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Remiero

Thermochromic Polymers in Food Packaging: A Comprehensive Systematic Review and Patent Landscape Analysis

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Abstract: This study addresses the gap in research on the application of thermochromic polymers (TPs) in food packaging and their potential for real-time temperature monitoring, aiding in the assessment of food quality and shelf-life. TPs exhibit a visible color change in response to temperature variations. A comprehensive systematic review (SR) across multiple engineering peer-review databases using predefined terms was conducted. Additionally, international patent databases were investigated using the same predefined terms. Independent experts reviewed the methodology to identify and address potential biases. A total of 288 eligible articles and 922 patents were identified. After a duplicate selection and extraction process according to the inclusion criteria, four related full-text publications were selected from the initial 288 articles, and five relevant patents were selected from the 922 patents. The qualitative review suggests that TPs hold significant promise as food packaging materials due to their unique physical properties. The study concludes that TPs offer valuable properties for the food packaging industry, meriting further investigation to exploit their benefits fully.

Keywords: stimuli-responsive materials; thermochromic polymers; food packaging; systematic review; patent landscape analysis

1. Introduction

Packaging is an essential component of the food industry. It facilitates the food products' conveyance, protection, preservation and information dissemination [1]. Materials such as plastic, metal, paper, and glass protect food products against deterioration and contamination during transportation and storage. Packaging also contains essential information such as nutritional content, batch numbers and expiration dates, and provides convenience to customers. In 2022, the food packaging market was estimated to be worth \$358.3 billion and is expected to reach \$592.8 billion by 2032 [2]. This growth is accredited to the increasing world population, urbanisation, and the expansion of international food trade, creating greater demand for food and convenience [3].

However, traditional packaging materials (plastic, metal, paper, and glass) cannot communicate their interactions with the surrounding environment and are thus considered passive [4]. Over 40% of plastics produced worldwide are used in packaging applications [5]. A significant challenge in today's material research is to develop functional intelligent materials that can actively adapt to changes in their surroundings. Current interdisciplinary research focuses on food packaging materials that can undergo purposeful change, playing an active part in how the structure or device works [6]. These materials, referred to as "stimuli-responsive" materials (SRMs) or "smart" materials (SMs), have an excellent ability to perceive and react to environmental changes or external stimuli [7]. Figure 1 provides a schematic representation of the behavior of innovative materials. With the

application of a stimulus (internal or external), innovative materials react in a predictable and repeatable manner. Such triggers include physically-dependent stimuli (e.g., temperature [8], electric fields [9], specific wavelength [10], ultrasound [11], magnetic fields [12], and mechanical deformation [13], chemically-dependent stimuli (e.g., pH [14], ionic strength [15], redox (reduction/oxidation) [16] and solvent [17], and biologically-dependent stimuli e.g., glucose [18], glutathione [19], and enzymes [20]. In addition, a key feature of intelligent behavior is the ability to return to the original state after removing a stimulus [21].



Figure 1. Schematic representation of innovative materials' behavior.

Chromogenic materials (CMs): These are characterised by the color-changing capability (CCC). CMs with CCCs can change color instantly in response to an appropriate stimulus [22]. After removing the stimulus, they regain their original color (Figure 3). Chromogenic materials play a central position in intelligent packaging. As a result, these materials can be used as additives for pioneering packaging applications. Chromogenic materials possess unique characteristics which inform customers of the surrounding environment of the food and ultimately allow for food waste reduction. According to their stimuli, they can be classified as having internal stimuli or external stimuli [23] (Figure 2);

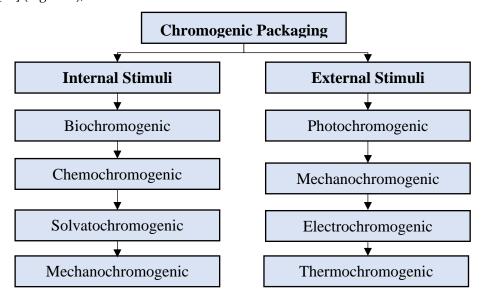


Figure 2. Chromogenic packaging stimuli.

Thermochromic materials (TMs): These are characterised by the color-changing effect (CCE). TMs with CCEs are stimuli-responsive materials that change color at specific temperatures, known as the "transition temperature" [24]. The transition temperature can change at various temperatures, depending on the specific material and application. TMs can change their color back and forth in response to changes in temperature. This change may be reversible or irreversible [25]. CCE is repeatable over ten times [26]. The CCE of TMs is a representative example of a defining quality of several TMs. (Figure 4).

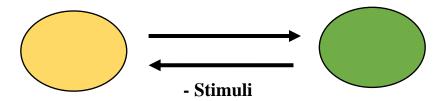


Figure 3. Schematic representation of color-changing material (CCM) after the application and removal of external stimuli.

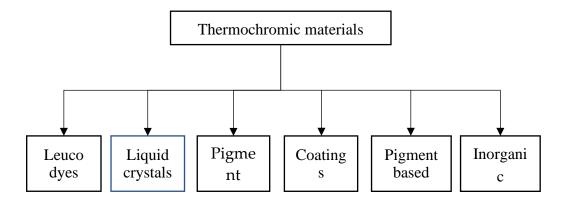


Figure 4. Color-changing materials (CCM) within thermochromic materials content.

Temperature-sensitive polymers are polymeric materials that exhibit CCC in response to an external temperature stimulus [27]. The first notable discovery of thermochromism was recorded in 1888 by Australian botanist Friedrich Reinitzer and the German crystallographer Otto Lehmann from the Rhenish-Westphalian Technical University in Aachen [28]. Various types of polymers, such as Polyurethanes (PUs) [29], Polyethlyeneterephthalate (PET) [30], Polystyrene (PS) [31], and Thermoplastic Elastomers (TPE) [32], and many more exhibit CCC.

The transition of specific chromophores or pigments between different energy states is the fundamental working principle for the CCC in thermochromic polymers (TPs). Chromophores are the chemical groups or molecules' within a material responsible for the color. In TPs, the chromophores are typically embedded in the polymer matrix and are colorless or transparent at low temperatures. As the temperature increases, the chromophores absorb energy and transition to a higher energy state, which results in a color change. The color change can be from one color to another or from a colorless state to a colored state, depending on the specific thermochromic polymer [33].

The color-changing effect has attracted attention for its potential application in packaging since its discovery in plastics [34]. In food packaging, introducing temperature indicators represented a noticeable paradigm shift in the quality control of perishable food due to the temperature sensing of products exposed to temperatures outside the acceptable range [35]. In pharmaceuticals, the use of thermochromic materials allows the monitoring of thermolabile pharmaceuticals, reducing the incidence of drug degradation due to exposure to temperatures that are too high or too low, ensuring stability and efficacy and that the drug will perform as expected [36]. These advancements in packaging benefit the end users of the product. Pharmaceutical packaging applications in which thermochromic materials can be used include pill bottles [37], blister packs [38], and insulin pens [39].

