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Recent Advances on Thermochromic Inks for Security Applications

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ABSTRACT

In the security printing industry, high complexity stimuli-responsive functional inks have been used to develop anti-counterfeit elements, such as authentication markers and encrypted patterns. Through these applications, they create more reliable products, reduce economic losses, help retain brand identity, and assist in identifying counterfeit products. Thermochromic inks, owing to their temperature-responsive color-shifts, have gained considerable interest in the last decade as functional inks in a variety of fields. Here, we outline the different types of thermochromic materials, their properties, development scales, and how they have been previously applied, with a focus on their use as security inks for anti-counterfeit elements. Despite promising functional properties, thermochromic materials are hindered from use by low stability, generally high toxicity, and a lack of printing optimization. In recent years, however, efforts have been made to enhance their durability and sustainability, along with an improved understanding of their thermochromic transitions and novel ink formulations, granting great promise for more diverse and specialized applications. Additionally, their integration alongside other functional materials has promoted the development of multilevel high-security elements with potential for future anti-counterfeit technologies. In this review, we highlight recent advancements in property enhancement, sustainable development, and scalability of thermochromic security inks.

1 | Introduction

1.1 | The Need for Anti-Counterfeit Technology

Counterfeit products are illegally manufactured and sold in violation of intellectual property rights by pretending to be genuine licensed products [1]. This practice is harmful to both consumers and manufacturers, with consumers being served lower-quality and potentially overpriced products that might evade safety or health regulations and bolster illicit activities, while manufacturers suffer a loss of market shares, economic growth, and reputability for their own original products [2, 3]. A report published in 2024 by the European Commission highlights a 77% increase in seized counterfeit products in 2023 compared to 2022, which itself had an estimated 5.8% of imported goods as counterfeit or pirated products in the EU [4]. There are growing indications that the manufacture and assembly of counterfeit goods is increasingly occurring within the EU itself, heavily relying on the digital domain to distribute these items and source components from criminal networks outside the EU [5]. To combat the spread of counterfeit products, manufacturers and retailers make use of marketing and advertising [6], customer awareness campaigns [7], more varied product differentiation, such as new features, functionalities, or anti-counterfeit technologies [7–9], all of which entail greater costs. Therefore, there is a pressing and ongoing demand for new, innovative, and scalable

FIGURE 1 | Types of security applications for functional, stimuli-responsive security inks.

security technologies to effectively combat the escalating threat of product counterfeiting in the global economy [10, 11].

Anti-counterfeiting, authentication, and encryption technologies exist as a direct response to counterfeiting, ensuring a product's design, functionality, or quality can be trusted, reducing economic losses and better ensuring public safety. Simpler anti-counterfeit strategies, such as security inks, watermarks, barcodes, and digital signatures, have seen widespread use in most industries but can suffer due to ease in duplication or lack of public awareness [12, 13]. The ideal solution is to develop anti-counterfeit technologies with a high level of security through a complex and non-replicable composition while retaining simple product application, quick authentication speed, high reliability, and ease in usability (especially for authentication purposes) [14]. As a result, the continuous advancements in the design and development of anti-counterfeit technologies with these properties present a significant opportunity to meet the growing demand in the market.

Functional security inks are a major sector of anti-counterfeit technology and are designed and developed using materials with variable physical, chemical, or optical properties, providing an enhanced security level by requiring specific external stimuli [11]. These changes can be visual, such as the overt optical variability in fluorescence, chromism, holographic, or iridescent inks, or more covert property changes, such as in magnetic or biometric inks [15, 16]. They are used in a wide array of security applications, as illustrated in Figure 1. This review is focused on thermochromic materials, which change their reflectance and absorbance in response to temperature variations, resulting in a perceived color-shift, and which are a staple in stimuli-responsive chromic ink types [17].

1.2 | Thermochromic Inks and Their Applications as Products

Thermochromic materials (TCMs) possess a color-changing effect triggered by a specific transition temperature range. This color variation can be reversible or irreversible, transitioning between colored and/or colorless states, and the transition can occur at a specific temperature value or over a wider range, depending on the particular properties of the material. They can be organic, inorganic, or organic–inorganic hybrids, with the transition mechanism behind the color-shift being reliant on changes in molecular structure, coordination geometry/number, crystalline structure, phase transitions, or band gap energies, depending on the material type and specific properties [18].

Reversible TCMs are the most widely used, finding applications in energy-efficient smart-windows and coatings [19–21], temperature sensors [22], intelligent textiles [15, 23], wearable medical devices and treatments [24, 25], heat storage systems [26], and as printed patterns on product labels [18, 27]. Irreversible TCMs on the other hand, find fewer, but more specialized uses, such as reliable temperature markers on products, namely their use in food or beverage packaging labels for quality control [28, 29] or for reporting maximum temperature variations in clinical procedures, such as thermal ablation devices, and machinery to facilitate testing and avoid malfunctions [30, 31].

For the security industry in particular, TCMs have found use as temperature markers and as functional security inks for authentication and encryption in anti-counterfeit technologies, as seen in Figure 2 [16]. Their practical advantage of not requiring a specialized reading device or electrical system while providing a visually identifiable and potentially measurable color-shift makes it highly interesting for development. Additionally, counterfeiting and forgery commonly resort to heat and steam-based processes, including the use of hot solvents, surface drying, and adhesive dilutants to remove quality certification seals or authentic labels, with the intent of reusing them to sell counterfeit or duplicate products [10]. TCMs with an irreversible color-shift, in particular, can be used to detect these processes, permanently damaging a label to prevent its re-use and/or invalidating its credibility, as well as marking it as having been tampered with to facilitate the detection of counterfeits. Reversible TC security inks, on the other hand, are used in encryption and authentication as intelligent sensing elements, including variable barcode tags [32] or integrated into complex patterns such as QR codes, to bolster multifunctionality in the design of anti-counterfeit elements [33].

Various security technology manufacturers distribute different types of commercial TC security inks or devices, such as those provided by Securikett, NanoMatriX, SpotSee (which recently acquired TMC and LCR Hallcrest LLC, two global leaders in temperature-responsive products), CromaticTechnologies Inc, SFXC, and QCR Solutions Corp. Some of these have had their products used in property enhancement research [36–38], printing process optimizations [39], and innovative applications [40]. Anti-counterfeit technology is also directly distributed, as seen in Figure 3, with BlindSpotz, developed by CTI, a dual-indicator sensor that can detect if a product has been subjected to excessively high or low temperatures, irreversibly changing color to indicate that the product might have lost integrity, been tampered with, or become unsafe to use [41]. Or the thermochromic smart-label solutions provided by Securikett, which can be used to detect tampering using heat, cold, or liquids, alongside physical cuts that damage the label to prevent peeling and reuse [42].

The thermochromic materials market has steadily grown each year, owing to more efficient production cycles and rising demand for specialized thermochromic components in security, medicine, and interactive/smart packaging [43]. Many studies also highlight

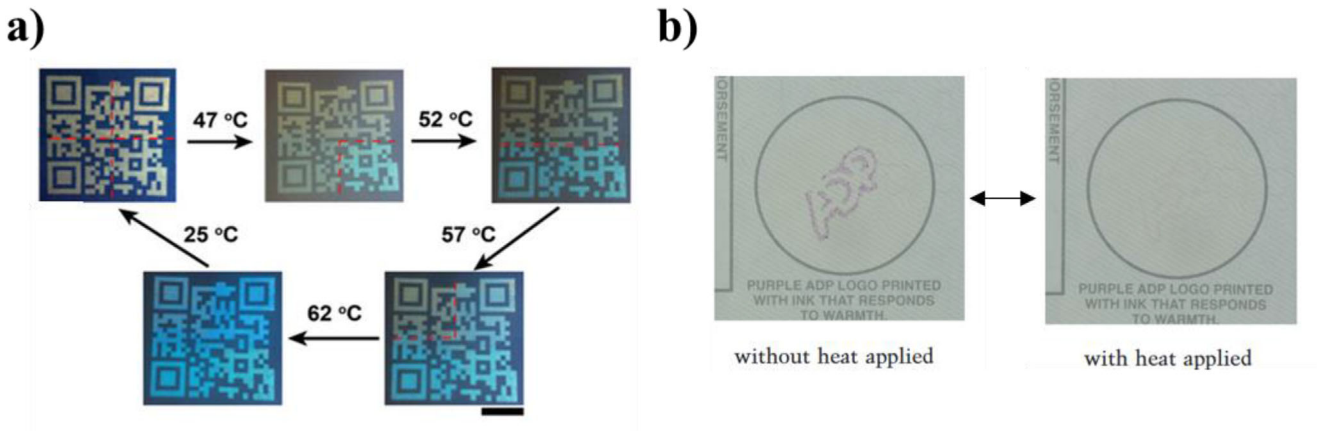

FIGURE 2 | Representative examples of thermochromic security elements: a) A tunable multilevel-encryption thermochromic QR code with distinct emissions at certain temperatures (Reused with permission from [34] Copyright 2020 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, all rights reserved) and b) a simple body-heat activated thermochromic logo for quick authentication (Reproduced with permission under a CC BY 4.0 open access license from [35] Copyright 2003 by the authors, all rights reserved).

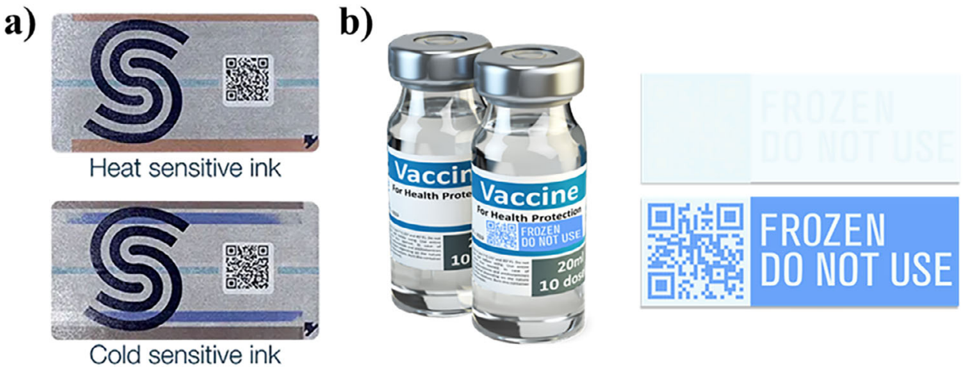

FIGURE 3 | a) Thermochromic sensors developed by Securikett for smart labelling solutions (Adapted with permission from [42], Copyright 2025 Securikett, all rights reserved) and b) Blindspotz freeze-warning technology for medicine and food product safety by Chromatic Technologies Inc (Adapted with permission from [41], Copyright 2024 by Chromatic Technologies Inc., all rights reserved).

thermochromic inks as becoming increasingly more relevant as value-adding to products, particularly for applications in smart packaging, security printing, or visual marketing strategies. In the last decades, research on TCMs has improved immensely through the advancement of nanotechnology and functional material development processes, facilitating the optimization of their properties and targeting them for specific applications [18]. This has also resulted in a gradual increase in interest in TCM research, as shown in Figure 4, which highlights the growing trend of articles and reviews focusing on TCMs in the last two decades. Additionally, alongside this growth, we can also identify a sharp rise in interest for applications targeted at thermochromic security technologies, particularly in the last five years. Innovations to enhance their properties will expand their range of applications, further driving their growing trend as versatile and desired smart materials. Previous works have highlighted advancements and goals in functional ink development as a whole within the field of anti-counterfeiting [15, 17, 44]. This review focuses specifically on outlining current TCMs and TC security ink technologies as well as the challenges they present, with an emphasis on the innovations that could be feasibly targeted toward the functional security ink industry.

2 | Classification of Thermochromic Materials and their Transition Mechanisms

2.1 | Organic-Based Systems

2.1.1 | Leuco Dyes

Thermochromic leuco dye systems are the most prominently used organic-based TCM, capable of reversibly transitioning from a colored state to a colorless one [45]. The color-shift occurs from the interaction between three components: A color former (the electron donor leuco dye), a developer (a proton donor weak acid), and an organic phase-change medium (a solvent, generally a fatty alcohol or ester) [46, 47]. The TC transition, as observed in Figure 5a, stems from a molecular structure change in the leuco dye-developer-solvent system. Below the solvents' melting, the color former leuco dye is induced into a colored lactone open-ring state when bonded to the proton-donor developers. Above the melting point of the organic solvent, the dye-developer bond breaks, being replaced with a dominant solvent-developer bond, which brings the color former leuco dye into its close-ring colorless state [48]. The transition temperature of the complex

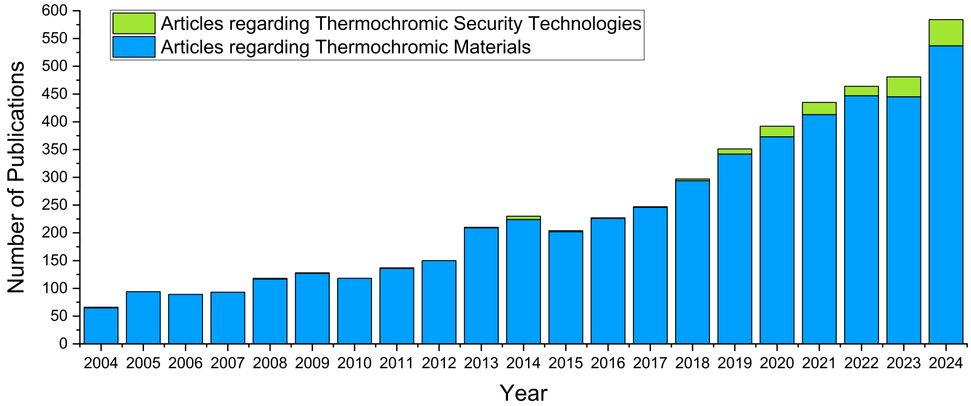

FIGURE 4 | Number of scientific papers regarding thermochromic materials and thermochromic security or anti-counterfeit technologies published in the last 20 years (Citation Report graphic data is derived with assistance from Clarivate Web of Science, Clarivate 2025, all rights reserved).

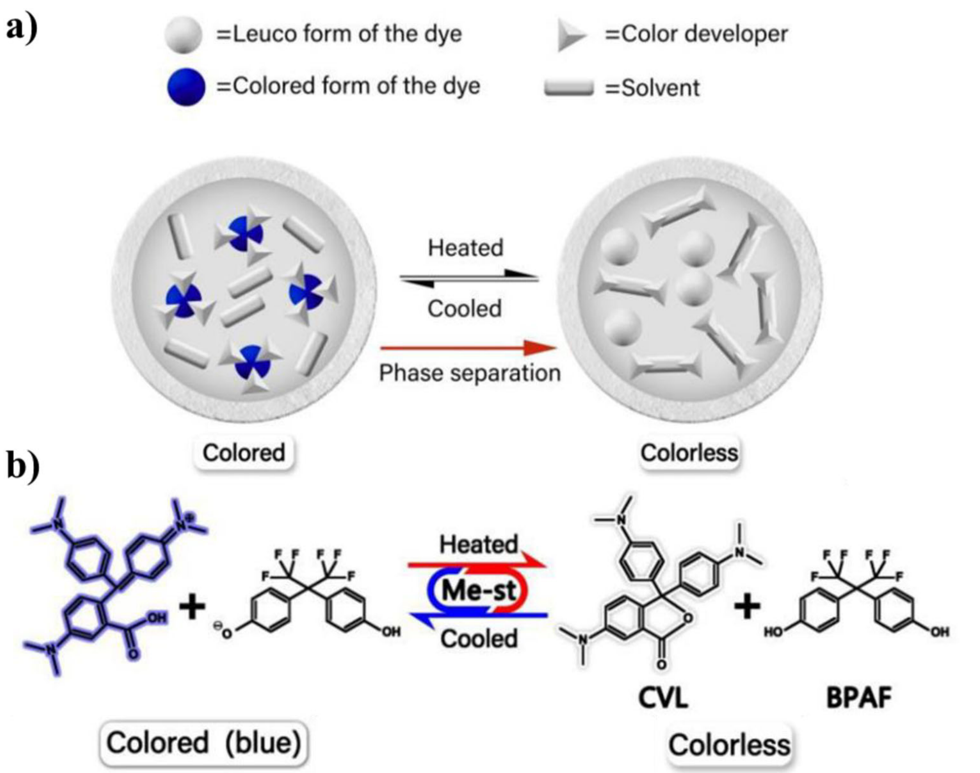

FIGURE 5 | a) Thermochromic transition behavior of a crystal violet lactone/bisphenol AF/methyl stearate (leuco dye/developer/solvent) system, b) Chemical formula of the reversible transition of crystal violet lactone with a Bisphenol AF (BPAF) developer. (Adapted with permission under a CC BY open access license from [50], Copyright 2023 by the authors, all rights reserved).

is primarily controlled by the melting and crystallization points of the long-chain organic solvent, although the structures of the color former, developer, and solvent can all influence the competitive binary interactions that permit the thermochromic effect. Furthermore, the acidity of the developer and basicity of the color former, as well as their solubility in the solvent, are all influential in the interactions between them [49].

Crystal violet lactone (CVL) is the most commonly used and studied leuco dye for thermochromic applications due to its bright color, high discoloration speed, and simple transition mechanism, shown in Figure 5b [51]. This simple yet efficient transition to the colored state is owed to the single requirement of the addition of a proton to the oxygen atom of the lactone's carbonyl group to achieve the brightly colored open-ring structure [49]. Derivates of Rhodamine B, another commonly used leuco dye alongside CVL, have also shown great promise for both reversible and irreversible TC applications, due to their multifunctional properties, showcasing strong fluorescence and chromism in response to pH, water content, electric fields, light, and of course, heat [52]. Additionally, the reversible nature of its color-shift can be controlled by the physical state of the system. In the presence of a solvent, the increase in temperature causes a proton transfer into the ring structure, leading to the color development of the system, which is fully reversible back to its initial state. However, in a solid state, with only the developer and rhodamine B derivate, an increase in temperature causes an irreversible shift to the open-ring molecular structure, retaining the transferred proton and colored state even after cooling [53]. These novel properties grant leuco dye-based TCMs higher versatility and potential for applications beyond their standard use as reversible heat-sensors.

