

Chameleonic Color-Variable Microbead Emitters for Multilevel Spectroscopic Encryption from Visible to Infrared Wavelengths

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Light emission mediated by whispering-gallery-mode (WGM) assumes distinct sharp resonant peaks, providing a unique spectral signature for photonic barcoding, that make them highly advantageous for compact microtagging systems. Most existing photonic barcodes are limited to visible wavelengths, restricting their tagging potential. Achieving precise control over light emission covering from the visible (VIS) to the near-infrared (NIR) region using “*a single object made of one kind of material*” has been a significant challenge. Here, we report a scalable, gram-scale synthesis of multicolor emitting carbonized polymer microspheres (MECPMs) via a facile hydrothermal process. These spherical microcavities, containing extended π -conjugated

polycyclic aromatic hydrocarbons (PAHs), exhibit broadband light emission spanning in the entire VIS-NIR wavelength region. Leveraging their excitation-dependent photoluminescence (PL), a single MECPM can show colour adaptable WGM-based white-light-emission (WLE) throughout the VIS-NIR region, generating both visible and invisible microbarcodes by simply adjusting the excitation wavelength. Compared with conventional polymer- or oxide-based microcavities, MECPMs demonstrate enhanced chemical stability against acids and organic solvents, remarkable spectral tunability, and powerful capabilities for photonic encryption. Their ecofriendly, metal-free “chameleonic” photonic functionality thus provides a robust platform for anti-counterfeiting, micro tagging and labelling, and next-generation photonic security systems. Our invention paves the way for innovative photonic encryption via information encoding and transmission across the VIS-NIR spectral regions.

1. Introduction

The rising demand for secure labeling and anti-counterfeiting solutions has led to significant advancements in miniaturized authentication technologies. Counterfeit goods pose a severe global economic challenge, resulting in financial losses of hundreds of billions of dollars annually while also undermining brand trust and consumer safety.^[1-3] To combat this issue, photoluminescent barcoding has emerged as a promising approach, enabling rapid, reliable identification through visible or near-infrared (NIR) emissions.^[4, 5] Various materials, including organic dyes,^[6, 7] rare-earth (RE) doped phosphors,^[8, 9] polymer microparticles,^[4, 10] RE-coordinated polymers and metal organic framework (MOF),^[11-14] quantum dots (QDs),^[15] carbon dots (CDs),^[16] and inorganic nanostructures,^[17] have been explored for barcode generation. Organic dyes are cost-effective and widely available but suffer from broad emission profiles, making them susceptible to counterfeiting.^[18-20] Semiconductor-based barcoding, particularly using one-dimensional segmented structures with tunable optical properties, which still suffers with broad emission spectrum and limited barcoding capacity.^[21-23] However, their toxicity and potential energy transfer issues when densely packed limit their applicability. RE-based luminescent materials, known for their photostability, long fluorescence lifetimes, and distinct emission lines, have also been utilized.^[9, 24] Despite these advantages, their relatively simple optical signatures make them vulnerable to duplication, and their reliance on RE elements raises concerns regarding sustainability and toxicity.^[21, 25, 26]

Photonic barcodes, leveraging optical resonances to encode data via distinct spectral signatures, present a powerful alternative for secure **microlabeling**.^[27, 28] These barcodes operate by precisely controlling the resonance frequencies of microresonators, enabling unique optical signals that are challenging to replicate.^[29] Among these, whispering-gallery-mode (WGM) microcavities stand out due to their exceptional light confinement properties. The curved geometry of these microstructures facilitates total internal reflection (TIR), generating well-defined optical resonances observable as sharp peaks in the photoluminescence (PL) spectrum through transverse electric (TE) or transverse magnetic (TM) modes.^[30, 31] Each mode serves as an individual “bar” in the barcode, allowing the generation of highly complex, miniaturized microbarcodes. Even slight variations in microcavity size, often below the diffraction limit, can induce detectable spectral shifts, leading to distinctive and complex optical signatures capable of carrying encoded barcode’s information.^[32] These amplified PL characteristics serve as unique identifiers when tagged on materials, making them highly suitable for unclonable anti-counterfeiting applications. Traditionally, fluorescent dyes,^[33-35] semiconductor quantum dots,^[36] π -conjugated polymers,^[37, 38] carbonized polymer microspheres,^[39] and nanographene ^[40] are employed as gain materials within spherical microcavities. **Recently, a strategy was developed to construct dual-stimuli-responsive photonic barcodes based on organic molecules and perovskite quantum dot (PQD)-doped polymer WGM microcavities for covert and overt anti-counterfeiting via light-controlled acidichromism or swelling-deswelling mechanisms.**^[7, 41] However, most photonic microbarcodes reported so far are limited to the visible wavelength region. Developing microcavities that emit across the *visible to near-infrared* (VIS-NIR) spectrum remains an ambitious challenge, that liberate us from relying on toxic narrow-bandgap semiconductors and synthetic metal complexes.

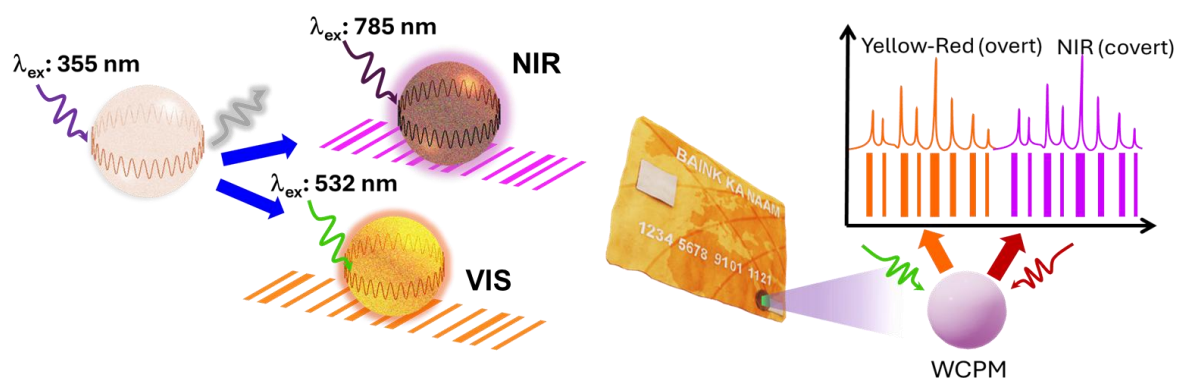


Figure 1. Schematic illustration of multilayer encrypted microbarcodes generation, enabling both visible to NIR photonics encoding for secure product authentication.

To address these these serious limitations, we propose a novel “Chameleon” microcavity architecture. This system allows the optical resonances of a single material to be dynamically tuned across the VIS-NIR range simply by varying the excitation wavelength. This concept draws inspiration from chameleons, which achieve dynamic color changes through the vertical arrangement of multiple pigment cells in their skin, responding to external factors such as light intensity, temperature, humidity, and even emotional states.^[42, 43] Similarly, this Chameleon-inspired microcavity enables the generation of encrypted photonic microbarcodes that function in both visible (overt) and near-infrared (covert) spectral regions. This approach introduces a promising direction for multilevel optical tagging and anti-counterfeiting technologies, an unexplored area with considerably rich application and high security.

