


 Cite this: *RSC Adv.*, 2025, **15**, 26353

## Substrate temperature modulated optical characterizations of $\alpha$ -CdIn<sub>2</sub>Se<sub>4</sub> thin films grown by pulsed laser deposition technique

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The current study examines the effect of substrate temperature ( $T_s$ ) on the optical characteristics of CdIn<sub>2</sub>Se<sub>4</sub> thin films grown by the pulsed laser deposition technique using a UV-Vis-NIR spectrophotometer. Transmittance maxima of CdIn<sub>2</sub>Se<sub>4</sub> thin films shift with a change in  $T_s$ , exhibit high absorption in the visible region, and depict an absorption coefficient ( $\alpha$ ) of  $\approx 10^7 \text{ m}^{-1}$ . Refractive index spectra of CdIn<sub>2</sub>Se<sub>4</sub> thin films controlled by  $T_s$  reflect crests at characteristic wavelengths ( $\lambda_c$ ) and a wavelength ( $\lambda$ ) higher than  $\lambda_c$ ; spectra display normal dispersion. The extinction coefficient ( $k$ ) of  $T_s$  tempered CdIn<sub>2</sub>Se<sub>4</sub> thin films decreases as  $\lambda$  increases and reaches a minimum at  $\lambda_c$ ; the contribution of free carrier absorption can explain the increase in  $k$  values after  $\lambda_c$ . The effect of  $T_s$  on the optical band gap energy ( $E_g$ ) of the CdIn<sub>2</sub>Se<sub>4</sub> thin films is discussed. For  $T_s$ -modulated CdIn<sub>2</sub>Se<sub>4</sub> thin films, dielectric constants, loss tangent, Urbach energy, Urbach absorption coefficient, and optical and electrical conductivities have been inferred. Peak values of the volume and surface energy loss functions of CdIn<sub>2</sub>Se<sub>4</sub> thin films were retrieved. A Fourier transform infrared spectrophotometer verified the purity of CdIn<sub>2</sub>Se<sub>4</sub> thin films formed at different  $T_s$ . The current study indicates that CdIn<sub>2</sub>Se<sub>4</sub> thin films are promising options for designing and developing upcoming high-efficiency opto-electronic devices. Implications are discussed.

Received 1st June 2025  
 Accepted 17th July 2025

DOI: 10.1039/d5ra03888j  
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## 1 Introduction

Due to their potential uses in thermo-electric materials,<sup>1</sup> semiconductor instrumentation,<sup>2</sup> switching devices,<sup>3,4</sup> photocatalysts,<sup>5-7</sup> photoconductors,<sup>8,9</sup> non-linear optical devices,<sup>10,11</sup> optical filters,<sup>12</sup> optoelectronics,<sup>13</sup> solar cells,<sup>14-17</sup> and more, ternary semiconducting compounds with the composition II-III<sub>2</sub>-VI<sub>4</sub> (where II = Zn, Cd, or Hg; III = Al, Ga, or

In; VI = S, Se, or Te) have been studied extensively over the globe since 1955.<sup>18</sup> II-III<sub>2</sub>-VI<sub>4</sub> group compounds are earth-abundant, less expensive, and less toxic; they are largely derived from the zinc blende structure using the Grimm-Sommerfel rule. According to Hahn *et al.*,<sup>18</sup> ternary semiconducting compounds of composition II-III<sub>2</sub>-VI<sub>4</sub> have a space group  $S_4^2(\bar{I}4)$ . They are thiogallates that crystallize into a tetragonal structure<sup>19</sup> and are distinguished from chemical and iso-electronic analogs with the structure of chalcopyrite and sphalerite by the presence of an ordered cation vacancy. As a result, compounds of composition II-III<sub>2</sub>-VI<sub>4</sub> are known as defective chalcopyrite (DC).

The II-III<sub>2</sub>-VI<sub>4</sub> family's CdIn<sub>2</sub>Se<sub>4</sub> has enthralled substantial contemplation from scientists over the orb due to its prerogatives in thermoelectric materials,<sup>20,21</sup> photoanodes,<sup>22</sup> heterojunction solar cells,<sup>23</sup> PEC solar cells,<sup>24-28</sup> photovoltaics,<sup>29</sup> etc. The structural, morphological, electrical, optical, mechanical, vibrational, and thermoelectric properties of CdIn<sub>2</sub>Se<sub>4</sub> in bulk and/or thin film form have been explored by various authors.<sup>30-34</sup> Experimental studies of the ordered-vacancy compound CdIn<sub>2</sub>Se<sub>4</sub> have revealed that its tetragonal structure can be deformed with a lattice constant ratio  $c/a$  of 1, 2, and 3, which are referred to as the  $\alpha$  (pseudo-cubic),  $\beta$ , and  $\gamma$  phases, respectively, depending on the synthesis conditions.<sup>31</sup> Because

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of their high absorption coefficient, optical band gap energy ( $E_g$ ) in the visible spectra, and relatively high photo-electronic sensitivity from the visible to the near-infrared spectrum range,  $\text{CdIn}_2\text{Se}_4$  thin films are a propitious entrant for photonic devices. As an upshot, research on their optical characteristics is of prodigious concern.

When depositing ternary semiconducting compounds with constituents that have varying vapor pressures, pulsed laser deposition (PLD) is an excellent choice. Comparing PLD technology to traditional vacuum-based thin film deposition methods, the former reduces processing times by enabling the growth of uniform and fast thin films. Of particular relevance is the ease with which heterojunctions and multilayer structures requiring precise control over film thickness can be produced using PLD technology. In the current study, the authors employed PLD to deposit  $\text{CdIn}_2\text{Se}_4$  thin films because it offers several benefits over other thin film deposition methods, such as outstanding thickness controllability, broad universality, high-level cleanliness in the deposited film, and reproducible maintenance of the target material's precise composition.<sup>35,36</sup>

In the current study, amorphous quartz glass (fused silica) was employed to deposit  $\text{CdIn}_2\text{Se}_4$  thin films for their optical characterization. Due to its excellent transparency in the UV-Visible-NIR spectra, quartz glass is perfect for examining the optical characteristics of thin films without the substrate affecting the results. Characterizing the thin film's intrinsic optical behavior is easier because quartz glass is non-crystalline and does not introduce diffraction patterns or crystallographic effects. Because it does not react with many solvents and can tolerate high processing temperatures, quartz is useful for post-treatment and film deposition procedures. The smooth surface of high-quality quartz glass usually aids in producing homogeneous films and reliable optical data.

