Research article

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Stretchable and self-healable organometal halide perovskite nanocrystal-embedded polymer gels with enhanced luminescence stability

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Abstract: Stretchable and self-healing polymer gels with luminescent property are very promising materials for next generation soft optical devices. This work presents the preparation of self-healing and luminescent polymer gels by simply blending organometal halide perovskite nanocrystals (OHP NCs) with poly(dimethylsiloxane)-urea copolymer (PDMS-urea). On the one hand, the obtained luminescent gels are not only flexible, stretchable and relatively transparent, they also exhibit excellent self-healing capability due to the reversible hydrogen bonding network in the PDMS-urea copolymer. On the other hand, the embedding of OHP NCs (MAPbBr₃ and MAPbI₃ NCs) inside the hydrophobic PDMS-urea gel greatly improved the photoluminescence stability of OHP NCs against water. Their applications as phosphors for LEDs have been demonstrated. Both the MAPbBr₃/PDMS-urea gel and MAPbI₃/PDMS-urea gel can fully convert the blue emission of GaN chip to green and red

emissions, respectively. These gels can be used as photoluminescent materials in flexible optical devices with good self-healing capability.

Keywords: luminescent; organometal halide perovskite nanocrystals; poly(dimethylsiloxane)-urea gel; self-healable; soft optical devices.

1 Introduction

Over the past few years, organometal halide perovskite nanocrystals (OHP NCs) have received great interest because of their unique properties, such as high photoluminescence efficiency, tunable emission wavelength in the visible light region, narrow emission bandwidth and easy processability [1–3]. This makes perovskite NCs very promising photoelectronic materials in light-emitting diodes, NC lasers and solar cells [4–6]. However, the inferior stability of HP NCs, which are moisture- and air-sensitive, has set a great challenge to their practical applications [7, 8]. To address the stability issue, a variety of methods, including protective coating [9, 10], alternative capping ligands other than oleic acid (OA)/oleylamine (OLAm) [11, 12] and polymer blends [13–16], have been employed with significant success achieved. For example, Huang's group reported the improved stability of MAPbBr₃ NCs through the protective coating of silica oxide [10]. Wang et al. utilised an alkyl phosphonic acid instead of oleic acid as ligand to grow phase-stable cubic perovskite NCs [11]. Luo's et al. group chose branched capping ligands as passivators and stabilisers for perovskite NCs [12]. Compared to the approaches based on the protective coatings and different capping ligands, blending with polymers to improve the stability of OHP NCs is much easier to handle [17]. The blends of the CsPbBr₃ NCs with the PMMA, PS and PBMA prepared by a one-pot strategy could maintain their quantum

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yield for more than 1 month in air [13]. Wang et al. prepared MAPbBr₃ NC/polymer composite films through the swelling-deswelling microencapsulation strategy and the resulting films showed ultrahigh stability against water and heat [15]. After boiling the samples in water for 30 min, the photoluminescence quantum yields (PLQYs) of the MAPbBr₃/PC and MAPbBr₃/PS showed decay rates of only less than 7% and 15%, respectively [15].

Self-healing gels are materials that possess the ability to automatically repair their structure and restore their original functionality [18, 19]. Due to this characteristic, self-healing materials can enhance the durability of devices and lower the cost of repairing the damaged devices, which makes them very attractive option in the fabrication of next-generation smart devices with self-repairing properties. Up to now, a large amount of self-healing materials have been developed and applied in coating, sensors, solar cells, supercapacitor and E-skin [20–23]. Among the multifunctional self-healing gels, luminescent self-healing gels combine the properties of self-healing and luminescence, offering the advantages of versatile shaping and patterning, excellent optical qualities and easy control of the refractive index. These advantages make them excellent candidates for various applications, such as sensing, imaging, fluorescent tags and photovoltaic conversion [19, 22, 24]. For example, the usage of PVA hydrogel embedded with inorganic quantum dots as holographic display medium has been illustrated [19]. The self-healing hydrogel containing Eu-polyoxometalate shows reversible luminescent response to acid and base vapours [24].

Among the different self-healing materials, poly(dimethylsiloxane)-urea (PDMS-urea) copolymers are a group of intrinsic self-healing polymers that have been widely investigated due to their tunable mechanical and self-healing properties [25–27]. This group of self-healing polymers can be synthesised through a simple on-step reaction, thus holding great promise for preparing self-healing composites with perovskite NCs. In the current work, we present the preparation of stretchable and self-healing polymer gels with high photoluminescence stability by embedding OHP NCs (CH₃NH₃PbBr₃ and CH₃NH₃PbI₃) in the PDMS-urea copolymer. The prepared luminescent composite gels are very flexible and stretchable, and show excellent self-healing capability to external damage. Furthermore, the stability of OHP NCs are greatly improved due to the protection of the hydrophobic gel. To the best of our knowledge, this is the first report on self-healable and luminescent polymer gels based on OHP NCs.

