Luminescence anti-counterfeiting: From elementary to advanced

Xiaowei Yu1 | Hongyue Zhang1,2 | Jihong Yu1,2

1 State Key Laboratory of Inorganic Synthesis and Preparative Chemistry, College of Chemistry, Jilin University, Changchun, P. R. China
2 International Center of Future Science, Jilin University, Changchun, P. R. China

Correspondence
Jihong Yu, State Key Laboratory of Inorganic Synthesis and Preparative Chemistry, College of Chemistry, Jilin University, 2699 Qianjin Street, Changchun 130012, P. R. China.
Email: jihong@jlu.edu.cn

Funding information
National Natural Science Foundation of China, Grant/Award Numbers: 21835002, 21621001; 111 Project, Grant/Award Number: B17020

Abstract
Luminescence anti-counterfeiting derives from the easily changeable luminescence behaviors of luminescence materials under the regulation of various external stimuli (such as excitation light, chemical reagent, heat, and mechanical force, etc.) and luminescence lifetime, which plays an important role in preventing forgery of currency, artworks, and product brands. According to the numbers of changes of anti-counterfeiting labels under various regulation conditions, luminescence anti-counterfeiting can be classified into three levels from elementary to advanced: single-level anti-counterfeiting, double-level anti-counterfeiting, and multilevel anti-counterfeiting. In this review, the recent achievements in luminescence anti-counterfeiting are summarized, and the regulation of various factors to anti-counterfeiting labels is discussed. Finally, existing problems, future challenges, and possible development directions are proposed in order to realize facile, quick, low-cost, environmentally friendly, and difficult-to-replicate advanced luminescence anti-counterfeiting.

KEYWORDS
anti-counterfeiting, down conversion, external stimuli, lifetime, luminescence, up conversion

1 | INTRODUCTION

Counterfeiting phenomenon widely exists in all around the world.[1-5] Though many efforts, such as issuing laws, have been made by governments and businesses to prevent counterfeiting, plenty of counterfeiting products in food, medicine, apparel, and electronic fields still spread over our daily life, which seriously influences the property safety, society stability, and even threats human health.[6-10] According to the previous reports, the global economic losses caused by counterfeiting may reach to 1.82 trillion USD in 2020.[11] Therefore, developing powerful anti-counterfeiting technologies is highly demanded.

To prevent counterfeiting, various anti-counterfeiting strategies have been explored, including watermarks, holograms, luminescence, barcodes, and two-dimensional codes.[12-14] However, traditional anti-counterfeiting strategies, such as watermarks, barcodes, and two-dimensional codes, and so on, are easily to be copied and difficult to effectively prevent counterfeiting. Among these anti-counterfeiting strategies, luminescence anti-counterfeiting has attracted growing attentions because of excellent optical properties of luminescence materials, such as high emission intensity, multiple emission colors, long emission lifetime, and various emission modes including photoluminescence, chemiluminescence, and mechanoluminescence.[12,15] At present, the most mature luminescence anti-counterfeiting technology is based on fluorescent anti-counterfeiting labels, which has been widely used in currencies around the world.[3,8] However, facing rampant counterfeiting phenomena, simple fluorescence anti-counterfeiting labels already cannot meet anti-counterfeiting demands. Therefore, advanced luminescence anti-counterfeiting strategies need to be constantly explored.

Carbon dots (CDs), semiconductor quantum dots, perovskites, organic dyes, and Ln3+-doped nanoparticles are all outstanding luminescence materials, and more importantly, their luminescence can be excited, quenched, or tuned by external stimuli such as light, heat, chemical reagents, and mechanical force. For example, the green fluorescence of CsPbBr3 perovskite can be generated by ultraviolet (UV) excitation and can also be quenched by water treatment,[16] while the luminescence color of some Ln3+-doped nanoparticles can be tuned by changing excitation wavelength.[17] Such stimulus-responsive properties of these materials make them become the ideal materials in advanced luminescence anti-counterfeiting. Apart from external stimuli, luminescence lifetime as an intrinsic characteristic of luminescence materials can also induce the luminescence to change from fluorescence to afterglow. Generally, according to the
numbers of changes of anti-counterfeiting labels under various regulation conditions, such as excitation light, luminescence lifetime, heat stimuli, mechanical stimuli, rotation stimuli, and chemical regents, and so on, luminescence anti-counterfeiting can be classified into three levels from elementary to advanced: single-level anti-counterfeiting, double-level anti-counterfeiting, and multilevel anti-counterfeiting (Figure 1). Significantly, the fast development of multilevel luminescence anti-counterfeiting strategies make them become the promising next-generation anti-counterfeiting technologies that can be applied into market.

Undoubtedly, many reviews involving luminescence anti-counterfeiting have been reported, but they are all mainly focused on the anti-counterfeiting materials, anti-counterfeiting printing technologies, or the studies before 2016.[3,8,18] The summarization about anti-counterfeiting level associated with regulation factors has not been reported. In this review, we classify the luminescence anti-counterfeiting strategies into three types according to the anti-counterfeiting level: single-level anti-counterfeiting, double-level anti-counterfeiting, and multilevel anti-counterfeiting. We present the recent developments in luminescence anti-counterfeiting by discussing the regulation of various factors to luminescence behaviors of anti-counterfeiting labels, and analyze existing problems and future challenges in luminescence anti-counterfeiting and point out the future developing perspectives.

2 | SINGLE-LEVEL LUMINESCENCE ANTI-COUNTERFEITING

Single-level luminescence anti-counterfeiting is the simplest luminescence anti-counterfeiting mode, whose anti-counterfeiting labels change only once under the regulation of external stimuli due to the single fluorescence characteristics of materials. It has been widely applied into various currencies and certificates.[3] However, many simple single-level luminescence anti-counterfeiting technologies are easily to be copied and counterfeited. Therefore, the diversity of single-level luminescence anti-counterfeiting materials needs to be continuously increased to avoid forgery. Recently, more and more materials with different luminescence modes have been applied into single-level luminescence anti-counterfeiting to improve the ability of single-level luminescence anti-counterfeiting and increase the difficulty of forgery.[19–34] According to the difference of luminescence modes, the single-level luminescence anti-counterfeiting can be divided into down-conversion (DC) photoluminescence anti-counterfeiting,[19,21–27,29,31] up-conversion (UC) photoluminescence anti-counterfeiting,[20,28,30,32,34] and chemiluminescence anti-counterfeiting.[33]

2.1 | DC photoluminescence anti-counterfeiting

DC photoluminescence anti-counterfeiting is realized by absorbing one high-energy photon and releasing one low-energy photon.[35] At present, the materials used in single-level DC photoluminescence anti-counterfeiting include organic luminogens,[19] graphene quantum dots,[21] Ln-carbon quantum dots,[22] cellulose nanofibrils/CdTe quantum dots,[23] carbon and oxygen co-doped hexagonal boron nitride,[24] nitrogen-doped carbon dots,[25] carbon dots/Ln-MOFs hybrids,[26] Ln-metal organic frameworks (Ln-MOFs),[29] Th3+/Eu3+-grafted melamine cyanurate,[31] and SrAl2O4:Eu2+.[30] As shown in Figure 2A, Gao et al.[36] prepared SrAl2O4:Eu2+(SAOE)@TPU films with DC photoluminescence by mixing and co-electrospinning SAOE phosphors and film-forming agent polyethylene-based thermoplastic polyurethane (TPU). The authors cut the films into shape of letters “SUST” and encapsulated them in transparent bases to prepare an anti-counterfeiting label that was almost invisible under sunlight, but emitted low-energy green fluorescence after absorbing high-energy UVlight. This work adopts transparent films to prepare an anti-counterfeiting label other than relies on traditional ink printing, which provides a facile route for preparing luminescence anti-counterfeiting labels. In addition, Feng et al.[19] adopted a kind of light-mediated ink-free screen-printing technology to prepare a fluorescence anti-counterfeiting label that was colorless under daylight, but displayed blue fluorescence under UV irradiation. This anti-counterfeiting strategy was in favor of designing more complex anti-counterfeiting patterns and decreasing cost by replacing expensive inks with UV lights. Though the single-level DC photoluminescence anti-counterfeiting is easily realized due to the diversity of DC photoluminescence materials and simpleness of anti-counterfeiting process, this anti-counterfeiting strategy is also more easily to be copied and forged, thus limiting the development of this anti-counterfeiting strategy.