In the security industry, leuco dye systems have been used primarily for quality control or authentication markers. By printing leuco dye-based TC ink without a developer onto a surface and applying it at a later date using a corresponding co-solvent, we can allow color to develop in a controlled environment to test the authenticity of a product [54]. Using this principle, leuco dye-based inks can be used in conjunction with conventional inks to imprint a hidden message or pattern into a substrate. This results in an exceedingly difficult to replicate security element, on account of requiring the exact material, density, and color hue of the specialized thermochromic pigments to match the conventional inks in an unnoticeable manner [55]. Another authentication method lies in integrating leuco-dye-based pigments with heat-emitting systems, reacting with their expected color-shift in response to sudden temperature variations. An example of this can be found in the Yb-doped Sr/Zr hybrid oxide particles developed by Molina-González et al. (2021), which, when combined with commercial thermochromic leuco dye pigments, formed a secure anti-counterfeit system activatable by a laser diode at a short near-infrared wavelength range [56]. The thermochromic response of this system to a very specific stimulus leads to a reliable high-security element providing easy access to authentication with the necessary equipment.

Moreover, stimuli-responsive photonic crystals with embedded leuco dye-based pigments have also shown great promise in sensors, smart-displays, and anti-counterfeit, owing to their high brightness, iridescence, quick response speed, and sensitivity. They are formed by the periodical arrangement of two or more media with different dielectric constants, and their low fabrication cost, non-volatility, and general environmentally friendly properties could result in a more sustainable alternative for anti-counterfeit information encryption elements in textiles [15, 33].

2.1.2 | Liquid Crystals

Thermochromic cholesteric or chiral liquid crystals are also a major type of TC ink. These compounds showcase a color-shift within a specific temperature interval, called the bandwidth, and remain colorless outside of this region [36, 57]. This color-shift is also dynamic, spanning the entire color spectrum from red to blue, with each color being attributed to a narrow temperature segment of the bandwidth. This effect is known as the color-play of the TC liquid crystals and distinguishes their color-changing properties from other TCMs [57]. This TC transition is triggered by a shift in the orientational structure, namely, the pitch, of the liquid crystals with temperature, and the way light reflects off the organized helical structures during phase transitions. For temperature values below the bandwidth, the TC will be in a transparent solid state, and for temperatures above the bandwidth, the TC liquid crystal will be in a transparent liquid state [58]. Only within the temperature bandwidth of the material, during the transition from the solid to the liquid state, or vice versa, the light reflected off the microencapsulated TC liquid crystals suffers interference in the visual spectrum range, resulting in the color-play effect, as shown with the fiber embedded with TC liquid crystal pigments in Figure 6a [59]. The bandwidth interval where this color-play effect occurs can span between 1°C to 30°C, loosely classified as narrow-bandwidths for short intervals around 1–2°C, or as wide-bandwidths for intervals higher than 5°C. Narrow-bandwidth TC liquid crystals in particular possess high sensitivity to temperature variations as small as 0.5°C and can provide a response time as short as 3 ms, making them ideal for high-sensitivity applications [60]. The beginning of the bandwidth, named the onset temperature, for conventional cholesteric TC liquid crystals, is adjustable and depends primarily on their specific crystalline compositions, with available operating values for the transition between −30°C to 150°C being widely available for commercial or industrial applications [61, 62]. Much like leuco dyes, they also require some form of encapsulation to guarantee isolation and prevent contamination from other ink components, which in this case could disturb the organized liquid crystal structure, critical for its intended optical properties [63].

Liquid crystal TC inks find less use compared to leuco dyes, due to higher processability costs and lower stability, owing to a high UV and chemical sensitivity of the helical structure [36]. The color-play effect and specified bandwidth of a TC liquid crystal can suffer significant unintended change from external interference, such as UV exposure, excessive heat, or chemical interactions with oxidative agents. Additionally, a black substrate or background, as shown in Figure 6b, is essentially required to absorb light that is not scattered by the liquid crystals themselves; otherwise, light reflected off the substrate can interfere with the color-play effect, greatly diminishing its visible effect [67]. The microcapsule structure, binder between microcapsules or ink formulation itself, can also act as a barrier for light, further interfering with the color-play effect, requiring a more careful consideration of their properties during development [65]. These limitations hinder the widespread application of this TCM type, especially when accounting for their higher processability cost when compared to leuco dye-based inks [61].

Despite these considerable drawbacks, some practical examples are still available, such as the TC liquid crystal substrates provided by industry giants in the field, including LCR Hallcrest, SPLinx, and Holo Solution Inc., which can be applied directly to product surfaces to provide heat-sensitive components. These commercially available TC liquid crystals are primarily based on esters of cholesterol or methyl-butyl-phenol esters, finding use as sensors, inks, and smart window components. Of note, Yang et al. (2022) showcased coatings based on TC cholesteric liquid crystal microcapsules mixed into printable coating slurries, which exhibited good temperature responsivity with a wide 12–42°C bandwidth and were deemed promising for use in complex-to-reproduce anti-counterfeit labels [68].

2.1.3 | Polydiacetylenes

Polydiacetylenes (PDA's), a class of conjugated polymers, are also very promising functional materials, due to their facile

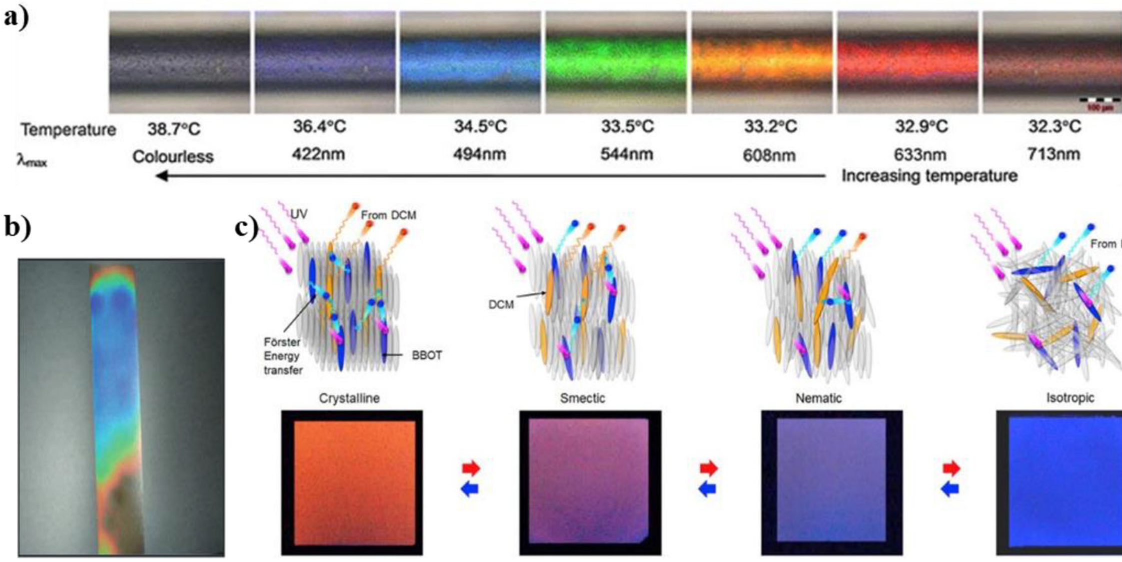

FIGURE 6 | a) A representative example of the color-play effect of a thermochromic liquid crystal-embedded fiber, showcasing the maximum reflected wavelengths throughout the bandwidth interval (Adapted with permission from [64] Copyright 2025 Springer Nature, all rights reserved), b) a thermochromic liquid crystal, demonstrating a range of activation phases for the color-play effect on black paper (Adapted with permission under a CC BY-NC 4.0 open access license from [65] Copyright 2018 by the authors, all rights reserved), and c) the mechanism for thermochromic liquid crystal structures based on dual embedded non-chromic dyes at different phases (Adapted with permission from [66] Copyright 2020 Elsevier Ltd., all rights reserved).

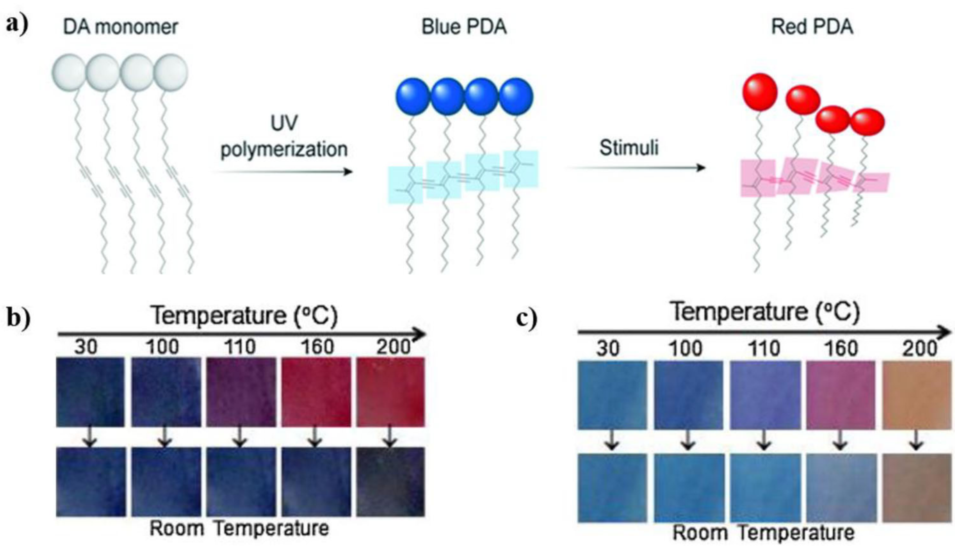

FIGURE 7 | a) Photopolymerization of DA monomers into the stimuli-responsive polydiacetylene conjugated polymer structures (Adapted with permission under a CC BY 3.0 open access license from [78] Copyright 2022 The Royal Society of Chemistry, all rights reserved); b) The reversibility of the thermochromic effect for poly(PCDA)/ZnO films within a a) polystyrene + toluene polymer matrix and a c) polyvinyl alcohol + water matrix (Adapted with permission from [73] Copyright 2015 Elsevier B.V., all rights reserved).

synthesis methods through diacetylene monomer assembly without by-products, diverse morphologies, biocompatibility, and their stimuli-responsive chromic properties, including TC properties in response to heat [69, 70]. Diacetylene monomers (DA) assemble into organized structures in solution and solid states through topo-polymerization induced by UV irradiation, without the need for chemical initiators or catalysts, into stimuli-responsive PDAs, as shown in Figure 7a. The color-shift of a PDA-based TCM is dependent on configuration and length [71], where heat or chemical stimulation results in a structural relaxation, causing a color transition conventionally from a room-temperature blue hue to a purple, red, or orange hue based on their specific absorption properties [72]. The wide transition ranges and diverse reversible or irreversible color evolution of PDAs with temperature distinguish them from other TCMs, providing great utility as temperature sensors.

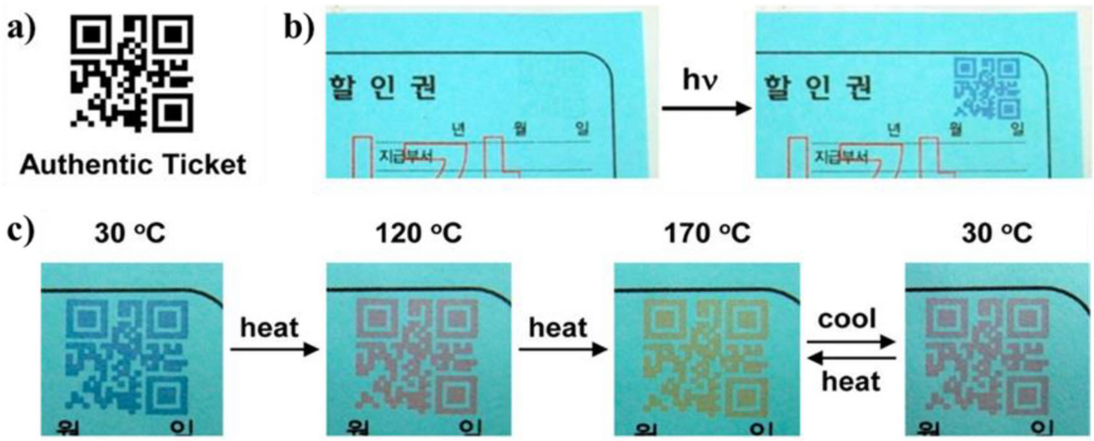

FIGURE 8 | a) A multifunctional QR code used for authentication, b) which can be revealed by UV irradiation after being printed through photopolymerization of the DA monomers, and c) provides a unique thermochromic response to specific temperature intervals (Adapted with permission from [79] Copyright 2013 American Chemical Society, all rights reserved).

For most pure PDA-based materials, such as 10,12-pentacosadiynoic acid (PCDA, one of the most widely studied, commercially available PDA's), this TC transition is irreversible, with the polymer structure retaining the high-temperature configuration even after cooling [70]. However, incorporating poly(vinyl alcohol) and poly(vinylpyrrolidone) into the PDA assembly process can yield reversible thermochromism, up to the degradation limits of the polymeric structure, by increasing the strength of side chain interactions, limiting PDA segmental movement [73]. Similarly, the presence of sodium or zinc ions or oxide nanoparticles can also promote reversible thermochromism through ionic interactions with PDA, limiting functional group movement [74]. Knowing this, reversible PDA-based TCMs are commonly prepared using zinc oxide (ZnO) nanoparticles, where diacetylene monomers can assemble upon their surfaces [72, 75]. These PDA and ZnO composites have highly tunable properties, such as specific color-tones or reversibility, through varying solvents and PDA chain length, as seen in Figure 7b,c, and have been applied toward diverse applications, for instance, in assisting medical treatments [76] or as colorimetric sensors in smart labels [77].

In the security industry, PDA-based TCMs have been developed as inks for encrypted elements, as shown in Figure 8, due to the initial transparent state of DA monomers, which facilitates hidden patterns in products. Over a decade ago, Yoon et al. (2011 and 2013) demonstrated how an inkjet compatible polymerizable DA suspension could be printed onto bank notes for anti-counterfeit, with and without the assistance of surfactant molecules to stabilize them [69, 79]. Their printable suspension was invisible in its initial state and could be revealed through UV-exposure, topo-polymerizing into the organized blue-hued PDAs at room temperature. The decrypted pattern could then be verified using a heat treatment, resulting in a standard blue-to-red PDA color-shift, which could be reversed and remained stable for over 100 cycles. More recently, Mergu et al. (2019) developed napthalimide-functionalized PDA TCMs in a polymer matrix, with a reversible green-to-red transition, adapted as both a thin film and a pen-to-paper wax-based pen for document identification and authentication, demonstrating an innovative color variation and a novel adoption of PDAs for security elements [80].

Despite sporadic studies showcasing the potential of PDA-based TCMs for anti-counterfeit, they have been seldom adopted for industrial production and practical products, likely due to considerably higher development costs when compared to their organic leuco dye counterparts. However, the unique TC properties of PDAs, with wide transition temperature ranges and potential for targeted irreversible color-shifts, grant them great versatility for prospective applications in the security industry as developments in this field continue.

Additionally, other macromolecular conjugated polymers, such as polyaniline, polythiophenes, and poly(phenylenevinylene) have demonstrated a degree of heat-responsive thermochromism, attracting attention as potentially novel functional materials. However, they have been found to be less practical than the aforementioned PDA-based TCMs, due to higher complexity in preparation, requiring chemical initiators, catalysts, or higher temperatures to undergo polymerization after deposition in a comparable fashion to PDAs [27], making them far less economically viable.

2.2 | Inorganic and Hybrid-Based Systems

The majority of inorganic TCMs exhibit their color-shift by means of a reversible or irreversible phase transition caused by a change in ligand geometry, crystalline lattice structures, coordination number of metal complex species, or band gap energy change for semiconductors [81]. Their TC transition points are considerably higher than organic-based TCMs, ranging from 70°C to over 900°C, but generally remain stable and structurally sound above 200°C, demonstrating far greater thermal and chemical durability than organic-based TCMs [18, 82]. Despite this, for purely inorganic TC complexes, their naturally high transition temperatures, generally nondegradable hazardous nature, complex manufacturing processes, and considerably higher cost are limiting factors for their widespread use and diminish interest in their research and development for TC applications [83, 84]. The European Commission, as of 2023, has identified several materials as strategic raw materials. These include vanadium, manganese, copper, cobalt, and nickel, which are strategic but

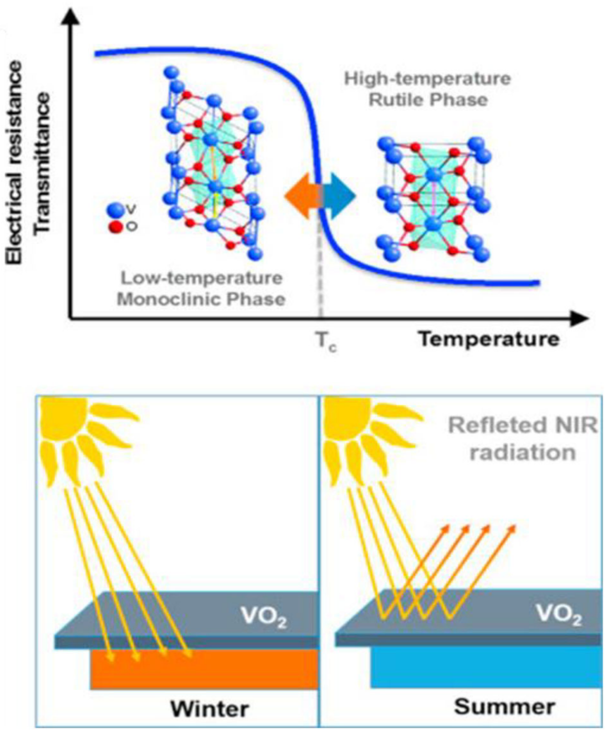

FIGURE 9 | Schematic of the crystalline structure transition mechanism and the resulting thermochromic change in reflectance of solar light for VO_2-coated smart surfaces (Adapted with permission from [21] Copyright 2016 Elsevier B.V., all rights reserved).

not critically rare. Additionally, rare earth elements, such as ytterbium, were classified as critical materials. Notably, all of these materials exhibit a degree of thermochromism in one or more forms. This further hinders their adoption in TCM development due to sourcing scarcity and/or the necessity for other fields with no suitable substitutes [85].

