuum deposition or thermal evaporation [7, 8], and their results should not be represented as direct evidence of nebulizer spray pyrolysis.

A central proposition of this review is that NSP should be evaluated as a coupled process–materials platform. A deposition recipe is useful only when it is connected to measurable film attributes and application-level outcomes. For gas sensors, surface adsorption and defect-mediated charge transfer may dominate; for transparent electrodes, carrier concentration, mobility and visible transmittance are critical; for photovoltaic absorbers, phase purity, carrier lifetime, interface quality and recombination become decisive.

2. Scope and literature basis

This critical narrative review draws on the publications identified in the author's supplied research record and on relevant published literature concerning nebulized or ultrasonic spray pyrolysis. Directly relevant NSP evidence includes solvent-dependent ZnO thin films [2] and Al-doped ZnO coatings [3]. Spray-pyrolysed CZTS literature is included as a chalcogenide case study [4–6]. The author's ZnTe and MgPc publications are incorporated as comparative thin-film studies of vacuum-deposited materials [7, 8]. A rare-earth-doped ZnO nanoparticle study prepared by co-precipitation is used only to illustrate dopant-related functional modification and is not treated as spray-pyrolysis evidence [9]. The expanded literature base includes comparative work on ZnO, AZO, SnO_2, CuO, TiO_2, Fe_2O_3, CdS and CZTS deposited by nebulizer, ultrasonic or conventional chemical spray pyrolysis [10–39].

The article is a synthesis of published findings, not a report of new experiments. No new measurements, datasets or device efficiencies are claimed. Quantitative values are attributed to the individual studies from which they originate and should not be interpreted as universal process limits. The added references include conventional chemical spray pyrolysis and ultrasonic spray pyrolysis as well as nebulizer spray pyrolysis; these are related aerosol-assisted thermal-deposition approaches, but they are not assumed to be experimentally identical. Each study should be interpreted according to its reported apparatus and deposition route.

3. Fundamentals of nebulizer-assisted spray pyrolysis

3.1 Aerosol generation and droplet transport

A nebulizer transforms a liquid precursor into a dispersed aerosol. Depending on the apparatus, atomization may be ultrasonic, pneumatic or based on another mist-generation mechanism. Droplet size and distribution influence evaporation rate, solute concentration, transport losses and the likelihood that droplets reach the substrate in a liquid, partially dried or dry-particle state. Fine droplets can promote more uniform delivery, although excessively rapid evaporation may lead to powder formation or material loss before the aerosol reaches the substrate. The influence of aerosol generation, substrate motion, deposition geometry and process variables has been examined in ultrasonic spray systems and related aerosol-based deposition studies [19, 21, 22, 26, 28].

The carrier gas transports the aerosol through the deposition zone. Flow rate, gas composition, nozzle geometry, spray distance and substrate dimensions influence residence time and spatial flux. A stable deposition process therefore requires more than a fixed solution concentration: it requires reproducible aerosol generation and transport.

3.2 Droplet-to-film conversion

The conversion of a droplet into a film-forming species can be described through overlapping stages: (i) solvent evaporation, (ii) solute concentration and possible precipitation, (iii) precursor decomposition, (iv) chemical reaction and nucleation, and (v) crystallite growth and film densification. Depending on thermal and chemical conditions, some decomposition may occur in flight, while other reactions take place after droplet impact. The balance between these pathways influences roughness, porosity, adhesion and composition. Experimental studies on SnO_2, ZnO and other oxide films show that deposition temperature and time affect crystallite size, preferred orientation, film thickness and surface morphology [19, 20, 26, 28, 29].

The heated substrate supplies the energy required for decomposition and reaction. A substrate temperature that is too low may leave incompletely converted precursor residues or poorly crystallized deposits. Excessive temperature may cause volatile-component loss, oxidation, interdiffusion or unwanted phase formation. The optimum temperature is therefore material- and precursor-specific rather than a universal value.