2.2 | UC photoluminescence anti-counterfeiting

UC photoluminescence anti-counterfeiting is realized by absorbing two or more low-energy photons and releasing one high-energy photon.[35] Compared with DC luminescence, UC luminescence possesses many advantages, such as mild excitation environment, various luminescence colors,
sharp emission bands, and long fluorescence lifetime.\cite{30,32}

Recently, Xie et al.\cite{30} designed a UC photoluminescence anti-counterfeiting label with the mixture of NaYF₄:Ln³⁺ hollow microtubes (UCHMs), ethanol, deionized water, and glycerol as ink, in which the UCHMs were prepared by hydrothermally treating precursor solution (Figure 2B). The anti-counterfeiting label was invisible under daylight, but could emit high-energy green fluorescence under the excitation of low-energy 980 nm laser. Moreover, the authors also proved the UC emission color of ink could be tuned by changing the composition of lanthanide. Therefore, the anti-counterfeiting label co-printed by these inks with different emission colors displayed multicolor luminescence under the excitation light, which greatly improved the safety performance of luminescence anti-counterfeiting. However, most of the materials used in single-level UC photoluminescence anti-counterfeiting involve expensive rare earth metal ions with high toxicity, which not only increases preparation cost of anti-counterfeiting label but also influences health of human.

2.3 | Chemiluminescence anti-counterfeiting

Besides the photoluminescence anti-counterfeiting, chemiluminescence anti-counterfeiting is also an excellent luminescence anti-counterfeiting strategy, whose luminescence is induced by chemical reagents and no excitation light source is required. Zuo et al.\cite{33} prepared a kind of water-soluble conjugated polymers (CPs) nanoparticles by supramolecular host–guest assembly strategy, in which the bis(2,4,5-trichloro-6-(pentyloxycarbonyl)phenyl) oxalate (CPPO) was doped into the CPs nanoparticles as energy donor and chemiluminescence substrate (Figure 2C). When CPPO-doped CPs encountered H₂O₂, the CPPO would be oxidized to high-energy intermediate 1,2-dioxetanediol that induced the generation of excited nanoparticles and triggered chemiluminescence. Moreover, the color of chemiluminescence could be regulated by adopting different guest molecules. Based on the unique luminescence properties of CPPO-doped CPs, the materials with different chemiluminescence colors were used as inks to write letters “RGB” as anti-counterfeiting label on paper. The anti-counterfeiting label was almost invisible under sunlight, but emitted bright multicolor fluorescence after treated by H₂O₂ in dark environment. Significantly, the anti-counterfeiting label prepared by chemiluminescence materials avoided the overreliance on excitation light source, which provided a novel idea for preparing luminescence anti-counterfeiting labels. However, the weak chemiluminescence intensity and uncommon chemical reagents in our daily life, such as H₂O₂, make the actual application of this anti-counterfeiting strategy become a challenge.

3 | DOUBLE-LEVEL LUMINESCEENCE ANTI-COUNTERFEITING

Compared with single-level luminescence anti-counterfeiting, double-level luminescence anti-counterfeiting is a more advanced luminescence anti-counterfeiting mode, in which the anti-counterfeiting labels will change twice under the regulation of external stimuli and luminescence lifetime, substantially promoting anti-counterfeiting level. Based on the category of regulation factors, the double-level luminescence anti-counterfeiting can be divided into three types: (1) regulated by excitation light;\cite{17,37–44} (2) co-regulated by excitation light and luminescence lifetime;\cite{45–58} (3) co-regulated by excitation light and...
3.1 Regulated by excitation light

Light is the most common stimuli source to trigger multiple changes of luminescence anti-counterfeiting labels, which includes various UV lights, visible lights, and infrared (IR) lights with different wavelengths and powers. Usually, two kinds of excitation lights are taken together to realize double-level luminescence anti-counterfeiting. Recently, Chen et al.\textsuperscript{[17]} developed a kind of anti-counterfeiting ink with excitation wavelength-dependent luminescence characteristic by mixing Ln\textsuperscript{3+}-doped NaLuF\textsubscript{4}/Y\textsubscript{2}O\textsubscript{3} composites and polyvinyl alcohol (PVA) aqueous solution, in which the Ln\textsuperscript{3+}-doped NaLuF\textsubscript{4}/Y\textsubscript{2}O\textsubscript{3} composites were synthesized by a hydrothermal process (Figure 3A). The anti-counterfeiting label screen-printed by anti-counterfeiting ink was hardly visible under daylight, but emitted red fluorescence under 254 nm excitation, and green or blue fluorescence under 980 nm laser. In addition to the excitation wavelength-dependent luminescence, some materials also possess excitation power-dependent luminescence characteristic. As shown in Figure 3B, Wang et al.\textsuperscript{[43]} painted a flower pattern on a Chinese Dehua porcelain with Er:KYb\textsubscript{2}F\textsubscript{7} NCs@glass as luminescence paint. When the pattern was exposed to a low-power laser (60 mW), the flower displayed red luminescence, while under the excitation of a high-power laser (250 mW), the red luminescence of the flower changed to yellow luminescence, which can be attributed to strong laser-induced photothermal effect leading to greatly increased electron population in the Er\textsuperscript{3+}^2H\textsubscript{11/2} excited state via laser-induced thermal activation from thermally coupled Er\textsuperscript{3+}^4S\textsubscript{3/2} state. Such dual-responsive properties to two kinds of excitation lights endowed the materials with double-level luminescence anti-counterfeiting ability.

3.2 Co-regulated by excitation light and luminescence lifetime

Luminescence lifetime is an intrinsic temporal characteristic of luminescence materials. Compared with traditional luminescence anti-counterfeiting based on luminescence intensity and color, luminescence anti-counterfeiting based on lifetime can effectively avoid luminescence overlap, background interference, and improve anti-counterfeiting ability, providing a new dimension for luminescence anti-counterfeiting.\textsuperscript{[62]} As shown in Figure 3C, Kalytchuk et al.\textsuperscript{[50]} adopted two kinds of CDs-based materials (CDs-f and CDs-s) with identical emission color (blue) but different fluorescence lifetimes (4.4 ns and 6.1 ns) as anti-counterfeiting inks to prepare anti-counterfeiting tags. The anti-counterfeiting tags displayed uniform blue fluorescence under the excitation of 365 nm UV light, but became a pattern of a character “R” under the detection of fluorescence lifetime imaging (FLI) system due to the difference of fluorescence lifetime other factors (e.g., heat, chemical reagents, and mechanical force).\textsuperscript{[59–61]}
lifetime of the two kinds of anti-counterfeiting inks. This work provided a new conceptual anti-counterfeiting strategy regulated by luminescence lifetime. Note that the short luminescence lifetime of these luminescence materials results in needing complicated FLI system to capture lifetime-regulated imaging process, which limits the development of this luminescence anti-counterfeiting technology. Therefore, the materials with long luminescence lifetime are highly desired to be developed.