The major exception to this lies in vanadium dioxide, VO_2, where, despite its classification as a critical raw material and a moderately volatile price, it has not seen a significant decline in use for research and technological advancements [85]. Vanadium dioxide has established itself as the most prominent inorganic TCM due to possessing a reversible metal-to-insulator TC phase transition at 68°C, which greatly affects its near-infrared reflectance, as represented in Figure 9 [86, 87]. This abrupt transition stands out due to a considerably low activation temperature when compared to alternative TC transition metal species, and even lower transition temperatures can be achieved by tungsten doping, though this can hinder functional properties and stability [88]. It is predominantly applied in smart-window and energy storage device development, with extensive research covering these topics [21, 89, 90]. Recently, however, innovations using VO_2-based materials have also resulted in some notable high-stability anti-counterfeit technology development, though primarily in a solid state as opposed to a functional ink. For instance, a reconfigurable laser scattering optical physical unclonable function (PUF), proposed and demonstrated by Gan et al. (2022), combined VO_2 nanocrystals with TiO_2 scattering nanoparticles, resulting in a diverse and complex structure with higher optical security than conventional PUF encodings [91]. While He et al. (2020)

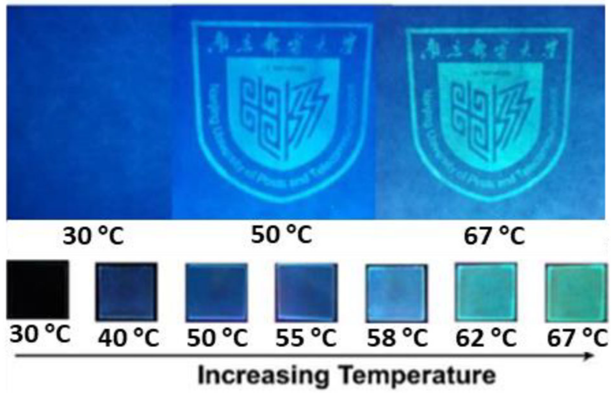

FIGURE 10 | Thermochromic luminescent pattern composed of iron (III) complexes within a polyethylene glycol matrix, forming a reversible multilevel anti-counterfeiting and high-security element (Adapted with permission from [101], Copyright 2020 The Royal Society of Chemistry, all rights reserved).

also demonstrate that thermochromically-active VO_2 films can be integrated into optically responsive absorbers to form switchable and tunable pixels using heat, which also hold suitable properties for optical anti-counterfeiting [92]. Novel applications for the TC properties of VO_2 and other inorganic compounds are an ever-evolving field, and it is likely that future developments will lead to additional applications in the security industry.

Inorganic–organic TC complexes have also gained increased interest in the last decade, providing varied and adjustable properties [93], whilst showcasing considerably lower and more versatile transition temperatures than purely inorganic TC complexes. Ionic liquids in particular are promising for the development of these complexes, providing a high thermal stability, nonvolatile, and negligible vapor pressure medium, which can greatly benefit the stability of functional pigments [94]. A notable property of ionic liquids lies in their modular nature, as the constituent ions of these so-called "designer solvents" can be changed, by directly varying the cation/anion species, functional groups, and alkyl chain lengths, to tailor their thermophysical properties for a given purpose [95, 96]. Nickel, cobalt, iron, and copper-based metal–organic and ionic liquid complexes have demonstrated reversible and irreversible thermochromism through changes in their coordination environment [93, 97–100].

This is still a relatively underdeveloped field compared to other TCMs, but functional inks prepared using these TC complexes have showcased high stability transitions with visually intense color contrast, ideal for long-term, reliable security elements. In particular, a thermochromic and luminescent polyethylene glycol and metal–ligand Fe(III) complex, printable by inkjet, was prepared by Liu et al. (2020) for security information printing, exhibiting three distinct colors and emission states between 30°C to 70°C, as observed in Figure 10. A remarkably low temperature range for an inorganic-based TC transition while providing a complex and difficult-to-clone multifunctional behavior, bolstering its security against forgery attempts [101]. Overall, there are numerous unexplored avenues toward developing purely inorganic and inorganic–organic hybrid TCMs for security applications, especially when considering their higher potential stability, lower

TABLE 1 | Summarized comparison of the properties of the outlined thermochromic material types (Data repurposed from [18, 36, 62, 82, 100, 102–106]).

	Leuco dye	Liquid crystal	PDA	Inorganic	Inorganic–organic hybrid
Transition reversibility	Reversible or Irreversible	Reversible	Reversible and/or Irreversible	Reversible or irreversible	Reversible or irreversible
Conventional transition temperature ranges (°C)	−15 to 60	−30 to 150	−18 to 250	70 to 500	−40 to 160
Transition temperature intervals (ΔT, in °C)	3 to 15	0.5 to 4 (Narrow bandwidth) 5 to 30 (Wide bandwidth)	20 to 200	2 to 400	2 to 100
Encapsulation	Required	Required	Possible	Possible	Possible
Relative stability	Low	Very Low	Moderate	Very high	High
Relative development cost	Low	High	Moderate	High	Moderate
Relative toxicity	Moderate	Low	Low	Very High	Moderate

reactivity, high color contrast, and quick response time compared to organic-based TCMs.

2.3 | Thermochromic Material Type Property Comparison

In Table 1, we summarize some of the properties of the mentioned TCMs to more easily compare their benefits and/or limitations. However, we note that adopting a specific material type for any given application has greater considerations besides the broadly outlined properties, such as compatibility with ink formulations, printing methods, and specific functionality. This includes the targeted transition temperature range and intended sensitivity of the TC ink and its associated color-shift.

Furthermore, in Figure 11, as a visual summary of this introductory chapter to TCMs, we have outlined the key properties and primary challenges in research and development for each of the highlighted TCM types.

2.4 | Literature and Market Trends of Thermochromic Inks

Given the recent growth in the TCM industry, a statistical analysis of literature and patents provides valuable insights into trends, innovations, and technological advancements in this field.

From the articles referenced throughout this review and the research conducted for this study, we have gathered a list that quantifies the types of TCMs used toward the development, research, and characterization of TC inks from a sum total of 100 articles under these topics (Supporting Information). The data for these claims can be observed in Figure 12; however, we should note that due to the scale of this statistical analysis, this represents only a partially representative group for this field of academic literature, and the following conclusions should be seen in a supportive sense to the wider study of this field, rather than a deterministic one.

From this study, we have found that leuco dye-based materials are undoubtedly the most widely used for novel developments in the field of printable TC inks, finding frequent use in the preparation of sensors and temperature markers for product packaging, and being subject to a wider variety of characterization and optimization processes for many distinct printing methods. For the other organic TCMs, while PDAs seem to have a similar overall presence in literature compared to TC liquid crystals, they have a higher presence for research targeted at security applications, especially when directed at encrypted information, likely due to more reliable stability over a wider temperature range. Although inorganic materials, namely vanadium, are studied to a larger degree than organics under the broader field of thermochromics, which primarily encompasses smart applications and energy storage devices, they are much more scarcely found in the development of printable inks when compared to organics. In recent years, we also find that the use of organic–inorganic hybrid compounds has increased steadily, as expected with the constant development of novel synthesis methods, material synergies, and applications in the ever-growing field of hybrid materials. The unspecified percentage attributed to "Other" TCM types can be loosely split between temperature-dependent aggregation-induced emissive particles, quantum dots (inorganic, carbon, and polymer dots), viologens, biopolymers, and composite microcapsules. All of which have showcased a degree of thermochromism but have not been adopted at a larger scale in the field of TC ink development.

2.4.1 | Intellectual Property of Thermochromic Security Inks

Additionally, an extensive report that explores the patent landscape regarding thermochromic ink compositions and their potential use as a security tool was conducted for this review.

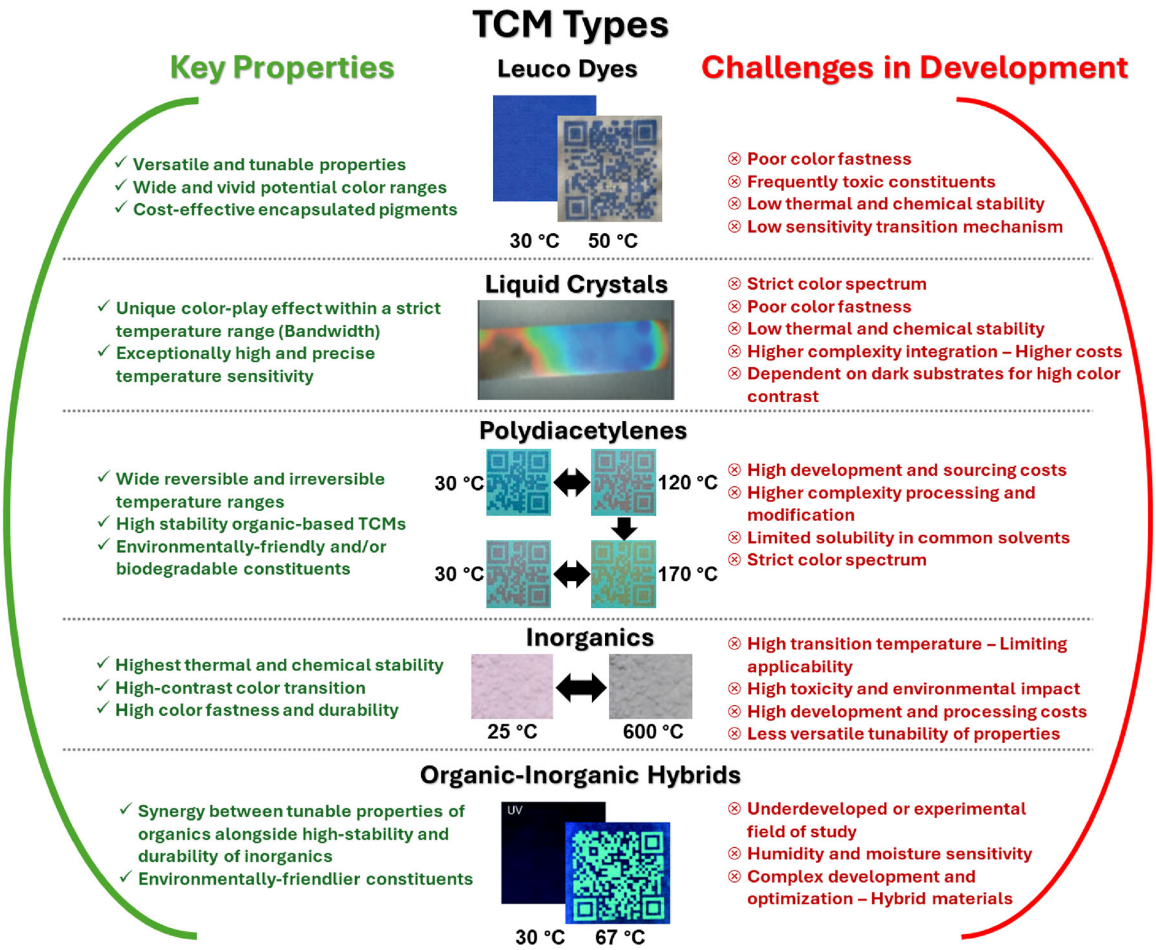

FIGURE 11 | Visual scheme of the key properties and primary challenges in development for each of the outlined thermochromic material types, including: Leuco dyes, represented by leuco dye-based ink imbedded fibers with an marking element (QR-code) (Adapted with permission under a CC BY 4.0 open access license [107] Copyright 2025 by the Authors, all rights reserved); Liquid Crystals, represented by a TC liquid crystal layer deposited on black paper (Adapted with permission under a CC BY-NC 4.0 open access license from [65] Copyright 2018 by the authors, all rights reserved); Polydiacetylenes, represented by a diacetylene-printed QR codes on paper substrates (Adapted with permission from [79] Copyright 2013 American Chemical Society, all rights reserved); Inorganic materials, represented by a chromium-doped alumina powders (Adapted with permission from [22]. Copyright 2018 Elsevier, all rights reserved); and Organic–Inorganic Hybrids, represented by a thermochromic luminescent iron (III)-based complex within a polymeric matrix to form an encrypted QR-code (Adapted with permission from [101], Copyright 2020 The Royal Society of Chemistry, all rights reserved).

In this search, the technical features considered were any functionally thermochromic inks or coatings with mentions of anti-counterfeit applications in patent titles, abstracts, or general claims. In particular, we looked to highlight inks with the existence of an encapsulation means and based on compositions including organic, transition metals, or liquid crystal components. The patent search was conducted in the commercial patent search engine *Patsnap*, where a sample of 3233 results was summarized into 46 individual patents that were considered the most relevant, originating primarily from Europe and the United States. Moreover, the patent families were confirmed using *Espacenet*, the free-access patent database provided by the European Patent Office, to prevent duplicate examinations of inventions with similar properties.

From the number of results obtained in the selected documents, it is worth noting that this technical field is currently the object of tremendous development. Several companies are working on and patenting new compounds that may incorporate TC ink compositions. However, we find that the vast majority of these representative patents, much like in research and development, are focused on reversible organic leuco dye-based systems. Instead of a high variability in the functional material choice, there is a trend in providing details for a specific chemical composition of a leuco dye color former, with a subsequent focus on outlining unique developers, solvents, encapsulants, or stabilizing agents to achieve desired ink properties. The use of metals to establish stable coordination complexes with functional properties is a known process that has found growing use in other functional security inks, but is still scarcely used to prepare TC inks. Instead, we find that inorganic or hybrid-based TCMs have been primarily developed as solid-state coatings for passive energy-regulating surfaces, with no practical example in security technologies.

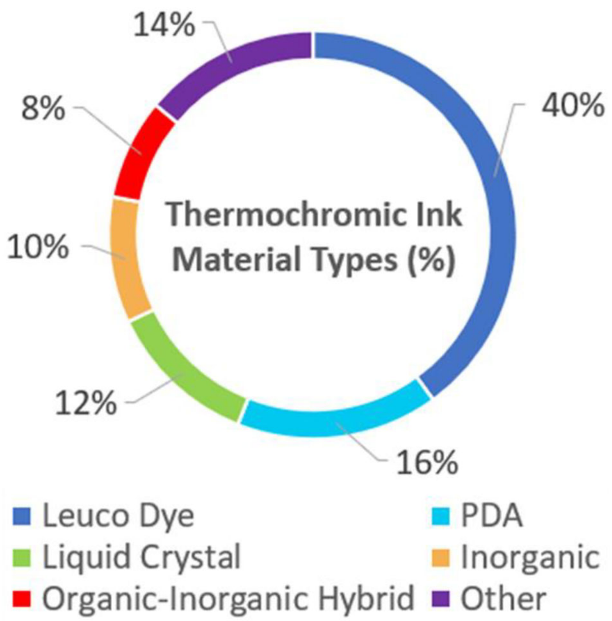

FIGURE 12 | Statistical percentage representing the presence of thermochromic material types used for the development, research, and optimization of TC inks for a sample of 100 articles in academic literature referenced and researched for this review.

Within organic TC pigments, most developers are based on weak acids, to enable a stable and low-reactive proton donor that changes the leuco dye to its protonated and colored form. Although stronger acids have been noted to promote this transition to be irreversible, this is seldom applied in leuco dye-based inks, as it can further hinder their relatively low chemical stability. These organic systems also benefit greatly from low reactivity solvents, with high molecular weight, neutral, and nonpolar compounds being ideal to minimize the degradation of the functional properties. Most listed solvents and/or co-solvents can be classified as aliphatic compounds, aldehydes, thiols, sulfides, ethers, ketones, esters, alcohols, acid amides, or, of course, water. They are chosen on the basis of stability, drying properties, and toward promoting the interaction of the color former leuco dye and the corresponding developer. Encapsulant agents or carriers are also always included to enhance the stability of the TC systems, and are primarily composed of waxes or aminoplast resins, with frequent use of melamine and/or urea-based compositions.

Unexpectedly, systems lacking traditional thermochromic transitions are also identified. In these, two complementary dyes are introduced into an ink formulation, in which one is dominant but easily degradable through heat or humidity. When the system is exposed to a relative excess of these stimuli, the dominant yet sensitive dye is decomposed, while the recessive, heat-resistant dye remains, irreversibly changing the color of the printed pattern. This allows the use of more conventional, perhaps cheaper dye pigments, with a greater focus on the development of specific encapsulant properties, granting finer control over decomposition at the expected temperature.