In the extant investigation, an attempt has been made to acquire exhaustive information on the effect of substrate temperature ( $T_s$ ) on the optical properties of pulsed laser deposited  $\alpha$ -phase  $\text{CdIn}_2\text{Se}_4$  thin films by engaging sophisticated techniques such as ultraviolet-visible-near infrared (UV-VIS-NIR) and Fourier transform infrared (FTIR) spectrophotometers by keeping  $\text{CdIn}_2\text{Se}_4$  thin film's thickness ( $d$ ) identical throughout the investigation. The authors believe that the contemporary enquiry on the hitherto unfabricated-unpublished-unreported,  $T_s$ -controlled optical characterizations of pulsed laser deposited  $\alpha$ -phase  $\text{CdIn}_2\text{Se}_4$  thin films should not only be an accumulation to the prevailing list but also lead to the further understanding of the optical properties of thin films and the research will afford optimized optical parameters for designing and developing of future novel semiconducting compound thin film opto-electronic device applications.

## 2 Experimental

For their optical characterizations,  $\text{CdIn}_2\text{Se}_4$  ternary semiconducting compound thin films were deposited using the PLD technique (model: Compex-Pro Excimer Laser 102F; make: Coherent, Germany). The source (target) material was a single-

phase  $\text{CdIn}_2\text{Se}_4$  (ref. 37) in a pellet form with 10 mm (diameter)  $\times$  3 mm (thickness) dimensions. The substrates were ultrasonically cleaned amorphous quartz glass (make: Blue Star, made: Polar Industries Corporation, India) with dimensions  $10 \pm 0.1$  mm (length)  $\times$   $3 \pm 0.1$  mm (breadth)  $\times$   $1.45 \pm 0.1$  mm (thickness). A  $\text{CdIn}_2\text{Se}_4$  pellet and amorphous quartz glass were mounted in a stainless-steel PLD chamber (make: Excel Instruments, India). Silver conductive adhesive paste (model: RS pro-RS186-3600, made: RS Components & Controls Limited, India) adheres the  $\text{CdIn}_2\text{Se}_4$  pellet to the target holder and amorphous quartz glass substrates to the substrate holder. The  $\text{CdIn}_2\text{Se}_4$  pellet was irradiated by a krypton fluoride (KrF) pulsed laser through a quartz lens. The plasma plume dimension at the  $\text{CdIn}_2\text{Se}_4$  pellet was optimized and confined using a quartz lens.  $45^\circ$  separates the substrate holder from the incident laser that cataracts on the target. The target was spun at six revolutions per minute (rpm) during laser ablation to avert mutilation from dropping laser stuck at a single spot incessantly on the  $\text{CdIn}_2\text{Se}_4$  target.

The thin films of  $\text{CdIn}_2\text{Se}_4$  were deposited on the amorphous quartz glass substrate at diverse  $T_s$ , vacillating from room temperature (RT) ( $\approx 300$  K) to 675 K ( $300$  K  $\leq T_s \leq 675$  K) using a calibrated chromel-alumel (Cr-Al) thermocouple abetted microcontroller-based temperature controller (make: Coherent, India). The  $T_s$  were sensed and controlled during the thin film deposition by mounting the chromel-alumel (Cr-Al) thermocouple on the quartz glass substrate surface. The thin film  $d$  and deposition rate was monitored and/or controlled by the *in situ* digital quartz crystal thickness monitor (model: DTM-101, make: Hind High Vacuum Co. Pvt. Ltd, India). During  $\text{CdIn}_2\text{Se}_4$  thin films' deposition, the evaporation rate ( $\approx 10$  nm s $^{-1}$ ) and the  $d$  of the deposits ( $\approx 100$  nm) were kept constant. Table 1 displays the PLD parameters optimized for synthesizing  $\text{CdIn}_2\text{Se}_4$  thin films.

The variable laser ablation duration was used to achieve consistent  $d$  at all  $T_s$ . It was observed that the thin films deposited at lower  $T_s$  require less laser ablation time during the synthesis of thin films, whereas thin films synthesized at higher  $T_s$  require more laser ablation time to achieve the same  $d$  as those deposited at lower  $T_s$  because of re-evaporation at high  $T_s$ .

The effect of the  $T_s$  ( $300$  K  $\leq T_s \leq 675$  K) on the optical properties of pulsed laser deposited  $\text{CdIn}_2\text{Se}_4$  thin films was scrutinized by recording room temperature (RT) ( $\approx 300$  K) transmittance ( $T(\lambda)$ ) spectra employing UV-Vis-NIR

Table 1 PLD parameters optimized for the synthesis of  $\text{CdIn}_2\text{Se}_4$  thin films

S. no.	Parameter	Value
1	KrF laser wavelength ( $\lambda$ )	$\approx 248$ nm
2	KrF laser energy	$\approx 255$ mJ
3	Repetition rate (frequency)	$\approx 04$ Hz
4	Source-to-substrate distance	$\approx 38$ mm
5	Substrate temperature ( $T_s$ )	$300$ K $\leq T_s \leq 675$ K
6	Thickness of thin films ( $d$ )	$\approx 100$ nm
7	Base vacuum	$\approx 1.00 \times 10^{-4}$ Pa
8	Deposition time (laser ablation time)	05-to-20 minutes



spectrophotometer (model: Lambda 1050+, make: PerkinElmer, USA) in the 400 to 900 nm wavelength ( $\lambda$ ) range. FTIR spectrophotometer (model: IRSpirit-X, make: Shimadzu, Japan) operated in mid-infrared mode at room temperature (RT) ( $\approx 303$  K) in the wavenumber ( $\bar{\nu}$ ) range 4000–400  $\text{cm}^{-1}$  with a resolution of 2  $\text{cm}^{-1}$  to spot the functional groups existing (if any) in the pulsed laser deposited  $\text{CdIn}_2\text{Se}_4$  thin films deposited at various  $T_s$ .

The thin film characteristics altered depending on numerous pre- and post-deposition conditions. All other PLD parameters itemized in Table 1 were held constant during the current experiments, except for  $T_s$  and deposition duration, to ensure the reproducibility of the findings.