TPs are more attractive than other CCMs due to their significant flexibility, durability, long-lasting color change, cost-effectiveness, versatility, and reusability [40]. Due to these benefits, TPs have enormous potential to enter many fields of application, such as automotive [41], windows [42], and packaging [43].

The packaging industry is scrutinised for contributing to waste and environmental degradation as the global community becomes more concerned with environmental sustainability. Non-

biodegradable materials and poor waste management of said materials can cause environmental hazards [44]. Traditional packaging materials, notably plastics, pose environmental concerns due to their enduring effects on ecosystems [45]. In this context, the potential of TPs to support sustainable practices takes on particular significance in this setting. The key characteristics that define the environmental impact of these polymers are biodegradability, composability [46], and recyclability [47]. A sustainable thermochromic polymer would signal changes in the product's environment and ensure that it does not leave an indelible mark on our planet by degrading/decomposing over time. It is crucial to compare TPs' functional advantages and environmental impacts while investigating their properties and uses in food packaging. There may be areas where material science innovation and ecological responsibility may coexist. If the polymer is biodegradable or recyclable, there are many potential environmental benefits of using TPs in food packaging (minimised environmental pollution, lower carbon footprint). These benefits could result in economic benefits and cost savings in the long run, primarily due to reduced food wastage [48].

TPs are widely applicable in the packaging industry [49]. However, the necessity for further research and development in this field is evident, as indicated by the increasing demand for more scholarly publications. As the significance and potential of TPs in food packaging become increasingly evident, a thorough analysis of the existing literature and patents is imperative to identify advancements, gaps, and future directions.

Therefore, this paper conducts a comprehensive systematic review (SR) of the current literature and a patent landscape analysis (PLA) on the prospective application of TPs in food packaging. The research results from the recent emergence of intelligent food packaging in the global community due to consumers' desire for convenient food products with maintained quality [50]. It allows the investigation of unexplored areas in the research where TPs can offer value by focusing on external thermochromic stimuli for the food packaging industry. This detailed examination of the literature and patent landscape ensures that the review provides a comprehensive overview of the current state of knowledge. This review benefits the food packaging industry, which seeks a thorough understanding of the topic. An SR identifies and retrieves international evidence that gives insight into the academic progress of the research area [51], whilst a PLA allows a view into innovative advancements in the field and can spotlight where there is potential for patenting new solutions. Intellectual property (IP) is an asset which should be maximised and offers a competitive advantage [52]. Insights from this review will inform decision-making processes in product development, design, and implementing tangible properties in food packaging. In addition, this study offers an overview of the packaging applications of TPs, highlighting promising trends and ideas that may encourage the widespread adoption of these materials. Whilst thermochromic inks and prints are available on the market, there is a notable gap in studies comparing these with TPs, which could pave the path for future research. Specifically, the review seeks to:

- i. Determine the state-of-the-art advancements in the applications of TPs in food packaging through SR;
- ii. Highlight organisations and geographical regions leading in introducing TPs for food packaging through PLA;
- iii. Identify challenges, limitations, and potential solutions in applying TPs for food packaging;
- iv. Recognize trends and potential future directions based on the current research path and patent activity;
- v. Understand the consumer perception and market demand for intelligent packaging solutions incorporating TPs.

2. Materials and Methods

2.1. Review Design

A rigorous and comprehensive systematic literature review and PLA were conducted following the Preferred Reporting Items for Systematic Review and Meta-Analysis (PRISMA) methodology. This provides a structured checklist and flow diagram to ensure the transparency and reproducibility

of the methods used [53]. A research synthesis was undertaken to identify and retrieve international evidence of using TPs in food packaging.

Systematic literature reviews are an essential aspect of work. They allow for understanding the scope and depth of the body of work already completed by evaluating pertinent literature [54]. In addition, they propose ways to advance the research by suggesting opportunities and solutions [55].

Finding all the patents pertinent to a topic is known as patent landscaping [56]. A PLA provides an overview of patenting activity, trends, and a factual foundation for a particular area. This PLA concentrated on the thermochromic food packaging area. PLAs allow for research to be conducted in order to see what is current in technological and industry terms. By generating a PLA, a graphical representation of the data is displayed [57]. PLAs are intended to answer important questions about which technologies are covered, which organisations own the patents, and which country they are held in. They also offer access to an extensive collection of technologically focused data. PLAs provide evidence of the emerging trends in technology, analyzing the exchange of knowledge and collaboration and comparing markets across nations and regions [58].

The results were appraised and synthesised to inform practice, policy and further research areas [59].

2.2. Definition of the Research Question

The two-element format strategy was used to frame the research question, "Do thermochromic polymers have potential applications in food packaging?". The key elements for P (population/phenomena) and O (outcomes) are shown in Table 1. The research question conformed to FINER (feasible, interesting, novel, ethical, and relevant) criteria [60].

Table 1. Population/phenomena – outcomes (PO) framework for the research question.

Format Strategy	Research Question
P (population/phenomena)	Thermochromic polymers
O (outcomes)	Potential application in food packaging

2.3. Eligibility Criteria

This review included journal articles concerning TPs' potential applications in food packaging since 2018. Items that focused on the application of thermochromic polymers in areas other than food packaging (e.g., textiles and medical devices) were excluded to keep the review focused and relevant. Studies not available in English or those that could not be accessed in full text were excluded if translation or access was not feasible within the scope of the review. Grey literature such as conference papers, reports, working papers, white papers, theses and dissertations were excluded from the review, as were editorials, letters, case reports, case series, and studies.

Patents related to TPs in food packaging (IPC: B65D1/00) were subsequently reviewed. Patents unrelated to the food packaging application of TP's were excluded from the review. No language restrictions were applied. Studies not available in English or those that could not be accessed in full text were excluded if translation or access was not feasible within the scope of the review. No date restrictions were applied to the patent search to get a comprehensive overview and identify any prior art.

2.4. Information Sources, Search Strategy and Study Selection

Three authors (CB, KD, AH) conducted a computerised database search on 7 November 2023 to detect all peer-reviewed articles containing data regarding using TPs in food packaging since 2018. The comprehensive research conducted to identify significant studies without constraints on the publication year, status, or language encompassed examining various sources. These included *Scopus, Science Direct, IEEE Xxplore®, PubMed®, SpringerLink, Wiley Online Library, ACS (American Chemical Society) Publications, Google Scholar and ProQuest.* The search strategy used in this review is described in Table 2.

Table 2. Materials science, engineering, chemical and biomedical database search strategy.