From our findings, ink patents actively targeting security applications are essentially focused on luminescent or near-infrared absorbing properties, primarily utilizing high-stability inorganic species. Despite this, some examples of practical thermochromic systems can be found, such as printing substrates made with halochromic leuco dye coatings for printing ticket elements or high security dual-functional anticounterfeit elements made with thermochromic components activated by magnetic heating. Otherwise, patents involving thermochromic inks might simply mention security printing as one of many different potential applications, but are reluctant to focus their claims as security technology, as they are marketed as commercial inks for (mostly) any type of application.

3 | Current Challenges in TCM Innovation and Optimization

While TCMs showcase great potential as functional components in commercial applications, their development is faced with significant challenges, which have been previously alluded to. Namely, concerns over the long-term stability of organic TCMs and factors that can lead to degradation of their functional properties; how TC inks can be implemented into scalable printing lines and which properties and parameters influence their compatibility with conventional printing processes; as well as addressing potential environmental and health hazards presented in conventional TCMs by finding greener, lower toxicity alternatives.

3.1 | Stability and Degradation of TCMs

TCMs, especially organic-based ones, can suffer degradation of their functional properties from light exposure, extreme temperatures, and certain solvents, which results in generally poor stability and limits their use and reliability for long-term applications [18, 108, 109]. This concern with TC ink durability and sensitivity to adverse environmental conditions is noted by every major manufacturer and establishes itself as a prominent challenge throughout innovation and enhancement in this field, with encapsulation of TCMs as the most widely adopted solution [82, 110, 111].

3.1.1 | Lightfastness and Photodegradation

Ultraviolet (UV) radiation causes significant damage to TC inks over time, namely from the breakdown of molecular bonds in constituent organic polymers or thermochromic species by photodegradation, which can lead to the irreversible loss of their functional TC properties [108]. This limits the time they can be exposed to natural light in an external environment, thereby limiting their potential applications. This sensitivity stands true for most organic and inorganic–organic hybrid TCMs but is especially harmful for liquid crystal-based TC inks, which exhibit exceedingly low lightfastness for their helical structures and can quickly lose functionality when directly exposed to UV [36]. Prolonged UV exposure can also irreversibly break the lactone rings in leuco dye-based systems, potentially forcing the system into an unintended fixed color state and tarnishing their balanced and reversible system [112]. On the other hand, PDA or inorganic-based TCMs exhibit far higher resistance to UV degradation, making them an ideal choice for applications exposed to harsher environments, as seen in passive smart-window advancements, where inorganic TCMs prevail.

3.1.2 | Chemical Stability

TCMs also suffer degradation when exposed to reactive agents such as oxygen in an open atmosphere or acidic species in rain [113, 114]. For organic TCMs, these reactive agents can, similarly to UV-radiation, break down molecular bonds, inhibiting the transition mechanisms for leuco dye systems, or damaging the PDA chain and liquid crystal organized structures. Leuco dye-based pigments in particular possess low resistance to common chemical agents, with significant color variations, bleaching, or bleeding occurring when exposed to ethanol and weak acids. While inorganic TCMs possess greater chemical stability and are less reactive, some TC transition metals, such as VO_2, will gradually oxidize into more thermodynamically stable phases when exposed to oxidizing agents, where their structures no longer exhibit the desired thermochromic properties, thereby making them less reliable and reducing their expected lifetime.

3.1.3 | Thermal Degradation

Despite their intended properties as temperature-responsive materials, prolonged temperature exposure can irreversibly affect the transition point temperature interval or intended color hue of a TC ink [47]. This is of particular notice for TCMs possessing functional groups more easily subject to decomposition, such as carbonates and hydroxides, as is the case in some leuco dye-based and PDA-based TCMs [108]. High temperatures can break hydrogen bonds in PDA vesicles, resulting in unintended irreversible color-shifts of the polymer structure [115] or degrade the capsules that isolate leuco-dye systems, exposing them to external oxidizing agents or reactive species, which could, as mentioned, damage their intended functionality.

3.2 | Printing Processes

When printing functional inks, it is important to consider both the quantity and quality of the print, as these factors are also important for conventional inks. However, it is equally important to evaluate how the quantity and quality of the ink affect the functionality of the printed layers [116]. Thermochromic ink compositions and behavior can differ significantly from conventional printing inks, requiring a thorough optimization of the printing parameters for any given method to achieve a high-quality end-product [117], especially when we consider the higher costs they demand, and the reliability mandated by anti-counterfeit elements. Security ink printing technologies can be divided into two major groups: contact printing, which involves physical contact between the substrate and an inked and/or patterned object, and noncontact (digital) printing methods, which use printer heads or nozzles to transfer the pattern onto the substrate [44]. The main contact printing processes adopted for security inks are offset lithography, flexography, intaglio/gravure printing, and screen printing, while the most prominent noncontact process is inkjet printing [118, 119].

General value ranges for some relevant properties of each of the mentioned printing techniques are outlined and compared in Table 2, although the presented values are not exclusive to thermochromic inks. Furthermore, for some properties, we provide values defining the theoretical limits of the various types of printing processes as outlined in literature, as well as the standard values found in a practical industrial environment shared by security industry professionals from the National Printing House of Mint (INCM). These values correspond to the average expected printing speeds obtained with different printing techniques and systems currently available at INCM. For further clarity, in Table 2, we indicate the specific printing equipment models together with their corresponding typical operating speed. Although higher print speeds are theoretically possible, as evidenced by literature values and manufacturers themselves, the actual average production speed depends on multiple factors, including the type and thickness of the substrate, complexity of the security features being printed, number of colors/layers, drying requirements, ink formulation, environmental conditions, operator training, desired resolution and print quality, as well as machines' maintenance state and overall condition. Therefore, printing speeds for these techniques should not be regarded as a fixed characteristic, but rather as the result of a balance between the equipment, the substrate, the ink, the resolution requirements, and the production environment itself. Altogether, different machines may reach very different effective speeds depending on how these factors are combined.

Offset lithography is the most widely adopted contact-based printing method for security printing, due to simultaneous high resolutions, consistent printing quality, and considerably high throughput [126]. Conventional wet offset relies on the preparation of a metal printing plate cylinder, chemically treated to allow a moisturized layer in non-image sections of the plate, originating from a water reservoir transported with rollers, and an ink layer in the outlined image sections, originating from ink rollers and their reservoir, as represented in Figure 13. The ink-patterned plate cylinder then transfers the ink to a rubber blanket cylinder, which can subsequently print the ink onto the intended substrate under pressure from an impression cylinder [122]. Due to the intended water–oil repulsion, which allows for precisely patterned depositions, offset inks are mostly limited to oil-based formulations, as opposed to the more environmentally friendly water-based inks compatible with alternative printing methods. Dry offset, on the other hand, replaces the need for water by applying a silicone-based coating to the non-image areas of the plate cylinder, or engraved reliefs on the image areas of a nylon-based plate [127, 128]. Dry offset patterned cylinders offer faster drying speeds, higher image quality, and even potential reusability, but incur significantly higher costs in setup and maintenance than the simpler wet offset, and are highly sensitive to dust particles, which can damage the engraved plates, resulting in printing defects [127]. Of note, while conventional offset is primarily used with absorbent substrates such as paper, dry offset allows for higher compatibility with a wide range of plastics and synthetic nonabsorbent surfaces (albeit still primarily dependent on ink properties) due to lower humidity, granting it higher versatility for different product types.

The optimization of offset printing for functional TC inks has seen considerable advancements in recent years, although its practical

TABLE 2 | Feature and property range comparison between the commonly used printing technologies for functional inks (Data repurposed from [120–124] and industry professionals at INCM [125]).

	Offset lithography	Gravure printing	Flexography	Screen printing	Inkjet printing
Printing form	Chemically treated Al plate	Engraved cylinder	Relief polymer plate	Stencil-coated mesh	Digitally outlined (noncontact)
Resolution (lines/cm)	100 (wet offset) 200 (dry offset)	100	60	50	60–250
Ink viscosity (Pa·s)	5–100	0.05–0.2	0.01–0.5	0.1–50	0.001–0.1
Throughput (m^2/s)	3–30	3–60	3–30	2–3	0.01–0.5
Print speed (m/min)	100–900[a)] 30–400[b)]	100–1000[a)] 15–300[b)]	100–700[a)]	10–15[a)] 20[b)]	15–500[a)] 15–150[b)]
Film thickness (µm)	0.5–10	0.01–12	0.005–8	0.2–100	0.01–20
Line width (µm)	10–15	10–50	20–50	50–150	1–20
General cost	Low	High	Low	Low	High
Roll-to-roll scalability	Yes	Yes	Yes	Yes	Yes

[a)]Theoretical value obtained from referenced literature on the limits of the various printing technologies.
[b)]Standard values defined by industry professionals at the National Printing House and Mint (INCM) for modern industrial printing line equipment, which include: a DG Press Vision 6, Komori GL 450, Heidelberg SM 102, KBA Super Simultan and KBA Simultan IV offset printers; a KBA Intagliocolor 8 gravure/intaglio printer; a SPS Vitessa Star sheet-fed screen-printer; a Truepress Jet520 HD+ and a Domino K600i web-fed Inkjet printer. All of which are used in printing lines with speeds below the manufacturer's specified limits, to guarantee compatibility with other production line components and provide reliable, high-quality prints at an industrial scale.

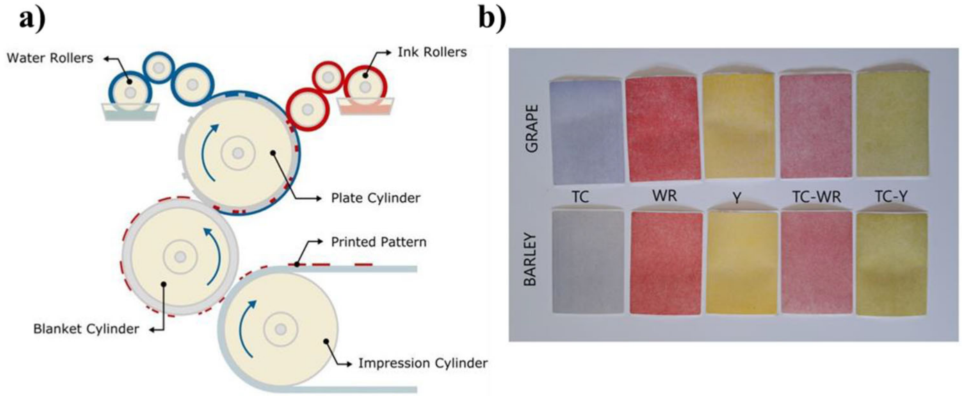

FIGURE 13 | a) Schematic diagram of an offset lithography printing process setup (Reproduced with permission under a CC BY 4.0 open access license from [130] Copyright 2015 by the authors, all rights reserved) and b) dry offset printed leuco dye-based thermochromic mixtures on environmentally friendly paper, using conventional red (WR) and yellow (Y) UV-curable offset inks (reproduced with permission under a CC BY 4.0 open access license from [131], Copyright 2023 by the authors, all rights reserved).

use in laboratories is scarce due to the complex and expensive setup. Enhancements in ink adhesion [38], ink tack stability [39], and printing parameters such as substrate types, ink quantities, and printing speed/pressure have greatly improved compatibility with this printing process, facilitating its application in realistic production lines [38, 129]. Additionally, while dry offset might be less common than conventional wet offset in the general printing industry, it is frequently used in the security industry for high-resolution patterns on specialized substrates, such as security paper for travel documents or polycarbonate for ID cards. High humidity and drying rates from wet offset could cause defects or hinder adhesion during the lamination process required for these products, so dry offset printing provides a water-free solution, albeit at higher maintenance, use, and setup costs. This dry offset method was investigated by Jakopčević et al. (2023), whose findings indicate that TC UV-curable inks can achieve edge quality and resolution that competes with conventional UV-curable prints [117].

In gravure (or intaglio) printing, the ink reservoir is directly accessed by a patterned printing plate cylinder, which directly transfers ink under pressure to the substrate, removing excess ink with a doctor blade to smooth the printed layer, as shown in Figure 14. This method produces high-resolution prints and can theoretically offer more cost-effective production than offset printing for exceedingly high-volume prints, due to faster

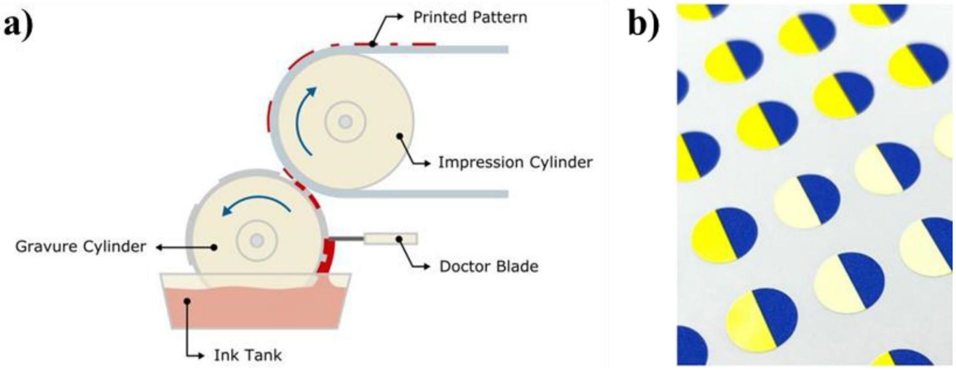

FIGURE 14 | a) Schematic diagram of a gravure printing process setup (Reproduced with permission under a CC BY-NC 3.0 open access license from [130] Copyright 2015 by the authors, all rights reserved) and b) MatriX-Thermochromic Inks-based temperature monitoring and anti-counterfeit sensors, compatible with gravure printing fabrication (Adapted with permission from [133], Copyright 2025 NanoMatriX Technologies Limited, all rights reserved).

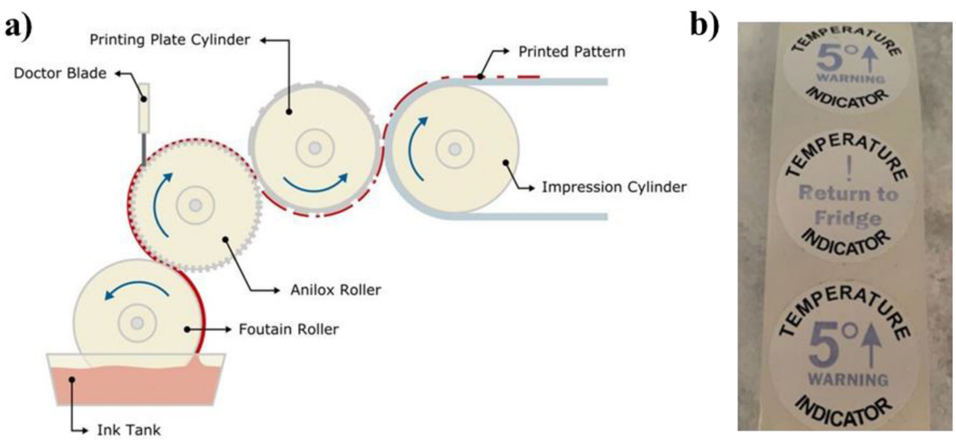

FIGURE 15 | a) Schematic diagram of a flexography printing process setup (Reproduced with permission under a CC BY-NC 3.0 open access license from [130] Copyright 2015 by the authors, all rights reserved) and b) a thermochromic safety label designed by GoldCrest Labels and developed specifically for flexographic printing (Reused with permission from [136] Copyright 2025 Goldcrest Labels, all rights reserved).

potential printing speeds and more consistent high-quality results. However, it is generally held back by considerably higher costs associated with preparing engraving cylinders compared to offset, which limits its use in production lines with high variability, which are commonly found in the field of security. Due to their similarities, just as in offset, gravure print speed and applied pressure can significantly affect quality, along with the geometry and spacing on the engraved plate cylinder [132]. For TCMs, despite the existence of commercially available TC inks designed to be compatible with gravure printing processes, such as the MatriX-Thermochromic Inks distributed by NanoMatriX [133], property enhancement research and printing optimization seldom use this printing process, likely due to the high costs in preparing printing plates and equipment maintenance, which are unrealistic in a lab environment.

Flexography uses flexible photopolymer plates, wrapped around a printing cylinder to transfer ink from an anilox roller, with engraved cells that hold ink, to a flexible substrate. The distance between cylinders allows for the ink to transfer only onto the raised segments that define the pattern of the photopolymer printing plate, which is then directly transferred onto the intended flexible substrate, as demonstrated in the schematic in Figure 15 [10]. This printing method is compatible with a greater type of substrates when compared to offset or gravure printing, allowing for the use of almost any flexible surfaces or material types. However, it cannot match the detail and color depth provided by offset, so it is instead primarily used in manufacturing high-volume packaging and labels, without the need for graphic-intensive or complex patterning [134]. Because of this, flexography finds fewer uses in the security industry apart from potentially simpler quality control sensors on flexible substrates. Despite this, this printing process incurs significantly lower set-up costs due to cheaper plate preparation costs and significantly less ink usage and waste, making it especially ideal for short-term and/or high variability printing requirements [135]. The development of novel TC inks targeted at flexographic printing in the literature is limited, but advancements in this printing method could provide an avenue for low-cost novel security elements for product packaging labels.