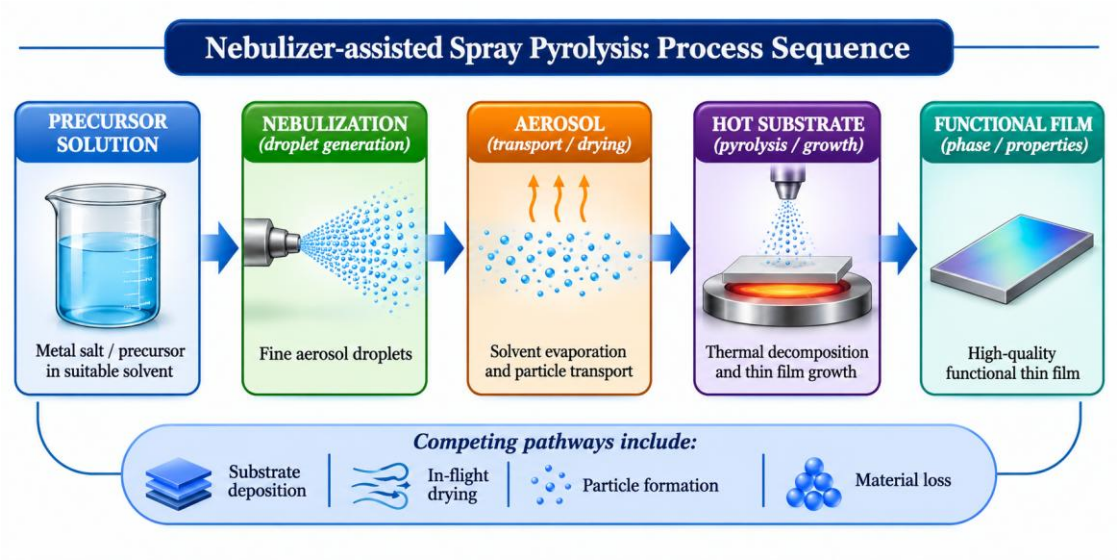


Figure 1. Schematic representation of nebulizer-assisted spray pyrolysis, from precursor solution to functional thin film. Competing pathways are process-dependent.

4. Process variables governing film formation

Process variable	Primary influence	Potential consequence if poorly controlled
Precursor identity and concentration	Complexation, decomposition, solute loading and deposition rate	Precipitation, non-stoichiometry or incomplete conversion
Solvent type and volume	Viscosity, surface tension, volatility and drying behaviour	Droplet instability, roughness, cracking or non-uniformity
Nebulizer frequency / atomization	Droplet size and aerosol flux	Broad droplet distribution and inconsistent coating
Carrier-gas flow	Aerosol residence time and transport	Overspray, deposition loss or uneven coverage
Nozzle-to-substrate distance	Evaporation time and droplet thermal history	Wet impact or in-flight powder formation
Substrate temperature	Pyrolysis, nucleation, crystallization and volatile loss	Poor crystallinity or decomposition / secondary phases
Spray rate and deposition time	Thickness, thermal cycling and material accumulation	Thickness gradients, cooling or cracking
Post-deposition annealing	Grain growth, defect redistribution and phase evolution	Volatile-element loss or unwanted reactions

5. Solvent engineering: a decisive NSP variable

Solvent selection is not merely a practical matter of dissolving precursor salts. It changes the physical properties of the liquid and the thermal history of each aerosol droplet. Solvent volatility, viscosity, surface tension, heat capacity and chemical interaction with precursor species can alter atomization, evaporation and solute redistribution. The solvent can therefore affect the deposited film's grain morphology, porosity, optical response and electrical behaviour. Related work on indium-doped ZnO and ZnO-based films further demonstrates that solvent composition, water content and precursor chemistry can influence electrical transport and film formation [27, 29].

Sebastian, Raj and co-workers examined ZnO films deposited by nebulizer spray pyrolysis using methanol, ethanol and deionized water [2]. Their study reported hexagonal ZnO, solvent-dependent optical gaps in the range 3.39–3.28 eV, and a higher ammonia response for the deionized-water-derived film under the reported test conditions. These results provide a direct example of how solvent choice can influence a functional NSP film. They should be interpreted within the reported deposition and sensor protocol rather than generalized to all ZnO compositions or nebulizer systems.