Database	Search Queries	Results
Scopus	(("thermochromic polymers" OR "thermochromic	35
	materials") AND "food packaging")	
Science Direct	(("thermochromic polymers" OR "thermochromic	15
	materials") AND "food packaging")	
IEEE Xplore®	(("thermochromic polymers" OR "thermochromic	100
	materials") AND "food packaging")	
PubMed®	(("thermochromic polymers" OR "thermochromic	1
	materials") AND "food packaging")	
SpringerLink	(("thermochromic polymers" OR "thermochromic	26
	materials") AND "food packaging")	
Wiley Online Library	(("thermochromic polymers" OR "thermochromic	7
	materials") AND "food packaging")	
ACS (American	(("thermochromic polymers" OR "thermochromic	6
Chemical Society)	materials") AND "food packaging")	
Publications		
Google Scholar	(("thermochromic polymers" OR "thermochromic	85
	materials") AND "food packaging")	
ProQuest	(("thermochromic polymers" OR "thermochromic	13
	materials") AND "food packaging")	
Total		288

Moreover, patent files on the utilisation of TP's in food packaging were identified through a systematic consultation of the following relevant databases and resources: *United States Patent and Trademark Office (USPTO) Database, Espacenet, Patentscope, and Canadian Intellectual Property Office (CIPO) Database.* The databases were consulted to identify patent files for using TPs in food packaging without restricting the timeframe applied.

Patent searches were conducted using International Patent Classification (IPC) with the code B65D (Rigid or semi-rigid containers having bodies formed in one piece; class hierarchy in Table 3). That was because each patent may submit several IPCs. These codes served the primary function of focusing the search and giving the authors an effective tool for researching and recovering patents. Table 4 describes the search strategy.

Table 3. International Patent Classification (IPC) class hierarchy.

Patent Classification	Title
В	Performing Operations; Transporting
Б	Transporting
65	Conveying; Packing; Storing; Handling Thin
63	or Filamentary Material
	Containers for storage or transport of articles
	or materials, e.g. bags, barrels, bottles, boxes,
D	cans, cartons, crates, drums, jars, tanks,
Б	hoppers, forwarding containers; accessories,
	closures, or fittings therefor; packaging
	elements; packages

Database	Search Queries	Results	
USPTO	(Thermochromic) AND (Polymer)	115	
	AND (Food) AND (Pack)		
Espacenet	(Thermochromic) AND (Polymer)	134	
	AND (Food) AND (Pack)		
Patentscope	(Thermochromic) AND (Polymer)	623	
	AND (Food) AND (Pack)		
CIPO	(Thermochromic) AND (Polymer)	50	
	AND (Food) AND (Pack)		
Total		022	

References were compiled and managed using a dedicated Reference Manager software (Mendeley v1.59.1), ensuring the removal of duplicates and irrelevant studies. Two researchers (CB and KD) independently selected and reviewed articles and patents, with a third author (AH) resolving any discrepancies. Further relevant studies were identified through a review of references within selected articles.

3. Results

3.1. Systematic Review

The flowchart (Figure 5) summarizes the selection process for articles. A total of 288 articles were initially recovered from the databases (Scopus, Science Direct, IEEE *Xplore*®, PubMed®, Springer Link, Wiley Online Library, ACS (American Chemical Society Publications), Google Scholar, and ProQuest); 63 articles were excluded due to duplicability, 184 articles were excluded because they did not correlate to TP's in food packaging. After removing duplicates and irrelevant articles, four papers were included in the final analysis. This lack of papers reflects the under-researched area TPs in food packaging, indicating a need for further research in this field. The selected studies are high-quality, well-conducted research that provides valuable insights and meaningful contributions to the review of the application of TPs in food packaging. The primary advantages identified include real-time temperature monitoring, enhanced food safety, and the potential to reduce food waste. Future research should focus on optimizing the cost-effectiveness of TPs and exploring their long-term stability under various environmental conditions.



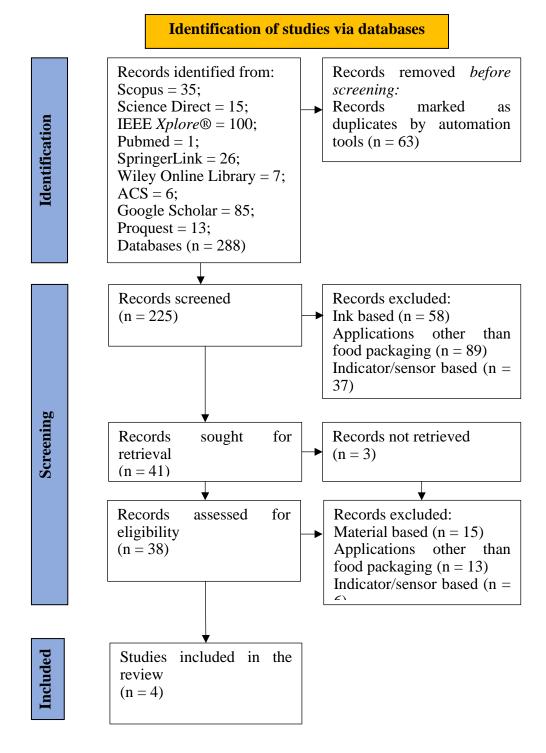


Figure 5. The selection process for articles.

A total of four papers were included in the analysis. This number reflects the under-researched area of TPs in food packaging, which has led to a need for more relevant studies. Due to the limited body of literature, there is a strong need for further research in this field due to the gap in existing knowledge. The quality of the studies included are high-quality, well-conducted studies that provide valuable insights and contribute meaningfully to the review. The PRISMA flow diagram for SRs, including searches of databases and registers, was the methodological technique used by the authors to present the study selection process visually. The criteria taken into account are shown in Figure 4. The flow diagram represents the three stages of an SR's information flow. It maps the quantity of identified, screened and included records. The identification phase allowed for the inclusion of

records identified through database searching, allowing for the removal of duplicates. Next, the remaining titles and abstracts were screened, and many records not to meet eligibility. After that, eligibility for full-text articles was evaluated, with more records being excluded due to inappropriate study design or outcome measures. The final included phase shows the number of studies included in the synthesis. Two reviewers (CB and KD) conducted the assessment separately, and prospective disagreements were discussed with a third researcher (AH).

The information in Table 5 was collated using predetermined data extraction forms from cited studies: article, author(s), year of publication, country, bibliographic source, technique, polymer, principal findings, and limitations of TPs acknowledged by the author(s).