The “Chameleon” microcavity based on white light emitting carbonized polymer microspheres (WCPM) are produced on a gram scale through a simple, hydrothermal synthesis process involving the crosslinking of natural peptides. During the formation, extended π -conjugated polycyclic aromatic hydrocarbons (PAHs) become embedded within the structure, enhancing their PL and enabling broadband emission from the VIS-NIR regions. This WCPM-based microresonator functions like optical “chameleons”, with tunable emission color that shift across the VIS-NIR range simply by changing the excitation wavelength. Furthermore, each microresonator can produce distinct emission peaks that serve as unique spectroscopic “fingerprints” serving as photonic barcode. Unlike traditional quantum dot- or dye-doped microresonators, a single WCPM microresonator can simultaneously generate both visible (overt) and NIR (covert) photonic barcodes (**Figure 1**). This dual-mode emission significantly improves data encryption and enables versatile, doubly secured secure information encoding that is invisible to the naked eyes. **This unique photonic behavior, combined with the random size distribution of the microbeads, generates complex and unpredictable spectral fingerprints, making the resulting optical tags extremely difficult to replicate compared with conventional fluorescent ink-based microcavities. Moreover, this WCPM-based system offers a rare-earth- and metal-free platform that is scalable, reliable, and well-suited for next-generation anti-counterfeiting and photonic security applications.**

2. Results and Discussion

The white light emitting (WLE) carbonized polymer microspheres (WCPM) were synthesized via hydrothermal techniques using citric acid (CA), natural peptide ϵ -poly-L-lysine (EPL), and perylenetetracarboxylic dianhydride (PTCDA) in aqueous solution (**Figure S1**). The -COOH

groups from CA and -NH₂ groups from natural peptides crosslinked to form spherical architectures, resulting in light yellow-colored carbonized polymer microsphere (YCPM).^[10] Under UV excitation, YCPM exhibited strong cyan emission and emitted multicolor light across the visible spectrum from cyan to red, depending on the excitation wavelength. To enhance yellow and red-light emission more and extend emission further into the NIR region, commercially available PTCDA dye molecules were introduced during synthesis. During the reaction, PTCDA converted to perylenetetracarboxylic diimide (PTCDI), extending π conjugation through di- and trimerization processes, narrowing the band gap, enhancing NIR absorbance and emission.^[44, 45]

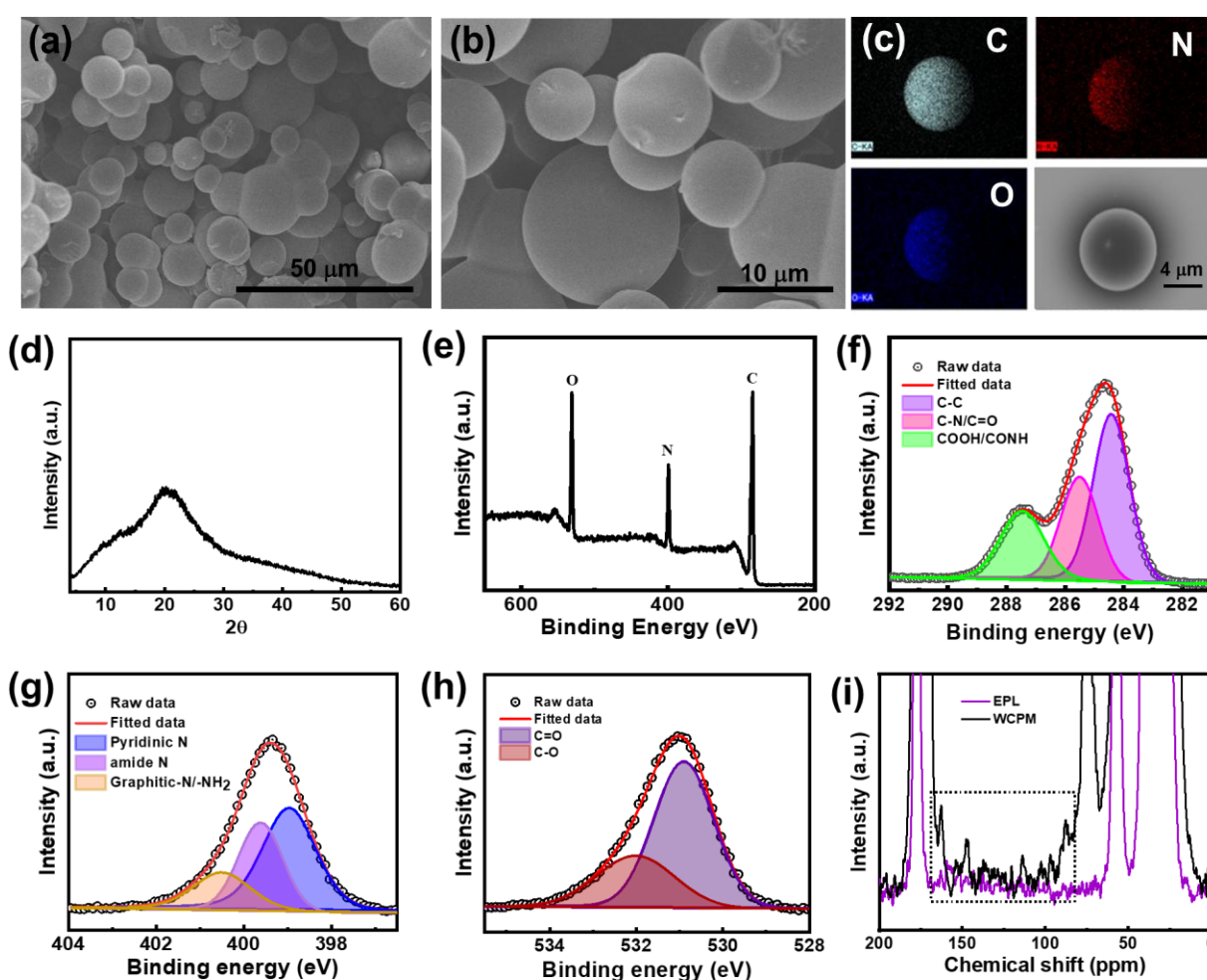


Figure 2. (a-b) low to high magnified SEM images of the WCPM microbead. (c) EDS mapping of single WCPM microbead. (d and e) XRD and XPS survey spectra of WCPM. (f-h) Deconvoluted HRXPS spectra of C, N, and O, respectively. (i) Magnified solid-state ¹³C CPMAS NMR spectra of WCPM and compared with the EPL.