### 3 Results and discussion

#### 3.1. UV-vis-NIR spectrophotometer analysis

The UV-VIS-NIR spectrophotometer was employed to derive optical spectra of  $T_s$ -modified  $\text{CdIn}_2\text{Se}_4$  thin films. UV-VIS-NIR spectrophotometer can produce optical spectra in a variety of modes, including  $T(\lambda)$ , reflectance ( $R(\lambda)$ ), and absorption ( $A(\lambda)$ ), depending on the materials' structure and characteristics. An opaque sample, such as pellet, bulk, etc., has awfully low transparency, often imminent to zero ( $\approx 0$ ), and the optical parameters are resolute when measured by consuming the diffuse reflectance spectra (DRS), which use the  $R(\lambda)$  spectra as per Kubelka-Munk (K-M) theory.<sup>38</sup> According to Jubu *et al.*,<sup>39</sup> the Tauc technique evaluates the optical characteristics of a transparent sample by using the absorption ( $A(\lambda)$ ) and/or  $T(\lambda)$  spectra of the UV-VIS-NIR spectrophotometer.

The current study analyzes  $\lambda$ -dependent room temperature (RT) ( $\approx 300$  K)  $T(\lambda)$  spectra, Fig. 1, for  $T_s$  tempered  $\text{CdIn}_2\text{Se}_4$  thin films using a UV-VIS-NIR spectrophotometer in the  $\lambda$  range of 400 to 900 nm.

The shift in  $\lambda$  dependent  $T(\lambda)$  maxima perceptible in Fig. 1 can be explained by the thin films of the  $\text{CdIn}_2\text{Se}_4$  formed at low  $T_s$  ( $T_s < 425$  K) have dispersed microcrystallites, random orientation, disordered grain boundaries, and are essentially

amorphous, which contributes to lower  $T(\lambda)$  and increased absorption ( $A(\lambda)$ ). Conversely,  $\text{CdIn}_2\text{Se}_4$  thin films grown at higher  $T_s$  ( $300$  K  $< T_s < 675$  K) have a stronger orientation, organized microcrystallites, starkly defined grain boundaries, and are essentially polycrystalline, all of which contribute to elevated  $T(\lambda)$  and diminished absorbing ( $A(\lambda)$ ) capabilities. When deposited at 675 K  $T_s$  ( $> 550$  K), thin films of the  $\text{CdIn}_2\text{Se}_4$  show an additional phase ( $\text{In}_2\text{Se}_3$ )<sup>40</sup> that could be the cause of the  $T(\lambda)$  and absorption ( $A(\lambda)$ ) divergence. The characteristic wavelengths ( $\lambda_c$ ) of  $T_s$  modified  $\text{CdIn}_2\text{Se}_4$  thin films with their  $T(\lambda)$  amplitude are revealed in Fig. 1. The  $T_s$  modulated  $\text{CdIn}_2\text{Se}_4$  thin films has high absorption in the visible region with a hump at  $750.00 \pm 58.50$  nm making it a good choice for light absorbing material.

Using eqn (1)–(3), Fig. 2 demonstrates how the optical parameters—the absorption coefficient ( $\alpha$ ), refractive index ( $\eta$ ), and extinction coefficient ( $k$ )—vary with  $\lambda$  for  $T_s$  curbed  $\text{CdIn}_2\text{Se}_4$  thin films.

$$\alpha = \frac{2.303 \times A}{d} \quad (1)$$

$$\eta = \frac{1 + \sqrt{R}}{1 - \sqrt{R}} \quad (2)$$

$$k = \frac{\alpha\lambda}{4\pi} \quad (3)$$

Eqn (1) shows that the  $d$  is  $\approx 100$  nm for  $\text{CdIn}_2\text{Se}_4$  thin films.

The  $\alpha$  scale depends upon radiation photon energy ( $h\nu$ ) and the structure of a thin film. Bulk and/or thin films with higher  $\alpha$  absorb photons more readily, which excite electrons into the conduction band (CB). The higher value of  $\alpha$  ( $\approx 10^7 \text{ m}^{-1}$ )<sup>32,41</sup> of  $T_s$  curbed  $\text{CdIn}_2\text{Se}_4$  thin films palpable in Fig. 2(a) supports the direct optical band gap ( $E_g$ ) nature of the semiconductor<sup>32</sup> and makes it a very good candidate for designing and developing opto-electronic devices.<sup>42</sup>

Fig. 2(b) shows  $R(\lambda)$  dependent  $\eta$  ( $= \frac{1 + R^{0.5}}{1 - R^{0.5}}$ ) spectra as a function of  $\lambda$  for  $T_s$ -controlled  $\text{CdIn}_2\text{Se}_4$  thin films. The dispersion curves of the  $\eta$  show crests at  $\lambda_c$ , which the oscillator model can explain. As per El-Nahass *et al.*,<sup>43</sup> at a  $\lambda$  higher than the  $\lambda_c$ , the spectral behavior of the  $\eta$  parades normal dispersion, which the single oscillator model can explicate. The  $\eta$  values deduced for  $T_s$  tempered  $\text{CdIn}_2\text{Se}_4$  thin films, stake well with the empirical relation  $\eta^4(E_g - 0.365) = 154$  ( $\eta \approx 3.27$ ) agreed by Reddy *et al.*<sup>31,44,45</sup>

The  $k$  quantifies the percentage of light energy lost through scattering and/or absorption per unit transit distance in thin films, reflecting the absorption of electromagnetic waves due to inelastic scattering events. Through empirical observation, the  $k$  is proportional to the  $\alpha$  ( $\propto \frac{\alpha\lambda}{4\pi}$ ). Fig. 2(c) shows the change in  $k$  with  $\lambda$  for  $T_s$  modified  $\text{CdIn}_2\text{Se}_4$  thin films. The  $k$  decreases with an increase in  $\lambda$  and reaches a minimum at  $\lambda_c$ ; the increment in  $k$  values after  $\lambda_c$  can be elucidated due to the free carrier

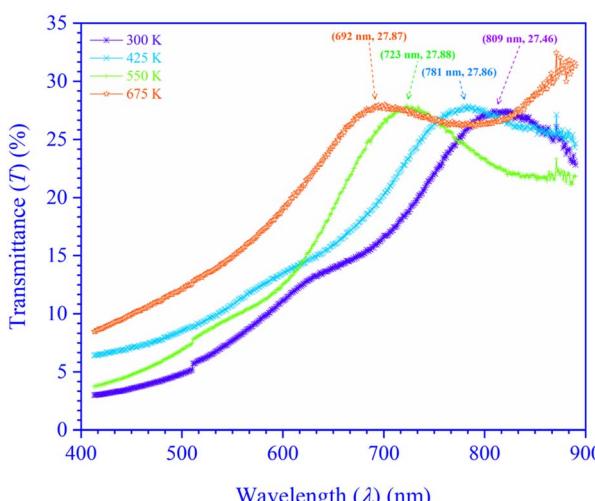


Fig. 1 Transmittance spectra of  $T_s$ -modulated  $\text{CdIn}_2\text{Se}_4$  thin films.