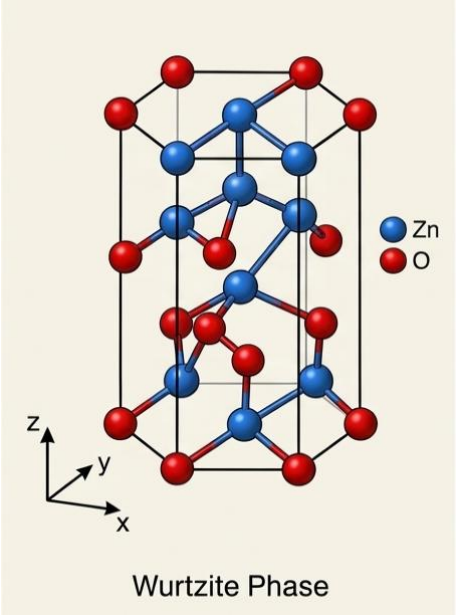
A related study by Deva Arun Kumar and co-workers investigated Al-doped ZnO films deposited by nebulized spray pyrolysis while varying solvent volume [3]. The reported films exhibited wurtzite structure, visible transmittance up to approximately 88%, and optical gaps between 3.34 and 3.28 eV for the investigated solution volumes. The study

also reported n-type conductivity and a carrier concentration of 3.54×10^{20} cm^{-3} under one reported condition. This work demonstrates the combined role of solvent volume, film structure and transport properties in transparent conducting oxide development.

6. Material systems and application-oriented properties

6.1 Zinc oxide and doped ZnO: transparent and sensing films

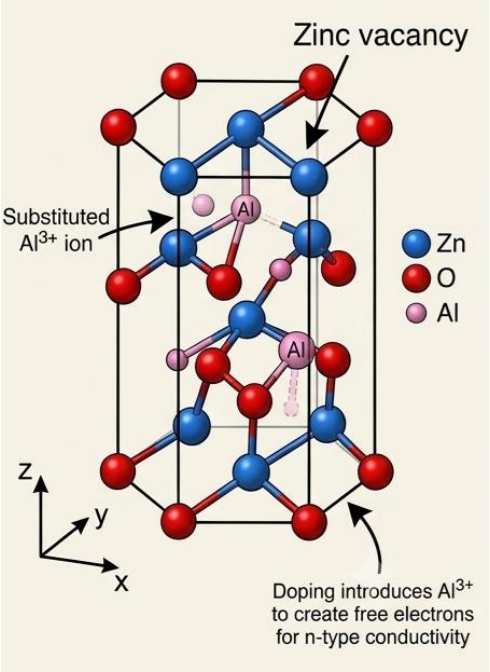
ZnO is a wide-band-gap II–VI semiconductor with a wurtzite structure. Its optical transparency and semiconducting behaviour support applications in transparent electrodes, UV optoelectronics and resistive gas sensors. In spray-deposited ZnO, precursor chemistry and thermal conditions determine crystallite development, oxygen-related defects, surface area and grain-boundary structure. For transparent conducting applications, high visible transmittance must be balanced against carrier concentration and mobility. For gas sensing, accessible surface and adsorption-related charge transfer become central. Additional nebulizer/ultrasonic spray studies have reported thickness-dependent ZnO sensing [17], Na-doped ZnO nanorods for ammonia detection [34], high-transparency ZnO coatings [28], and Mg-doped ZnO films [38]. Other spray-derived ZnO studies have examined photocatalysis and the effects of precursor preparation and deposition temperature [20, 26].



The solvent-dependent NSP study by Sebastian et al. [2] connects solvent choice with structural, optical, photoluminescence and ammonia-sensing measurements. It also illustrates why sensing performance should be reported together with gas concentration, operating temperature, response definition, response/recovery time and humidity conditions.

Dopant incorporation offers another route to property modification. The rare-earth-doped ZnO nanoparticle study by Sebastian et al. [9] reported Tb-, Er- and Yb-modified ZnO prepared by co-precipitation, with structural, optical and ammonia-sensing characterizations. Since that work used co-precipitation rather than NSP, it is cited here as evidence that dopant identity can modify ZnO properties, not as proof of nebulizer deposition. Future NSP studies could investigate dopant incorporation using controlled precursor ratios and verify dopant location and oxidation state using complementary compositional methods.