Generally, long afterglow materials can avoid the use of FLI system, and are more suitable to be utilized in lifetime-regulated luminescence anti-counterfeiting.[45–49] Lin’s group developed a kind of pure CDs-based long afterglow materials with dual-emissive property of fluorescence and room temperature phosphorescence (RTP) by a facile and quick microwave-assisted process.[49] The aqueous solution of CDs was directly used as anti-counterfeiting ink to print the portrait of Chairman Mao on a banknote as anti-counterfeiting label. After complete drying, the anti-counterfeiting label was colorless under ambient conditions. Under the stimuli of UV light, the anti-counterfeiting label displayed bright blue fluorescence, while after the UV light was turned off, the blue fluorescence immediately turned to green RTP. However, the stability of bare CDs is poor, so that their luminescence is easily to be quenched after exposed to ambient conditions for a long time. Notably, Yu’s group confined CDs in zeolitic or open-framework matrices by a solvothermal or hydrothermal synthesis method and developed a kind of CDs-based composites with dual emissions of fluorescence and thermally activated delayed fluorescence (TADF) or RTP.[52,57] As displayed in Figure 3D, the anti-counterfeiting pattern co-painted by CDs@zeolite composites and benzil displayed dual-color fluorescence under the stimuli of UV light, including blue fluorescence of petals and green fluorescence of leaves, while after the UV light was turned off, the green fluorescence of leaves vanished, and the blue fluorescence of petals turned to naked-eye visible blue TADF. The whole process was visible to naked eyes and did not rely on the FLI system, which was proved to be a more effective and facile luminescence anti-counterfeiting mode compared with the above-mentioned luminescence anti-counterfeiting co-regulated by excitation light and short fluorescence lifetime. In addition to the CDs-based long afterglow materials, many luminescence materials with naked-eye visible afterglow emission can also realize the lifetime-regulated luminescence anti-counterfeiting, such as organic phosphors,[46] dye-encapsulated metal-organic frameworks,[53] graphene quantum dots,[45] and ordered thin films.[47] The diversity of long afterglow materials greatly promotes the development of lifetime-regulated luminescence anti-counterfeiting technology.

### 3.3 Co-regulated by excitation light and other factors

Apart from luminescence lifetime, mechanical force, chemical reagents, and heat are the main external stimuli factors to trigger or change the luminescence of anti-counterfeiting labels.[59–61] As shown in Figure 4A, Wu et al.[60] designed a luminescence anti-counterfeiting label by embedding “ML” pattern prepared by solidified Sr3Al2O6:Eu3+ (SAOE)/polydimethylsiloxane (PDMS) composites film into PDMS/TiO2 matrix, in which the white TiO2 powder played a role of background to make sure the embedded “ML” pattern invisible under daylight or dark. Under the radiation of UV light, the “ML” pattern displayed bright red fluorescence. Interestingly, the “ML” pattern could also emit intense red fluorescence after undergoing a stretch stimulus, due to the characteristic emission of Eu3+ induced by tunneling transfer of carriers from traps to Eu3+-O2- charge transfer band (CTB) and the transfer of electrons in CTB to excited energy levels of Eu3+. The fluorescence spectra in Figure 4A proved the two kinds of luminescence. Such dual-responsive luminescence properties to photon stimuli and stretch stimuli endowed the materials with double-level luminescence anti-counterfeiting ability.

In addition to the mechanical stimuli-induced luminescence, the luminescence of some materials is responsive to the stimuli of chemical reagents. For instance, Yuan et al.[61] prepared an anti-counterfeiting label on a banknote with the mixture of B-doped CDs (B-CDs) and PVA solution as ink. Under the excitation of 365 nm UV light, the anti-counterfeiting label displayed pink fluorescence, but emitted blue fluorescence after treated by acid, which derived from acid-induced formation of metastable structure in CDs producing a new energy level. However, due to the reversibility of the metastable structure, blue fluorescence of the anti-counterfeiting label treated by HCl could be restored to pink fluorescence after re-treated by alkali. The double changes of anti-counterfeiting label induced by excitation light and chemical reagents realized the double-level luminescence anti-counterfeiting.

Furthermore, heat is also an important stimuli factor in luminescence anti-counterfeiting owing to its quenching or enhancing effects on luminescence.[59,63] As proved in Figure 4C, Lei et al.[59] painted characters “Er” and “Ho” on paper by the inks prepared by hydrophilic Yb/Er:Na3ZrF7 and Yb/Ho:Na3ZrF7 nanocrystals. At room temperature, the characters “Er” and “Ho” emitted weaker red and yellow fluorescence under 980 nm laser, respectively, while when the temperature was increased to 413 K, the luminescence intensity of the two characters was obviously enhanced due to the released electrons (RE) from defect state (D) participating in the UC process. The dual-emissive characteristic of weak fluorescence at room temperature and thermal-induced strong fluorescence demonstrates these materials can realize the double-level luminescence anti-counterfeiting. However, the anti-counterfeiting strategies based on emission intensity are vulnerable to background interference, and some subtle changes of emission intensity cannot be observed by naked eyes, needing expensive instruments to detect, which limits the development of such anti-counterfeiting strategies.
lifetime, heat stimuli, chemical reagents, mechanical force, and so on. Based on the category of regulation factors, multilevel luminescence anti-counterfeiting can be divided into five types: (1) regulated by excitation light;[64–80] (2) co-regulated by excitation light and luminescence lifetime;[81–92] (3) co-regulated by excitation light and heat stimuli;[35,63,93–96] (4) co-regulated by excitation light and chemical reagents;[16,97–105] and (5) co-regulated by other factors.[106–121]

4.1 Regulated by excitation light

Multilevel luminescence anti-counterfeiting regulated by excitation light is easily realized by tuning excitation wavelength, excitation power, irradiation time, and irradiation mode, which is an efficient anti-counterfeiting strategy that does not require other external stimuli.

Sun et al.[75] designed an anti-counterfeiting label whose luminescence could be regulated by excitation wavelength. The anti-counterfeiting label displayed fluorescence with different colors when excited by lights with different wavelengths, thus realizing multilevel luminescence anti-counterfeiting. Moreover, two kinds of excitation lights can also be used in superimposition to form a new excitation light source.[68,70] For example, Chen’s group prepared white anti-counterfeiting labels by using the mixture of Tm:KYbF7@glass powder, CsPbBrxI3-x perovskite nanocrystals@glass powder, and commercial blank white screen-printing ink as anti-counterfeiting ink.[70] Interestingly, the white anti-counterfeiting labels showed blue UC fluorescence of Tm:KYbF7@glass under the excitation of near infrared (NIR) light and red DC fluorescence of CsPbBrxI3-x perovskite nanocrystals@glass under the excitation of UV light (Figure 5A). Specially, when the anti-counterfeiting label was excited by superimposed NIR light and UV light, it displayed pink mixing emissions of blue UC fluorescence and red DC fluorescence. In this strategy, the superimposition of two kinds of excitation lights provides a new light source for triggering the change of anti-counterfeiting label, decreasing the demands to excitation lights with other wavelengths.

Similarly with excitation wavelength, excitation power can also regulate the luminescence of anti-counterfeiting labels.[67,79,80] Li et al.[67] developed an excitation-power-dependent color-tunable anti-counterfeiting film and
successfully realized multilevel luminescence anti-counterfeiting. However, traditional luminescence anti-counterfeiting strategies regulated by excitation wavelength or excitation power are easy to be copied. Therefore, more advanced multilevel anti-counterfeiting technologies need to be explored.

Luminescence anti-counterfeiting regulated by irradiation time is a kind of emerging dynamic luminescence anti-counterfeiting strategy. With irradiation time prolonging, the emission color of anti-counterfeiting label is constantly changing. For instance, Jin et al. adopted cellulose derivative C-imidazole salt (Im$^+$)-spiropyran (SP)/N, N-dimethylformamide (DMF), negatively charged CDs (NCDs)$@$C-Im$^+$-SP/DMF, and CDs$@$cellulose derivatives/H$_2$O as anti-counterfeiting inks to print a full-color airplane pattern on paper. The anti-counterfeiting pattern displayed blue fluorescence under 365 nm UV light, but with irradiation time prolonging, the blue fluorescence of anti-counterfeiting pattern gradually changed to red fluorescence owing to the convert of SP groups from a ring-closed form to a ring-open form and the changes of energy transfer process and efficiency between CDs and SP groups (Figure 5B). Such dynamic luminescence behaviors induced by irradiation time endowed the materials with advanced dynamic luminescence anti-counterfeiting ability.