Figure 2 a-b show the SEM images of WCPM and it shows the microbeads diameter range from 5-15 μ m and smooth surface similar to YCPM (**Figure S2**). The Energy-dispersive X-ray

spectroscopy (EDS) of a single microbead indicate that it is composed of mainly carbon (C), oxygen (O), and nitrogen (N) (**Figure 2c**). **Figure 2d** shows X-ray diffraction (XRD) pattern of the microbeads and shows a broad peak at 20 degree at 2θ indicating the formation of amorphous structures after the crosslinking reaction with EPL peptide. The X-ray photoelectron (XPS) survey spectra, providing insights into elemental composition (**Figure 2e**). **The XPS survey spectra indicate that the surface atomic composition consists of approximately 68.8% C, 13.55 % N, and 17.65% O.** The deconvoluted HRXPS spectrum of WCPM reveals various functional groups. The C-1s spectrum shows three distinct peaks: C-C/C=C, C-N/C-O, and COOH/CONH (**Figure 2f**).^[39, 46] The N 1s spectrum features peaks at 399.0 eV (C-NH₂, C-N-C), 399.7 eV (amide), and 401.0 eV (amine/graphitic N), indicating presence of pyridinic and graphitic nitrogen along with amide, and amine bonds (**Figure 2g**).^[46-48] In the O 1s region, peaks at 531.3 eV and 532.8 eV correspond to N-C=O and O-C=O bonds, confirming oxygen functionalities in WCPM (**Figure 2h**). The solid-state nuclear magnetic resonance (ssNMR) characterization using ¹³C revealed distinctive features in the magnified cross-polarization, magic angle spinning (CPMAS) NMR spectra of pure EPL and WCPM (**Figure 2i**). In the ¹³C CPMAS NMR spectra comparing WCPM to EPL raw materials, notable peaks appeared around ≈ 29 and 40 ppm, corresponding to sp³-hybridized CH₂ sequences in aliphatic carbons, which differed from EPL.^[49] Additionally, a broad peak at 173 ppm indicated a cross-linked structure involving -COOH groups from CA and -NH₂ groups from EPL, contributing to the formation of WCPM structures (**Figure S3**). The broadening observed in the aliphatic carbon (CH₂) spectral profile supports the of cross-linking between CA and EPL, while the upfield shifts of the amorphous phase reflect conformational heterogeneity.^[50] Additionally, new signals appearing at 114 ppm suggest the presence of sp²-hybridized carbon atoms associated with PAHs. A lower chemical shift at 88 ppm points to =CH carbons, potentially influenced by the electron-withdrawing effects of two nitrogen atoms located at a two-bond distance from the imide bond within PTCDI. Furthermore, the ¹³C CP-MAS NMR spectra reveal sharp peaks at approximately 148 and 163 ppm, likely attributed to sp²-hybridized carbon atoms linked to cyclic imide bonds originating from PTCDI.^[51] The ¹⁵N CPM NMR also reveals a broad peak at -263 ppm, indicative of the amide bonding configuration (**Figure S4**).^[52, 53] These characteristics indicated the conversion of PTCDI from PTCDA dye molecules, highlighting the structural transformations observed in the WCPM material.

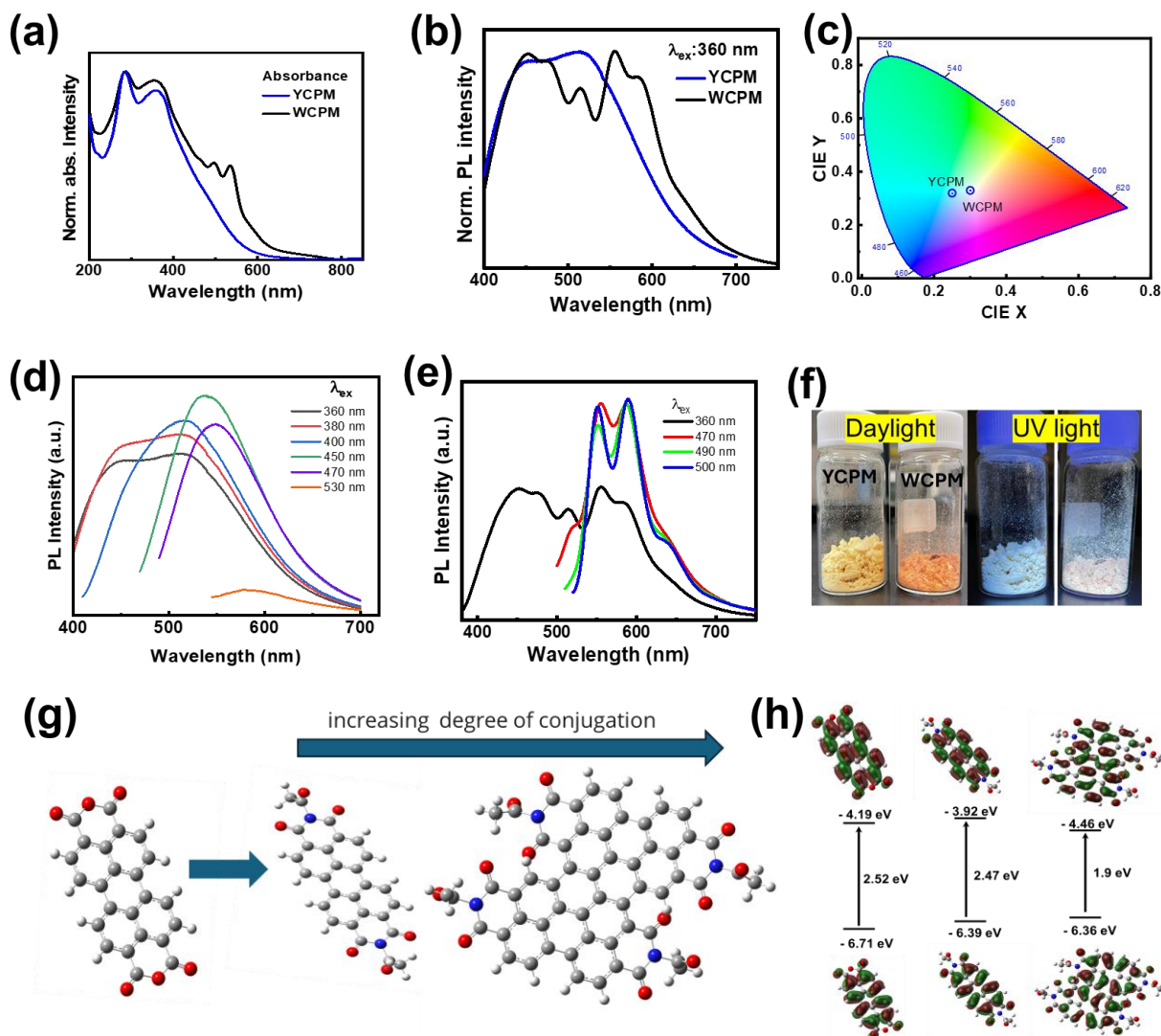


Figure 3. (a) UV-vis-NIR absorbance spectra comparing YCPM and WCPM. (b and c) Comparative PL spectra and corresponding CIE diagram of WCPM and YCPM. (d and e) Excitation wavelength-dependent PL spectra of YCPM and WCPM. (f) Photographs of WCPM and YCPM under daylight and 365 nm UV light. (g) Schematic illustration of the in-situ formation of PAHs from PTCDA within the WCPM matrix. (h) DFT calculation results for PAHs model with increasing conjugation, showing computed energy diagrams of HOMO-LUMO energy gaps.