Table 5. Enumeration of thermochromic polymers (TPs) used in food packaging with bibliographic sources.

sources.			
Article / Author(s) / Year of publication / Country	Source / Technique / Polymer	Principle Findings	Limitations of TPs acknowledged by the Authors
Green Thermochromic Materials: A Brief Review / [61] / 2022 / USA	Advanced Sustainable Systems / Extrusion / PBA, PS	This review discusses the phases, forms, and chemical structures of green thermochromic materials, which are highly sought after for temperature sensing applications. Most current thermochromic materials are nondegradable, toxic, and nongreen.	Toxicity. Non-degradable. Lack of research. UV susceptible.
Chromogenic Polymers and Their Packaging Applications: A Review / [23] / 2020 / South Korea / USA	Polymer Reviews / Extrusion / PVC, PVA	This review paper provides an overview of chromogenic polymers and their potential uses in packaging. It also discusses recent advances in overcoming common challenges and describes prospects, market trends, and academic research related to chromogenic polymers.	Weak color- changing capability. Susceptible to mechanical stress. Vulnerable to chemicals. Non- biodegradable, non-renewable, non-re-usable.
High-performance reversible thermochromic composite films with a wide thermochromic range and multiple colors based on micro/ nanoencapsulated phase change materials for temperature indicators / [62] / 2023 / China	Composites Science & Technology / Extrusion, blown film / PLA	Reversible thermochromic composite films made with thermochromic microcapsules and polylactic acid can potentially be temperature sensors. Their characteristics, including morphology, thermal, mechanical, thermochromic, and optical properties, and their resistance to thermal cycling were researched.	screw extrusion
Packaging Technology and Engineering Pharmaceutical,	· ·	This book covers the chemistry, physics, materials science, engineering, and therapeutic aspects of many different	Validated integrity, stability, and performance concerning

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Medical and Food	PET, PETE, HDPE,	packaging materials,	sterilization
Applications / [63] / 2020 /	LDPE	emphasizing the applicability of	f methods.
UK		various packaging science and	Toxicity.
		technology aspects.	Reduced shelf life.

The papers listed in Table 5 provide a comprehensive overview of thermochromic materials' properties and mechanisms, their green alternatives, and potential applications, including food packaging. Table 6 details the relevance of these papers to this paper's overall objective.

Table 6. Relevancy of selected articles to the research.

Article / Author(s) / Year of Publication / Country	Applicability to the Research Question
Green	Thermochromic Materials: Overview of types, chemical structures,
Thermochromic	mechanisms, and applications of thermochromic materials.
Materials:	Food Packaging: Emphasis on the need for non-toxic, biodegradable alternatives
A Brief Review [61]	to conventional thermochromic materials in food packaging.
	Temperature Sensing: Potential use of thermochromic materials for
	temperature monitoring in food packaging.
	Green Chemistry and Sustainability: Focus on developing and applying green
	thermochromic materials for sustainable packaging solutions.
Chromogenic	Overview of Chromogenic Polymers: Comprehensive overview of chromogenic
Polymers and Their	polymers, which include thermochromic materials.
Packaging	Applications in Packaging: Focus on thermochromic materials in packaging,
Applications: A	which provide visual indicators of temperature changes to signal spoilage or
Review [23]	quality changes in foods.
	Thermochromic Materials Focus: Discussion on types, mechanisms, and
	potential applications of thermochromic polymers in packaging contexts.
	Functionality in Packaging: Emphasis on enhancing packaging performance
	through freshness, temperature control, and safety indicators.
High-performance	Thermochromic Polymers Focus: Development of thermochromic composite
reversible	films using thermochromic microcapsules and PLA; color change in response to
thermochromic	temperature, relevant for food packaging.
composite films with	Application Potential in Food Packaging: They can be used as temperature
a wide	indicators in food packaging, providing visual cues for temperature history,
thermochromic range	spoilage, and storage conditions.
and multiple colors	Mechanical and Thermal Properties: Analysis of tensile strength, elongation at
based on micro/	break, thermal stability, and phase change enthalpy; crucial for assessing
nanoencapsulated	durability and stability.
phase change	Manufacturing Techniques: Using blown film extrusion for scalable production
materials for	is essential for commercial food packaging applications.
temperature	Environmental Considerations: The use of biodegradable PLA is crucial for
indicators [62]	minimizing environmental impact, as it supports sustainable and eco-friendly
	packaging solutions.
Packaging	Materials Science and Chemistry: Characteristics of polymeric materials,
Technology and	including thermochromic pigments.
Engineering	Food Packaging Applications: Focus on food-specific packaging requirement to
Pharmaceutical,	safeguard perishable goods' quality and safety.
Medical and Food	Environmental Impact and Sustainability: Discussion on recyclability and
Applications [63]	environmental impact of packaging materials,

The patent selection procedure is summarized in the flowchart (Figure 6). The PRISMA flow diagram for SRs, including searches of databases and registers, was the methodological technique used by the authors to present the study selection visually. The criteria taken into account are shown in Figure 6. The flow diagram represents the three stages of an SRs information flow. It maps the quantity of identified, screened and included records. The identification phase allowed for the inclusion of records identified through database searching, allowing for the removal of duplicates. Next, the remaining titles and abstracts were screened, causing many records to not meet eligibility. After that, eligibility for full-text articles was evaluated, with more records being excluded due to inappropriate study design or outcome measures. The final included phase shows the number of studies included in the synthesis. Two reviewers (CB and KD) conducted the assessment separately, and prospective disagreements were discussed with a third researcher (AH).

The identification stage initially resulted in 922 database patents (USPTO, Espacenet, Patentscope, and CIPO). Data clean-up and grouping were carried out on the data to correct errors/inconsistencies and group synonymous entries. This organisation was achieved manually using pivot tables, conditional formatting and group technology categories based on established classification systems. In total, 102 patents were eliminated due to duplicity.

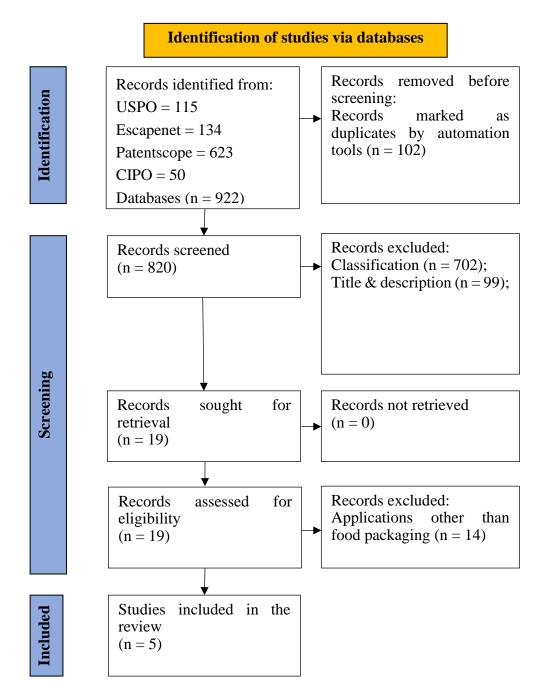


Figure 6. The selection process for patents.