2 Experiments

2.1 Synthesis of the PDMS segmented urea copolymer

PDMS-urea was synthesised according to the previously reported method [28, 29]. To a deoxygenated, anhydrous solution of tolylene-2,4-diisocyanate (0.17 g, 1.0 mmol, Sigma-Aldrich, USA) in dried toluene (5.0 ml), PDMS-NH₂ (M_n = 2500, 2.6 ml, 1.0 mmol, TCI, Japan) was added, and the mixed solution was stirred for 24 h under nitrogen atmosphere. The resulting viscous solution was stored under nitrogen until further use.

2.2 Synthesis of OHP NCs

For a typical synthesis of CH₃NH₃PbBr₃ (MAPbBr₃) NCs [30], 0.2 mmol of CH₃NH₃Br (laboratory synthesised), 0.2 mmol of PbBr₂ (Aladdin, China), 1 ml of oleic acid (OA, Aladdin, China) and 40 µl of 1-octylamine (OTAm, Aladdin, China) were dissolved in 10 ml of DMF under magnetic stirring at room temperature to form a clear precursor solution. Then, 1.8 ml of the precursor solution was injected into 30 ml of toluene under vigorous magnetic stirring and kept stirring for 10 s to give a green colloid solution. Next, 5 ml of the colloid solution was centrifuged (10 min, 8000 rpm/min) and the NCs that settled on the bottom of the tube were collected and redispersed in 5 ml toluene for the ultraviolet–visible spectroscopy (UV-Vis), photoluminescence (PL), X-ray diffraction (XRD) and transmission electron microscope (TEM) characterisation.

The CH₃NH₃PbI₃ (MAPbI₃) NCs were synthesised according to the previous reported methods with some modifications [31, 32]. The precursor solution was prepared by dissolving PbI₂ (0.09 mmol, Aladdin, China), CH₃NH₃I (0.1 mmol, laboratory synthesised) and OTAm (20 µl, Aladdin, China) in 200 µl of DMF and 6.8 mL of acetonitrile. The precursor solution was added dropwise into 10 ml of toluene containing 200 µl oleic acid under vigorous stirring. The precipitates were collected by centrifugation using the same method as MAPbBr₃ NCs and collected for further use.

2.3 Preparation of the OHP NC/PDMS-urea composites

A certain amount of OHP NCs (MAPbBr₃ or MAPbI₃) was blended with 1 ml of PDMS-urea solution under stirring

for 10 min to give a uniform solution. The resulting solution was casted onto a home-made Teflon mould. The solvent was evaporated under ambient conditions to form OHP NC/PDMS-urea gels, and the gels were further dried in a vacuum oven at room temperature for 30 min. For the MAPbBr₃/PDMS-urea gels, MAPbBr₃ NCs loading ratios were changed from 0.4 and 0.8 wt.% to 1.2 wt.%. For the MAPbI₃/PDMS-urea gel, only a 0.8 wt.% MAPbI₃ NCs loading ratio was used. A blank PDMS-urea gel was also prepared for comparison. The OHP NC/PDMS-urea gels used for UV-Vis and PL characterisation have a thickness of 0.8 mm and width and length of 15 mm. The gel samples used for tensile test have a cylindrical shape with diameter of 3 mm and length of 20 mm.

2.4 Self-healing of the OHP NC/PDMS-urea composite gels

For the self-healing study, the blank copolymer and the composite films containing different OHP NCs contents with dimensions of 15 × 15 × 0.8 mm (length × width × thickness) were used. The films were placed on a glass substrate and cut using a knife. Then, the damaged samples were kept in ambient conditions or at elevated temperatures and the self-healing process was monitored. The PL spectra for self-healing analysis were taken from the self-healing region.

2.5 Characterisation

The UV-vis absorption and photoluminescence spectra were collected using a TU 1901 (Persee, China) and a PerkinElmer LS 55 (PerkinElmer, USA) fluorescence spectrofluorimeter, respectively. The morphology of the MAPbBr₃ NCs was characterised on a transmission electron microscope (JEM-2100F, Jeol, Japan). The XRD patterns of the OHP NCs and the OHP NC/PDMS-urea composites were recorded on a Bruker AXS D8 (Bruker, Germany) powder diffractometer using Cu K α radiation ($\lambda = 0.154$ nm). The absolute photoluminescence quantum yields were determined using a fluorescence spectrometer with an integrated sphere (HORIBA FluoroMax-4 spectrofluorimeter, USA) excited at a wavelength of 450 nm. Tensile measurements were performed at 25 °C with an HY-0580 (Hengyi, China) instrument. The molecular weight of the PDMS-urea was determined using Malvern Viscotek GPC Max System (Malvern, USA) with a refractive index detector, a 270 Dual Detector (light scattering and viscometer), and G+ T6000M column at 35 °C.