Screen printing is based on the transfer of ink, with a particularly high viscosity, through a patterned screen mesh using pressure provided by a rubber-edged blade (known as a squeegee),

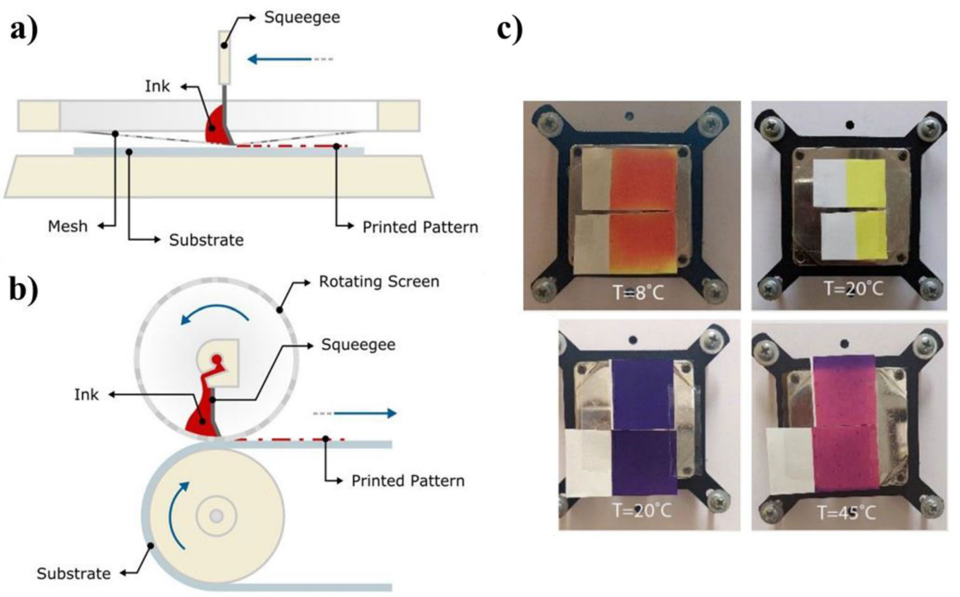

FIGURE 16 | Schematic diagram of a) a flexographic printing process, a b) screen printing flat-bed process (Reproduced with permission under a CC BY-NC 3.0 open access license from [123, 130] Copyright 2015 by the authors, all rights reserved) and c) prints of commercial leuco dye-based thermochromic UV-cured inks screen printed onto two paper substrates (Adapted with permission under a CC BY-NC 4.0 open access license from [47] Copyright 2023 by the authors, all rights reserved).

resulting in a characteristically thicker layer compared to other printing processes [119]. There are two common types of screen printing: flatbed screen printing and rotary screen printing, as illustrated in Figure 16. Flatbed screen printing, as the name suggests, utilizes a flat screen to transfer ink onto an equally flat substrate. This method can be conducted either manually or automatically, providing straightforward access to the screen-mesh. In contrast, rotary screen printing is an automated process featuring a stationary squeegee that relies on the continuous rotation of the screen itself to transfer the patterned ink. This results in significantly higher printing speeds and consistency, ideal for high-volume production lines. However, despite its prevalent use in industrial-scale applications, it also entails higher costs in setup, maintenance, and the replacement of screen-meshes due to altered patterns or ageing. For either type, applied pressure, screen specifications, squeegee properties, and angle can significantly affect how ink is transferred through the screen onto the substrate and its resulting resolution [137].

Optimization of intelligent functional inks in screen-printing has previously been explored, particularly because of the methods' ease of application in a laboratory setting. Key parameters that significantly influence printing quality include ink quantity, squeegee angle, and applied force [116]. Furthermore, TCMs, in particular organic pigments, have been previously applied to textiles using screen printing, as shown in [138], and have demonstrated high stability of their thermochromism within the embedded fibers [23], showing great promise for the development of thermochromic textiles. While adaptable to a wide range of substrates, screen printing is less practical for fabricating higher security elements due to lower potential resolution and slower printing speeds when compared to other contact printing processes. These properties limit the complexity and scalability, so screen printing has primarily been used for printing security elements onto textile substrates, which require a higher quantity of ink to guarantee the embedded fibers retain their intended functionality, or as a simple method to prepare proof-of-concept tests in a lab environment [15, 139, 140].

Inkjet printing is an additive noncontact technique that involves expelling low-viscosity inks from printheads onto a substrate surface. There are two main types of inkjet printing—continuous inkjet systems, which electrically deflect droplets as they fall to form the intended pattern, recirculating excess ink back into the system, as shown in Figure 17a, and modern drop-on-demand systems, which only generate droplets as they are needed to form the pattern, controlled by a thermal or piezoelectric mechanism, as shown in Figure 17b [141]. As a digitally controlled process with no reliance on patterned plates or screens, it provides reliably low variability between prints and far greater ease in changing intended printing patterns [79, 142]. Additionally, it also provides minimal ink waste when compared to contact-based printing methods, especially with the use of drop-on-demand printing heads, contributing to a more sustainable process [142]. The complete interaction between the printhead, ink, and substrate is influenced by multiple factors, with the printhead nozzle type, diameter, ink flow stability, the inverse Ohnesorge number, and speed as the most directly controllable equipment parameters [143]. The main drawback of inkjet lies in its considerably lower printing speed, and thereby lower throughput, which can impact its cost-effectiveness for large-volume prints when compared to contact printing methods.

In the security industry, because inkjet printing provides exceedingly higher resolutions and more reliable quality than other conventional contact-based printing methods, it enables the development of higher complexity anti-counterfeit systems and is largely considered the method of choice for printing security

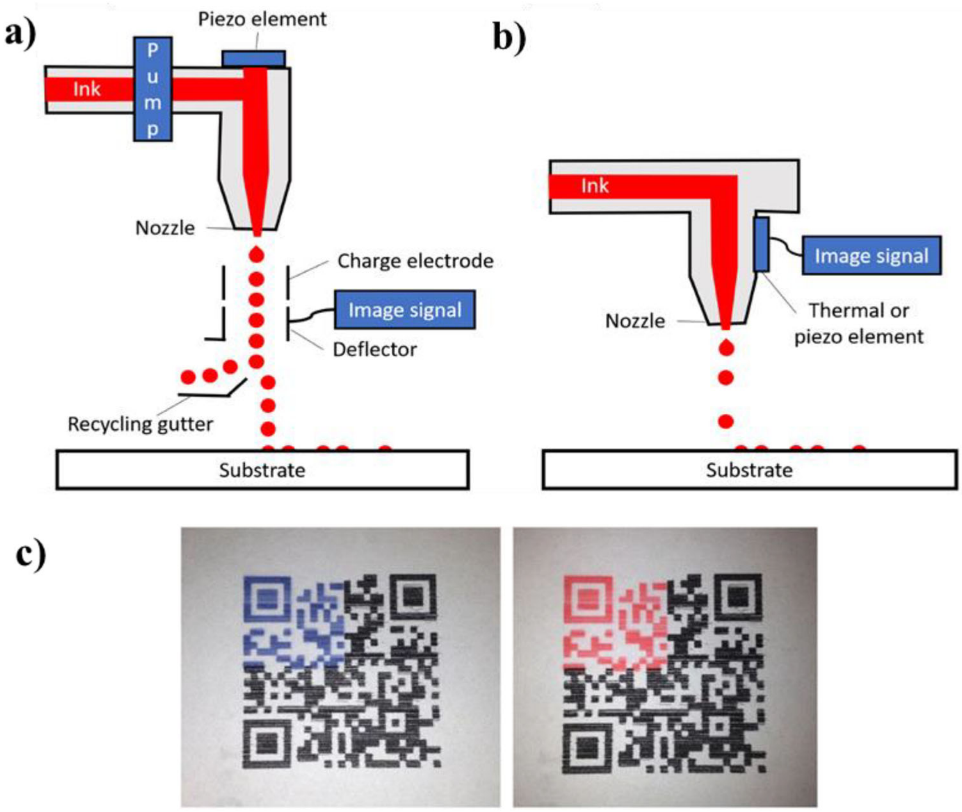

FIGURE 17 | Schematic diagram of Inkjet printing processes, namely a) a continuous inkjet system and a b) drop-on-demand inkjet system (Reused with permission under a CC BY 4.9 open access license from [122] Copyright 2021 by the authors, all rights reserved) and c) a Poly(PCDA)/ZnO fabricated using inkjet printing on paper as a thermochromic segment of a QR code (Reproduced with permission from [144] Copyright 2013 Elsevier B.V., all rights reserved).

elements alongside wet and dry offset lithography [44]. Advancements in TC ink depositions using inkjet technology, such as with the PDA-based QR code sensors developed by Wu et al. (2014) [144], as seen in Figure 17c, have provided some insights into the use of this method to develop high-quality TC anti-counterfeit elements, but would greatly benefit from a modern optimization of printing parameters using novel TC inks.

Defining the right printing technique for an intended ink and product is a crucial step in the development of novel, scalable applications. Even with the established differences between the outlined printing methods, production costs from maintenance, required ink quantity, printing volume, engraving cylinder setup, and expected energy consumption, as well as chosen material type, intended print quality, market demand, and the availability of technology are all factors that would have to be considered when establishing an industrial printing line. Overall, despite efforts to understand the effects of printing parameters on functional inks for each of the aforementioned printing methods, targeted research on optimizing TC inks for scalable processes is still lacking.

Additionally, high-security applications such as bank notes can make sequential use of several of these processes, with multiple steps using offset, gravure, and screen printing to obtain all the intended identifiers and security features, as is the case for euro notes [145]. This further emphasizes the importance of optimizing TC security inks for multiple printing methods and facilitating their seamless integration into standard printing lines. The current shortage in studies toward scalable development using TC inks makes this a relevant focus for innovation and an eventual necessity for their adoption at an industrial level.

3.2.1 | Ink Properties for Scale-Up

In order to have meaningful advancements in new TCM fabrications, their resulting ink needs to be compatible with large-scale production, as the end goal of thermochromic material research generally entails using them to develop new products. Otherwise, investment in innovative or enhanced properties cannot be adopted to a wider degree, even when reaching promising results. Scalability is therefore a critical goal for newly developed TC inks to ensure they can be adapted into an industrial production line.

The scalability and suitability of any given TC security ink, besides depending on the specific parameters of the chosen printing method, as previously outlined, are also dependent on a multitude of factors inherent to the ink itself. This includes the inks' nature (liquid solution, gel, solid powder, colloid, dispersed or dissolved, etc.), viscosity, density, base solvent (solution or water-based), particle size, and drying requirements. Auxiliary additives (waxes, drying agents, fillers, inhibitors) and ink thinners can also affect the absorbency or adherence of the ink to a given substrate [146]. All these properties and processes need to be considered when optimizing a security ink for a given printing process, to guarantee a reliable end-product.

3.2.1.1 | Viscosity. The viscosity of an ink is an integral property when choosing and optimizing a printing process, as it quantifies how easily the ink can flow and can limit its applicability. Viscosity can impact the sharpness of printed patterns, with excessive viscosity not permitting a smooth flow of ink, resulting in uneven coatings or defects, and insufficient viscosity causing unneeded spread of the ink, which can cause blurred edges in the print [117]. A target viscosity range always exists, as exemplified in Table 1, within which an ink is ideal for an individual printing system, and functional TC security inks are no different in this aspect. Choosing the correct TCM concentration, solvent, and thickening additives to correctly control the viscosity of a TC printing ink is an essential step in optimization.

Contact printing methods, which rely on printing rollers, require inks with sufficient viscosity and tack to guarantee proper adherence to the corresponding rollers and substrates [39]. However, this viscosity cannot be too high as it would hinder ink transfer between rollers, nor can it be too low, which would unnecessarily extend drying speed or harm printing resolution. A balanced control of viscosity is thereby critical to achieving high-quality prints, which can be attained using additives or by varying the TCM concentration within the ink formulation.

Noncontact methods, such as inkjet, are typically limited to lower viscosity inks due to the requirement of higher fluidity to assist in forming droplets and reduce aggregation/drying of the ink within the nozzle systems. Lower viscosity when using TCM-based inks generally entails a decreased concentration of the functional component, which can lead to reduced color intensity in the final print and contribute to slower drying. Advancements in inkjet printhead technologies, allowing for higher viscosity inks, have also benefited the adoption of this printing method for TC inks, by permitting higher color intensities and drying speeds competitive with conventional contact printing processes [142]. As mentioned, PDA-based TC composites for security prints have been previously manufactured using inkjet printing [79, 144]. However, to the best of our knowledge, a rigorous study of their compatibility with other printing methods has not yet been conducted.

3.2.1.2 | Particle Sizes. Particle size is a primary concern when optimizing a TC ink toward a printing process, as the microcapsules in organic-based TCMs are typically an order of magnitude larger than conventional ink or dye pigment particles [147]. Particle agglomeration can become a concern when these larger organic pigments are involved, as their resulting agglomerates, arising from lower mobility and higher collisions (despite lower surface energy when compared to smaller particles) can gradually damage printing equipment or negatively impact resolution [148]. TC microcapsules with smaller sizes also showcase higher color-change sensitivity due to higher surface areas for heat transfer, which can affect the expected temperature transition point or color-shift rate of the printed ink patterns.

Screen-printing, which relies on the space between the printing meshes to transfer inks onto substrates, can require a lower mesh count if particle sizes exceed a third of the mesh hole dimensions [149], resulting in an even lower maximum printing resolution. Notably, pigment particle size is of greater concern in inkjet printing, where the larger particle sizes can cause clogging within the printing nozzles through particle aggregation or drying after prolonged use [150]. Printing inks chosen for inkjet cannot form aggregates larger than the nozzle diameter, which can be challenging to achieve when nozzles range from 10 to 150 μm, while individual TC pigments can reach upward of 20 μm [151]. Consequently, optimizing TC printing inks targeted for inkjet printing requires a significant focus on particle size reduction, which can be achieved by controlling the synthesis rate of encapsulating shells for TC leuco dye, liquid crystal, or silica-coated systems. For PDA-based TCMs, the introduction of assembly-promoting and hydrophilic groups can promote the formation of higher-concentration supramolecules with smaller particles. This is exemplified by Yoon et al. (2013), who developed TC PDA-based inks for paper-based multilevel anti-counterfeit barcode systems, where they successfully limited particle aggregate sizes to under 1 μm during operation (from an initial 20 μm), finding great compatibility with the 20 μm-sized inkjet printing nozzles chosen for their process [79].

3.2.2 | Pre and Post Treatments, Quality Control, and Printing Regulations

3.2.2.1 | Pre-Treatments and Drying Methods (Post-Treatment). Additional surface treatments, ink dispersion, and the drying process can significantly affect the efficiency of a production line, potentially hindering the scalability of a given TC ink. Ideally, scalable printing inks should have a short drying duration (at a minimum, a couple of meters per minute in a continuous system, although this varies by printing technique) to be easily adaptable within efficient roll-to-roll production lines. To achieve this, the drying rate of organic solvent-based inks is conventionally faster and requires lower energy than water-based inks; hence, they are preferably chosen in larger-scale industrial systems that focus on efficient and cost-effective printing lines, especially when using nonabsorbent substrates [16]. However, water-based inks provide considerable benefits of their own, making the optimization of drying technologies for water-based inks a major focus of innovation [152].

The use of UV-curing for conventional printing inks during the post-deposition drying process is commonly adopted to increase manufacturing efficiency. However, due to the previously mentioned UV sensitivity of TCMs, this drying step may prevent their use in certain production lines prepared for UV exposure. For these cases, it is critical to determine if the exposure intensity and duration would have lasting effects on the stability and functionality of the printed TC ink. As well as whether UV-resistant components are necessary to prevent functional degradation, and how their introduction could affect other properties of the printed ink itself. Furthermore, during ink formulation, auxiliary treatments, such as surface modifications, to render pigments more hydrophilic or hydrophobic, have previously been shown to improve stability in TC inks [112]. Both in retaining the stable suspension of TC particles within the solvent, as well as in raising the surface energy of particles to enhance adherence to a substrate during printing. Careful consideration of additives and treatments is therefore essential for optimizing a newly developed TC ink toward scalable production.

3.2.2.2 | Quality Control. As a final step in optimization, post-printing quality control is critical to identify color uniformity, transition contrast, alignment, or any printing defects that might occur when printing functional inks, just as with any conventional printing procedure. This is especially true in security printing, where a high degree of quality control is required to ensure that anti-counterfeit elements operate correctly and possess the necessary stability to retain functionality for their intended purpose. For gravure, flexography, and inkjet printing, especially, the lower potential thickness of the printed layer, and therefore the lower quantity and/or concentration of TCM species, requires a higher attention to the resulting color contrast between or throughout transition states [117]. This is necessary to ensure an effective and clear visual indication of the color-shift, while minimizing necessary film thickness to reduce production costs.

3.2.2.3 | Label and Ink Regulations. In the context of the European Union, the application of printing inks onto labels or products is subject to specific regulations. Most regulatory actions in regard to printed inks focus on ensuring their safe use on food-related products. For food packaging in particular, we note that materials containing Bisphenol A, which is frequently used as a developer within leuco dye-based systems, are restricted in their use for applications that come into direct contact with foodstuffs, in accordance with Regulation (EU) 2024/3190 [153]. For example, Bisphenol A, a common developer in leuco dye-based TC inks and dyes, has already been recognized, as of 2021, as a very great concern substance by the European Chemicals Agency, due to endocrine-disrupting properties [154]. This does not directly impede their use as a component in functional inks for label features, as they are applied to the non-food contact side of the material or product in subject, but it can hinder their general adoption due to risk and concerns with contamination.

While, according to our findings, functional security inks have no specific regulations, the European Printing Ink Association has established guidelines for safe handling, storage, and material selection of UV printing inks, intended to be cured by exposure to UV, and frequently used for printed security elements [155]. Additionally, for the general large-scale manufacturing of inks, including functional security inks, toxic solvents are largely avoided or outright banned due to environmental and health concerns. They also provide guidelines that describe the risks associated with the use of inks and coatings intended to be exposed to high temperatures, which can likewise be used to identify the safety requirements required with the use of TCMs. Additionally, information on laboratory safety is provided, establishing common practices in changing existing inks or preparing novel ink formulations [156], as well as recommended procedures for storage and transportation, in accordance with ISO 45001 and/or ISO 14 001 guidelines [157].