In the screening stage, 702 remaining patents were disregarded because they were unrelated to the relevant classification (IPC: B65D1/00). After reading the titles and descriptions of the patents, 99 patents were eliminated based on their titles and descriptions. This resulted in 19 patents assessed for complete eligibility. Of the 19, 14 were removed due to being irrelevant. The remaining five results were tabulated using predefined data extraction forms. Due to stringent inclusion criteria being employed, numerous studies were excluded. The following information was organized from the included patents: title, publication number, publication kind, applicants, publication country, technique, polymer, and a summary of the invention (Table 7). The countries with the most relevant patent applications were the United States (three patents) and Japan (two patents).

Table 7. Patents included in the patent landscape analysis (PLA).

Title / Publication	Applicants /	Technique /	Invention Overview	Limitations of TPs
No./ Publication	Publication	Polymer		Acknowledged by
Kind	Country			the Author(s)
Advanced multi-	Segan	Injection	The invention integrates a	 Biocompatible, biodegradable,
element consumable-	Industries,	molding,	thermochromic masterbatch into	landfill
disposable products /	Inc,	extrusion /	a high-volume food packaging	
[64]	USA	PET, PP,	consumable using various	3. Stability.
/ 20180003565 /		PETE, PS,	polymer processing techniques	
B2		HDPE, PVC,	and polymer materials.	
		LDPE		
High-efficiency	Brighton	Blow	The invention relates to a	1. Embrittlement
polymeric sterilant	Development,	molding,	method for sterilizing a	post-
container assembly /	LLC,	extrusion /	$thermochromic\ film/fiber\ inside$	sterilization.
[65]	USA	POM	a sealed container, and	
/ 20180071419 / B2			irradiating between 1 - 200 kg	
			and then heated from 20 $^{\circ}\text{C}$ – 90	
			°C for a period.	
Modified silicone	PPG	Extrusion /	Coating compositions	1. Non-toxic.
coating composition /	Industries	Silicone,	comprising a thermochromic	2. Resistance to blemishes.
[66]	Ohio, Inc.	acrylic,	pigment applied to the	Diemones.
/ 20190112496 /	USA	latex, PE.	interior/exterior of the	
B2		PP. PET	packaging.	
Cooked state	TOYO	Extrusion,	The invention is a heat-sensitive	a) Reaction to
indicator material,	BOSEKI,	injection	material made of a polymer and	different forms of heating.
food packaging	Japan	molding /	a thermochromic dye. This	or reading.
material, cooked state		PE, PC, PA,	material changes color when	
indication method,		PAN, PAA,	exposed to a specific	
and heat cooking		PMMA,	temperature for a set time,	
method / [67] /		PAM, PAS,	serving as an indicator for	
WO2014/129405 / B2		PPO, PPS,	cooked food and packaging.	
		PVC, PS		
Container, device and	Takuya	Blow	This invention aims to prolong	1. Inaccuracy to
method for optimally	Asano,	molding,	the shelf life of food with a resin	temperature change.
preserving and	Japan	extrusion,	having a thermochromic	2. Recyclability.
providing beverage,		injection	function that changes color	3. Function
food, medicine,		molding /	depending on the temperature.	deteriorates.
commodities and		PP		4. Toxicity.
artistic cultural assets				
/ [68]				
/ JP2005138906A				

The patents listed in Table 7 provide a comprehensive overview of thermochromic materials' properties and mechanisms, their green alternatives, and potential applications, including food packaging. Table 8 details the relevance of this patents to this paper's overall objective.

Table 8. Relevancy of selected patents to the research.

Title	Applicability to the Research Question
Advanced	Thermochromic Applications: Thermochromic dyes and colorants are integrated into
multi-	consumables for temperature indication and quality monitoring.
element	Material Science and Chemistry: Exploring chemical compositions and additives,
consumable-	including thermochromic materials, enhancing functionality in food packaging.
disposable	Manufacturing and Production: Innovative processes, e.g. co-extrusion, for
products	producing thermopolymer-based packaging with thermochromic features.
[64]	Environmental Considerations: Use of bio-compatible, biodegradable, and landfill-
	degradable materials alongside thermochromic elements for sustainable packaging.
	Functional Features: Practical applications of thermochromic materials for visual
	indicators of temperature changes, enhancing freshness and safety monitoring.
High-	Thermochromic Materials: Incorporation of thermochromic materials into
efficiency	polymer fibers for visual temperature indicators relevant to food packaging.
polymeric	Advanced Polymeric Materials: Polyoxymethylene (POM) and copolymers are
sterilant	suitable for forming thin fibers and non-woven mats in packaging applications.
container	Environmental and Safety Considerations: Development of bio-compatible and
assembly	functional materials, including thermochromic properties, for sustainable packaging.
[65]	Manufacturing Techniques: Processes, e.g. blowing and gamma irradiation, for high-
	performance thermopolymer packaging materials with added features.
	Sterilization and Food Safety: Focus on low-heat sterilization processes with
	formaldehyde release, crucial for maintaining sterility and food safety.
Modified	Thermochromic Materials: Special effect compositions include pigments that
silicone	produce effects such as thermochromism.
coating	Advanced Polymeric Materials: Utilization of enhanced polymers relevant for
composition	protective and functional food packaging.
[66]	Environmental and Safety Considerations: Focus on BPA-free, formaldehyde-
	reduced compositions that provide safe and non-toxic coatings for food packaging.
	Manufacturing Techniques: Describes application techniques to food packaging.
	Sterilization and Food Safety: Coatings are designed to maintain integrity under
	various conditions, e.g. acidic foods, ensuring the safety and longevity of packaging.
Cooked state	Thermochromic Materials: Describes a polymer-dye system that irreversibly
indicator	changes color at specific temperatures, indicating a cooking state.
material, food	Polymer Composition: Utilizes polymers like PET and PETG for stability and
packaging	functionality in food packaging applications.
material, cooked	Application: Used in packaging materials to indicate proper cooking temperature
state indication	without additional tools.
method, and	

heat cooking	Environmental Considerations: Designed for food safety with non-toxic materials
method [67]	suitable for food contact.
	Functionality and Safety: Provides clear, irreversible color change to ensure proper
	cooking, enhancing consumer safety.
Container,	Thermochromic Polymers: Use of thermochromic materials for color change
device and	indicators in food packaging.
method for	Material Science: Development of containers with thermal insulation and
optimally	temperature indication properties.
preserving and	Innovative Design: Includes functional elements like heat-insulating materials and
providing	thermochromic labels.
beverage, food,	Production Techniques: Use of heat-shrinkable films and specialized inks for
medicine,	packaging applications.
commodities	Environmental Considerations: Focus on recyclable and eco-friendly materials in
and artistic	packaging.
cultural	
assets [68]	

4. Discussion

4.1. Overview

This comprehensive SR and PLR critically and unbiasedly examined the use of TMs in food packaging by scrutinizing academic articles and patents in this field of study. A commonality among the selected four articles and five patents chosen as relevant research compositions is their focus on TPs for food packaging applications. Improvements in intelligent packaging designed to convey information about the current state of the product temperatures are predicted to become a significant share of the food packaging market. The use of thermochromic inks is well-documented in the application of intelligent packaging. However, the introduction of thermochromic additives during the polymer processing manufacturing phase must be investigated.