In addition to excitation wavelength, excitation power, and irradiation time, irradiation mode is also an alternative factor to regulate the luminescence of anti-counterfeiting...
Usually, irradiation mode includes steady irradiation, dynamic scanning, focusing irradiation, and defocusing irradiation. Different irradiation modes can induce different luminescence behaviors of anti-counterfeiting labels. As illustrated in Figure 5C, Liu et al. \cite{72} designed a two-dimensional (2D) anti-counterfeiting pattern utilizing Mn$^{2+}$-activated core-shell nanoparticles with different lanthanides compositions. Under the steady irradiation of 980 nm laser, the whole anti-counterfeiting pattern displayed multicolor luminescence. However, under the dynamic scanning of 980 nm laser, the “i” region of anti-counterfeiting pattern displayed a green main emission spot, while other regions (ii–vi) showed an additional green tailed emission. Due to the difference of lanthanides compositions in “ii–vi” regions, the color of main spot emission in “ii–vi” regions varied from red (region “ii”) to white (region “vi”). Moreover, when changing the excitation wavelength from 980 nm to 808 nm, whether it was dynamic scanning or steady irradiation, both the “iv” and “v” regions displayed comparable emission states with those obtained with 980 nm as excitation wavelength. The multiple emission states of anti-counterfeiting pattern indicated the anti-counterfeiting strategy was difficult to be copied and counterfeited. Furthermore, focusing irradiation and defocusing irradiation can also induce different fluorescence emissions for anti-counterfeiting label. Utilizing this characteristic, Huang et al. \cite{65} developed a high-level anti-counterfeiting label that could display five kinds of luminescence behaviors.

Notably, the luminescence of materials can not only be triggered or regulated by light but also be quenched by light \cite{64,69,71,76,77}. Sometimes, the quenching process is accompanied with discoloration of materials, and the quenched luminescence and changed color of materials can be restored to the initial state after undergoing the irradiation of visible light. Such multiple changes induced by light benefit for realizing multilevel luminescence anti-counterfeiting. Guo et al. \cite{64} demonstrated the effectiveness of this strategy by adopting 3-(1,2,2-triphenylvinyl) benzofuran (3-TPVF) solution as anti-counterfeiting ink. The anti-counterfeiting pattern painted by anti-counterfeiting ink was invisible under daylight, but emitted blue fluorescence under the excitation of 365 nm UV light. However, with irradiation time increasing, the blue fluorescence of anti-counterfeiting pattern gradually weakened due to the occurrence of photo-induced cyclization reaction, and after turning off the UV light, an obvious yellow trace appeared on paper. Notably, the quenched fluorescence and yellow trace of anti-counterfeiting pattern could be restored to initial state after irradiated by white light because of the occurrence of cycloreversion reactions, illustrating a kind of photo-induced nondestructive anti-counterfeiting process. The multiple changes of anti-counterfeiting label including photo-luminescence, light-induced luminescence quenching, and photo-chromic, and afterglow of organic materials can be restored to the initial state after undergoing the irradiation of IR light because of the occurrence of cyclization process and realized naked-eye visible multilevel luminescence anti-counterfeiting. As displayed in Figure 4B, the CDs@zeolite composites with dual emissions of TADF and TADF lifetime co-realized five-level luminescence anti-counterfeiting and avoided the use of complex time-gating device. However, complex synthesis process, low quantum efficiency, and short afterglow lifetime of organic afterglow materials limit their applications in luminescence anti-counterfeiting. Recently, Yu’s group prepared CDs@zeolite composites with long RTP and long TADF lifetime and high quantum efficiency by a facile and quick hydrothermal or solvent-free thermal crystallization process and realized naked-eye visible multilevel luminescence anti-counterfeiting \cite{90,91}.

As shown in Figure 6A, it is a typical luminescence anti-counterfeiting strategy co-regulated by excitation wavelength and fluorescence lifetime. Liu et al. \cite{85} exploited two kinds of colorless anti-counterfeiting inks prepared by LiYbF$_4$:Y@LiGdF$_4$:Tb/Tm@LiYF$_4$:A (A = Eu, Tb)@LiGdF$_4$:Ce core/tri-shell nanoparticles (Eu-CSSS and Tb-CSSS) to print a flower pattern on paper as anti-counterfeiting label (petal: Eu-CSSS ink, leaf: Tb-CSSS ink). Under the irradiation of UV light, steady-state NIR, and the co-irradiation of UV light and NIR, the anti-counterfeiting label displayed fluorescence with different colors and intensities, respectively. Notably, under the detection of NIR time-gating device, the anti-counterfeiting label displayed lifetime-regulated fluorescence, whose color was different from the above-mentioned three kinds of fluorescence. Hence, the four-level luminescence anti-counterfeiting technology co-regulated by excitation wavelength and fluorescence lifetime is achieved. However, due to the short luminescence lifetime of materials, complex time-gating device was needed to monitor the lifetime-regulated imaging process, which limited the actual application of this anti-counterfeiting strategy in commercial products.

Yang et al. \cite{89} designed an anti-counterfeiting process by employing three kinds of phosphorescent Zn-based organic-inorganic hybrid metal halides (OIHMs) with different emission colors and lifetimes as anti-counterfeiting materials, in which two fluorescence imaging processes regulated by excitation wavelength together with three naked-eye visible phosphorescence imaging processes regulated by lifetime co-realized five-level luminescence anti-counterfeiting and avoided the use of complex time-gating device. However, complex synthesis process, low quantum efficiency, and short afterglow lifetime of organic afterglow materials limit their applications in luminescence anti-counterfeiting. Recently, Yu’s group prepared CDs@zeolite composites with long RTP and TADF lifetime and high quantum efficiency by a facile and quick hydrothermal or solvent-free thermal crystallization process and realized naked-eye visible multilevel luminescence anti-counterfeiting \cite{90,91}. As displayed in Figure 6B, the CDs@zeolite composites with dual emissions of RTP and TADF, pure TADF material, and the mixture of CDs and starch were utilized to design a 2D code pattern. Under the irradiation of UV light, the whole 2D code pattern displayed blue fluorescence. Interestingly, when turning off UV light for 0.1 s, only the parts composed of TADF material and CDs@zeolite composites could display afterglow emission, while the part composed of mixture of CDs and starch did not show afterglow. When the UV light was turned off for 1 s, the part composed of CDs@zeolite composites still emitted afterglow due to the longer afterglow lifetime of CDs@zeolite composites than that of TADF material. Such triple changes demonstrated naked-eye visible multi-level luminescence anti-counterfeiting process co-regulated by excitation light and afterglow lifetime. This luminescence anti-counterfeiting strategy was simpler and more convenient.
compared with the luminescence anti-counterfeiting co-regulated by excitation light and short fluorescence lifetime.

Irradiation time is also an effective regulation factor in multilevel luminescence anti-counterfeiting, which can not only regulate fluorescence of materials but also regulate phosphorescence of materials. Huang et al. developed three kinds of organic crystal powder whose phosphorescence emissions were closely associated with irradiation time. The three rectangular frames painted by three kinds of crystal powder emitted blue fluorescence under the irradiation of UV light (Figure 6C). However, after turning off the UV light, only the rectangular frame painted by carbazole (CZ) could display orange phosphorescence, while other parts did not emit phosphorescence. Interestingly, when prolonging photo-activated time to 10 s and 120 s, the orange phosphorescence of rectangular frames painted by 9-isopropyl-9H-carbazole (CZIP) and 2-(9H-carbazol-9-yl)ethanol (CZEO) could be observed, respectively, which derived from light-induced strong interactions between two molecules suppressing their movement and decreasing nonradiation decay rates. Such multiple phosphorescence emissions regulated by irradiation time prove these materials are excellent multilevel luminescence anti-counterfeiting materials.