3.3 | Sustainability

Thermochromic materials and inks often contain or are encapsulated within toxic substances, which can make them potentially hazardous due to low degradability and persistence in water systems. This can lead to environmental pollution and disrupt ecosystems. As a result, their use is limited in applications such as food packaging, medical sensing devices, or as exposed security inks in documents and product labels [110]. Primarily due to concerns about the potential leaching of toxic components, such as those found in the widely adopted leuco dyes, including the popular crystal violet lactone [158], their associated developers [159], or metal ion byproducts from inorganic or hybrid compositions. The toxic nature of the majority of commercial TCMs raises concerns in a world increasingly focused on sustainable and environmentally friendly production, so the development of low-toxicity and bio-based TC inks has also established itself as a focus of research and innovation, as well as a driving force in the functional ink market.

3.3.1 | Reducing Toxicity and Volatile Organic Compound Use

The most direct way to develop lower toxicity security inks is by substituting the use of volatile organic compound (VOC) solvents (or those that emit them as byproducts), which account for the majority of a printing ink constitution (varying by functional and additive material content) and possess highly toxic, reactive or carcinogenic properties, commonly found throughout the printing industry as primary or auxiliary ink constituents [160]. Due to their polluting nature, efforts to reduce VOC emissions are an important challenge in the printing industry [161]. Water-based ink formulations provide safer and cleaner storage and fabrication processes, due to low volatility and low flammability, and result in less polluting, potentially even VOC-free, environmentally friendly inks [17]. The main disadvantages of using water as the primary solvent are that, compared to conventional VOC solvents, they have lower drying rates, which slow down manufacturing and require higher energy costs for evaporation, especially on less absorbent substrates with lower surface tensions [16]. To mitigate these disadvantages, cosolvents, such as acrylic resins, can be used to increase viscosity or vapor pressure, improving the base drying qualities of water-based inks, as demonstrated by Bhagya et al. (2022) with their novel water-based, eco-friendly TC and fluorescent inks for anti-counterfeit applications [162]. Significant efforts to integrate TCMs into water-based inks continue to be made in the literature, and the higher adoption of water as the primary ink solvent will greatly facilitate the development of sustainable TC printing inks.

Another method to tackle toxicity lies in reducing the toxic nature of the TCMs themselves, either by optimizing their existing properties and functional groups, or substituting them with greener alternatives [110]. An example of this lies in biopolymers, such as cellulose derivates, which have been found to possess a degree of thermochromism and have also been prepared as frameworks for integrating existing TCMs [68, 163]. Encapsulation materials for TC pigments are also an avenue toward this goal, with biopolymers and inert metal oxides both finding great success in forming nontoxic capsules through emulsion synthesis [164, 165]. Together, these “greener” TCM developments can significantly expand their use in industries currently dissuaded from adopting them due to environmental or public health concerns, subsequently giving rise to new opportunities and applications.

Apart from this, it is of note that the low quantity of TC ink applied to a given substrate in the security industry, normally as a simple patterned element, corresponds to a very low percentage of the total substrate surface area on a product. Therefore, it is mostly economically unviable to attempt to recycle, reuse, or otherwise recover the functional inks during their disposal. Efforts have been made by Vukoje et al. (2016) to improve the removal of conventional leuco dye-based TC inks from paper substrates using chemical deinking flotation. However, their deinkability was deemed quite poor, with a high adherence to the paper substrates, and any improvement in these efforts was minimal [166]. Therefore, we reiterate that it is of significant importance in modern innovation to resort to lower-toxicity materials, efficient printing processes, and focus on water-based ink solvents, when possible, to minimize the environmental impact of these functional inks during development, use, and disposal.

4 | Current Advances in Thermochromic Security Ink Development

4.1 | Property and Stability Enhancement

4.1.1 | Microencapsulation to Enhance Durability

Considering the outlined property limitations inherent to TCMs, it is essential to protect them from external agents and direct light exposure to ensure the development of stable and durable inks. Enhancing durability presents a significant challenge in TCM research, as consistency and reliability are paramount to their purpose, especially when intended for rigorous applications such as functional security inks. The encapsulation or coating of TCMs is the most straightforward and widely adopted method of isolating them from external agents [18, 114]. Organic TCM systems—such as leuco dyes, liquid crystals, and PDAs—are often designed with a protective shell, encapsulated by a stable and/or inert polymeric or inorganic capsule, that form composite thermochromic pigments [84, 147]. The most common materials used for the encapsulation of these TC particles include thermosetting resins or polymers (such as urea, melamine, and formaldehyde compounds), gellan-gum Arabic, epoxy resins, or inert silica shells [146]. Likewise, organic and inorganic TCMs have frequently been encapsulated within inert metal oxide shells, using materials such as silica to not only enhance chemical and mechanical stability, but also improve their optical performance [113]. The incorporation of UV-absorbing elements into the encapsulation materials of TCMs, or even directly into the primary solvent of the printing ink as additives, has also shown great promise in stabilizing and protecting TC components [108]. Zinc oxide and titanium oxide nanoparticles have been previously incorporated into protective polycaprolactone (PCL) encapsulants by Vukoje et al. (2022), showcasing greatly improved stability and thereby longevity of leuco dye-based TC pigments, through the partial absorption of incident UV radiation [112]. Overall, microencapsulation has been shown to greatly prolong the expected lifecycle of TC pigments [111], with a wide range of physical and/or chemical methods being adopted toward

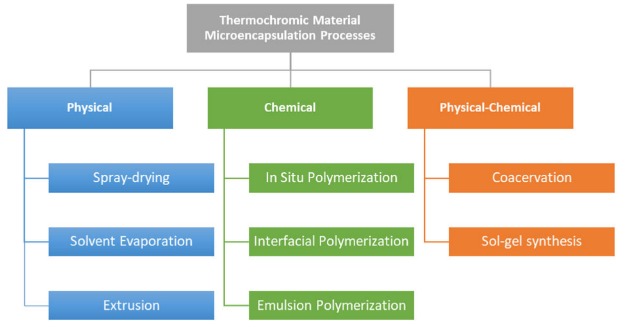

FIGURE 18 | Diagram highlighting the most widely used methods for encapsulation, which have been applied to thermochromic pigments.

this purpose, as summarized in Figure 18 The most common processes include in situ, interfacial, and emulsion polymerization [167, 168], complex coacervation, sol–gel, extrusion, solvent evaporation and spray drying [84, 169, 170].

4.1.1.1 | Chemical Encapsulation Processes. In situ polymerization is a simple and preferred chemical technique for the industrial production of TC microcapsules. With this method, a mixture of TC compounds, developer (if necessary), and other additives is directly added to an aqueous solution, resulting in an oil-in-water emulsion. Water-soluble thermosetting resins or polymers, which act as the encapsulating materials, are simultaneously added to this solution, and polymerize directly at the oil–water interfaces of the suspended mixture after a pH or heat-activated stimulus, creating uniform shells around the dispersed compounds. All polymerization occurs in this continuous aqueous phase, requiring vigorous stirring to guarantee [54]. This process is widely adopted for leuco dye-based TC security ink development due to ease in fabrication while providing high yield, low costs, efficient and consistent results, which facilitates scalability. The most commercialized melamine–formaldehyde-based microcapsules used to encapsulate leuco dye systems are commonly prepared using this process and can be demonstrated by the in-depth studies carried out by Geng et al. (2018) on CVL core-based microcapsules, as visualized in Figure 19 [171]. Its predominance as an encapsulating agent in this field is primarily due to low material cost, robust sealing capabilities, good chemical stability, and simple fabrication process [50]. However, the release of formaldehyde through gradual degradation of the shell layers presents both environmental and health hazards, making this an ideal area for innovation through the development, or optimized use of, equally robust and nontoxic encapsulation materials.

In interfacial polymerization, polymerization of the organic microcapsules occurs on a layer between two immiscible phases, generally two liquids, making up an organic and an aqueous two-phase system. Instead of the oil-based mixture directly emulsifying within the aqueous phase as with in situ polymerization, the resin or polymer shell forms around the TC cores at the oil–water interface, which are then dispersed into the aqueous phase as core–shell structures [172]. Similar to in situ polymerization, it can also provide high yield while being easy to adapt, modify, and repeat. Despite higher potential control of the synthesized shell properties due to lower variability, it typically incurs higher costs from stricter solubility and property requirements, as well as from

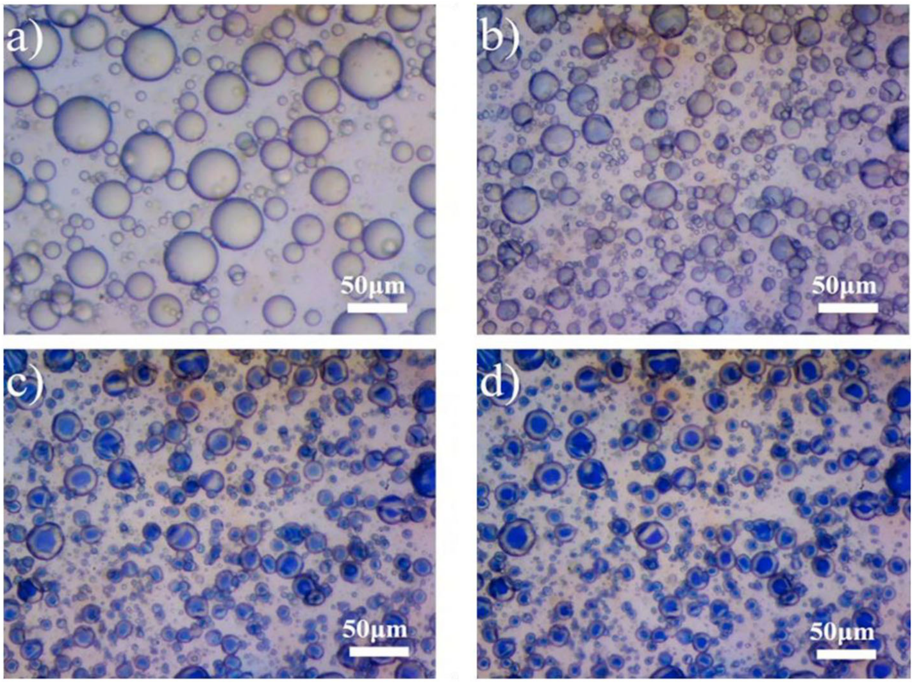

FIGURE 19 | Real-time observation of in situ polymerization of CVL-based thermochromic microcapsules with methylated melamine–formaldehyde at various periods after initial mixture, namely: a) 25s; b) 45s; c) 80s; d) 92s (Reused with permission from [171] Copyright 2018 Elsevier Ltd., all rights reserved).

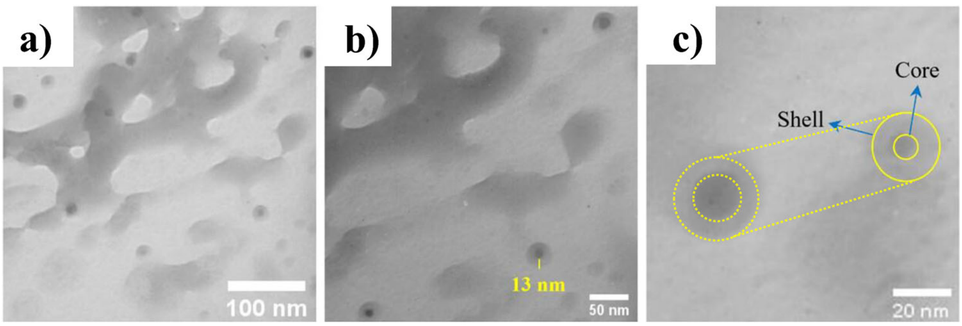

FIGURE 20 | TEM images of CVL-based thermochromic fluorescent pigments coated with a poly(methyl methacrylate) shell using emulsion polymerization at different resolutions (Adapted with permission from [176], Copyright 2025 American Chemical Society, all rights reserved).

a more complex subsequent purification to isolate the produced functional particles [173].

Emulsion polymerization involves the dispersion of core thermochromic materials in an aqueous solution, forming the standard oil-in-water emulsion of core droplets with the help of surfactants and vigorous stirring [174]. The encapsulating polymer monomers can then be introduced, with polymerization triggering around the dispersed core materials by a water-soluble initiator. The use of this process to produce solid-state TC powders can be found, for instance, in the work done by Zeng et al. (2024), where the prepared TCMs are integrated into glass technologies to develop smart window devices [175]. The proven success in preparing encapsulated TCMs using this process has made it a staple alongside in situ polymerization, as exemplified by Masoumi et al. (2025), who prepared reversible CVL-based nanometric-sized capsules with a high-transparency, stable, and nontoxic poly(methyl methacrylate-*co*-methacrylic acid) shell for anticounterfeit inks, as seen in Figure 20, proving the encapsulation techniques' reliability for specialized applications [176].

4.1.1.2 | Physical Encapsulation Processes. While chemical methods are dominant in the industrial production of functional security ink pigments, due to higher reliability and chemical robustness, there is a growing interest in hybrid or physically assisted encapsulation methods due to higher compatibility with lower-cost sustainable practices and bio-friendly materials. Physical encapsulation methods include spray-drying, extrusion, and solvent evaporation as the most frequent.

Spray-drying is a process where an oil–water emulsion containing the TC and encapsulating materials is sprayed using an atomizer into a drying chamber. Inside, a gas stream at a specific temperature rapidly evaporates the remaining solvent, and the solid particles are filtered for collection. While it presents provable scalability and has been effectively used for preparing encapsulated particles, it generally has lower efficiency due to loss in product within the drying chambers' walls [177], and has seen scarce use in developing TC pigments.

Extrusion is another cost-effective physical method, where the functional materials are added to a high viscosity hydrocolloid solution, which is then forced through a thin nozzle into a curing solution, resulting in relatively large particles (generally >100 µm) [178]. This higher particle size can, as mentioned previously, hinder some subsequent printing processes, but extrusion can nevertheless be an option to obtain reliably homogeneous microcapsules, with efficient reproducibility and without the need for potentially toxic organic solvents or temperature requirements [179].

With solvent evaporation, the encapsulating polymers and core materials are both dissolved and stirred in a water-immiscible organic solvent. The solution is added dropwise into an aqueous solution with a suitable stabilizer, such as poly(vinyl alcohol), resulting in loose and porous microparticles that harden over time by evaporating the aqueous solvent and can be collected for further processing into ink formulations [180]. While this cost-effective method results in reliable and highly stable microcapsules, the limited control over particle sizes and low quantity of prepared TC pigments per batch limit its applicability at a larger scale [82].

4.1.1.3 | Physical–Chemical Encapsulation Processes. Coacervation is a hybrid physical–chemical method, which can be classified as simple or complex coacervation. For simple coacervation, a single coating polymer is dissolved in an aqueous solution with a TC oil-phase emulsion. By adjusting the temperature or pH of the solution, the dissolved polymer loses solubility and deposits around the TC particles, hardening into an encapsulating layer as it cools and/or is assisted by crosslinking agents. With complex coacervation, two oppositely charged polymers are used, which interact electrostatically under ideal conditions (temperature, pH, etc.) to form the encapsulating shell. The gelatine-based shell materials commonly prepared using coacervation have relatively low mechanical strength when compared to the resins used in standard polymerization techniques, but are simple to scale-up and do not require any specialized solvents or apparatus [181]. TC cholesteric liquid crystal microcapsules have previously been prepared using complex coacervation by Guan et al. (2018), where a precise control of the emulsification and reaction conditions enabled low variation for particle sizes, exhibiting excellent TC behavior within their transition bandgap [182]. This technique is also ideal in maintaining the stability of biopolymers during the encapsulation process [183], garnering greater interest for TC particle systems which include, for example, gelatine or cellulose derivate-based encapsulants, in the effort to promote sustainable development [180, 184].

Encapsulation through a sol–gel process entails a transformation of a "sol" or solid particles subject to hydrolysis and/or polymerization reactions, forming a suspended colloidal solution, which is converted into a solid or gel-like state. This process is frequently adopted for microencapsulating TC particles with silica, through the use of tetraethoxysilane (TEOS) as a precursor or other inorganic oxides [185, 186], such as with the silica-coated vanadium dioxide core–shell structures developed by Wang et al. (2016) [169]. The inorganic coatings produced using this process are chemically inert and showcase greatly improved durability when compared to polymeric alternatives.

4.1.2 | Innovative Materials and Property Control

Recent research has directed attention toward optimizing the synthesis of TCMs, particularly in the hopes of better controlling transition temperatures and thermochromic behavior. Lynn et al. (2019) demonstrate this with their PDA and indium/gallium-doped zinc oxide nanocomposites with a reversible color-shift at 70°C for gallium and undoped particles, and an irreversible color transition at 50°C for indium-doped particles [75], as can be seen in Figure 21, providing new insights into how to control the TC behavior of PDA through the modification of the ZnO nanosubstrates they are frequently built upon. With PDA's proven disposition for information encryption [69, 79, 80], the capacity to control their intended TC behavior can be used to create cyphers with tunable behaviors, to assist in forming highly secure and difficult-to-replicate authentication markers.

Similarly, the development of entirely new TC inks from underutilized, yet promising, TCMs can also lead to broader applications in anti-counterfeit. Over a decade ago, gold and silver nanorod and polymeric ionic liquid composites were shown, by Tollan et al. (2008), to possess irreversible thermochromism between 25°C to 200°C, associated with a permanent change in plasmonic resonance as the nanorods reduce in size and gradually transform into nanospheres from heat, as is represented in Figure 22 [106]. This gradual structural transition is directly associated with a color-shift, which can be used to determine the maximum temperature the nanoparticles were subjected to. Since then, gold and silver nanoparticles' TC properties have been seldom adopted for functional TC ink development, with research involving them instead focusing on their dynamic luminescent properties as nanoclusters [187] or as elements for multifunctionality alongside other established TCMs [188]. However, their highly stable irreversible transition has the potential to serve as the basis for developing novel and high-reliability temperature-sensing anti-counterfeit markers.