The articles studied in-depth aid in understanding the current use of TPs in intelligent food packaging applications. The studies investigate the development of plastic food packaging (including a thermochromic pigment) that can change color in response to temperature changes. The studies found that intelligent packing systems for plastic food packaging applications are familiar. For example, literature referenced in Sadeghi et al. [23] as far back as 2014 discussed the use of TMs in intelligent food packaging applications and highlighted the potential benefits such as the perfectly chilled beverage and the onsite detection of unsafe food [69]; [70]. Overall, developing state-of-theart TPs in food packaging involves innovative packaging systems that can monitor and communicate information about temperature changes to consumers visually and intuitively.

The four chosen studies (Table 5) concentrate on TPs and their potential applications in packaging. Classifications, mechanisms, chemistry, recent progress and challenges related to thermochromic food packaging are discussed. Confirmation that thermochromic characteristics are currently added to sectors other than food packaging is acknowledged, but more research is needed to incorporate thermochromic additives into smart food packaging [61]. Recognition that a benefit of thermochromism is its ability to serve as an optical marker of the quality and freshness of food products [23]. While Cheng et al. (2023) acknowledged the potential for thermochromic films in applications such as packaging, they did not specifically mention food packaging. However, due to the high-quality, well-conducted study, the research obtained valuable insights and contributions

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into TPs in packaging. They also referenced ten papers on food packaging [62]. Other benefits of thermochromic polymeric food packaging materials are their flexibility and ease of modification [63].

4.2. Attractiveness of Thermochromic Polymers in Food Packaging

The selected studies highlight various TPs' unique properties for food packaging applications. They clearly understand the material's performance metrics, essential for assessing their suitability for different uses. Fundamental properties and quantitative values include:

- i. Transition temperature range.
- ii. Color variations and optical properties.
- iii. Thermal and mechanical properties.
- iv. Phase transition enthalpy.
- v. Durability

4.2.1. Transition Temperature Range

TPs are sensitive to temperature changes within a specific range, resulting in color changes and heat absorption release, making them valuable for high-performance applications [62]. Crosby (2022) highlights the behavior of Vanadium Dioxide (VO₂), which undergoes a phase transition at room temperature, changing from a brown-yellow color and can be modified to enhance transparency and alter its color properties. Liquid crystal materials, such as hydroxypropyl cellulose (HPC) hydrogels, exhibit thermotropic transitions, shifting from transparent at 20°C, translucent at 45°C, to opaque at 50°C - the film's luminescence changes from 86.1% at 20°C to 48.7% at 45°C. Thermochromic fiber with starch-graft-poly(butyl acrylate/styrene) transitions from purple below 31°C to pink above 31°C [61]. The addition of transition metal oxides such as titanium oxide (Ti₂O₃), vanadium dioxide (VO₂), vanadium (III) oxide (V₂O₃), vanadium (v) oxide (V₂O₅), Iron(II,III) oxide (Fe₃O₄), and beta'-Molybdenum Oxide (Mo₉O₂₆) can enable adjustable and reversible phase transition properties. For example, VO₂ can significantly reduce the transition temperature from 70°C to 25°C by incorporating tungsten ions, demonstrating its tunability for specific applications [23]. Liquid crystals (thermotropics) display various color changes depending on temperature [63].

4.2.2. Color Variation and Optical Properties

Specific compounds allow for a range of color changes. Examples include copper mercury iodide (Cu₂_22HgI₄), which transitions from red to brown; Indium(III) oxide, which changes from yellow to brown; and bis(dimethylammonium)tetrachloronickelate, which changes from green to yellow at 118°C [63]. TPs containing a mix of blue and yellow thermochromic microcapsules displayed varied colors, such as green, yellow, and colorless, while maintaining a light transmission of over 80% at 750 nm across all temperatures [62].

Based on structural modifications, conjugated polymers like polydiacetylene exhibit temperature-dependent color transitions, ranging from blue to red, pink, or magenta. Hydrogels, including poly(N-isopropyl acrylamide) (PNIPAM), demonstrate thermochromic and thermotropic behaviors, with reversible color changes driven by phase transitions. **Additionally, n**anohydrolgels modified with chitosan exhibit reversible thermochromism, transitioning from a loose, flexible coil above 37°C to aggregated molecules below 4 °C, creating UV-blocking properties, effectively preventing UV light transmission when the modules aggregate at low temperatures. PLA-Anthocyanidin complex shifts color from dark red at around 25°C to violet at around 45°C, with a thermotropic transition from cloudy to clear at 60°C [61].

For specific food packaging applications, irreversible color changes are desirable. Crystallized diacetylenic compounds, such as 2,4-hexadiyn-1,6-bis(alkylurea), can change color irreversibly based on environmental factors. The color-changing mechanism is linked to the diacetylenic monomer properties in the solvent after variations in temperature and time. Polydiacetylene (PDA), for instance, shows color changes from blue to red due to phase transitions induced by temperature variations [23].

4.2.3. Mechanical and Thermal Properties

TPs prepared with TCMs and PLAs demonstrated significant mechanical properties, notably achieving an approximately 536% increase in elongation at break compared to standard PLA films. This substantial improvement indicates that TPs are more flexible and resilient under stress, making them highly suitable for food packaging.

In addition to their mechanical strength, the thermal cycling durability was also notable, with the films maintaining excellent stability even after 60 thermal cycles [62]. This durability underscores the robustness of these materials, ensuring that they retain their functional properties despite repeated exposure to varying temperatures, which is essential for applications in fluctuating thermal environments.

The glass transition (Tg) and melting temperatures (Tm) of various TPs are critical for determining their usability in different applications. Differential scanning calorimetry (DSC) analysis can identify these thermal transitions, which are crucial for understanding the material's behavior under various thermal conditions. The degree of crystallinity, including the formation of crystallites or spherulites, significantly influences the mechanical properties of the polymers, such as opacity and brittleness, of the polymers. A higher degree of crystallinity typically results in increased brittleness and reduced transparency, impacting the material's overall application potential [63].