Figure 3a shows a comparative analysis of the absorbance spectra of YCPM and WCPM across the ultraviolet-visible to near-infrared (UV-VIS-NIR) wavelength range. The YCPM primarily absorbs in the UV region, exhibiting distinct peaks at 280 nm and around 370 nm. These peaks correspond to π - π^* and n - π^* electronic transitions associated with C-O and C-N bonding structures, which originate from blue light-emitting molecular fluorophores formed inside the

microspheres. Additionally, a lower-energy absorption band is observed, attributed to $n-\pi^*$ transitions involving C=O and C=N bonds present in the cross-linked amide structures.^[54-56] In contrast, the WCPM exhibits new, strong absorption peaks at 500 and 536 nm, confirming the incorporation of PTCDI fluorophores into the microstructures. Furthermore, additional weak absorption bands extending into the red to NIR wavelength region are observed in the tail of the spectrum compared with YCPM. Recent studies have reported that the formation of larger PAHs through PTCDI-based di- and trimerization processes leads to VIS-NIR absorbance in CD-based nanostructures.^[44, 45] This extended π -conjugation in PTCDI-derived PAHs contributes to the enhanced VIS-NIR absorbance and emission observed in WCPM compared with YCPM. The UV-excited emission spectra of WCPM exhibit an extended emission range from yellow to red wavelengths, leading to WLE, in contrast to YCPM (**Figure 3b**). Under UV excitation, YCPM emits cyan light, while WCPM exhibits WLE, as confirmed by emission spectra and the International Commission on Illumination (CIE) coordinates. The CIE coordinate for WCPM is (0.30, 0.33), as shown in Figure 3c corresponding to WLE. YCPM shows excitation wavelength-dependent emission, where the intensity of visible light emission at longer wavelengths gradually decreases as the excitation wavelength increases (Figure 3d). The WCPM exhibits a broad emission under UV (360 nm) excitation compared to the YCPM, resulting in single-component WLE (**Figure 3e**). Moreover, the WCPM also displays excitation wavelength-dependent emission across the VIS-NIR region when the excitation wavelength is varied (**Figure S5**). Interestingly, in the excitation range of approximately 470-500 nm, the emission becomes excitation-independent, showing strong yellow and red emissions peaks at 551 nm, 588 nm, and 636 nm, respectively (**Figure 3e** and **Figure S5**). These spectral features indicate the *in situ* chemical transformation of PTCDA into PTCDI-based molecular fluorophore within the microspheres during their formation (**Figure S6**). **Control experiments revealed that the hydrothermal reaction between PTCDA and EPL led to the in-situ formation of yellow-emissive PTCDI-based molecular fluorophores through chemical transformation. These fluorophores exhibited broad absorption bands centered at 496 and 526 nm and displayed excitation-independent yellow-to-red emission with characteristic peaks at 537, 577, and 629 nm under UV excitation (Figures S7). The PL quantum yield (PLQY) of WCPM in the solid-state is approximately 30 % by 500 nm excitation (Figure S8) at solid state, whereas PTCDA molecules alone exhibit emission with a PLQY $\sim$ 3% at N-methyl-2-pyrrolidone (NMP) solution and < 1 % at solid state.^[57] The fluorescence lifetime imaging microscopy (FLIM) analysis of individual WCPM microbeads was performed under two different excitation wavelengths (405 and 500 nm) from the same position (Figure S9). The results revealed**

average decay times (τ_{av}) of approximately 4.92 ns and 2.55 ns, respectively, with a narrow lifetime distribution, indicating uniform optical characteristics of the microbeads. The time-resolved photoluminescence (TRPL) spectra of PTCDI-EPL in the colloidal state exhibit τ_{av} of 3.13 and 3.21 ns for the emission peaks at 540 nm and 580 nm, respectively, under 361 nm laser excitation. Remarkably, these lifetimes increase to 30.4 and 20.4 ns in the WCPM system (**Figure S10**). This significant enhancement can be attributed to the spatial confinement of PTCDI-based fluorophores within the microcavity, which effectively suppresses aggregation-induced quenching and π - π energy transfer and promotes higher PLQY in the solid state.^[58] Due to the incorporation of PTCDI-based PAHs within the microbeads facilitates Förster resonance energy transfer (FRET) due to their close packing, enabling efficient energy transfer and resulting in the observed WLE in WCPM. The TRPL spectra of blue emission peak (~ 454 nm), τ_{av} of YCPM decreases from 4.80 ns to 2.96 ns for the case WCPM, indicating ~ 38 % of FRET energy transfer from YCPM based framework to PAHs during the UV light excitation (**Figure S11**).^[33] A digital photograph of YCPM and WCPM under daylight shows a noticeable color shift, with YCPM appearing light yellow and WCPM appearing reddish color materials and emit cyan and WLE under 365 nm UV excitation (**Figure 3f**). **Figure 3g** shows the schematic representation of the formation of PTCDI-based PAHs. This process extends the π -conjugation through dimerization and trimerization and confined inside the microstructures, resulting in material with unique “chameleon-like” emission behavior from VIS-NIR wavelength region by varying the excitation light source. To gain deeper insight into the optical behavior, Density Functional Theory (DFT) calculations are being carried out on PAH structures. These calculations include geometric optimization and energy level analysis, revealing HOMO-LUMO energy gaps of 2.47 eV and 1.90 eV, respectively (**Figure 3h**). The experimental and computational results clearly show a red shift in the absorbance spectra, indicating that the WCPM can absorb light at longer wavelengths, from the visible to the NIR region. This behavior comes from the formation of PTCDI-based fluorophores and extended PAH structures inside the microbeads. These species act as light-emitting centers with different energy gaps, leading to excitation wavelength-dependent PL across the VIS-NIR range. The visible emission mainly originates from isolated PTCDI-like fluorophores, while the NIR emission is produced by larger PAH networks with extended π -conjugation.