4.1.2.1 | Advanced Characterization Techniques. Characterizing the properties of TCMs allows for a deeper understanding of their inherent properties and is critical for the development of innovative ink formulations. In literature, we find frequent use of differential scanning calorimetry (DSC), UV–Visible spectroscopy, Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), energy-dispersive spectroscopy (EDS), and thermogravimetric analysis (TGA) to characterize thermal, structural, optical and chemical behavior [18].

With DSC, by measuring the heat-flow of a sample during heating or cooling can be verified the existence of a phase transition or transformation in a sample, along with its the heat-flow as an endo or exothermic reaction. This can be used to identify the thermal stability, activation range, and reversibility

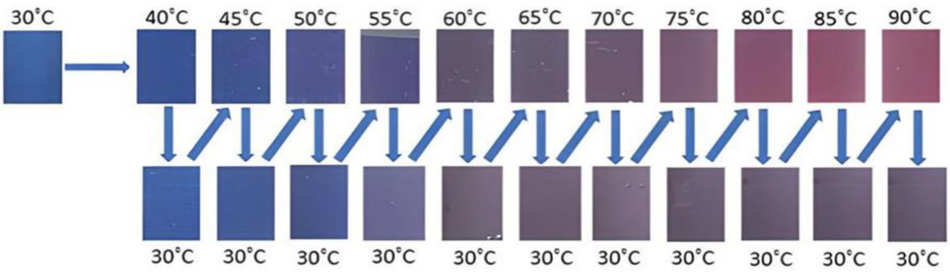

FIGURE 21 | Thermochromic reversibility of poly(PCDA)/InZnO nanocomposite (15:85 ratio of In:ZnO), showcasing the possible irreversibility at low temperatures by controlling doping concentrations (Reproduced with permission under a CC BY 3.0 open access license from [75], Copyright 2019 IOP Publishing Ltd, all rights reserved).

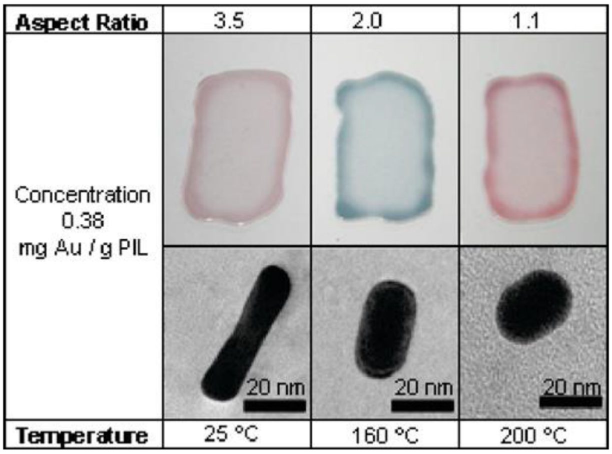

FIGURE 22 | Images showcasing the color-shift of gold nanorod/ poly(1-vinyl-3-ethylimidiazolium) bis(trifluoromethanesulfonimide) films at different temperatures and the corresponding TEM images of the transformation of the nanorod structures throughout this irreversible shift (Adapted with permission from [106], Copyright 2008 American Chemical Society, all rights reserved).

for the characteristic phase transition in TCMs. Furthermore, this technique is frequently used alongside TGA to measure the thermal decomposition limits of novel TCMs, both in air and inert nitrogen atmospheres. UV–visible spectroscopy is essential in measuring the optical behavior of TCMs to quantify color changes and measure the corresponding light absorption shift during the TC transitions [189]. Fourier transform infrared spectroscopy (FTIR) is also commonly used for organic-based TCMs, as it can determine the vibrational frequencies characteristic of molecular bonds, critical in identifying functional groups through a quick and nondestructive process [190, 191]. For instance, Rossi et al. (2021) describe how the shift in specific peaks during a FTIR measurement can be correlated to the opening of the lactone ring present in leuco dye systems [37]. Although FTIR generally offers higher sensitivity measurements, temperature-dependent Raman spectroscopy is yet another process by which we can measure the vibration peaks of TCMs, especially for inorganic materials, where it can provide more details to identify the crystalline structures throughout the transition states [192, 193]. These measurements help to determine which functional groups or structures are involved during the color-shift of these TC compounds, providing insights into how to potentially influence the transition mechanism.

SEM imaging is a high-resolution imaging technique that allows for a direct visualization of the morphology and surface of TC systems. The homogeneity, porosity, aggregation, pigment sizes, and thickness of a printed layer can be observed, as well as the effects of any potential ink additives on these properties [194]. Notably, the uniformity and thickness of encapsulant layers can be identified to determine if their synthesis conditions were ideal. EDS is primarily used in tandem with SEM and is used to measure the weight percentages for the elemental composition of the printed samples or dry pigments. When used with higher resolutions, EDS can also be used to identify the composition of the encapsulant layer for TC pigments and affirm if they have an expected and uniform distribution of the intended constituents [37]. TEM, similar to SEM, is another type of microscopy analysis, providing even higher resolutions that can visualize particles at a nanometric scale (0.2 nm for a conventional TEM), providing detailed information on the interfaces, sizes, and uniformity of encapsulated TC pigments [195, 196], as was previously observed in Figure 20.

There are several approaches in the literature on how to determine the dynamic color changes in TCMs, such as through the use of CIELAB color space coordinates to determine the difference in color contrast or total color difference between transition phases [197]. The use of climate and ageing chambers alongside colorimetry analysis tools can be used to determine the influence of heat and UV exposure on the color of a TC system. The controlled environment within these devices can simulate long-term exposures in a shorter timeframe and is beneficial toward estimating the expected lightfastness and durability of printed TC films before scale-up [108]. Additionally, the hysteresis loop for reversible TCMs can also be evaluated through various parameters, including variations in the width of the hysteresis loop (associated with the transition temperature interval), discoloration, contrast, and rate of the color change. In literature, Panák et al. (2015) corroborate the importance of a method for identifying the dynamic thermal and color changes in TCMs, contributing to this by outlining these critical parameters and defining a system for the analysis of leuco dye-based TCMs [48].

Advancements in computational modeling and simulation have also provided new methods to predict the behavior of leuco dye molecular interactions, DA monomers, and subsequent TC properties of these materials, which will also serve to bolster research efficiency [198, 199]. Computational screening can also

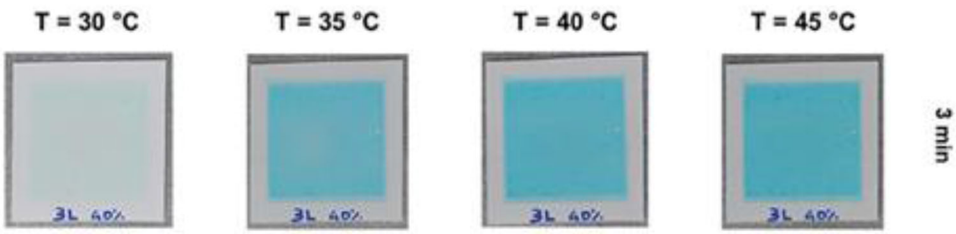

FIGURE 23 | Thermochromic behavior of films composed of three layers of inks composed of 40 wt.% $[Bmim]_2[NiCl_4]$ complexes 10 wt.% in hydroxypropyl cellulose and 50 wt.% water, using flexographic printing (Adapted under a CC BY-NC 4.0 open access license from [202] Copyright 2025 by the authors, all rights reserved).

serve as a time-efficient method for analyzing potential molecular structures and facilitating the design of novel TCMs, as demonstrated using the simulation protocol developed by Deichmann et al. (2024), which identified two ideal leuco dyes out of more than 1600 potential structures [199].

As the functionalities for these analytical techniques continue to improve, with higher accuracies, quicker measurements, in situ thermal monitoring, and real-time visualization of the samples, so too will our understanding of TCMs continue to expand, facilitating their development for specialized applications.

4.1.3 | Ionic Liquid Integration

As previously mentioned, ionic liquids are highly promising organic frameworks for the development of hybrid inorganic–organic TC complexes. Their combination of beneficial and modifiable properties, ecologically friendly nature, and non-flammability has prompted an increase in research and development of ionic liquid-based advanced materials in recent years [200], including for stimuli-responsive films and inks [201]. Within the field of TC ionic liquids, Fernandes et al. (2019) [97] and Morais et al. (2025) [202] both showcase comparable nickel chloride and bis(1-butyl-3-methylimidazolium) complexes ($[Bmim][NiCl_4]$) featuring high stability TC transitions, from a high transparency tetrahedral configuration to a high contrast blue octahedral configuration, between 30°C and 85°C in solution, and at a narrower 30–40°C as a printed thin film, as observed in Figure 23. The ionic liquids' interaction with the transition metal species creates a dynamic and stable complex in which the reversible change in coordination number of the nickel ion can occur, induced by temperature, resulting in the intended color-shift [201].

To further grasp their potential, we note that ionic liquid-based composites have also found great promise in other functional security ink developments (non-thermochromic), also based upon their high stability and sustainable properties. This is exemplified by the novel eco-friendly films produced by Cruz et al. (2023), composed of sodium alginate and [Bmim][Eu(tta)4], which showcase consistent photoluminescent properties suitable for anti-counterfeit security elements [203]. Similarly, luminescent PVDF and [Bmim][Eu(tta)4] composite inks have also been prepared for the first time by Correia et al. (2022) and printed successfully as a proof-of-concept for anti-counterfeit applications using both blade-coating and screen-printing methods [204]. Studies such as these demonstrate the advantages ionic liquids bring toward the development of security-focused functional systems, emphasizing the need for additional research into how they could be used as frameworks and enhancers for thermochromic ink formulations.

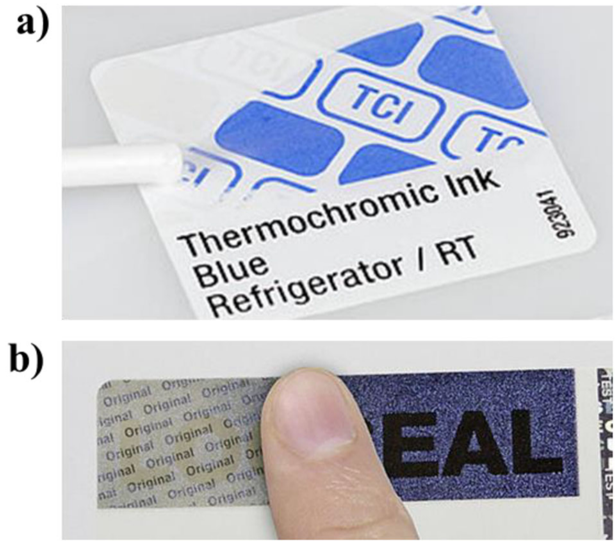

FIGURE 24 | Bisphenol A-free leuco dye-based ink formulations developed by Schreiner Group(r) for a) cold and b) heat-activated semi-covert anticounterfeit features (Reproduced with permission from [205], Copyright 2025 Schreiner Group, all rights reserved).

4.2 | Sustainable Development

4.2.1 | Biocompatible and Biodegradable Thermochromic Materials

As mentioned, the primary goal of sustainable development in commercial TCM ink development is to reduce the use of toxic and environmentally hazardous materials, while transitioning to more biodegradable and low cytotoxicity ones. An example of this can be seen in the latest generation TC inks developed by Schreiner Group, as seen in Figure 24, which showcase leuco dye-based formulations with much lower toxicity by replacing the conventional Bisphenol-A developer [205]. Resorting to water instead of volatile organic compound (VOCs) releasing organic formulations as the base solvent in TC security ink formulations is the most direct way to minimize their environmental impact. The interest in the use of water and resin emulsion-based security inks has grown alongside the interest in eco-friendly formulations, offering a viable cost-effective alternative to conventional organic solvents, which are frequently flammable, explosive, toxic, or polluting in nature [206]. Considerable efforts have been made in recent years to improve the usability of water-based inks, from additive binders for improved adhesion to polymeric substrates [207] to optimizing the techniques and factors that affect the preparation of functional materials within water-based inks and coatings [208, 209].

Another method by which sustainable TC inks are being developed is through the use of natural dyes and biopolymers as functional components, such as those directly provided by foods and plants, including anthocyanin-based pigments or curcumin membranes [150, 210, 211]. An example of this can be found in the anthocyanidin-based red-cabbage extract inks prepared by Alshammari et al. (2022), which showcase a gradual color-shift from a blue–purple hue to a vivid red between 25°C and 65°C when in the presence of ferrous sulphates, resulting in green organic–inorganic TC formulation. The high thermal stability, durability, and self-healing nature of this ink proved ideal for low-cost TC anticounterfeiting applications and showed potential for medical applications due to exceedingly low cytotoxicity [150]. PDA-based TCMs have also been shown to possess naturally high biocompatibility, which has resulted in their extensive use in medical research and temperature-sensing food packaging concepts compared to other conventional TCMs [78]. Their low cytotoxicity, along with proven use in anti-counterfeit markers, makes them ideal for security ink developments focused on sustainability, despite incurring higher costs than their simpler organic leuco dye-based counterparts. Furthermore, ionic liquid mediums can also serve as a direct replacement for VOCs in an ink formulation, replacing the toxic byproduct-releasing properties of conventional solvents and/or encapsulation materials with their nonvolatile and negligible vapor pressure nature, allowing them to act as a "green" framework for sustainable TCM ink research and development [201].

4.2.2 | Cellulose Derivates

The use of cellulose derivates in the development of printing inks has been a highly pursued scientific effort in recent years, due to their natural abundance, biodegradability, and excellent mechanical properties [212]. Moreover, it can be seen as another method of improving the eco-friendly properties of TC ink formulations. For instance, nanocellulose-based films doped with black-to-leuco thermochromic particles were recently developed by Jaiswal et al. (2024), showcasing reversible thermochromism around room temperature values [213]. These thin films, prepared using suspension casting as passive room-cooling elements, retained stable TC behavior of the leuco dye pigments within the nanocellulose films, which enhanced the cooling of the system and provided a sustainable medium. Likewise, Morais et al. (2025) also integrated cellulose derivates into their inorganic–organic hybrid TC complex-based ink formulations, in order to assist in fine-tuning viscosity as an ink additive, while providing improved mechanical and thermal behavior to the passive TCM component [202]. Cellulose-based substrates can also provide TC ink prints with an enhanced color stability throughout their ageing when compared to conventional thermoplastic substrates, as shown in the tests conducted by Kulčar et al. (2023), which provided significant insights into understanding the influence of substrates for long-standing TC ink prints [214].

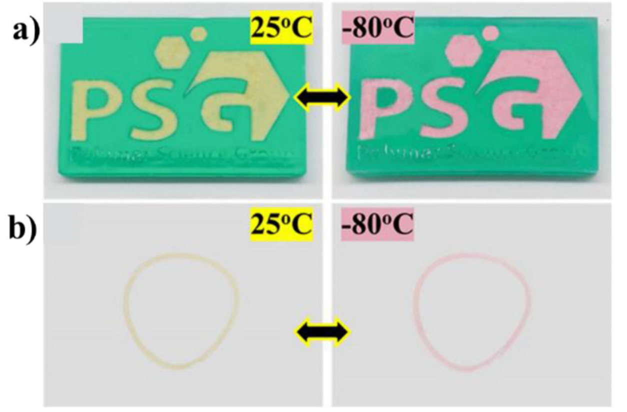

FIGURE 25 | Nontoxic thermochromic microcapsules a) cast into a 3D-printed PDMS film and b) screen-printed as an ink onto paper (Reproduced with permission from [164] Copyright 2020 American Chemical Society, all rights reserved).

The proven value of cellulose derivates as a green platform for TCM developments, as additives to control viscosity and enhance stability, and for reducing costs of ink formulations shows great promise for security applications.

4.2.3 | Sustainable Microencapsulation

The use of biodegradable, nonreactive, and/or nontoxic microcapsule materials for the development of TC pigments is also a direct way to improve their sustainability. Inert or nonreactive encapsulants, such as silicon-based shells, can provide high-performance functionality while replacing environmentally harmful substances. As an example of this, Liu et al. (2020) developed a nontoxic chlorophenol red-water binary system, encapsulated within inert silicon shells, which resulted in 1 μm-sized pigments with stable reversible thermochromism between 25°C and −80°C. These eco-friendly microcapsules were then successfully developed into ink formulations for screen-printing and as additives for PDMS films, as seen in Figure 25, providing a novel nontoxic alternative to conventional leuco dye-based commercial inks without compromising their TC capabilities [164]. Future efforts in developing sustainable encapsulating agents for TC pigments will continue to focus on reducing environmental impact and improving cost effectiveness while retaining their inherent functionality.

Green encapsulants, including chitosan, alginate, gelatin, gum arabic, and cellulose derivatives, have found use in recent years in the preparation of environmentally friendlier and biodegradable TC pigments [182, 184, 211]. Much like the previously mentioned functional biopolymers, these encapsulants can also be supplied from natural sources, providing a sustainable and cost-effective solution for the encapsulation of leuco dye or liquid crystal systems. Most can be dispersed in water, facilitating VOC-free ink formulations, and can be used to directly replace more harmful encapsulating compounds such as formaldehyde-based synthetic resins. The works of Tözüm et al. (2023 and 2024) highlight these green encapsulants, successfully encapsulating conventional leuco dye systems using both a chitosan/sodium alginate and a gelatin/gum arabic using complex coacervation [184, 215]. Both of these natural TC microcapsule systems were further used to prepare functional fibers and subjected to multiple heat and wash cycles, where they showcased reasonable durability and functionality, demonstrating the potential of biopolymers in isolating TC systems.