4.2.4. Phase Transition Enthalpy

Thermochromic materials, known as polymorphs, can exist in various crystalline forms, which significantly influence their thermal and optical properties. The specific crystalline structure of a TM determines its behavior under thermal stress, including the temperatures at which it transitions between different states. These polymorphs exhibit distinct transition temperatures and associated enthalpy changes (ΔH), as demonstrated by differential scanning calorimetry (DSC) data [63]. DSC analysis is a critical tool for understanding the thermal properties of TMs, as it measures the heat flow associated with phase transition.

For example, the heat absorbed during the melting process (Δ Hm) and the heat released during crystallization (Δ Hc) were measured across various samples. These measurements are essential for determining the material's thermal storage capacity and stability. One notable instance is thermochromic microcapsules (TCMB), which exhibited a Δ Hm of 126.29 J/g and a Δ Hc of 123.14 J/g. These values suggest that TCMBs possess significant thermal storage capabilities, crucial for their performance in applications requiring precise thermal management [62]. The ability of these materials to store and release heat efficiently makes them highly valuable in applications such as smart packaging, where maintaining specific temperature ranges is critical for preserving product quality.

4.2.5. Durability

Films produced have demonstrated excellent thermal cycling durability, retaining thermochromic and latent heat storage-release properties after 60 thermal cycles. This stability and repeatability in color-changing ability are essential for successfully implementing TMs in practical food packaging applications where consistency and reliability are paramount [62].

Crosby (2022) discusses how the stability of materials such as PDA (polydiacetylene) enables the material to return to its original structure after multiple heating and cooling cycles, which is essential for practical applications where durability is a concern. It is also noted that the microencapsulation of TPs can protect the chromophore from environmental degradation, including UV exposure and oxidations, thereby enhancing the durability of TPs. The hydroxypropyl cellulose (HPC) films were subjected to durability testing under UV light to simulate outdoor conditions. The films demonstrated good reversibility and durability over 10 days, indicating potential for longer-term applications. The color change of thermochromic fiber with starch-graft-poly(butyl acrylate/styrene) is repeatable over 100 cycles, further illustrating the robustness and practicality of TPs in the food

packaging industry, where materials must withstand repeated use without compromising their performance [61].

4.3. Challenges and Considerations for Thermochromic Polymers in Food Packaging

Based on the limitations of TPs acknowledged by the author(s) in the selected research articles and patents, a summary of the central gap of existing knowledge presented by the SR and PLA include:

- i. Advanced manufacturing processes;
- ii. Functional performance;
- iii. Longevity and stability;
- Environmental impact and sustainability;
- v. Toxicological safety.

4.3.1. Advanced Manufacturing Processes

Although the studies examined reference high costs associated with TPs [71], they lacked detailed explanations of the specific adjustments needed for standard production processes. Integrating TPs into existing machinery lines may require significant modifications, such as changes in equipment settings, material handling procedures, quality control measures and waste recycling. These modifications to production and manufacturing processes are critical for ensuring that the enhanced properties of TPs are consistently achieved without compromising the efficiency or reliability of the production process. However, the literature needs to elaborate on what those adjustments may entail or how these modifications should be implemented, leaving a gap in practical knowledge for manufacturers.

Additives can improve or deteriorate a final product's performance, appearance, or stability. Different concentrations of additives could affect the integrity of the base material. [72]. Cheng et al. (2023) was the only previous study that discusses the preparation of thermochromic films, in this case, using thermochromic microcapsules and polymers and the extrusion blown film method. The influence of different mass ratios of microcapsules and polymers was discussed, along with the potential for applications in areas such as packaging. Queries surrounding the functionality of TPs post-twin-screw extrusion due to high shear forces were also expressed. Studies versus single-screw versus twin screw are warranted [62].

4.3.2. Functional Performance

Functionality deterioration concerns new innovative polymer materials [68]. In specific food packaging scenarios, TMs' desire to have excellent temperature sensitivity is crucial [73]. However, during this comprehensive SR, the impact of repeated subjection to multiple cycles of temperature change was found to be under-researched. This raised concerns about the immediate functionality of TPs, particularly in maintaining consistent color-changing properties.

Chemical sensitivity is a consideration when assessing the functionality of TPs in different food packaging environments [62]. Exposure to certain chemicals could alter the polymer structure, affecting its color-changing properties or leading to degradation. Thermochromic food packaging chemicals could impact the color-changing abilities of TPs.

The mechanical integrity of TPS in food packaging allows the packaging to withstand stress, thereby preventing any physical product deterioration. Questions of the susceptibility of TPs to mechanical stress are evident [23]. Additionally, the appearance of plastic food packaging is essential for creating a positive customer experience and ensuring the product's marketability - resistance of TMs to blemishes in an area for review [66].

TPs operate by changing color in response to a thermal stimulus. Nonetheless, the extent to which TPs exhibit a uniform response to distinct heating modalities, such as water or air exposure, remains presently undisclosed [67]. In addition, inaccuracies in temperature changes require further study [68].

4.3.3. Longevity and Stability

Discussions stating that plastics are affected by factors, such as yellowing observed in PS, polyolefins, PU and PC, suggest that similar degradation could affect TPs. Concerns about the long-term reliability of TMs, particularly under UV exposure and repeated temperature cycles, highlight issues like UV susceptibility and weak color-changing capabilities [23].

Stability refers to the ability of plastic food packaging to maintain its physical and chemical properties over time. This stability includes resisting changes in color, texture, structure, and overall integrity. While TPs offer unique features, they also introduce an additional layer of complexity to food packaging. The reduced shelf life of TPs incorporated with polymers is something to consider [63]. The patents scrutinized as part of this review also reference stability limitations [64].

Food packaging is subjected to various sterilization systems to inactivate viable microorganisms that may pose a health risk to food consumers [74]. High temperatures may be utilized depending on the sterilization method [75]. The studied documents have references to the effects of high-temperature sterilization. Much of the discussion is centered around the impact of sterilization on the mechanical properties of the packaging materials, e.g. thickness, weld integrity, and material. Accelerated ageing studies have shown that certain polymers may experience some type and level of degradation over time, particularly those subjected to specific environments, e.g. temperature. This results in the alteration of their (mechanical, optical, thermal, and functional) properties, thus altering their performance and shortening their life cycle [76] [77]. However, the effects of different forms of sterilization on the optical characteristics of pigmented TPs, e.g., discoloration, are currently unresearched [63]. Embrittlement post-sterilization is acknowledged in the patent review as requiring scrutiny [65].

The performance and longevity of TPs in various environmental conditions, including exposure to UV radiation and temperature fluctuations, require further rigorous testing to address their impact on the stability, recyclability, and overall environmental impact [78].