6.2 Aluminium-doped ZnO: transparent conducting oxide functionality

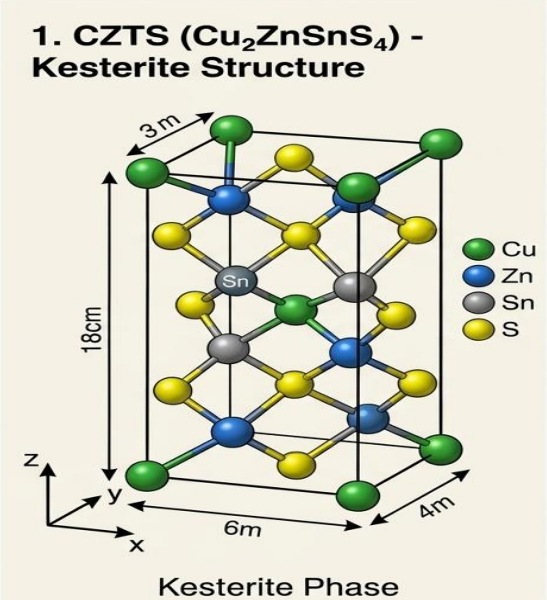


AZO is an important ZnO-based transparent conducting oxide. Substitutional aluminium can act as a donor, increasing electron concentration when incorporated appropriately, although segregation or secondary-phase formation may limit the expected benefit. The NSP work of Deva Arun Kumar et al. [3] provides a relevant benchmark for solvent-volume-dependent AZO film properties. It reports wurtzite structure, high visible transmittance and Hall-derived electrical properties. Future comparisons should include sheet resistance, Hall geometry, carrier mobility, thickness and optical transmittance over a defined wavelength interval. The broader transparent-conducting-oxide literature includes ZnO–SnO_2 binary films prepared by ultrasonic spray pyrolysis, with composition-dependent optical and electrical properties [22]. Indium- and nitrogen-modified ZnO films provide further examples of dopant- and thickness-dependent transport [27, 29].

6.3 Cu_2ZnSnS_4: spray-pyrolysed photovoltaic absorber films

Cu_2ZnSnS_4 (CZTS) is a kesterite-structured quaternary sulphide investigated as an earth-abundant thin-film photovoltaic absorber. Its potential derives from suitable optical absorption and a band gap in the solar-relevant range. Spray pyrolysis provides a solution-based route to deposit the multicomponent absorber, but simultaneous control of copper, zinc, tin and sulphur makes the process chemically more demanding than deposition of a binary oxide. Further spray-based CZTS studies have examined precursor sulphur concentration, non-equilibrium growth, morphology, optical parameters and absorber-layer optimization [11–13, 36].

The author's earlier review by Raj, Sebastian and Rajendran [4] surveyed spray-pyrolysed CZTS films for solar-cell applications. Subsequent studies have emphasized precursor stoichiometry, substrate temperature, sulphur availability, post-deposition sulphurization, secondary-phase identification and device interfaces [5, 6]. For CZTS, XRD alone is insufficient to establish phase purity because diffraction features of kesterite and competing sulphide phases can overlap. Raman spectroscopy, elemental mapping and surface-sensitive chemical analysis are valuable complements [40].



The central challenge is to connect film-level measurements—such as band gap, absorption coefficient, crystallite size and morphology—to complete device metrics. A favourable optical gap does not establish high photovoltaic efficiency. A credible device report requires a defined architecture, active area, calibrated illumination, current–voltage characteristics, open-circuit voltage, short-circuit current density, fill factor and stability information.

6.4 ZnTe and MgPc: comparative thin-film systems and route compatibility

ZnTe and magnesium phthalocyanine (MgPc) are included because they form part of the author's broader thin-film research record and offer useful comparisons in optical absorption, annealing response and photoconductivity. However, the cited studies prepared these materials by vacuum deposition or thermal evaporation, not by nebulizer spray pyrolysis. Raj and Xavier reported ZnTe and MgPc films formed by vacuum deposition and investigated their optical and photoconductive response following annealing [7]. A separate study investigated the structural and optical properties of thermally evaporated MgPc films [8].