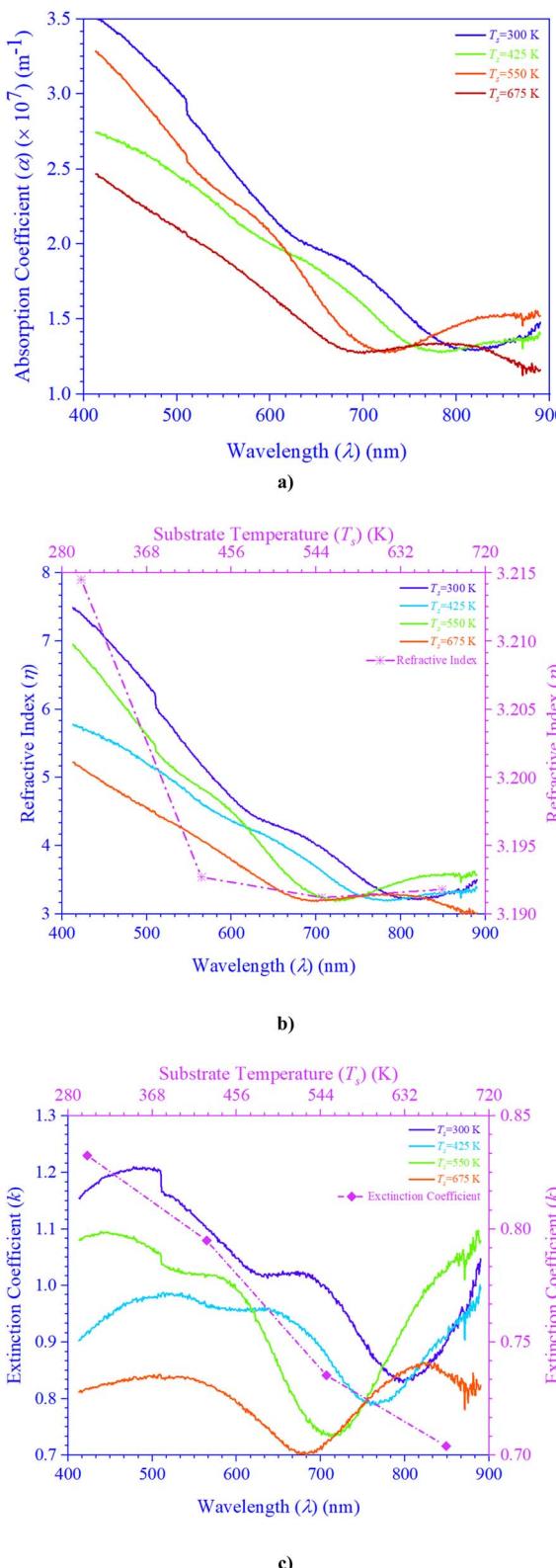


Fig. 2  $T_s$  modulated  $\text{CdIn}_2\text{Se}_4$  thin films' (a) absorption coefficient, (b) refractive index, and (c) extinction coefficient.

absorption contribution; the behavior agrees well with the results reported by El-Nahass *et al.*<sup>43,45</sup>

The course used to synthesize the bulk and/or thin film, the thickness of the bulk and/or thin film ( $d$ ),  $T_s$  at which thin films were/are grown, *etc.*, could all be causative factors to the variation in the  $\alpha$ ,  $\eta$ , and  $k$  values for the bulk and/or thin film  $\text{CdIn}_2\text{Se}_4$ .

To reach the lowest energy optically excited state, the incident photon energy ( $h\nu$ ) is designated by the optical band gap ( $E_g$ ). The Tauc technique construes the  $E_g$  of  $T_s$ , moderated  $\text{CdIn}_2\text{Se}_4$  thin films. The optical  $\alpha$  is correlated to the  $E_g$  for interband transitions in a semiconductor, adjacent to the  $E_g$  by engaging eqn (4).<sup>46</sup>

$$(\alpha h\nu)^{\frac{1}{n}} = A(h\nu - E_g) \quad (4)$$

In eqn (4), the transition probability is symbolized by  $n$  ( $=2, 1/2, 2/3$ , and  $1/3$  for direct allowed, indirect allowed, direct forbidden, and indirect forbidden transitions, respectively), and  $A$  is a constant that is contingent on the nature of the transition.

Photons have less energy than the band gap; therefore, materials in any form—bulk or thin film—cannot absorb light beneath it. Fig. 3 shows the plot of  $(\alpha h\nu)^2$  *vis-à-vis* photon energy ( $h\nu$ ), which is used to find the  $E_g$  values of  $T_s$  curbed  $\text{CdIn}_2\text{Se}_4$  thin films. The direct  $E_g$  values may be articulated by extrapolating the straight segment of the graphs at the values where  $(\alpha h\nu)^2$  tends to zero ( $=0$ ).

The  $E_g$  values of the thin films grown below  $550\text{ K}$   $T_s$  ( $<550\text{ K}$ ) *i.e.* at  $300\text{ K}$  and  $425\text{ K}$  are found to be  $\approx 1.53\text{ eV}$  and  $\approx 1.59\text{ eV}$ , which are slightly less than the  $\text{CdIn}_2\text{Se}_4$ 's reported direct  $E_g$  value  $\approx 1.67\text{ eV}$  attainable for pure  $\alpha$ -phase  $\text{CdIn}_2\text{Se}_4$  crystal derived by optical absorption method at  $293\text{ K}$  temperature.<sup>19</sup> Nevertheless,  $\text{CdIn}_2\text{Se}_4$  thin films grown at  $550\text{ K}$   $T_s$  ( $\approx 550\text{ K}$ ) have an  $E_g$  value ( $\approx 1.72\text{ eV}$ ) which is more or less equal to the reported bulk value. At higher  $T_s$  ( $>550\text{ K}$ ), the higher value of the  $E_g$  ( $\approx 1.79\text{ eV}$ ) can be explained by the dissociation of  $\text{CdIn}_2\text{Se}_4$  thin film.<sup>40</sup> The  $E_g$  value increases with the  $T_s$  increase.