While green encapsulants generally possess lower thermal stability and mechanical durability than their synthetic counterparts, which can limit their use in more intensive manufacturing processes, advancements in bio-based resins and composites have been gradually increasing their competitiveness as a suitable alternative [216, 217]. Additionally, with the focus on sustainable development in modern research, the use of these materials is under frequent focus, and novel techniques and formulations could facilitate their integration and/or enhance their expected properties in order to achieve competitive functionality as encapsulants for TC inks.

4.2.4 | Printing Optimization and Cost-Effective Development

4.2.4.1 | Printing Optimization of Thermochromic Inks. Composition and behavior of functional TC inks can vary greatly from conventional printing inks, meaning a thorough optimization of the preprint, printing, and drying processes is needed in order to guarantee high-resolution and high-quality patterned products [10]. Over the last few years, research aimed at enhancing the printed quality of functional security inks has increased, with offset lithography and inkjet being the primary focus of optimization. As an example of this, the optimization of dry offset printing using TC inks was carried out by Jakopčević et al. (2023), where they determined that TC inks could achieve quality prints competitive with conventional inks, and even provided lower raggedness and blurriness when printing horizontal lines, despite having a rougher surface due to larger microcapsule sizes [117].

The curing process for the oil-based offset inks is primarily done using tungsten-based lamps, which can rapidly degrade substrates such as conventional polycarbonates due to a wide emitted spectrum, limiting the types of substrates compatible with a majority of established production lines. In recent years, this has been mitigated through the transition to LED lamps, providing a more sustainable and lower operating cost curing process compatible with a wider array of substrates, at the cost of slightly lower production line efficiency from lower light intensity [218]. However, since ink formulations need to be similarly adjusted toward the strict wavelengths emitted by LED-based systems, the transition to these more versatile offset production lines has been slow.

The incorporation of inorganic TC and ionic liquid complexes into sustainable biopolymer matrixes as inks for screen printing has been explored by Cruz et al. (2024), as seen in Figure 26, who demonstrated that the ink formulation can possess suitable adhesion and long-term stability on a variety of substrates [194].

As mentioned, TC particle size reduction remains integral in facilitating printing process compatibility for functional TC inks, especially when referring to organic-based pigments. For this, a novel room-temperature solution-mixing method for preparing PDA and zinc oxide composites with TC and luminescent properties was developed by Chanakul et al. (2021), resulting in smaller PDA/ZnO nanocomposite particle sizes (~60 nm), highly suited for standard printing methods and providing an improved potential for larger-scale adoption of PDA-based TCMs [219].

The continuous effort in establishing the necessary considerations required for efficient printing of TC inks will facilitate their integration into existing production lines and bolster the interest in functional ink applications. Therefore, concern remains in expanding our current understanding of optimizing printing parameters such as flexography, screen-printing, and gravure printing for modern thermochromic solutions.

4.2.4.2 | Process Cost-Reduction and Lower-Cost Material Integration. The material cost of some TCMs can also serve as a limiting factor for large-scale production, particularly in the case of liquid crystal and inorganic-based TCMs. Larger particle-sized organic TC pigments may also incur additional material costs, from requiring more wasteful printing processes such as screen-printing and gravure printing. From a business standpoint, a reduced cost in raw materials and processes would greatly improve the appeal of thermochromic ink development for scalable production lines. This is yet another reason that the property enhancement of leuco dye-based systems, due to their considerably lower production costs, or the simplification of other TCMs' development processes, could further bolster the interest in and competitiveness of this growing market.

4.3 | Multichromic Inks for Enhanced Complexity and Functionality

As TC ink development leads to more reliable, durable, and sustainable materials, multifunctionality surges as an effective way to further improve the security level of these functional printing inks. The ability to simultaneously respond to a variety of stimuli in addition to temperature greatly improves its complexity and further complicates clonability, while broadening application potential. In the effort to develop multifunctional inks possessing thermochromism, fluorescent and photoluminescent materials showcase the highest compatibility due to comparable optical properties born from structural and/or chemical transitions. TCMs exhibiting paramagnetic, shape-memory, and energy-storage properties have also been noted in literature [100, 220, 221]. These properties prove difficult to adopt in conventional security applications, but could provide a platform for unique and novel multifunctional sensors using specialized equipment.

Quantum dots, and carbon dots in particular, due to their excellent fluorescent and tunable optical properties as luminophores, have received great interest in the development of multifunctional security technology [222]. Multicolor carbon dots with temperature-dependent fluorescence were achieved by Wang

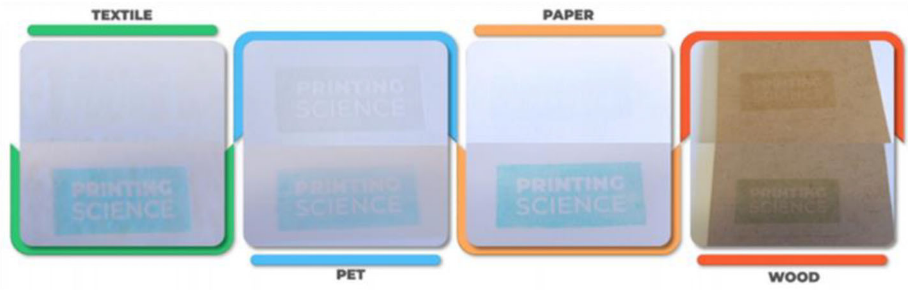

FIGURE 26 | Poly(L-lactic acid and a thermochromic [Bmim][$NiCl_4$] complex (40% wt.) screen-printed onto various substrates (Textile, Polyethylene Terephthalate, Paper, and Wood) to demonstrate its compatibility (Adapted with permission from [194] Copyright 2024 by the authors, all rights reserved).

et al. (2024) by using a mix of optically variable carbon dot particles to generate a full visible color range [223]. These carbon dot inks yielded inkjet-deposited patterns where the emissive fluorescence intensity and color tone could be controlled by varying the temperature of the substrate, resulting in printable patterns with multistage encryption. Xia et al. (2024), on the other hand, demonstrate the potential compatibility of quantum dots alongside established TCMs by synthesizing PDA and zinc oxide quantum dot composites in a water-based solution, showcasing excellent simultaneous thermochromic and fluorescent behavior with a novel, environmentally friendly solution [224]. Likewise, Yang et al. (2019) report the synthesis of polymer dots with multifunctional properties. Their research underscores the complex temperature-dependent fluorescence exhibited by these materials, which demonstrates significant potential for anticounterfeiting applications. Furthermore, the ability to incorporate encrypted elements on both flexible substrates and conventional paper enhances the versatility and applicability of these polymer dots in established industrial printing lines [11]. All these examples can be directly seen in Figure 27, where the color shift of the security patterns can be observed with and without UV exposure at various temperatures.

Fluorescent cellulose-based anti-counterfeit technology has also found considerable interest in the wave of sustainable development, such as with the UV-absorbent cellulose-based fluorescent films extensively highlighted by Quan et al. (2023) [225]. Although not directly exhibiting TC properties, similarly prepared UV-absorbent functional biopolymers could instead be used to protect UV-sensitive TC particles, to simultaneously develop a complex, multifunctional response to both heat and light using a sustainable medium. Organic luminogens that exhibit aggregation-induced emission have also shown thermochromic behavior, with temperature-dependent shifts in emissive strength and wavelength of their fluorescent properties, particularly at temperatures below 0°C. This is demonstrated by Lin et al. (2015), who developed stimuli-responsive powders that could undergo reversible emission for double-layered encryption and display their potential as TCMs verifiable at negative temperatures [226].

5 | Conclusions

In response to the growing prevalence of counterfeit products, the interest in developing novel high-security anti-counterfeit technologies has increased significantly. The development of higher forms of security technology has evolved to better provide secure product identity, preserve market growth, and guarantee quality and safety in products. In this field, thermochromic security inks, which exhibit temperature-responsive color changes, have gained increasing interest over the last decade, showcasing various approaches in information encryption, authentication patterns, and anti-counterfeit technologies as a whole.

In this review, we examined a diverse range of organic, inorganic, and organic–inorganic hybrid thermochromic materials, each with distinct transition mechanisms. Leuco dyes are organic-based materials that rely on a temperature-dependent molecular rearrangement of a dye-developer-solvent system. Their ease in fabrication, strong color contrast, and reversibility make them the most widely employed thermochromic material in a variety of fields. However, low stability, even when encapsulated, makes them less advised in rigorous applications, such as expected in anti-counterfeit technologies. Thermochromic liquid crystals offer color variations over the visible spectrum when transitioning from a crystalline phase to an isotropic liquid phase, remaining colorless outside of this transition. This response offers precise temperature sensitivity with a vivid color shift, making them valuable as high-end temperature-sensing elements within a controllable temperature range. Despite this, they are highly sensitive to light, humidity, or mechanical strain, which can irreversibly damage their functional properties, requiring effective encapsulation for reliable application. Polydiacetylene conjugated polymers showcase thermochromism due to a structural rearrangement of the distance between polymer chains. This transition occurs over a much wider temperature range and can be tailored to exhibit a reversible and/or irreversible color shift. They showcase the highest stability of conventional organic TC materials, making them ideal candidates for reliable high-security anti-counterfeit elements. Inorganic TCMs, such as transition metal oxides, exhibit high stability transitions arising from structural transitions and changes in coordination numbers. But regardless of their promising properties, most inorganic TCMs' transition temperatures occur at unreasonably high temperatures or possess high-toxicity components, making them unsuitable for printed elements. However, recent successes in the development of inorganic–organic hybrids have resulted in promising, high-stability systems with tunable properties, ideal for the creation of novel TC security technologies.

Despite promising functional properties, thermochromic materials suffer significant challenges, such as degradation from UV

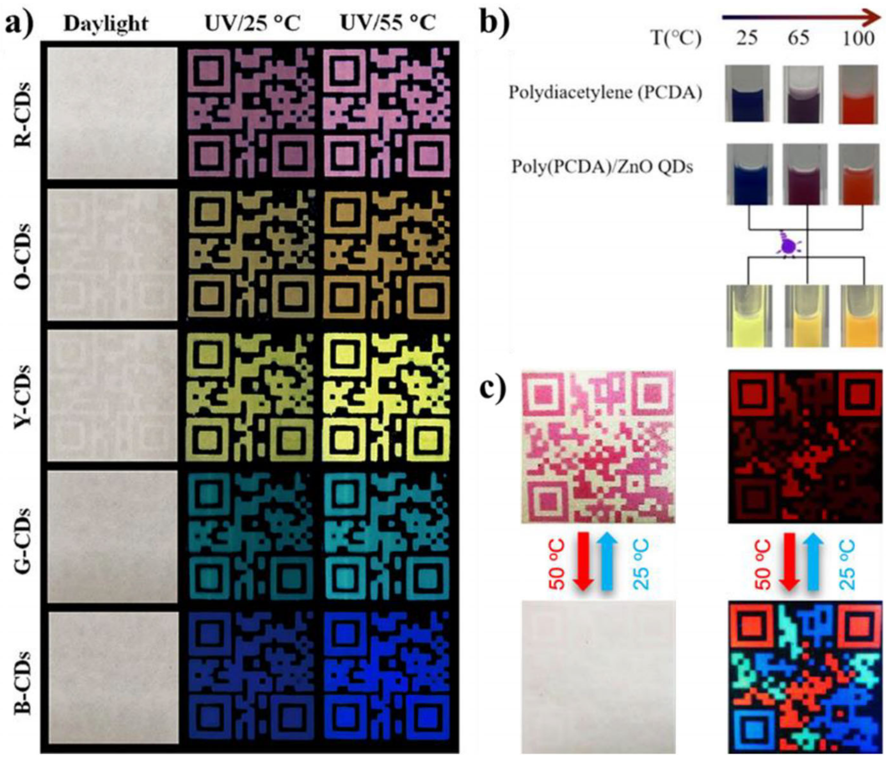

FIGURE 27 | Examples of novel multifunctional quantum dot-based systems showcasing thermochromism: a) Multicolor quantum dot-based QR-code encryption showcasing temperature-dependent fluorescence under daylight, room temperature UV exposure and 55°C UV exposure (Adapted with permission under [223] Copyright 2023 Published by Elsevier B.V. on behalf of Chinese Chemical Society and Institute of Materia Medica, Chinese Academy of Medical Sciences, all rights reserved); b) Poly(PCDA) and fluorescent zinc oxide quantum dot composites showcasing thermochromism at varying temperatures under conventional sunlight and under strong UV exposure (Adapted with permission from [224] Copyright 2024 Elsevier Ltd., all rights reserved); c) Semiconducting polymer dot-based QR-code encryption under daylight (left) and UV light (right) at varying temperatures, demonstrating multilevel encryption with RGB fluorescence revealed by sufficient heat (Adapted with permission under a ACS public use license from [11] Copyright 2019 American Chemical Society, all rights reserved).

radiation and oxidative agents, or being commonly composed of or used alongside toxic compounds, limiting their applicability and long-term effectiveness. However, recent advancements in thermochromic material research have led to significant improvements through a greater understanding and more precise control over transition mechanisms and color reversibility, broadening their versatility and expanding their use toward security applications. The development of higher durability inks and films through novel formulations and innovative encapsulation methods has enhanced their stability, performance, and longevity, resulting in more reliable printed patterns better suited for anticounterfeit elements. Simultaneously, the strive for environmentally friendly thermochromic inks has led to a focus on developing water-based TC inks free of volatile organic compounds, as well as replacing hazardous components with bio-based materials such as cellulose derivatives. These innovations align with the growing demand for sustainable development in the printing industry. Additionally, the integration of stable thermochromic materials with other stimuli-responsive systems has allowed for the development of multifunctional security elements, offering higher complexity, tunability, and greater reliability in combating counterfeiting or tampering.

Beyond property improvements, the adoption of thermochromic security inks at an industrial scale, by enhancing their compatibility with conventional printing methods, is an underdeveloped yet crucial focus. Achieving ideal printing equipment parameters, ink composition, and formulation properties, such as viscosity, drying behavior, and stability, is essential to ensure long-term durability and high-quality printed patterns. However, further optimization is required to guarantee cost-efficient printing, and future progress in this field should facilitate its incorporation into modern production lines in the security printing industry.

As increasingly diverse, durable, sustainable, and complex thermochromic security inks are developed, alongside the growing interest in the industry and our understanding of nanofabrication, polymer chemistry, and printing optimization, higher security thermochromic patterns with tailored properties will be developed. These continued advancements in thermochromic functional ink technology will contribute to raising product security and strengthening protections against counterfeit threats for both consumers and manufacturers.

6 | Future Perspectives

6.1 | Production and Development Costs

Thermochromic ink formulations can potentially incur high costs, owing to expensive raw materials or synthesis processes for TC pigments, which can in turn inhibit their scalability or cost-effective adoption. No matter the reliability of the end-product thermochromic security elements, if, theoretically, their preparation and manufacturing demands higher costs than the corresponding economic impact of counterfeit products themselves, their use is resultantly unjustified for large-scale production.

Knowing this, a high manufacturing or material cost in the development of TC security inks can potentially be justified by the quality and reliability of the developed security element. Reducing the functional material quantity or the printed film thickness can greatly help minimize costs; however, a high degree of functionality must still be assured to guarantee that the deposited TC patterns serve their security functions. For luxury items or critical documentation, expensive TC anti-counterfeit technology that assists in verifying authenticity can still become a worthwhile investment, despite incurring additional costs in manufacturing, due to a higher importance allocated to brand image, security, and reputation. However, for more "conventional" anti-counterfeit elements, a focus on cost-reduction will always be integral in the overarching development of TC security inks for scalable production.

6.2 | Life-Cycle-Assessment

To our knowledge, an extensive life-cycle-assessment (LCA) of the various types of thermochromic inks, from their preparation to disposal, has not been conducted. This systematic outline would benefit our understanding of their potential environmental impacts, waste generation throughout the printing process, and potential risks during and after manufacturing. The environmental footprint associated with the extraction of core components in inorganic TCMs is of particular note, as most are listed as critical raw materials and can incur high extraction costs. TCMs, alongside their potential binders, solvents, and additives used in functional ink formulations, also require a careful evaluation of their long-term degradability and byproduct emission before being adopted into large-scale production. Careful consideration of all these factors would assist in focusing development targets and innovations toward achieving the sustainability goals set out by the United Nations, particularly goals 9, in fostering sustainable innovations, and 12, to ensure sustainable development processes.

6.3 | Novel Encapsulation Processes

While widely adopted techniques like in situ or emulsion polymerization dominate the encapsulation of thermochromic material, less utilized methods such as coacervation, spray drying, and sol–gel encapsulation present promising avenues for innovation. These alternative processes can feature higher versatility in preparing biocompatible, porous, and tunable microstructures that can potentially enable greater control over the final properties. As security inks require a great degree of reliability, this finer control of TC properties and the expected color-contrast means improvements to standard and novel encapsulation methods will be pivotal in further advancing TC security inks into robust tools for printing security elements and protecting products.

Author Contributions

The original manuscript draft, methodology, and investigation were performed by Duarte B. Oliveira. Conceptualization, review, and editing were performed by Duarte B. Oliveira, Andreia Araújo, Marta Corvo, and Emanuel Carlos. Supervision and validation were performed by Andreia Araújo, Marta Corvo, and Emanuel Carlos. All authors examined, commented, and have given approval to the final version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

Data Availability Statement

The authors have nothing to report

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.
Supporting file: admt70590-sup-0001-SuppMat.docx

Biographies

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