These studies are relevant to the general science of functional thin films: they show that post-deposition thermal treatment can modify structural order, optical absorption and photocurrent. They do not demonstrate that MgPc or ZnTe can be deposited by NSP under the same precursor and thermal conditions. MgPc is a molecular organic semiconductor, and its solubility and thermal stability require separate assessment before any aerosol-based solution route is proposed. ZnTe formation by spray pyrolysis would require suitable soluble precursor chemistry, control of tellurium-source reactivity and volatility, and verification of phase composition. Such routes should be presented as research opportunities until supported by direct experimental evidence.

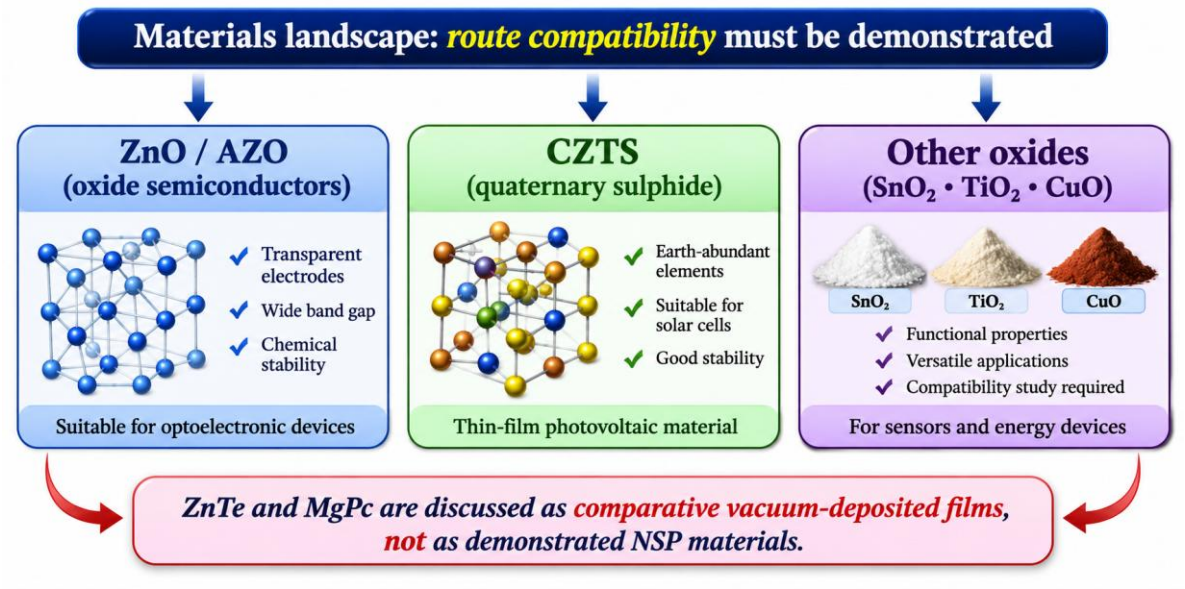


Figure 2. Materials landscape. ZnO/AZO and CZTS have direct spray/nebulizer-spray literature; ZnTe and MgPc are included as comparative PVD thin-film systems.

7. Structure–property relationships and characterization

A rigorous NSP study should not rely on a single characterization technique. X-ray diffraction (XRD) identifies crystalline phases, preferred orientation and peak broadening. Raman spectroscopy probes vibrational modes and can help discriminate phases with overlapping XRD signatures. Scanning electron microscopy (SEM) reveals surface coverage, grain morphology and cross-sectional thickness, while energy-dispersive X-ray spectroscopy (EDS) provides local elemental information. Atomic force microscopy (AFM) quantifies surface topography and roughness. UV–visible spectroscopy assesses transmittance and absorption; photoluminescence (PL) can probe radiative defect-related transitions; and Hall measurements provide carrier concentration, mobility and resistivity. Comparable spray-derived systems—including SnO_2, ZnO, CuO, Fe_2O_3 and CdS—have used combinations of XRD, Raman spectroscopy, electron microscopy, optical spectroscopy and electrical measurements to relate processing conditions to film properties [10, 15, 16, 19, 20, 25, 30, 31].

The interpretation of defect-related optical signals requires care. Visible PL bands are often discussed in relation to oxygen vacancies or other native defects, but a single emission band rarely establishes a unique defect assignment. Complementary methods such as XPS, controlled-atmosphere annealing, temperature-dependent PL and electrical measurements can strengthen such interpretations.