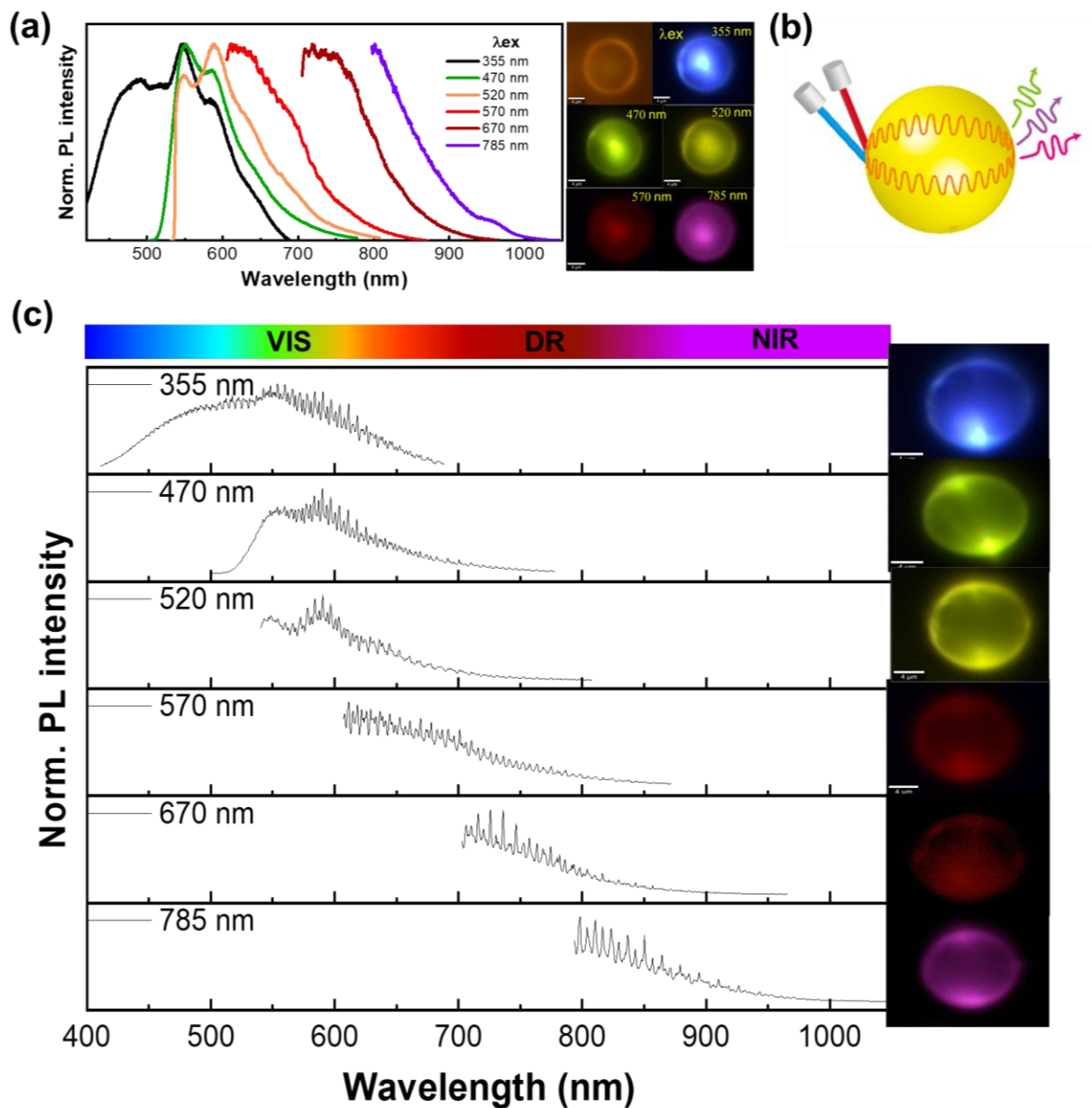


Figure 4. (a) Excitation wavelength-dependent μ -PL spectra from VIS-NIR wavelength from a single microbead and corresponding μ -PL images excited with UV-VIS-NIR laser sources at their center. (b) Schematic illustration of chameleon microcavity. (c) Wavelength adaptable microresonator behavior of a microbead via changing the excitation laser source via excitation at their right side and show the multicolor of WLE to deep red-NIR, and NIR emission (scale bar 4 μ m).

The μ -PL properties of individual microsphere were investigated by varying the excitation wavelength across the UV-vis-NIR spectrum. When the same WCPM microbead was exciting using different wavelengths (355, 470, 520, 570, 670, and 785 nm), a broad range of light

emissions was observed, spanning from the visible region to deep red (DR)-NIR and the NIR wavelength range. **Figure 4a** shows the μ -PL spectra and images of the microbead when excited at various laser wavelengths at their center. The corresponding μ -PL spectra demonstrates wavelength-adaptable light emission from the visible to DR-NIR and the NIR region. The CIE coordinates indicate that individual microbeads exhibit WLE under UV excitation, with the color gradually changing from green to yellow, red, deep red (DR), DR-NIR, and NIR (**Figure S12**). In contrast, the comparative μ -PL spectra of YCPM microbeads show orange emission when excited with a 532 nm laser, with an emission center wavelength at 600 nm (**Figure S13**). On the other hand, WCPM microbead displays three distinct emission centers at 545 nm, 590 nm, and 636 nm, attributed to the formation of PTCDI-based PAHs integrated into microbeads. This μ -PL spectra of individual WCPM microbead exhibit color-adaptable emission from the visible to NIR region, depending on the excitation wavelength, demonstrating a chameleon-like fluorescence behavior (**Figure 4b**). **Notably, when a single microbead (diameter $\sim 12\ \mu\text{m}$) is excited at the edges, WGM-based light amplification is observed across the entire visible spectrum (including white, green, yellow, red, and deep red-NIR) and into the NIR region.** This light amplification is achieved simply by changing the excitation laser wavelength, without modifying the microcavity itself, solely changing its color flexibly like a chameleon (**Figure 4c**). Multiple microbeads ($7.2\ \mu\text{m}$ and $9.8\ \mu\text{m}$) exhibit chameleon-like microresonator behaviors, where the WGM emission patterns are strongly dependent on the bead diameter (**Figure S14**). In contrast, the YCPM microresonator's emission is restricted to the shorter visible wavelength region (**Figure S15**). Surprisingly, unlike organic polymer-based microresonators, the WCPM microbeads exhibit high stability against organic solvents, acid treatments, and prolonged laser exposure (**Figure S16**). Overall, this eco-friendly, rare-earth-metal-free spherical microarchitecture, composed of a single material, exhibits color-adaptable WGM-mode emission spanning the entire VIS-NIR spectral region. This performance significantly surpasses that of previously reported spectral microresonators (**Table S1**), whose emissions are typically limited by doped resonant PL enhancement due to WGM confinement and confined to either the visible or NIR range. This chameleon-like microresonance, combined with high stability, makes these materials highly promising for practical use in encrypted photonic microbarcodes and secure authentication applications.