Tetrahydrofuran (THF) was used as the eluting solvent with a flow rate of 1.0 ml min⁻¹. The system was calibrated with a polystyrene standard. The copolymer solution was filtered through a 0.22- μ m poly(vinylidene fluoride) millipore filter before injection. The thermogravimetric analysis (TGA) measurements were performed on a Mettler Toledo TGA/SDTA851e (Mettler Toledo Star, China) thermogravimetric analyser under a nitrogen atmosphere with a heating rate of 10 °C/min.

3 Results and discussion

The PDMS-urea copolymer was synthesised from the PDMS-NH₂ and tolylene-2,4-diisocyanate (TDI) according to the published method [28, 29], as shown in Scheme 1A. After the reaction, the viscous solution was casted onto a Teflon mould and further dried in a vacuum oven for 30 min. The resulting copolymer is colourless and transparent. The measured number-average molecular weight of the copolymer is 13,500 with the polydispersity index of 1.98. Additionally, the PDMS-urea copolymer contains a large amount of urea units, enabling the formation of reversible hydrogen bonds between the polymer backbones (Scheme 1B), resulting in the copolymer flexibility and self-healing capability.

The CH₃NH₃PbBr₃ (MAPbBr₃) NCs were synthesised by using the solvent-induced re-precipitation method with some modification [30]. The obtained MAPbBr₃ NCs are nanoparticles with sizes ranging from 5 nm to 12 nm, based on the TEM (Jeol, Japan) characterisation (Figure 1A). The clear interference fringes from the lattice plane presented in the HRTEM image (the inset in Figure 1A) illustrate the good crystalline order of the prepared MAPbBr₃ NCs. Figure 1B presents the UV-Vis absorption and PL spectra of the MAPbBr₃ NCs dispersed in toluene. The UV-Vis spectrum shows an absorption onset at 540 nm and a narrow emission peak at 520 nm with a full-width at half-maximum (FWHM) of 26 nm observed in the PL spectrum. The PLQY of the prepared MAPbBr₃ NCs is 40%, as measured with an excitation wavelength of 450 nm.

The luminescent MAPbBr₃/PDMS-urea gels were prepared by dispersing the collected MAPbBr₃ NCs into the PDMS-urea solution in toluene (Scheme 1C). Figure 1C gives the UV-Vis and PL spectra of the MAPbBr₃/PDMS-urea gel containing 0.8 wt.% of the MAPbBr₃ NCs. A sharper absorption onset at 532 nm is observed for the MAPbBr₃ NCs embedded in the polymer gel. The emission peak of the gel is located at 535 nm with a FWHM of 25 nm, which is red-shifted about 15 nm compared to

Scheme 1: (A) Synthesis of the PDMS-urea copolymer; (B) Proposed arrays of hydrogen bonding of the PDMS-urea copolymer and (C) Schematic illustration of the preparation and structure of the OHP NC/PDMS-urea gels.

Figure 1: (A) The TEM image of the MAPbBr₃ NCs. Inset: the HRTEM image; (B) UV-Vis absorption and PL spectra of the MAPbBr₃ NCs dispersed in toluene, Inset: photographs of the solutions for the MAPbBr₃ NCs under ambient light and UV light of 365 nm; (C) UV-Vis absorption and PL spectra of the MAPbBr₃ (0.8 wt.%) / PDMS-urea gel, Inset: Photograph of the MAPbBr₃ (0.8 wt.%) / PDMS-urea gel under UV light of 365 nm and (D) Photograph of the stretched MAPbBr₃ (0.8 wt.%) / PDMS-urea taken under 365 nm UV light.

that of MAPbBr_3 NCs dispersed in toluene. These results indicate that Ostwald ripening occurred and led to the narrower size distribution of NCs during the gel formation process. The imine groups contained in the polymer chain may contribute to the size optimisation due to the weak coordination between the nitrogen and the lead atoms. The measured PL quantum yield (PLQY) of this gel is 23.8%, which is lower than that of the MAPbBr_3 NC solution due to the entrapment of the latter in the solid matrix. The gels containing 0.4 and 1.2 wt.% of MAPbBr_3 NCs were also prepared to investigate the effect of the NCs content on the gel's optical properties (Figure S1). Clearly, the PL intensity of the gels first increases with the increase of the MAPbBr_3 NCs content. When the content reaches 1.2 wt.%, the PL intensity decreases due to the aggregation of the NCs. On the other hand, the increase of the MAPbBr_3 NCs content causes a continuous red-shift of the emission wavelength.