As discussed in Section 4.1, the difference of irradiation mode can trigger many novel luminescence for anti-counterfeiting label, such as main spot emission and tailed emission. Such novel fluorescence emission regulated by irradiation mode can be combined with phosphorescence emission to co-realize multilevel luminescence anti-counterfeiting. For example, Liu et al. adopted three kinds of Ce/Eu, Ce/Tb, and Yb/Tm/Ce/Mn co-doped phosphors to create a 2D covert pattern as anti-counterfeiting label
(Figure 6D). Under the steady irradiation of 254 nm, the whole anti-counterfeiting label displayed dual-color fluorescence, while under the steady irradiation of 980 nm laser, only the area “iii” composed of Yb/Tm/Ce/Mn co-doped phosphors emitted white fluorescence. In both cases, after ceasing excitation, the area “iii” displayed green persistent luminescence, while other areas did not show persistent emission. Moreover, under the dynamic scanning of 980 nm laser, the area “iii” displayed a unique circle-shaped emission with green tail and whitish head. These interesting luminescence phenomena suggest adopting irradiation mode and lifetime to regulate the luminescence of anti-counterfeiting label is also an outstanding choice.

4.3 Co-regulated by excitation light and heat stimuli

Heat stimuli can not only induce luminescence enhancement but also result in thermoluminescence (ThL), luminescence quenching, change of luminescence color, and discoloration of luminescence materials, which is undoubtedly an important stimuli factor in multilevel luminescence anti-counterfeiting.[35,63,93–96]

Sang et al.[35] designed an anti-counterfeiting pattern composed of four kinds of rare earth-ion-doped LiNbO3 materials (Figure 7A). Under the excitation of 365 nm UV light and 980 nm NIR light, the anti-counterfeiting pattern displayed luminescence with different colors, respectively. Moreover, under the heat stimuli, the area composed of LiNbO3:1%Pr-0.1%Er composites displayed obvious red ThL, while other areas did not show any luminescence. In this anti-counterfeiting strategy, the introduction of ThL added a new anti-counterfeiting mode for realizing multilevel luminescence anti-counterfeiting.

In addition to ThL, heat stimuli can also bring many interesting changes for anti-counterfeiting labels.[63,93–96]
et al.\textsuperscript{[95]} utilized three kinds of polymer dots solution as anti-counterfeiting inks to write word “CHEMISTRY” on paper (Figure 7B). At 298 K, the letters were light purple under daylight, and only part letters could emit bright fluorescence under 365 nm UV light, while the fluorescence of other letters was weaker. But when increasing the temperature to 323 K, due to thermal-induced conversion of chemical structure from ring-closed conformation to ring-opened conformation, the letters became colorless under daylight, and all the letters could emit bright fluorescence under 365 nm UV light because of the heat-induced luminescence enhancement. Interestingly, the luminescence and color of anti-counterfeiting label could be restored to initial state when temperature was decreased to 298 K, because the conversion of chemical structure of polymer was reversible, which indicated the anti-counterfeiting label could be reused. Similarly, Qi et al.\textsuperscript{[94]} also adopted excitation light and heat stimuli to induce changes of anti-counterfeiting label. But differently, in this work, heat stimuli played a role of restoring luminescence and color of anti-counterfeiting label. The multiple changes of anti-counterfeiting label co-regulated by excitation light and heat stimuli illustrated the effectiveness of this anti-counterfeiting strategy.

4.4 Co-regulated by excitation light and chemical reagents

Similarly with heat stimuli, chemical reagents can also induce a series of changes for anti-counterfeiting labels. Huang et al.\textsuperscript{[99]} adopted 3-mercaptopropionic acid (MPA) to replace oleyl amine (OAm) and oleic acid (OA) ligands capping on the surface of CdS:Ag quantum dots (QDs) to improve water solubility of CdS:Ag QDs, and designed an anti-counterfeiting pattern “BIT” with the mixture of hydrophilic CdS:Ag QDs, alkaline water and ethylene glycol as anti-counterfeiting ink (Figure 7C), in which alkaline water provided alkaline environment that benefited for the dispersion of QDs and avoided fluorescence quenching, and ethylene glycol played a role of modifying viscosity of luminescent ink. The anti-counterfeiting pattern was invisible under daylight, but displayed bright red fluorescence under 365 nm UV light. When immersing the anti-counterfeiting pattern into methanolic Ag\textsuperscript{+} solution, the fluorescence of anti-counterfeiting pattern was quickly quenched due to the fast cation exchange between Ag\textsuperscript{+} and Cd\textsuperscript{2+}. Interestingly, after again immersing the anti-counterfeiting pattern into toluene containing Cd\textsuperscript{2+} and tributyl phosphate, the quenched fluorescence could be restored to red fluorescence due to the reverse cation exchange between Ag\textsuperscript{+} and Cd\textsuperscript{2+}. These luminescence phenomena proved multilevel luminescence anti-counterfeiting strategy co-regulated by chemical reagents and excitation light had been successfully implemented. Furthermore, researchers also explored other chemical reagents, such as CHCl\textsubscript{3} vapor,\textsuperscript{[98]} diluted sodium hydroxide solution together with diluted acetic acid,\textsuperscript{[104]} CH\textsubscript{3}NH\textsubscript{3}Br together with water,\textsuperscript{[16]} ClO\textsuperscript{-} together with SCN\textsuperscript{-},\textsuperscript{[101]} and oxygen, etc.\textsuperscript{[97]} to regulate the luminescence of anti-counterfeiting label. However, these chemical reagents are not common in our daily life, and the toxicity and dangerousness of these chemical reagents are also non-ignorable, which would limit the development of this luminescence anti-counterfeiting strategy.

Similarly with other chemical reagents, water, a kind of common, safe, and nontoxic chemical reagent, can also regulate the luminescence of anti-counterfeiting label\textsuperscript{[106,102,103,105]} Recently, Yang et al.\textsuperscript{[103]} prepared CDs with the property of aggregation-induced emission (AIE) via one-pot solvothermal treatment, and painted an anti-counterfeiting label by employing hydrophobic CDs (H-CD) solution as ink (Figure 7D). Dry anti-counterfeiting label was white under white light, and displayed blue fluorescence under 365 nm UV light, but did not display any luminescence under 254 nm UV light; this was because H-CD monomers could not be excited by 254 nm but could be excited by 365 nm. Interestingly, after treated by water, the anti-counterfeiting label displayed pink fluorescence under 365 nm UV light, but displayed red fluorescence under 254 nm UV light. The detail mechanisms were that the H-CD aggregates on the surface of filter paper and the H-CD monomers in filter paper were simultaneously excited to emit red and blue fluorescence, respectively, thus displaying hybrid pink fluorescence under 365 nm UV light; while under 254 nm UV light, only the H-CD aggregates on the surface were excited, thus resulting in red emission. Recently, Yu et al.\textsuperscript{[105]} designed a CsPbBr\textsubscript{3}-based anti-counterfeiting label, in which excitation light and water were used as stimuli sources to regulate the luminescence of anti-counterfeiting label, ultimately, realizing triple-level luminescence anti-counterfeiting. The successes of these anti-counterfeiting strategies demonstrate water is indeed an outstanding stimuli factor that can replace other complex chemical reagents to regulate the luminescence of anti-counterfeiting label.

4.5 Co-regulated by other factors

In addition to excitation light, luminescence lifetime, heat stimuli, and chemical reagents, many other regulation factors, such as rotation speed and mechanical stimuli have been also explored to regulate the luminescence of anti-counterfeiting label.\textsuperscript{[106,121]} For example, Dai et al.\textsuperscript{[106]} utilized NaErF\textsubscript{4}:10Yb\textsuperscript{3+}/NaYF\textsubscript{4}:NaYbF\textsubscript{4}:20Ca\textsuperscript{1+}/Tm\textsuperscript{3+}/NaYF\textsubscript{4} nanocrystals (NCs) to stamp an anti-counterfeiting pattern whose luminescence was closely associated with rotation speed and excitation power (Figure 8A). As excitation power was increased from 5 to 11 W cm\textsuperscript{-2}, the emission color of anti-counterfeiting pattern gradually changed from red to purple due to the excitation power-dependent luminescence characteristics of such materials. Notably, when rotated at 600 and 900 rpm, the anti-counterfeiting pattern displayed bright multicolor trajectories with different lengths, respectively. The introduction of this novel regulation factor endowed the luminescence anti-counterfeiting strategy with great innovativeness.