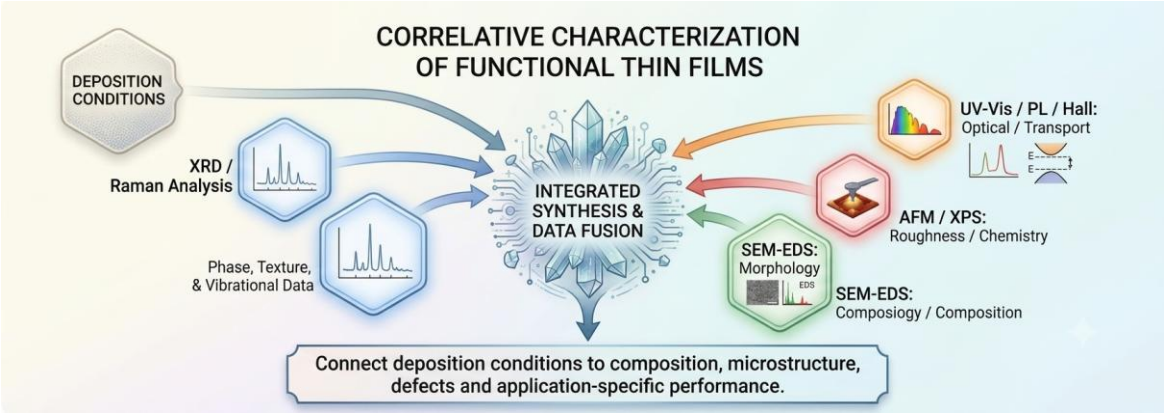


Figure 3. Recommended correlative characterization workflow for nebulizer-spray-pyrolysed functional thin films.

8. Application-specific optimization

8.1 Gas sensing

For metal-oxide gas sensors, adsorption of oxygen and target-gas molecules changes the near-surface carrier concentration and therefore the measured resistance. Film porosity, grain size, surface defects, operating temperature, humidity and electrode geometry all affect response. Sensitivity values should be compared only when response definition, concentration range, temperature and test atmosphere are equivalent. The ZnO solvent study [2] provides a specific ammonia-sensing example, but its numerical response should not be generalized to other gases or test conditions. The gas-sensing literature on spray-derived oxides includes SnO_2 for NO_2 and H_2S detection, Cu-doped SnO_2, doped CuO for hydrogen sensing, and ZnO-based sensing layers [10, 15–18, 31–34]. A review discusses how spray parameters, doping and heterostructure formation influence sensor response, selectivity, stability and response/recovery behaviour [14].

8.2 Transparent conducting electrodes

Transparent conducting films must simultaneously exhibit low electrical resistivity and high optical transmittance. AZO deposited by NSP is a relevant system because it combines an oxide host with a donor dopant and can be processed from solution [3]. Optimization should account for the trade-off between carrier concentration, mobility, free-carrier absorption, thickness and surface roughness. Ultrasonic spray pyrolysis has also been used to prepare ZnO–SnO_2 systems and highly transparent ZnO films, supporting its relevance to transparent-electrode development [22, 28].

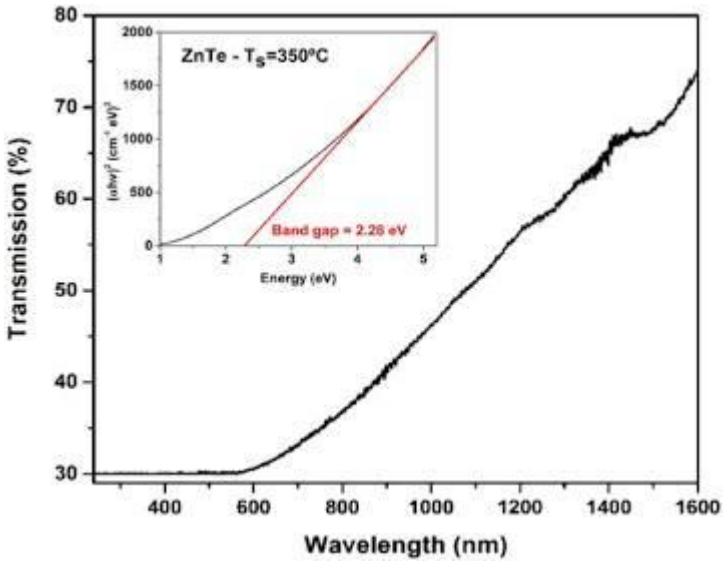
8.3 Photovoltaic absorbers and interfaces