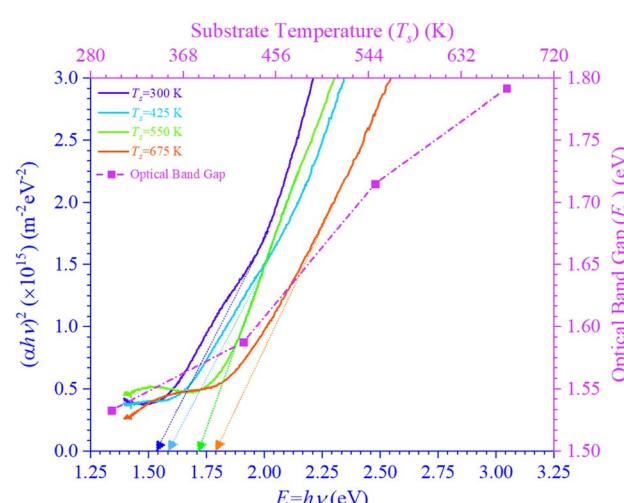


Fig. 3 Dependency of  $(\alpha h\nu)^2$  on the photon energy for  $T_s$ -controlled  $\text{CdIn}_2\text{Se}_4$  thin films.



As the  $T_s$  of  $\text{CdIn}_2\text{Se}_4$  thin films rose, as ostensible in Fig. 1,  $T(\lambda)$  changed toward higher energy ( $=h\nu$ ), signifying that the optical band gap energy widened concurrently. The increase in  $E_g$  with increase in  $T_s$  may be explained due to the higher  $T_s$  providing atoms greater energy to rearrange into well-ordered crystal structures, they diminish grain boundaries and dislocations, which typically bring localized energy states within the  $E_g$ . Larger grain sizes in polycrystalline films frequently result from higher  $T_s$ ; dominating grain growth improves electronic properties and widens the  $E_g$ .<sup>47–51</sup> The  $E_g$  value construed in the present investigation advocates that 550 K  $T_s$  deposited  $\text{CdIn}_2\text{Se}_4$  thin films are hypothetically adequate for thin film solar cell applications.<sup>52</sup>

There is diversity in the literature about the type of electronic transition and the corresponding  $E_g$  values. The  $E_g$  values of  $T_s$  tempered  $\text{CdIn}_2\text{Se}_4$  thin films determined in the present investigation differ when compared with the literature survey,<sup>23,24,45,52,53</sup> the incongruities explicated may be due to the individual or combined consequence of some factors like incongruent nature of the melting of  $\text{CdIn}_2\text{Se}_4$ , innumerable methods employed for synthesizing the compound and/or thin film deposition,  $T_s$ , an aberration in bulk and/or thin film's stoichiometry, charge impurities at the grain boundaries, lattice strain present in bulk and/or thin film, extent of structural disorder, etc.

The imaginary (IDC) ( $\varepsilon_i$ ) and the real (RDC) ( $\varepsilon_r$ ) dielectric constants are portions of the complex optical dielectric function; the IDC epitomizes the absorption of energy ( $E$ ) from an electric field ( $F_e$ ), which may be pronounced due to the motion of a dipole, while the RDC embodies the capacity of materials to lessen the speed of light ( $c$ ); IDC and RDC have unswerving kin with the  $\eta$  and  $k$ . Fig. 4 displays the relationship between energy and the changes in the IDC ( $=2\eta k$ ) (Fig. 4(a)), RDC ( $=\eta^2 - k^2$ ) (Fig. 4(b)), and loss factor/dissipation factor/loss tangent ( $\tan \delta$ ) ( $=\frac{\varepsilon_i}{\varepsilon_r}$ ) (Fig. 4(c))) for  $T_s$  modified  $\text{CdIn}_2\text{Se}_4$  thin films.

It is evident from Fig. 4 that IDC varies from  $0.89 \times 10^2$  to  $0.75 \times 10^2$ , RDC from  $-37.69$  to  $19.12$ , and  $\tan \delta$  from  $-2.37$  to  $-3.92$  with a change in  $T_s$ .

The negative values of the real dielectric constant ( $\varepsilon_r$ ) indicate that the semiconductor materials reveal metallic behavior.<sup>54</sup>

The rapport between  $E_g$  ( $=h\nu$ ) and the vicissitudes in the natural logarithm of  $\alpha$  ( $\ln \alpha$ ), optical conductivity ( $\sigma_o$ ), and electrical conductivity ( $\sigma_e$ ) is revealed in Fig. 5. An alteration in optical state occurs when the valence band (VB) tail becomes occupied, and the conduction band (CB) edge becomes unoccupied, as revealed by eqn (5).<sup>55</sup>

$$E_U \ln \frac{\alpha}{\alpha_0} = h\nu \quad (5)$$

In this context,  $\alpha_0$  represents an Urbach absorption coefficient (constant), and  $E_U$ , which stands for Urbach's energy, determines the slope of the exponential edge and can be seen as the width of the tail of localized states in the forbidden energy gap. The thermal vibrations of the lattice form the basis of the  $E_U$ .