4.3.3. Environmental Impact and Sustainability

In 2022, Crosby and Netravali proposed the development of green TMs as an alternative to toxic or non-degradable materials commonly used in thermochromic systems. The article emphasizes the pressing need for more environmentally friendly options while summarizing the state of research in this new field [61]. Since this field is in its infancy, there are gaps in the current state of research that have yet to be investigated, such as the risk of toxic, hazardous, or non-degradable components in food packaging addressing the potential sustainability of TPs, such as their biodegradability or recyclability, is a welcome addition.

Providing clear evidence of bio-compatibility, biodegradability, composability, and landfill-degradable properties would align with the growing consumer and regulatory demand for reducing environmental impact. With its vast reach and influence, the food packaging industry holds immense potential for driving improvements in environmental sustainability and product efficacy. However, this study did not uncover evidence regarding potential technological barriers to the incorporation of TMs. For instance, existing recycling facilities might not have the capability or need to sort and recycle these products. Downcycling of products into less valuable items may need to be considered. The examined patents address biocompatibility, biodegradability, landfill degradability [64], and recyclability [68], recognizing them as potential limitations of TMs that must be addressed.

4.2.4. Toxicological Safety

Concerns related to the toxic effects of substances on human health from the migration of substances into packaging are increasing due to the chemical composition of polymers [79]. The integration of TPs food packaging introduces complexities, warranting careful consideration to mitigate cytotoxicity risks as substances may leach into food under various conditions, such as changes in temperature, pH, or fat content, e.g. VCM, a plasticizer from PVC-based heated have been studied due to its slight toxicity and well-documented teratogenic, mutagenic, and carcinogenic

effects. Leuco dyes, essential for TPs, contain toxic compounds like Bisphenol A, are associated with neurotoxicity, immunotoxicity, and endocrine disruption.

Crosby (2022) highlights Seeboth et al.'s 2003 development of a nontoxic, thermochromic polymer-dye complex using amorphous PLA and anthcyanidin dye. This complex exhibited reversible color and thermotropic transitions, changing from dark red at room temperature (25°C) to violet at 45°C, then cloudy at 45°C to clear at 60°C [61].

Polymers in food-related applications must comply with stringent regulatory standards set by agencies such as the U.S. Food and Drug Administration (FDA) and the European Food Safety Authority (ESPA) to minimize health risks.

The EU's Directive 2004/37/EC (Carcinogens and Mutagens Directive - CMD), is designed to protect workers from exposure to carcinogens or mutagens. Migration between packaging and food is typically measured using high-performance liquid chromatography (HPLC) due to its precision in detecting low concentrations. Toxic polymers have large implications for the safe disposal/recycling of food packaging [63].

A commonality among the reviewed patents included the concern related to the toxicity of TPs materials in food packaging, For example, PVC found in clear food packaging can release toxic side compounds, and polystyrene (PS), which can be found in meat trays, can leach potential toxic compounds into food [64]; [66] [68].

5. Summary and Future Prospective

The potential benefits of intelligent packaging include integrating functionality into the food packaging interface. Integrating TPs into food packaging offers significant potential to enhance food quality and safety monitoring. The unique color-changing properties of TPs allow for real-time temperature monitoring, which is crucial for maintaining the integrity of perishable foods. However, in addition to the concerns surrounding the use of TPs in food packaging, in today's world, the environmental impact of food packaging is also a significant concern, particularly regarding the ecological effects of these materials, including the generation of microplastics and issues related to biodegradability and recyclability.

Future research should focus on several key areas to address the challenges discussed in Section 4.3 and align with the global Sustainability Development Goals (SDGs),.

Economic and cost considerations.

The economic feasibility of TPs requires a full assessment encompassing factors such as the cost of production, scalability, and market reception. Research should investigate strategies to decrease manufacturing expenses, such as employing cost-effective raw materials or implementing more streamlined production methods [80]. There needs to be a discussion on the manufacturability of the color change using various polymer processing techniques. Further consideration should be made in determining their integrity and stability (UV, shelf life, temperature sensitivity, chemical resistance, repeatability) before this material can be utilized in applications such as food packaging since stability is an essential attribute of food packaging.

b. Sustainability and Life Cycle Assessment (LCA)

Comprehensive life cycle assessments (LCAs) are essential to fully understand the total environmental impact of TPs throughout their entire lifecycle - from raw material extraction to their eventual disposal, often referred to as a "cradle to grave" analysis. LCAs provide a detailed evaluation of each stage in the lifecycle and the potential for recycling, composting or other end-of-life scenarios. These assessments are vital for identifying TPs environmental footprint and comparing them to alternative materials used in food packaging [81].

c. Development of biodegradable alternatives.

The environmental impact of food packaging is a significant concern in today's world. Addressing the potential sustainability of TPs, such as their biodegradability or recyclability, would be a relevant addition. Given the growing concern over microplastics and the unsustainability of synthetic polymers, there is an urgent imperative to advance the development of biodegradable TPs. Polymers such as chitosan polylactic acid (PLA) and other bio-based materials present promising

alternatives to synthetic polymers in thermoplastic applications, offering comparable functional properties with enhanced environmental sustainability benefits [61].

d. Regulatory compliance and safety

As TPs are used in food packaging, they must comply with rigorous regulatory standards to ensure they do not pose health risks to humans. The European Union's Regulation (EU) No 10/2011 includes a list of authorized substances for manufacturing plastic materials in contact with food [82]. This regulation delineates precise requirements regarding plastic materials' safety, purity, and migration limits, ensuring that they do not present any risk to human health when utilized in food packaging. European Commission to regulate materials in food contact materials (FCMs) [83]. Future research should include toxicity assessments and migration studies to ensure that the materials used are safe for direct food contact

e. Aligment with SDGs

With the ongoing increase in global plastic production, there is a corresponding rise in national and international commitments to mitigate (micro)plastic pollution. The UN SDGs are a universal call to action to end poverty, protect the planet, and ensure that everyone enjoys peace and prosperity [84]. There are 17 universal goals for 2030 for national, regional and global sustainable development [85]. The development of sustainable TPs aligns with several SDGs, particularly those related to ensuring sustainable consumption and production patterns (SDG 12), taking urgent action to combat climate change and its impacts (SDG13) and conservation and sustainability use of the oceans, seas, and marine resources for sustainable development [86].

6. Conclusion

Based on the current literature and patent trends, thermochromic polymers' (TPs) distinctive characteristics make them extremely promising for the food packaging sector. These materials can improve food quality, sustainability, and safety by providing real-time temperature monitoring. However, further research is needed to explore and exploit their benefits in food packaging applications fully. By answering these questions, the review will define the current state of TPs in food packaging and provide a roadmap for future research and development endeavors in this domain.

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