For CZTS, the relevant target is not merely a crystalline absorber but a complete heterojunction with appropriate band alignment, limited recombination and efficient carrier collection. Film continuity, grain-boundary chemistry, secondary phases and buffer-layer compatibility must be assessed. The best reported kesterite cell across all deposition methods should not be confused with the best spray-pyrolysed pure-CZTS device. Spray-deposited CdS buffer layers and CZTS absorbers provide additional examples of solution-processed photovoltaic thin films, although their performance must be evaluated within complete device architectures [11–13, 30].

8.4 Photoconductive and optical films

The ZnTe and MgPc vacuum-deposition studies [7, 8] demonstrate the value of studying annealing-dependent absorption and photoconductivity. These findings can guide the selection of characterization metrics for other film systems, but route-specific claims must remain separate. For organic molecular semiconductors, thermal processing may influence molecular packing and crystallinity; for inorganic semiconductors, it may affect grain growth, stoichiometry and defect populations. Spray-derived TiO_2, Cu_xO and α-Fe_2O_3 films have also been investigated for optical, photocatalytic and photoelectrochemical functions [23, 25, 35].

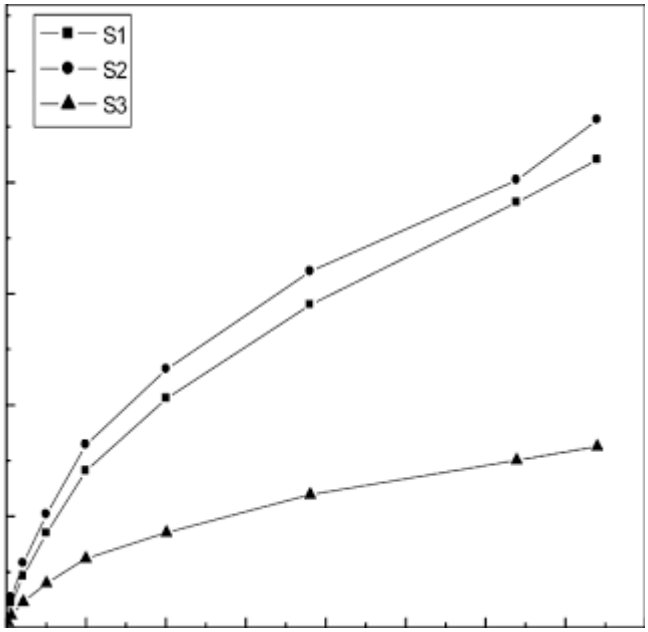
Optical Transmission and Tauc Plot for Vacuum-Deposited ZnTe Thin Film



Optical transmission spectrum and Tauc plot inset for vacuum-deposited ZnTe.. Source: ResearchGate / Optical transmission of the ZnTe film grown at 350 °C.

This graph displays the experimental optical transmission spectrum of a vacuum-deposited zinc telluride (ZnTe) thin film grown at 350°C. The inset shows the standard Tauc plot—$(\alpha h\nu)^2$ plotted against photon energy ($h\nu$) in eV—where extrapolating the linear region to the horizontal axis yields the direct optical band gap of 2.26 eV.

Photoconductivity Response Curves



Photoconductivity versus intensity curves for vacuum-deposited film samples.. Source: ResearchGate

This plot shows experimental photoconductivity measurements recorded across different thin film preparation conditions, illustrating the rise in photocurrent as illumination intensity increases.

Reference Citations

1. **Mahalingam, T., et al.***"Fabrication and optical characterization of zinc telluride thin films."*
2. **Raj, C. S. A., & Xavier, F. P.** *"Photoconductivity studies of hybrid ZnTe/MgPc thin films prepared by physical vapour deposition." Archives of Applied Science Research*.