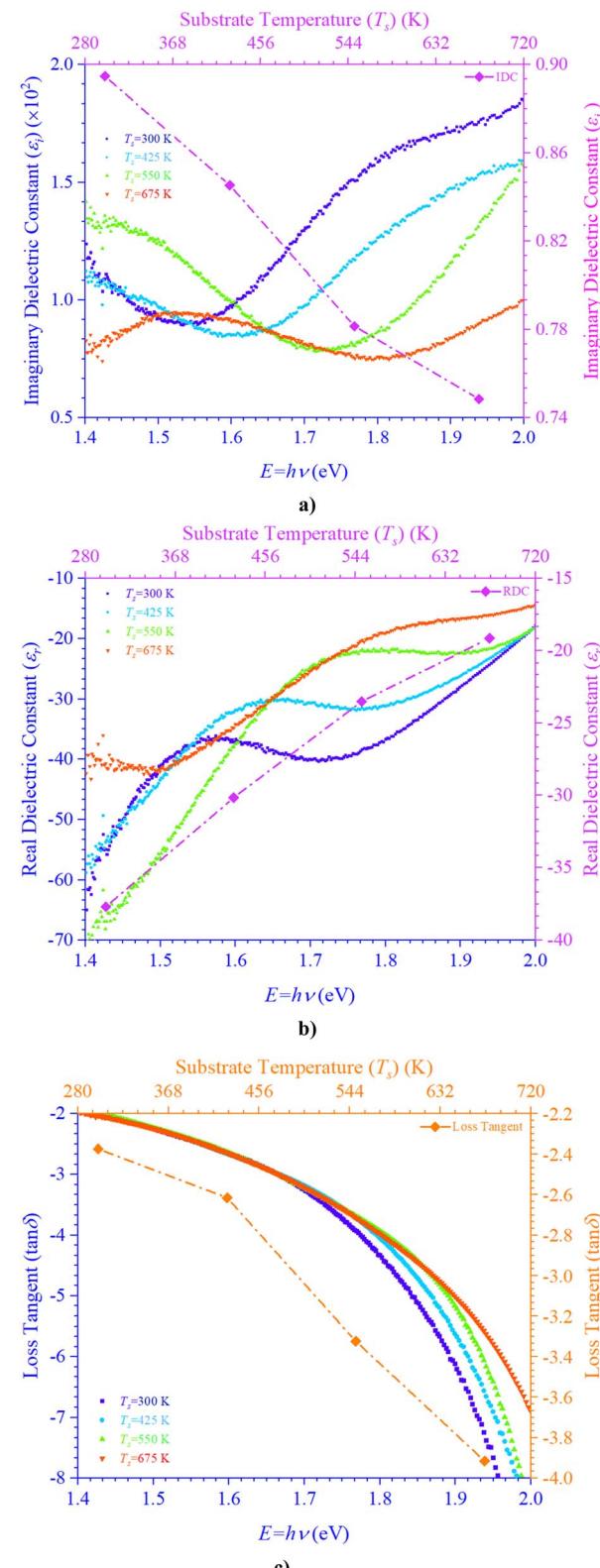


Fig. 4 Variation in (a) IDC, (b) RDC, and (c)  $\tan \delta$  of  $\text{CdIn}_2\text{Se}_4$  thin films as a function of energy.

The values of the  $E_U$  and  $\alpha_0$  attained from the  $\ln \alpha$  against photon energy ( $h\nu$ ) plot varies from  $1.38$  to  $1.97$  eV and  $4.75 \times 10^6$  to  $5.73 \times 10^6 \text{ m}^{-1}$ , respectively, with a change in  $T_s$ .



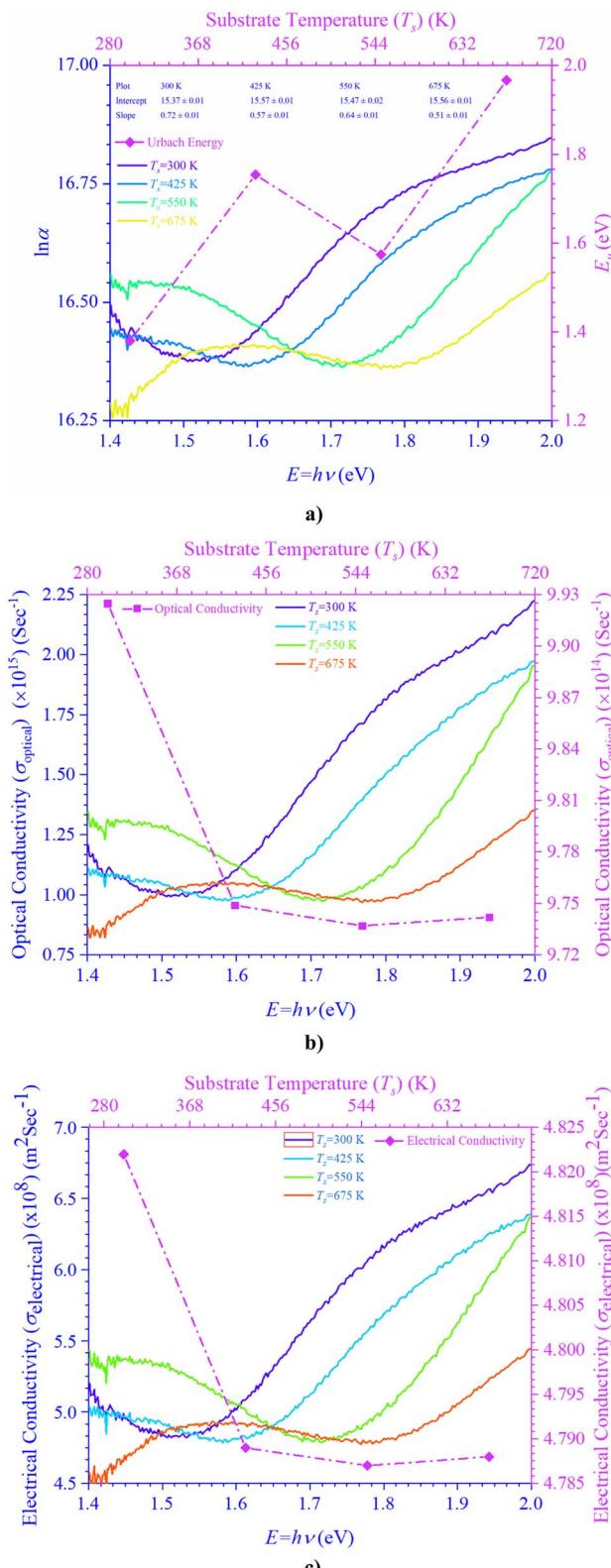


Fig. 5 Variation in (a) Urbach's energy, (b) optical conductivity, and (c) electrical conductivity of  $\text{CdIn}_2\text{Se}_4$  thin films as a function of energy.

The high density of localized states inside the  $E_g$ , as implied by the enormous  $E_u$  value, reveals numerous structural flaws in sample.<sup>55</sup> The powerful probes,  $\sigma_o$  ( $= \frac{\alpha\eta c}{4\pi}$ ) and  $\sigma_e$  ( $= \frac{2\pi\sigma_o}{\alpha}$ )

represent the mobility of the charge carriers induced by the alternating electric field of the passing electromagnetic waves,<sup>55</sup> and are employed in investigating the electrical properties of different materials. The values of  $\sigma_o$  and  $\sigma_e$  for  $T_s$  curbed  $\text{CdIn}_2\text{Se}_4$  thin films extracted at  $\lambda_c$  are plotted in Fig. 5(b) and (c), respectively.