All the above discussions are about luminescence anti-counterfeiting regulated by no more than two kinds of factors. In order to improve the ability of luminescence anti-counterfeiting, usually, more than two kinds of factors are simultaneously adopted to regulate the luminescence of anti-counterfeiting labels. For instance, Ma et al.\textsuperscript{[113]} cut Y\textsubscript{3}Al\textsubscript{2}O\textsubscript{12}:Ce\textsuperscript{3+}/polydimethylsiloxane (YAG:Ce\textsuperscript{3+}/PDMS) and Ba\textsubscript{0.5}Sr\textsubscript{0.5}Si\textsubscript{2}O\textsubscript{2}N\textsubscript{2}:Eu\textsuperscript{2+}/PDMS (BSSON:Eu\textsuperscript{2+}/PDMS)
elastomers into strips with different widths, and then put them into a Petri dish full of PDMS precursor to be cured at 353 K for 2 h, finally, obtaining the anti-counterfeiting device with bar codes (Figure 8B). After stimulated by folding, the strips composed of YAG:Ce$^{3+}$ could emit yellow ML due to phonon-assisted mechanical energy transfer processes, while the strips composed of BSSON:Eu$^{2+}$ did not display any luminescence (the left of Figure 8B). After irradiated by 365 nm UV light for 5 min to fill energy in traps, the strips composed of BSSON:Eu$^{2+}$ could also emit green ML after stimulated by folding (the middle of Figure 8B), owing to the release of energy stored in deep traps followed by transfer to luminescence centers. Interestingly, the strips composed of BSSON:Eu$^{2+}$ could display afterglow emission (the right of Figure 8B), because part energy stored in deep traps would be transferred to shallow traps, thus producing spontaneous emission, namely, ML afterglow. The triple emissions co-regulated by mechanical stimuli, UV irradiation, and luminescence lifetime accomplished three-level luminescence anti-counterfeiting process. Moreover, excitation light, heat stimuli together with chemical reagents, or excitation light, heat stimuli together with luminescence lifetime, or excitation light, luminescence lifetime together with chemical regents, or excitation light, heat stimuli, luminescence lifetime together with mechanical stimuli, etc., can also be simultaneously adopted to realize multilevel luminescence anti-counterfeiting. As displayed in Figure 8C, it is a typical luminescence anti-counterfeiting co-regulated by four kinds of factors. Zhang et al. encapsulated NaNbO$_3$:Pr$^{3+},$ Er$^{3+}$ particles into TPU to prepare anti-counterfeiting film. After charged by white light for 15 s, the anti-counterfeiting film displayed bright red long-lifetime afterglow arising from the $^1D_2^2\rightarrow\text{H}_4$ transition of Pr$^{3+}$ dopants. When writing letters “QDU” by a pen on anti-counterfeiting film, one could clearly observe letters “QDU” displayed bright ML that was stronger than the afterglow of background, which arose from the tri-coupling of piezoelectricity, trap levels, and Pr$^{3+}$ centers. When the anti-counterfeiting film contacted with ice (273 K, $\approx$1 s), due to the release of remaining trapped
electrons becoming difficult under low temperature, the afterglow of anti-counterfeiting film would be quenched, but when the anti-counterfeiting film again contacted with heat source ($\approx$306 K, $\approx$1 s), due to the thermal-induced emptying of remaining traps, the quenched afterglow could be restored. Moreover, under the excitation of 980 nm NIR laser, the anti-counterfeiting film displayed bright green luminescence, and with laser power density increasing, luminescence intensity of anti-counterfeiting film gradually increased, but after ceasing the excitation of 980 nm NIR laser, the anti-counterfeiting film did not display afterglow emission. When irradiated by UV light with different wave-lengths, the anti-counterfeiting film displayed luminescence with different colors, and after ceasing the irradiation of UV light, the anti-counterfeiting film displayed long-lifetime red afterglow. The multidimensional luminescence performance co-regulated by luminescence lifetime, mechanical stimuli, cold/hot stimuli, and excitation light proved the complex multilevel luminescence anti-counterfeiting strategy was extremely difficult to be copied and counterfeited.

5 | SUMMARY AND OUTLOOK

In recent years, the studies on luminescence anti-counterfeiting technologies have made a great progress. Various luminescence anti-counterfeiting strategies regulated by excitation light, luminescence lifetime, heat stimuli, chemical reagents, mechanical stimuli, and rotation speed are adopted to prevent counterfeiting phenomenon. In this review, we classify the anti-counterfeiting strategies according to anti-counterfeiting level and present the state-of-the-art research on luminescence anti-counterfeiting from elementary to advanced stage. The regulation of various factors to luminescence behaviors of anti-counterfeiting labels has been discussed. However, to realize the practical application of these luminescence anti-counterfeiting strategies in our daily life, there are still many problems and challenges to be addressed.

First, due to the proliferation of counterfeiting commodities, simple luminescence anti-counterfeiting technology already cannot meet anti-counterfeiting demands, and more multilevel luminescence anti-counterfeiting strategies need to be constantly explored. Second, though the materials with tunable luminescence behaviors under various regulation conditions can realize multilevel luminescence anti-counterfeiting, high cost, complex preparation process, low yield, poor stability, and low luminescence efficiency limit the mass production and long-term use of these materials. Therefore, it is highly desired to develop new types of luminescence anti-counterfeiting materials to low preparation cost, simplify synthesis methods, and improve luminescence performance and stability. Third, some regulation factors, such as excitation lights with different wavelengths and powers, chemical reagents, or rotating devices with adjustable rotation speed, are mainly used in laboratory but not common in our daily life. Therefore, though the multidimensional luminescence co-regulated by various factors can realize the advanced luminescence anti-counterfeiting that is particularly difficult to be copied and counterfeited, the practical application of these anti-counterfeiting strate-gies in our daily life remains limited. Therefore, more novel anti-counterfeiting materials whose luminescence can be easily regulated by simple factors, such as common excitation light and luminescence lifetime, still need to be explored. In particular, the materials that can display multiple emissions including naked-eye visible afterglow emission may become the promising next-generation anti-counterfeiting materials due to the facile and quick anti-counterfeiting process and advanced anti-counterfeiting ability.

In the future, with the deepening of research to luminescence anti-counterfeiting, we believe more exciting anti-counterfeiting materials with low cost, high yield, high luminescence efficiency, high stability, and easily regulated luminescence performance under simple regulation conditions will be discovered, and more advanced luminescence anti-counterfeiting technologies based on these materials will be applied into commercial products to guarantee the authenticity of these products and avoid counterfeiting. We hope this review can provide some guidelines for realizing facile, quick, low-cost, environmentally friendly, and difficult-to-replicate advanced luminescence anti-counterfeiting technologies and promoting practical applications of these anti-counterfeiting technologies in our daily life.