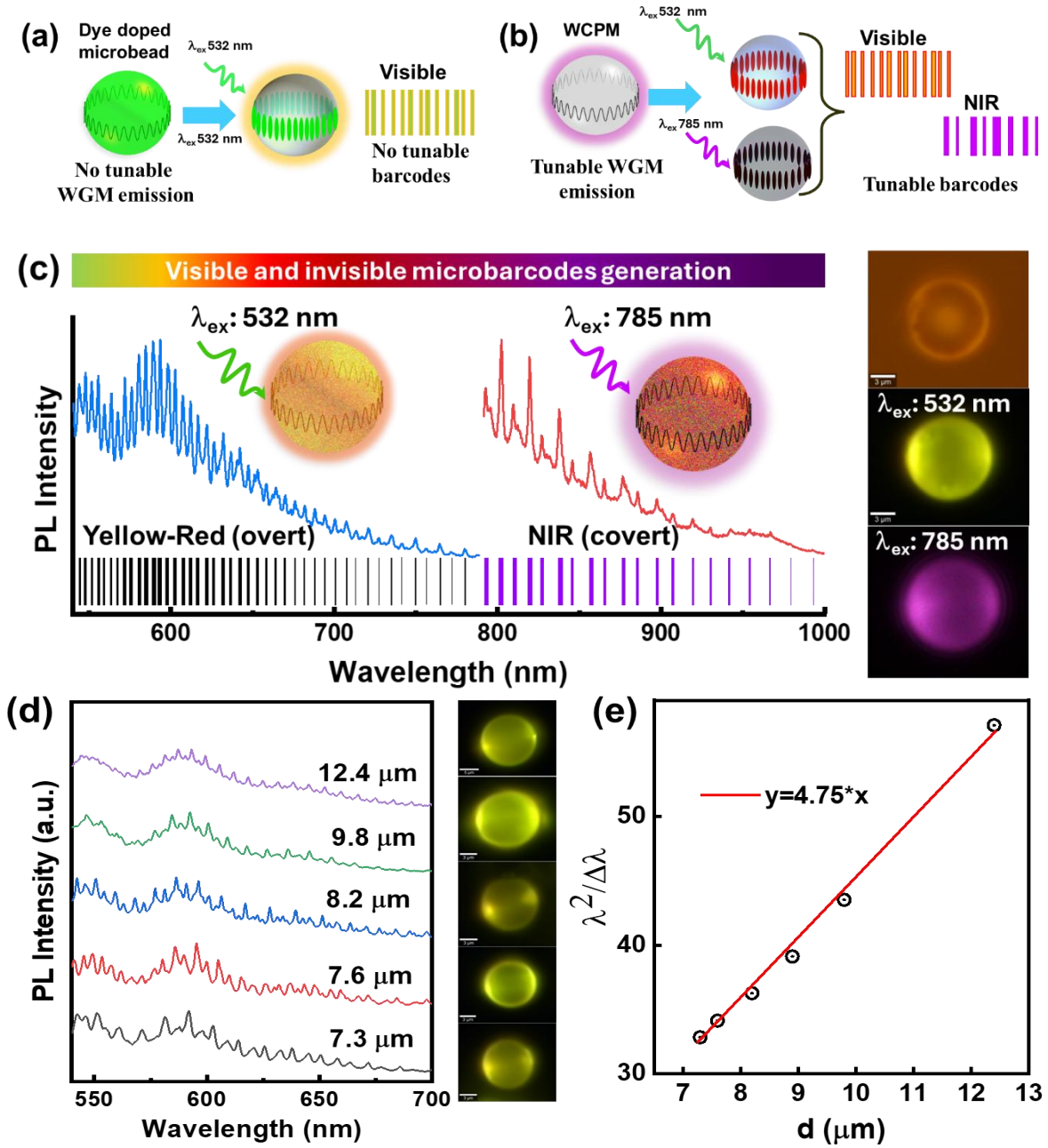


Figure 5. (a and b) Comparison of schematic illustration of visible microbarcodes generated by conventional dye-doped microresonators versus visible and invisible microbarcodes by WGM-based microresonators. (c) Vis-NIR WGM-assisted microbarcode generation, along with corresponding bright-field optical images and m-PL images obtained using 532 nm and 785 nm laser excitation of same microresonators. (d) WGM emissions from individual WCPM microbeads of varying sizes, along with their corresponding μ -PL images by 532 nm excitation (scale bar 3 μ m). (e) Correlation between $\lambda^2/\Delta\lambda$ and the d of microcavity. Correlation between $\lambda^2/\Delta\lambda$ and the d of microcavity and corresponding linear fit to the function $\lambda^2/\Delta\lambda = n_{eff}\pi d$ (redline).

Each synthesized microbead exhibits excitation-dependent emission spanning the visible to NIR range when excited by lasers (**Figure 4**). This property enables the generation of both overt (visible) and covert (invisible) microbarcodes using WGM resonance under different laser excitations. **The microbeads' high sphericity, smooth surface, and high refractive index allow efficient light confinement through total internal reflection, forming TE and TM optical modes.** These modes generate unique spectral barcodes, offering novel microscale optical tagging-in contrast to conventional 1D barcodes, which require larger centimeter-scale dimensions. Typically, conventional dye-doped (rhodamine 6G) polystyrene microresonators generate a single microbarcode corresponding to the emission wavelength of the gain medium (**Figure 5a** and **Figure S17**). In contrast, the WCPM exhibit multiple microbarcodes spanning both visible and NIR regions (**Figure 5b**). This VIS-NIR emissive fingerprints allows them to function as overt and covert barcodes for secure product tagging. **Figure 5c** shows the WGM emission in the visible to the NIR wavelengths, as well as the corresponding photonic microbarcode patterns obtained from the same single WCPM microbead when excited at 532 nm and 785 nm. The μ -PL images in both VIS and NIR wavelength regions show the bright ring patterns confirm light confinement at their perimeter due to WGM resonance at both wavelength regions. This approach enables the generation of dual VIS and invisible microbarcodes from individual microbeads. **The simplest expression for the WGM resonance positions is given by $m\lambda = n_{\text{eff}}\pi d$, which corresponds to the first-order term of the asymptotic expansion of the Mie solution (Airy function approximation),**^[59, 60] where m is an integer, λ the resonant wavelength, n_{eff} the effective refractive index of the resonant mode, and d the microcavity diameter.^[27, 61, 62] WGM emission occurs when the optical path length matches an integer multiple of λ . Each microbead, with its unique size and fixed refractive index, exhibits distinct sharp peaks, forming a spectral fingerprint. This WGM-regulated emission spectrum enables the identification of microbeads, that functioning similar to barcodes used for product recognition, making them ideal for unclonable anti-counterfeiting applications. **Figure 5d** shows the optical modulation of WGM resonance, where spectral variations correspond to differences in microbead size, along with their respective μ -PL images under 532 nm laser excitation at the edges. The free spectral range (FSR), defined as the wavelength difference between optical modes (TE/TM), is inversely related to the microresonator diameter.^[30] As the microcavity diameter increases from 7.3 μm to 12.4 μm , the FSR gradually decreases from 10.6 nm to 6.1 nm. **Figure 5e** presents the linear relationship between $\lambda^2/\Delta\lambda$ and d , with a fitted slope of $n_{\text{eff}}\pi = 4.75$. Given $\pi = 3.14$, the effective refractive index n_{eff} is determined to be 1.51. By varying the microcavity diameter, a wide range of unique barcodes can be generated. Moreover, the coding capacity/security is significantly

enhanced by extending the emission spectrum from vis-NIR through excitation wavelength tuning. This capability, which is not available in traditional dye-doped microcavities, allows for more versatile and high-density encoding.