The MAPbBr_3 /PDMS-urea gel is not only luminescent and flexible, but also presents excellent stretchability. Figure 1D shows that the gel with a length of 3 cm can be stretched up to 15 cm, or up to 500% of its original length, without breaking. To further examine the mechanical property of the gels, tensile tests were performed at room temperature. The corresponding stress-strain curves for the gels with different MAPbBr_3 NCs loading ratios of 0.4, 0.8 and 1.2 wt.% are shown in Figure S3. As can be clearly seen, increasing the loading ratio of the MAPbBr_3 NCs enhances the tensile strength of the MAPbBr_3 /PDMS-urea gel while also decreases the elongation of the gel until it breaks. Compared to the blank PDMS-urea gel, the addition of the MAPbBr_3 NCs improves the stress tolerance of the gel although it also weakens its extensibility. Additionally, the luminescent MAPbBr_3 /PDMS-urea gels are

relatively transparent. The gel film containing 0.4 wt.% of MAPbBr_3 NCs with a thickness of 0.8 mm displays 89% light transmittance over the visible to near-IR region (Figure S4).

XRD measurements were carried out to investigate the effect of the gel formation on the crystal structure of the MAPbBr_3 NCs. As shown in Figure 2A, the XRD patterns of the MAPbBr_3 NCs, respectively show the characteristic diffraction peaks of cubic MAPbBr_3 at 14.9° , 30.1° , 33.8° , 43.2° and 45.8° . The broad peaks at 12.1° and 21.2° observed in the PDMS-urea copolymer reveal the amorphous nature of the PDMS-urea copolymer, which is beneficial to the flexibility and self-healing capability [33]. The characteristic peaks of the MAPbBr_3 NCs become more obvious with the increasing MAPbBr_3 NCs concentration for the composite gels. The peak locations are the same as those of the MAPbBr_3 NCs, thus illustrating no phase change of MAPbBr_3 NCs after the gel formation. On the contrary, the broad peak at 12.1° decreases with the increase in the loading ratio of the MAPbBr_3 NCs, which could be due to the decline in the order packing of the polymer chain with the addition of MAPbBr_3 NCs.

The Fourier-transform infrared (FTIR) analysis (Figure 2B) was also performed to investigate the interaction of the MAPbBr_3 NCs with the PDMS-urea polymer molecules. The PDMS-urea polymers exhibit three peaks at 1565 , 1640 and 3325 cm^{-1} , which are assigned to the $-\text{NH}-\text{CO}-$ (amide), $-\text{C}=\text{O}$ (carbonyl) and $\text{N}-\text{H}$ stretching vibrations of the urea groups, respectively [34, 35]. Compared to the frequencies of the free $\text{N}-\text{H}$ ($3445\text{--}3450\text{ cm}^{-1}$) and $-\text{C}=\text{O}$ ($1690\text{--}1700\text{ cm}^{-1}$) bands, the observed frequency shifts of the $\text{N}-\text{H}$ and $-\text{C}=\text{O}$ bands indicate the formation of strong hydrogen bonds between the urea groups [34, 35]. The FTIR spectra of the composite gels

Figure 2: (A) XRD patterns of the MAPbBr_3 QDs, PDMS-urea gel and MAPbBr_3 /PDMS-urea gels with different loading ratios of the MAPbBr_3 QDs and (B) the FTIR spectra of the PDMS-urea gel and MAPbBr_3 /PDMS-urea composites with different loading ratios of the MAPbBr_3 QDs. Inset: enlarged FTIR spectra in the wavenumber range of $1680\text{--}600\text{ cm}^{-1}$.