ACKNOWLEDGMENTS

We thank the National Natural Science Foundation of China (Grant 21835002 and 21621001), and the 111 Project (B17020) for supporting this work.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

REFERENCES

1. Y. Gu, C. He, Y. Zhang, L. Lin, B. D. Thackray, J. Ye, Nat. Commun. 2020, 11, 516.
2. W. Hong, Z. Yuan, X. Chen, Small 2020, 16, 1907626.
3. W. Ren, G. Lin, C. Clarke, J. Zhou, D. Jin, Adv. Mater. 2020, 32, 1901430.
4. M. Tan, F. Li, X. Wang, R. Fan, G. Chen, ACS Nano 2020, 14, 6532.
5. M. K. Tsang, G. Bai, J. Hao, Chem. Soc. Rev. 2015, 44, 1585.
6. R. Arppe, T. J. Sørensen, Nat. Rev. Chem. 2017, 1, 0031.
7. J. Deng, L. Deng, Z. Guan, J. Tao, G. Li, Z. Li, Z. Li, S. Yu, G. Zheng, Nano Lett. 2020, 20, 1830.
8. P. Kumar, S. Singh, B. K. Gupta, Nanoscale 2016, 8, 14297.
9. R. Li, Y. Zhang, J. Tan, J. Wan, J. Guo, C. Wang, ACS Appl. Mater. Interfaces 2016, 8, 9384.
10. A. F. Smith, S. E. Skrabalak, J. Mater. Chem. C 2017, 5, 3207.
11. Y. Liu, Y. Zheng, Y. Zhu, F. Ma, X. Zheng, K. Yang, X. Zheng, Z. Xu, S. Ji, Y. Zheng, T. Guo, L. Qian, F. Li, ACS Appl. Mater. Interfaces 2020, 12, 39649.
12. Y. Fan, X. Jin, M. Wang, Y. Gu, J. Zhou, J. Zhang, Z. Wang, Chem. Eng. J. 2020, 393, 124799.
13. D. Li, L. Tang, J. Wang, X. Liu, Y. Ying, Adv. Opt. Mater. 2016, 4, 1475.
14. J. Liu, H. Rijckaert, M. Zeng, K. Hausseraete, B. Laforce, L. Vincze, L. Van Driessche, A. M. Kaczmarek, R. Van Deun, Adv. Funct. Mater. 2018, 28, 1707365.
15. A. M. Kaczmarek, Y. Liu, C. Wang, B. Laforce, L. Vincze, P. Van Der Voort, K. Van Hecke, R. Van Deun, Adv. Funct. Mater. 2017, 27, 1700258.
16. D. Zhang, W. Zhou, Q. Liu, Z. Xia, ACS Appl. Mater. Interfaces 2018, 10, 27875.
17. X. Chen, W. Yao, Q. Wang, W. Wu, Adv. Opt. Mater. 2020, 8, 1901209.
18. W. Yao, Q. Tian, W. Wu, Adv. Opt. Mater. 2019, 7, 1801171.
19. Z. Feng, W. Zhao, Z. Liang, Y. Lv, F. Xiang, D. Sun, C. Xiong, C. Duan, L. Dai, Y. Ni, ACS Appl. Mater. Interfaces 2020, 12, 11005.
20. A. L. Hagström, H. L. Lee, M. S. Lee, H. S. Choe, J. Jung, B. G. Park, W. S. Han, J. S. Ko, J.H. Kim, J.H. Kim, ACS Appl. Mater. Interfaces 2018, 10, 8985.

21. W. Huang, X. Li, X. Sun, D. Ying, Y. Feng, Y. Tang, P. Zhou, L. Wang, Q. Zhang, Chem. Eng. J. 2021, 405, 1265174.

22. P. Li, J. Zeng, B. Yang, Z. Cheng, J. Xu, W. Gao, K. Chen, Carbohydr. Polym. 2020, 247, 116721.

23. X. Li, Y. Hu, Carbohydr. Polym. 2019, 203, 167.

24. F. Liu, A. Nattestad, S. Naficy, R. Han, A. Angeloski, X. Xu, B. Yang, Angew. Chem. Int. Ed. 2018, 57, 125451.

25. P. Long, Y. Feng, C. Cao, Y. Li, J. Han, S. Li, C. Peng, Z. Li, W. Feng, Adv. Funct. Mater. 2018, 28, 1800791.

26. Q. Su, C. Lu, X. Yang, Carbon 2019, 152, 609.

27. L. Xue, S. Liu, Y. Geng, S. Zhu, S. A. T. Redfern, Y. Song, T. Feng, W. Xu, B. Yang, Angew. Chem. Int. Ed. 2018, 57, 23933.

28. B. Wang, Y. Yu, H. Zhang, Y. Xuan, G. Chen, W. Ma, J. Li, J. Yu, Angew. Chem. Int. Ed. 2019, 58, 18443.

29. Y. Wang, J. Yang, M. Fang, Y. Yu, B. Z. Tang, Z. Li, Mater. Horiz. 2020, 3, 449.

30. L. Lei, D. Chen, C. Li, F. Huang, J. Zhang, S. Xu, J. Mater. Chem. C 2018, 6, 5427.

31. C. Wu, S. Zeng, Z. Wang, F. Wang, H. Zhou, J. Zhang, Z. Ci, L. Sun, Adv. Funct. Mater. 2018, 28, 1803168.

32. K. Yuan, Z. Zhang, X. Li, R. Qin, Y. Cheng, L. Li, X. Yang, X. Yu, Z. Lu, H. Liu, Chem. Eng. J. 2020, 397, 125487.

33. Y. Lu, J. Zhuo, R. Zhang, Y. Liu, D. Liu, E.M. Goldys, X. Yang, P. Xu, A. Sunna, J. Lu, Y. Shi, R.C. Leif, Y. Huo, J. Shen, J.A. Piper, J.P. Robinson, D. Jin, Nat. Photonics 2013, 8, 32.

34. L. Xu, J. Chen, J. Song, J. Li, J. Xue, Y. Dong, B. Cai, Q. Shan, B. Han, H. Zeng, ACS Appl. Mater. Interfaces 2017, 9, 26556.

35. S. Gao, S. Zhou, J. Chen, P. Guo, R. Ding, H. Sun, H. Feng, Z. Qian, ACS Appl. Mater. Interfaces 2020, 12, 42410.

36. H. Huang, J. Chen, Y. Liu, J. Lin, S. Wang, F. Huang, D. Chen, Small 2016, 20, 2000708.

37. K. Jin, X. Ji, T. Yang, J. Zhang, W. Tian, J. Yu, X. Zhang, Z. Chen, Chem. Eng. J. 2021, 406, 126794.

38. F. Li, X. Wang, Z. Xia, C. Pan, Q. Liu, Adv. Funct. Mater. 2017, 27, 1700571.

39. X. Li, C. Yang, Y. Yu, Z. Li, J. Liu, X. Guan, Z. Zhang, D. Chen, ACS Appl. Mater. Interfaces 2020, 12, 18705.

40. Z. Li, G. Wang, Y. Ye, B. Li, H. Li, B. Chen, Angew. Chem. Int. Ed. 2019, 58, 18025.

41. J. Lin, C. Yang, P. Huang, S. Wang, M. Liu, N. Jiang, D. Chen, Chem. Eng. J. 2020, 390, 125214.

42. Z. Lin, H. Wang, M. Yu, X. Guo, C. Zhang, H. Deng, P. Zhang, S. Chen, R. Zeng, J. Cui, J. Chen, J. Mater. Chem. C 2019, 7, 11515.

43. X. Liu, Y. Wang, X. Li, Z. Yi, R. Deng, L. Liang, X. Xie, D. T.B. Loong, S. Song, D. Fan, A.H. Ali, H. Zhang, L. Huang, X. Liu, Nat. Commun. 2017, 8, 899.

44. Y. Liu, Z. Zhang, S. Nie, C. Liu, J. Mater. Chem. B 2019, 7, 6337.

45. X. Shen, B. Song, B. Fang, A. Jiang, S. Ji, Y. He, Chem. Commun. 2018, 54, 4947.

46. T. Sun, B. Xu, B. Chen, X. Chen, M. Li, P. Shi, F. Wang, Nanoscale 2017, 9, 2701.

47. W.K. Tsai, Y.S. Lai, P.J. Tseng, C.H. Liao, Y.H. Chan, ACS Appl. Mater. Interfaces 2017, 9, 30918.

48. J. Wang, Y. Gao, J. Zhang, H. Tian, J. Mater. Chem. C 2017, 5, 4571.

49. J. Wang, J. Ma, J. Zhang, Y. Fan, W. Wang, J. Sang, Z. Ma, H. Li, ACS Appl. Mater. Interfaces 2019, 11, 35871.

50. S. Wang, J. Lin, H. Ye, J. Chen, C. Yang, F. Huang, D. Chen, Chem. Eng. J. 2020, 394, 124889.

51. W. You, D. Tu, R. Li, W. Zhang, Chem. Nanoscale 2019, 12, 1417.

52. L. Gu, H. Shi, L. Bian, M. Gu, K. Ling, X. Wang, H. Ma, S. Cai, W. Ning, L. Fu, H. Wang, S. Wang, Y. Gao, W. Yao, F. Hua, Y. Tao, Z. An, X. Liu, W. Huang, Nat. Photonics 2019, 13, 406.