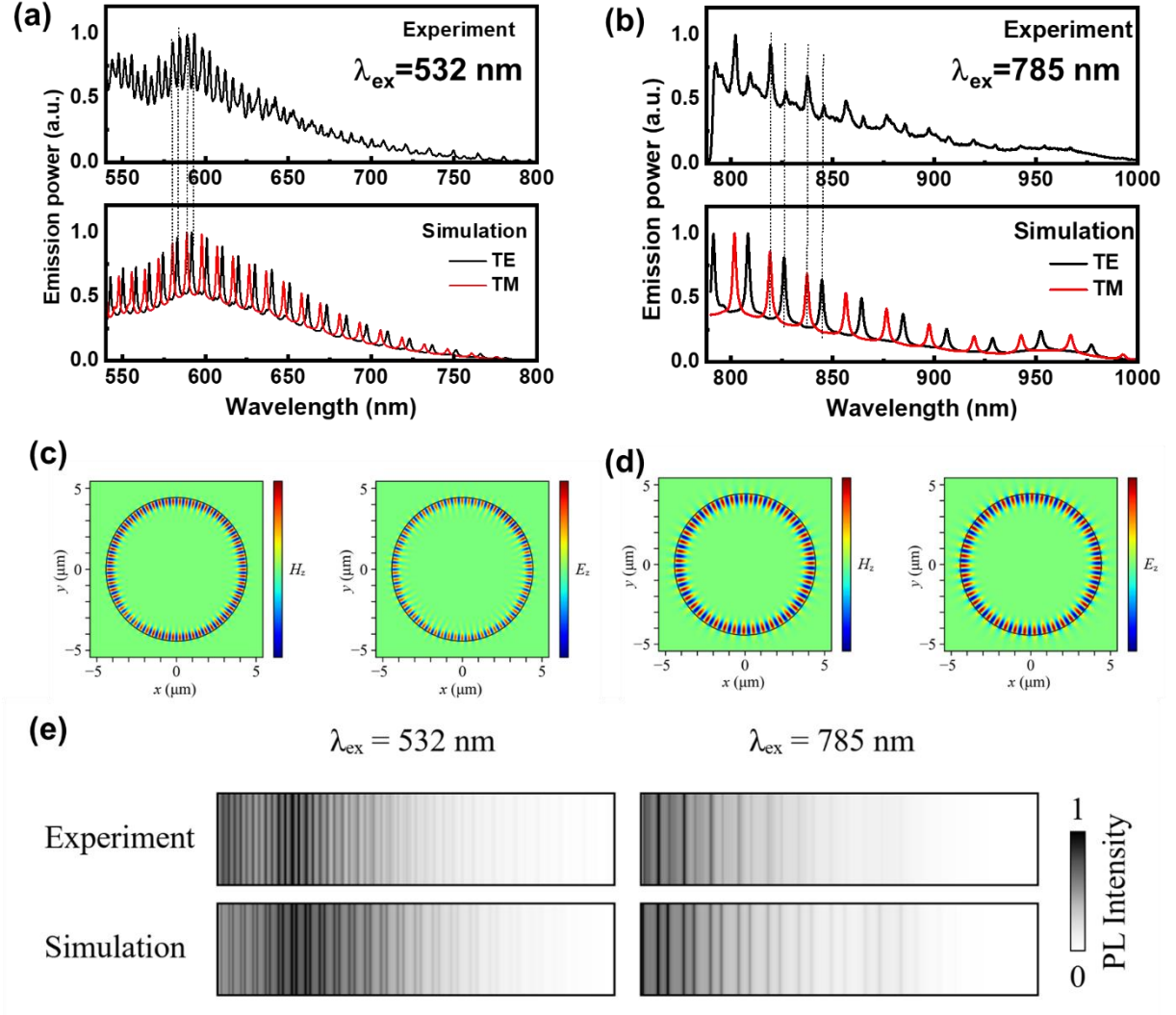


Figure 6. (a,b) Experimental and simulated μ -PL spectra of an individual WCPM ~ 8.9 μm microbeads excited at 532 and 785 nm. (c-d) Electromagnetic field distributions of TE and TM resonances in the VIS and NIR wavelength region. Comparison of wavelength-adaptable photonic barcodes for experiment (upper) and simulation (lower). (e) Comparison of visible and invisible wavelength-adaptable photonic microbarcodes for experiment (upper) and simulation (lower).

The μ -PL spectra across two distinct spectral regions, along with the corresponding field profiles, were simulated using the finite-difference time-domain (FDTD) method.^[16] The emission from the WCPM was modeled as a dipole source placed near the edge of the sphere, and the optical power was monitored via a transmission box enclosing the dipole. The power obtained, normalized to the source power, represents the emission rate enhancement due to the WGM resonance effect. The enhanced WGM emission spectra were then obtained by multiplying this factor by the background emission. The simulated WGM resonances in the yellow red (visible) and wavelength regions, along with background emissions from 8.9- μ m WCPM microbeads, were analyzed under excitation with two different laser wavelengths. The simulated spectra exhibit strong agreement with the experimentally measured emission spectra obtained under 532 nm and 785 nm laser excitation (**Figure 6 a, b**). Here, the broad fluorescence envelope corresponds to the background spectrum observed in the experiment when the excitation spot was focused at the center of the sphere. These results confirm the presence of both TE and TM modes within the WCPM microbeads due to the strong light confinement. The variation in optical path lengths for TE and TM modes naturally leads to differences in mode separation, resulting in nonuniform spectral spacing of the resonant peaks. Note that the linewidth of the simulated peaks depends on the extinction coefficient k of the WCPM. Accordingly, the k value was determined such that the linewidth in the simulation matched that in the experiment, yielding $k = 0.002$. **Figure 6c** show the mode profiles at WGM resonance peaks of 591.92 nm and 597.91 nm, showing the H_z field distribution for the TE mode and the E_z field distribution for the TM mode, respectively. These profiles highlight the fundamental TE and TM mode characteristics with a radial mode number of 1. Similarly, **Figure 6d** show the mode profiles at WGM resonance peaks in the NIR region (808.60 nm and 819.25 nm), represent the H_z field distribution for the TE mode and the E_z field distribution for the TM mode. Both the experimental and simulated WGM emission patterns demonstrate the potential for constructing photonic barcodes in the visible and NIR spectral regions (**Figure 6e**). This tunability is achieved by simply varying the excitation wavelength from 532 nm to 785 nm, offering a powerful pathway for advanced photonic security and encryption applications.