The  $\sigma_e$  is reliant on  $\sigma_o$ , while  $\sigma_o$  is based on the  $\alpha$ . As light absorption grows,  $\sigma_o$  and  $\sigma_e$  climb, enabling electrons to acquire more energy and become free for conduction.<sup>56</sup>

Fig. 6 portrays IDC and RDC reliant volume (VELF) ( $= \frac{\epsilon_i}{\epsilon_r^2 + \epsilon_i^2}$ ) (Fig. 6(a)) and surface (SELF) ( $= \frac{\epsilon_i}{(\epsilon_r + 1)^2 + \epsilon_i^2}$ ) (Fig. 6(b)) energy loss functions depict electron and optical transitions in  $T_s$ , moderated

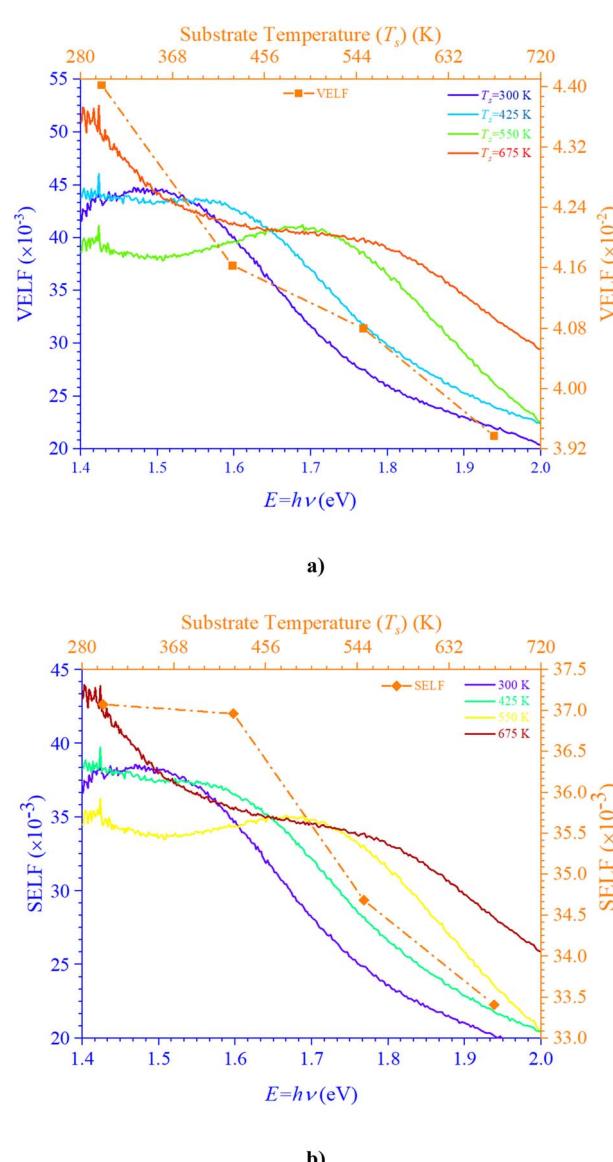


Fig. 6 Variation in (a) VELF and (b) SELF of  $\text{CdIn}_2\text{Se}_4$  thin films as a function of energy.

$\text{CdIn}_2\text{Se}_4$  thin films. The VELF and SELF are used to weigh the energy loss rates of electrons as they move across most of the surface. The peak values of VELF and SELF for  $T_s$  curbed  $\text{CdIn}_2\text{Se}_4$  thin films extracted at  $\lambda_c$  vary from  $44.03 \times 10^{-3}$  to  $39.37 \times 10^{-3}$  and  $37.08 \times 10^{-3}$  to  $33.41 \times 10^{-3}$ , respectively.

### 3.2. FTIR spectrophotometer analysis

The room temperature (RT) ( $\sim 300$  K) FTIR spectra of  $T_{\text{S}}$ -controlled  $\text{CdIn}_2\text{Se}_4$  thin films recorded in the 4000 to  $400\text{ cm}^{-1}$  wavenumber ( $\bar{\nu}$ ) range is offered in Fig. 7.

In synthesizing  $\text{CdIn}_2\text{Se}_4$ , only 5N (99.999%) pure Cd, In, and Se were employed; no other chemicals or chemical routes were used, and a non-chemical approach was used to deposit  $\text{CdIn}_2\text{Se}_4$  thin films.

A peak perceived in  $T_s$  tempered  $\text{CdIn}_2\text{Se}_4$  thin films between 880–892  $\text{cm}^{-1}$  wavenumber ( $\bar{\nu}$ ) can be construed due to bending  $\text{C}=\text{C}$  and/or  $\text{C}-\text{H}$  functional groups and alkene and/or 1,2,4-trisubstituted and/or 1,3-disubstituted class. A peak seized between 757 and 760  $\text{cm}^{-1}$  wavenumber ( $\bar{\nu}$ ) can be interpreted due to stretching  $\text{C}-\text{Cl}$  and halo compound functional group, and/or bending  $\text{C}=\text{H}$  functional group and 1,2-disubstituted or monosubstituted class. The absorption peaks due to  $\text{C}=\text{C}$ ,  $\text{C}-\text{H}$ , and/or  $\text{C}-\text{Cl}$  functional group/s habitually parade a stout absorption edge in the FTIR spectra if existing in the  $T_s$  modified  $\text{CdIn}_2\text{Se}_4$  thin films. The absorption edges are evident in the  $T_s$  controlled  $\text{CdIn}_2\text{Se}_4$  thin films' FTIR spectra between 880–892  $\text{cm}^{-1}$  and 757–760  $\text{cm}^{-1}$  wavenumbers ( $\bar{\nu}$ ), but they are frail; henceforth, it may be concluded that the  $T_s$  moderated  $\text{CdIn}_2\text{Se}_4$  thin films are intrinsic and free from any functional group.<sup>21,57</sup> The shift in wavenumber ( $\bar{\nu}$ ) dependent absorbance (A) maxima conspicuous in Fig. 7 can be explained by the amorphous, polycrystalline, and polyphase nature of the  $T_s$  controlled  $\text{CdIn}_2\text{Se}_4$  thin films. Peaks in FTIR spectra of  $T_s$  tempered  $\text{CdIn}_2\text{Se}_4$  thin films at wavenumbers ( $\bar{\nu}$ )  $3437.74 \pm 49.87 \text{ cm}^{-1}$ ,  $1658.41 \text{ cm}^{-1}$ , and  $1597.77 \pm 23.32 \text{ cm}^{-1}$  are detected and dispensed by the instrument, but their amplitude is impending almost to zero ( $\simeq 0$ ), so they are not identified and indexed.