53. Q. Huang, H. Gao, S. Yang, D. Ding, Z. Lin, Q. Ling, Nano Res. 2020, 13, 1035.

54. Q. Huang, X. Mei, Z. Xie, D. Wu, S. Yang, W. Gong, Z. Chi, Z. Lin, Q. Ling, J. Mater. Chem. B 2019, 7, 2530.

55. C. Lin, Y. Zhuang, W. Li, T. Zhou, R. Xie, Nanoscale 2019, 11, 6584.

56. S. Liu, L. Yan, Q. Li, J. Huang, L. Tao, B. Zhou, Chem. Eng. J. 2020, 397, 125451.

57. X. Liu, Q. Ji, J. Hu, C. Li, M. Chen, J. Sun, Y. Wang, Q. Sun, B. Geng, ACS Appl. Mater. Interfaces 2019, 11, 30146.

58. C. Shi, X. Shen, Y. Zhu, X. Li, Z. Pang, M. Ge, ACS Appl. Mater. Interfaces 2019, 11, 18548.

59. C. Yang, L. Gu, C. Ma, M. Gu, X. Xie, H. Shi, H. Ma, W. Yao, Z. An, W. Huang, Mater. Horiz. 2019, 6, 984.

60. S. Yang, B. Zhou, Q. Huang, S. Wang, H. Zhen, D. Yan, Z. Lin, Q. Ling, ACS Appl. Mater. Interfaces 2020, 12, 1419.

61. H. Zhang, J. Liu, B. Wang, K. Liu, G. Chen, X. Yu, J. Li, J. Yu, Mater. Chem. Front. 2020, 4, 1404.

62. H. Zhang, K. Liu, J. Liu, B. Wang, C. Li, W. Song, J. Li, L. Huang, J. Yu, ACS Chem. Chem. 2020, 2, 118.

63. M. Gimelth, H. Thomas, F. Sries, S. Reineke, Sci. Adv. 2019, 5, eaaaw7310.

64. J. Du, L. Sheng, Q. Chen, Y. Xu, W. Li, X. Wang, M. Li, S.X.A. Zhang, Mater. Horiz. 2019, 6, 165.

65. Q. Qi, C. Li, X. Liu, S. Jiang, Z. Xu, R. Lee, M. Zhu, B. Xu, W. Tian, J. Am. Chem. Soc. 2017, 139, 16036.
AGGREGATE

J.C. Yang, Y.C. Ho, Y.H. Chan, ACS Appl. Mater. Interfaces 2019, 11, 29341.

X. Yang, Q. Li, M. Tang, Y. Yang, W. Yang, J. Hu, X. Pu, J. Liu, J. Zhao, Z. Zhang, ACS Appl. Mater. Interfaces 2020, 12, 20849.

L. Chen, Y. Chen, H. Fu, Y. Liu, Adv. Sci. 2020, 7, 2000803.

Z. Gao, Y. Han, F. Wang, Nat. Commun. 2018, 9, 3977.

W. Huang, M. Xu, J. Liu, J. Wang, Y. Zhu, J. Liu, H. Rong, J. Zhang, Adv. Funct. Mater. 2019, 29, 1808762.

T. Jiang, X. Wang, J. Wang, G. Hu, X. Ma, ACS Appl. Mater. Interfaces 2019, 11, 14399.

C. Li, J. Hai, S. Li, B. Wang, Z. Yang, Nanoscale 2018, 10, 8667.

V.K. Singh, R.K. Chitumalla, S.K. Ravi, Y. Zhang, Y. Xi, V. Sanjairaj, C. Zhang, J. Jang, S.C. Tan, ACS Appl. Mater. Interfaces 2017, 9, 33071.

H. Yang, Y. Liu, Z. Guo, B. Lei, J. Zhuang, X. Zhang, Z. Liu, C. Hu, Nat. Commun. 2019, 10, 1789.

P. Yang, Z. Zhu, T. Zhang, M. Chen, Y. Cao, W. Zhang, X. Wang, X. Zhou, W. Chen, Carbon 2019, 146, 636.

X. Yu, L. Wu, D. Yang, M. Cao, X. Fan, H. Lin, Q. Zhong, Y. Xu, Q. Zhang, Angew. Chem. Int. Ed. 2020, 59, 14527.

X. Dai, K. Wang, L. Lei, S. Xu, Y. Cheng, Y. Wang, Nanophotonics 2020, 9, 1519.

L. Deng, X. Wang, J. Am. Chem. Soc. 2020, 142, 13558.

K. Jiang, X. Gao, X. Feng, Y. Wang, Z. Li, H. Lin, Angew. Chem. Int. Ed. 2020, 59, 1263.

K. Jiang, Y. Wang, C. Cai, H. Lin, Adv. Mater. 2018, 30, 1800783.

Y. Lei, W. Dai, J. Guan, S. Guo, F. Ren, Y. Zhou, J. Shi, B. Tong, Z. Cai, J. Zheng, Y. Dong, Angew. Chem. Int. Ed. 2020, 59, 16054.

T. Ma, T. Li, L. Zhou, X. Ma, J. Yin, X. Jiang, Nat. Commun. 2020, 11, 1811.

X. Ma, C. Xu, J. Wang, H. Tian, Angew. Chem. Int. Ed. 2018, 57, 10854.

Z. Ma, J. Zhou, J. Zhang, S. Zeng, H. Zhou, A.T. Smith, W. Wang, L. Sun, Z. Mater. Horiz. 2019, 6, 2003.

Y. Sun, J. Liu, X. Pang, X. Zhang, J. Zhuang, H. Zhang, C. Hu, M. Zheng, B. Lei, Y. Liu, J. Mater. Chem. C 2020, 8, 5744.

Z. Wang, Z. Zhang, L. Tao, N. Shen, B. Hu, L. Gong, J. Li, X. Chen, X. Huang, Angew. Chem. Int. Ed. 2019, 58, 9974.

Z. Wang, C. Zhu, Z. Wei, Y. Fan, M. Pan, Chem. Mater. 2020, 32, 841.

S. Yang, D. Wu, W. Gong, Q. Huang, H. Zhen, Q. Ling, Z. Lin, Chem. Sci. 2018, 9, 8975.

Y. Yang, Q. Li, X. Yang, W. Yang, R. An, T. Li, Y. Zhou, H. Zhang, J. Zhao, Z. Zhang, J. Mater. Chem. C 2020, 8, 3308.

S. Xu, W. Wang, H. Li, J. Zhang, R. Chen, S. Wang, C. Zheng, G. Xing, C. Song, W. Huang, Nat. Commun. 2020, 11, 4802.

J. Zhang, C. Pan, Y. Zhu, L. Zhao, H. He, X. Liu, J. Qu, Adv. Mater. 2018, 30, 1804644.

X. Zhang, J. Zhao, B. Chen, T. Sun, R. Ma, Y. Wang, H. Zhu, D. Peng, F. Wang, Adv. Opt. Mater. 2020, 8, 2000274.

AUTHOR BIOGRAPHIES

Xiaowei Yu is currently a Ph.D. student under the supervision of Prof. Jihong Yu at Jilin University. Her main research is focused on the design and synthesis of luminescence materials within zeolitic matrices and their application in optical fields.

Jihong Yu received her Ph.D. in inorganic chemistry from Jilin University in 1995 and worked as a postdoctoral fellow first at the Hong Kong University of Science and Technology and then at Tohoku University in Japan during 1996–1998. She has been a full Professor in the Chemistry Department, Jilin University, since 1999. Her main research interest is in the designed synthesis and application of zeolitic nanoporous materials in energy, environment, and other emerging fields.

How to cite this article: Yu X, Zhang H, Yu J. Luminescence anti-counterfeiting: From elementary to advanced. Aggregate. 2021;2:20–34. https://doi.org/10.1002/agt2.15