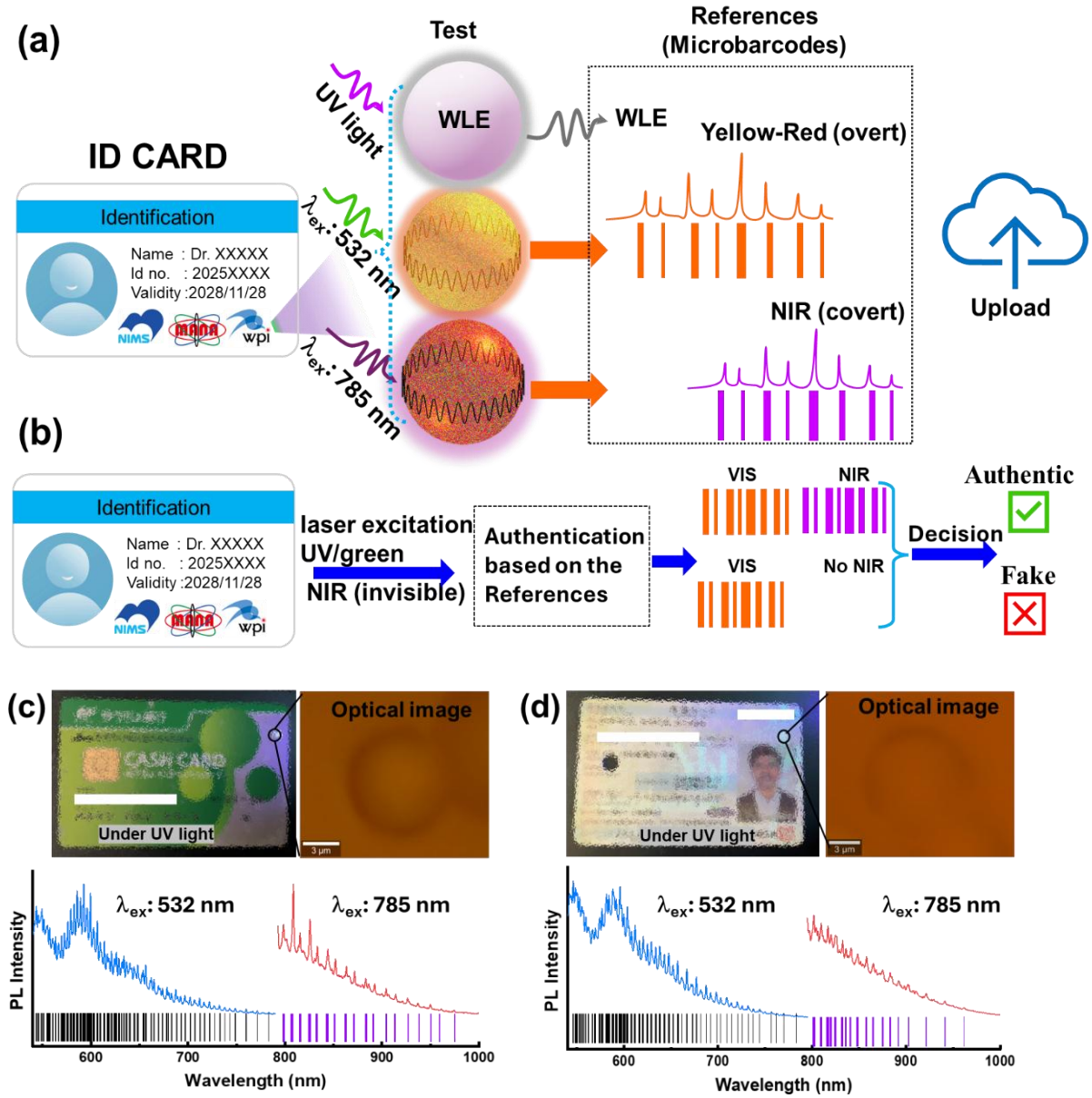


Figure 7. (a and b) Schematic diagram of the potential application of WCPM based encrypted photonics architecture with vis and NIR microbarcodes generation for secure product tagging application. (c and d) Simple demonstration of VIS-NIR microbarcode generation from single microbeads for authentication on a tagged bank card and resident card by varying the excitation wavelength between 532 nm (VIS) and 785 nm (NIR).

Figure 7a and **b** illustrate a proposed application of encrypted visible and invisible barcode generation using these chameleon-like WCPM microcavities for product tagging and authentication in information security. These microbeads are embedded within an identity card, debit/credit card, serving as a unique identification tag that is nearly impossible to replicate due to the distinct barcode patterns they produce. To verify authenticity, three tests are conducted

by varying the excitation source: UV light, a visible 532 nm laser, and an invisible 785 nm laser. A single microbead generates corresponding barcodes in the visible and NIR regions, as shown in **Figure 7a**. The recorded barcode data is securely uploaded to the cloud as a reference. For authentication, an unknown identity card undergoes the same three tests. If the microbeads in the card emit white light and generate the expected visible and NIR barcodes that match the reference data, the card is confirmed as authentic. Otherwise, it is identified as counterfeit (**Figure 7b**). As a proof of concept, we have integrated WCPM microbeads into a used real bank card and a used Japanese residence card (**Figure 7c and d**). In both cases, under UV illumination, 532 nm and 785 nm laser excitation (**Figure S18**), the microbeads exhibit white light emission and produced distinct visible and NIR spectral barcodes due to WGM, confirming the authenticity of the cards. This microbead-based security system, capable of generating both visible and invisible microbarcodes, significantly enhances security down to the microscale region. It offers a promising and powerful approach for advanced anti-counterfeiting measures, addressing global security challenges.

3. Conclusion

In this work, we developed white-light-emitting carbonized polymer microsphere (WCPM)-based WGM microresonators, representing a significant advancement in photonic barcoding for secure labeling and multilevel anti-counterfeiting applications. The WCPM design enables a scalable, gram-scale, metal-free synthesis with strong PL, low toxicity, and excellent chemical stability against acids and organic solvents, setting them apart from conventional polymer- or oxide-based microcavities. The extended π -conjugation in WCPMs allows broadband emission spanning the VIS-NIR region. Compared with traditional quantum dots and dye-doped microresonators, WCPMs demonstrate superior spectral range, stability, and chemical robustness. By utilizing the excitation-wavelength-dependent, color-variable emissions of the microbeads, both visible (overt) and NIR (covert) microbarcodes can be generated, enabling complex and tunable optical signatures for enhanced security. By incorporating these microresonators into multilevel encrypted microbarcode systems, we establish a secure and versatile anti-counterfeiting strategy. Encoding information across multiple spectral regions adds an additional layer of encryption, making unauthorized replication highly challenging. Overall, this work advances photonic security technologies and offers a practical, sustainable platform for real-world applications.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Whispering-gallery-mode microresonators with sharp resonant peaks enable photonic barcoding. Inspired by chameleon skin, a rare-earth-free microcavity integrates π -conjugated polycyclic aromatic hydrocarbons (PAHs), achieving VIS-NIR optical resonance in a single microbead. Dynamic microbarcode shifts via excitation changes enhance encrypted optical tagging and anti-counterfeiting, advancing photonic security with visible and invisible labeling for next-generation secure authentication.

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Chameleonic Color-Variable Microbead Emitters for Multilevel Spectroscopic Encryption from Visible to Infrared Wavelengths