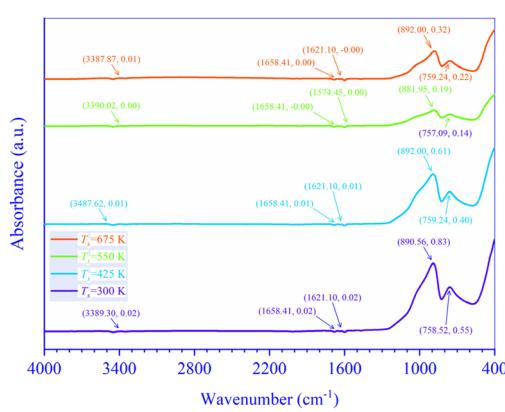


Fig. 7 FTIR spectra of  $T_s$  modified CdIn<sub>2</sub>Se<sub>4</sub> thin films

## 4 Conclusions

The optical characterization of the PLD technique deposited  $\approx 100$  nm thick  $\text{CdIn}_2\text{Se}_4$  thin films on amorphous quartz glass at mixt  $T_s$  was carried out by recording RT  $T(\lambda)$  spectra in the 400 to 900 nm  $\lambda$  range using a UV-Vis-NIR spectrophotometer. A shift in  $T_s$  modulated  $\text{CdIn}_2\text{Se}_4$  thin film's  $T(\lambda)$  maxima is evident and exhibits high absorption in the visible region with a hump at  $750.00 \pm 58.50$  nm and depicts  $\alpha$  of the order of  $\approx 10^7$   $\text{m}^{-1}$ . The  $\eta$  values deduced for  $T_s$  tempered  $\text{CdIn}_2\text{Se}_4$  thin films ( $\approx 3.27$ ). The  $k$  decreases with an increase in  $\lambda$  and reaches a minimum at  $\lambda_c$ . Because of their amorphous nature, the  $E_g$  of  $\text{CdIn}_2\text{Se}_4$  thin films grown below 550 K  $T_s$  is slightly lower than the published value; while the polycrystalline thin films of  $\text{CdIn}_2\text{Se}_4$  made at a  $T_s$  of 550 K have an  $E_g$  value that is approximately equal to the reported bulk value, the polyphase structure of the  $\text{CdIn}_2\text{Se}_4$  thin film at higher  $T_s$  explains greater values of the  $E_g$ . For  $T_s$ -modulated  $\text{CdIn}_2\text{Se}_4$  thin films, IDC, RDC, and  $\tan \delta$ , dependent on  $\eta$  and  $k$  have all been determined and compared with the reported data. The values of the  $E_U$ ,  $\alpha_0$ ,  $\sigma_o$ ,  $\sigma_e$ , VELF, and SELF of  $\text{CdIn}_2\text{Se}_4$  thin films limited by  $T_s$  were retrieved at  $\lambda_c$ . The purity of  $\text{CdIn}_2\text{Se}_4$  thin films formed at different  $T_s$  was verified by recording the FTIR spectra. The conclusions drawn from our extensive research include that the  $T_s$ -dependent optical parameters obtained in this study will aid in developing upcoming thin film electronic devices in which  $\text{CdIn}_2\text{Se}_4$  thin film is one of the semiconducting materials. The impact of thin film  $d$  on the optical characteristics of  $\text{CdIn}_2\text{Se}_4$  thin films deposited by PLD has not been investigated by researchers worldwide, despite the conclusion that this is a topic of future research. Research on compounds based on the II-III<sub>2</sub>-VI<sub>4</sub> group in silicon-based microelectronic devices is underway. However, no attempt has been made to create a junction between silicon and  $\text{CdIn}_2\text{Se}_4$ , so the results of this study may provide insight into the development of new silicon-based microelectronic devices in the future. Additionally, the current research suggests that  $\text{CdIn}_2\text{Se}_4$  thin films are potential choices for designing and developing future high-efficiency opto-electronic systems.

## Data availability

The authors declare that the data supporting the findings of this study are available/included in the paper's main text.

## Author contributions

All writers contributed to the study conception and design. Material preparation, data gathering, and analysis were accomplished by S. D. Dhruv, Tanvi Dudharejiya, Sergei A. Sharko, Aleksandra I. Serokurova, Nikolai N. Novitskii, D. L. Goroshko, Parth Rayani, Jagruti Jangale, Vanaraj Solanki, P. B. Patel, U. B. Trivedi, J. H. Markna, Bharat Kataria, and D. K. Dhruv. D. K. Dhruv wrote the first draft of the manuscript, and all authors commented on previous versions. All authors read and permitted the final manuscript. S. D. Dhruv: methodology. Tanvi Dudharejiya: conceptualization. Sergei A. Sharko:

investigation. Aleksandra I. Serokurova: formal analysis. Nikolai N. Novitskii: *visualization*. D. L. Goroshko: writing-review & editing. Parth Rayani: software. Jagruti Jangale: validation. Vanaraj Solanki: data curation. P. B. Patel: resources. U. B. Trivedi: resources. J. H. Markna: software. Bharat Kataria: project administration. D. K. Dhruv: writing-original draft, supervision.

## Conflicts of interest

The submitted work is the authors' original research work and has not been communicated elsewhere for publication. On behalf of all co-authors, I declare no conflict of interest.

## Acknowledgements

The work was supported by the Department of Science & Technology (DST), Government of India, New Delhi (File number: DST/INT/BLR/P-36/2023) and Belarusian Republican Foundation for Fundamental Research (File number: F23INDG-005) under the India-Belarus Programme of Co-operation in Science & Technology (Joint Research Project). The authors thank the Sophisticated Instrumentation Centre for Applied Research and Testing (SICART), Vallabh Vidyanagar-388120, Anand, Gujarat, India, for providing discounted UV-Vis-NIR and FTIR spectrophotometer facilities.

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