9. Comparative assessment of the materials systems

Material system	Route supported by cited work	Key property focus	Application direction
ZnO	Nebulizer spray pyrolysis [2]	Wurtzite structure; solvent-dependent optical response and PL	Ammonia sensing; optoelectronics
Al-doped ZnO	Nebulized spray pyrolysis [3]	Transmittance, band gap, carrier concentration and resistivity	Transparent conducting electrodes
CZTS	Spray pyrolysis [4–6]	Kesterite phase, composition, secondary phases, absorption	Thin-film photovoltaics
ZnTe	Vacuum deposition in cited author study [7]	Annealing-dependent optical absorption and photoconductivity	Comparative photoconductive films
MgPc	Thermal evaporation in cited author study [8]	Molecular ordering, absorption and photoconductivity	Organic optoelectronic films
REE-doped ZnO	Co-precipitation in cited study [9]	Dopant-dependent optical and sensing response	Context for dopant engineering; not NSP evidence

10. Reproducibility, scale-up and reporting standards

The comparison of spray-deposited films across laboratories is often complicated by incomplete reporting of nebulizer characteristics, precursor chemistry, gas flow, substrate temperature and deposition geometry. A minimum report should specify: (i) precursor identity, purity and concentration; (ii) solvent and solution preparation; (iii) nebulizer type and operating conditions; (iv) carrier gas and flow rate; (v) nozzle-to-substrate distance; (vi) substrate cleaning and temperature; (vii) spray duration, duty cycle and solution feed rate; (viii) film thickness; and (ix) post-deposition annealing atmosphere and schedule.

Scale-up introduces additional challenges. Temperature gradients across a larger substrate can produce non-uniform decomposition, while aerosol depletion and nozzle geometry can create spatial variation in material flux. Reproducibility should therefore be assessed through thickness mapping, composition mapping and property measurements at multiple substrate positions. Replicate samples and uncertainty estimates are needed before claiming process robustness.

11. Research opportunities and future directions

1. Develop solvent–precursor compatibility maps that connect viscosity, volatility and complexation with droplet drying and film morphology.
2. Use controlled nebulizer frequency, carrier-gas flow and nozzle geometry to narrow droplet-size distributions and improve coating uniformity.
3. Establish a robust NSP processing window for ZnO and AZO that balances visible transparency, carrier mobility and gas-sensing surface activity.
4. Extend spray-based processing to multicomponent chalcogenides such as CZTS using explicit control of cation ratios, sulphur activity and volatile-element retention.
5. Investigate aerosol-compatible precursor chemistries for less conventional systems, including telluride materials, but report them as exploratory until direct phase and composition evidence is obtained.
6. Assess whether solution-compatible molecular semiconductors can be processed by aerosol deposition without chemical degradation or loss of molecular order; MgPc should not be assumed to be spray-processable merely because it is a thin-film material.
7. Combine XRD, Raman mapping, SEM–EDS, XPS, AFM, optical spectroscopy and electrical measurements to distinguish phase purity from apparent crystallinity.
8. Connect film-level improvements to complete application metrics: calibrated sensor response, transparent-electrode figure of merit, or photovoltaic current–voltage and stability measurements.

12. Conclusions

Nebulizer-assisted spray pyrolysis is a versatile route for depositing functional thin films, particularly where solution-processable inorganic precursors can be converted into stable coatings through controlled aerosol transport and

thermal reaction. The literature on ZnO and AZO demonstrates that solvent composition and solution volume can influence structure, optical response and electrical behaviour. Spray-pyrolysed CZTS extends the platform to multicomponent photovoltaic absorbers, where stoichiometry, phase purity, sulphur retention and interfaces become especially demanding. The author's ZnTe and MgPc studies provide useful comparative evidence on annealing, optical absorption and photoconductivity, but their vacuum-deposition routes must be clearly distinguished from NSP. A reliable materials-development strategy should connect precursor chemistry and aerosol physics to phase constitution, morphology, defects and application-level performance. The field will benefit from standardized process reporting, correlative characterization, spatial uniformity assessment and careful separation of demonstrated deposition routes from proposed future extensions.

Author Contributions

C. S. A. Raj conceptualized the review, identified and synthesized the relevant literature, critically interpreted the materials and processing relationships, prepared the schematic figures and tables, and drafted and revised the manuscript. The author approved the final manuscript.

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Competing Interests

The author declares no competing interests.

Data Availability

No new datasets were generated or analysed in this review. The information discussed is available in the cited publications.

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