Figure 3: The process of self-healing for the MAPbBr₃/PDMS-urea composite films with different perovskite QDs loading. (A) PDMS-urea gel; (B) MAPbBr₃ (0.4 wt. %)/PDMS-urea; (C) MAPbBr₃ (0.8 wt. %)/PDMS-urea and (D) MAPbBr₃ (1.2 wt. %)/PDMS-urea.

are very similar to that of the PDMS-urea copolymer, and no significant changes are observed in the relative band intensities. These results reveal that the embedding of the MAPbBr₃ NCs in the PDMS-urea copolymer does not interfere with the formation of hydrogen bonds between the polymer chains, which is crucial for maintaining the self-healing capability of the PDMS-urea copolymer. The

thermostability of the gels was characterised by the TGA measurements (Figure S5). The TGA curves show initial weight losses of about 1.1% and 1.0% between 116°C–245°C and 153°C–240°C for the blank PDMS-urea gel and the MAPbBr₃ (0.8 wt. %)/PDMS-urea gel, respectively. These initial weight losses should correspond to the evaporation of the solvents. The degradation between 240°C and

Figure 4: The PL spectra of MAPbBr₃/PDMS-urea gels with different MAPbBr₃ NC loading ratios before and after self-healing. (A) 0.4 wt.% of MAPbBr₃ NCs; (B) 0.8 wt.% of MAPbBr₃ NCs and (C) 1.2 wt.% of MAPbBr₃ NCs. (D) Evaluation of the PL stability of the MAPbBr₃ (0.8 wt. %)/PDMS-urea gel by immersing the gel into water at room temperature.

440°C is attributed to decomposition of the urea bonds and the soft PDMS segments, followed by the carbonisation of the aromatic unit of the TDI between 440°C and 600°C [36]. These results indicate the considerably good thermostability of the gel.

The reversibility of the hydrogen bond presented in the urea unit endows the PDMS-urea gel with great self-healing capability [34]. The influence of the MAPbBr₃ NCs on this capability was evaluated by using the blank PDMS-urea copolymer and the composite gels with different MAPbBr₃ NCs loadings. The thicknesses of the tested film samples were controlled to be about 0.8 mm by using the

Teflon mould. The films were cut by using a knife to make a scratch with a depth of about 0.4 mm. The damaged samples were then kept in ambient conditions, and the self-healing process was monitored. The photographs taken at different times are presented in Figure 3. For all the samples, the scratches completely disappeared 15 min after cutting, implying the full recovery of the gels. Compared to the blank PDMS-urea gel, the self-healing rates of the tested MAPbBr₃/PDMS-urea gels show no distinct difference. All these results illustrate that the MAPbBr₃ NCs have no obvious influence on the self-healing capability of the PDMS-urea gel in the tested MAPbBr₃ NC loading range.

Figure 5: (A) UV-Vis absorption and PL spectra of MAPbI₃ NCs dispersed in toluene. Insets: photographs of the MAPbI₃ NCs dispersed in toluene under ambient light (top) and UV light of 365 nm (bottom); (B) Evaluation of the PL stability of the MAPbI₃ NCs dispersed in toluene after the addition of water at room temperature; (C) UV-Vis absorption and PL spectra of the MAPbI₃ (0.8 wt.)/PDMS-urea gel, Insets: photographs of the MAPbI₃ (0.8 wt.)/PDMS-urea gel under ambient light (top) and UV light of 365 nm (bottom); (D) Evaluation of the PL stability of the MAPbI₃ (0.8 wt.)/PDMS-urea gel immersed in water at room temperature. (E) The self-healing process of the MAPbI₃/PDMS-urea gel with 0.8 wt.% MAPbI₃ NCs [36].

We further investigated the effect of temperature on the self-healing rate by choosing the MAPbBr₃ NC (0.8 wt. %)/PDMS-urea gel as a representative. As depicted in Figure S6, the damaged gel can fully recover within 10 min and 5 min at 40°C and 50°C, respectively. This demonstrates that the higher temperature leads to the faster self-healing rate due to the accelerated movement of polymer molecular segment.

To determine whether the self-healing process affected the PL properties of the MAPbBr₃/PDMS-urea gel, the PL spectra were taken from the same region of the samples before cutting and after the self-healing process. The results are given in Figure 4A–C. As can be seen, the PL intensities of the three tested samples show little differences before and after the self-healing process, indicating the good PL stability of the MAPbBr₃/PDMS-urea gels.

The MAPbBr₃ NCs are quite sensitive to moisture. The PL intensity of the MAPbBr₃ NCs dispersed in toluene decreases to 36% of the initial value after the addition of water for 2 h (Figure S7). We investigated the PL stability of the MAPbBr₃/PDMS-urea gel by immersing the sample containing 0.8 wt. % of the MAPbBr₃ NCs into water at room temperature. Figure 4D presents the changes of PL intensity with time. In the initial 6 h, the PL intensity decreased relatively fast. After 7 days, the PL intensity decayed to 67% of the initial value, and then kept stable. The first decrease of the PL intensity could be due to the degradation of the MAPbBr₃ NCs located on the surface of the gel. After the complete decomposition of the MAPbBr₃ NCs on the gel surface, the hydrophobic gel prevents the further decomposition of the inner MAPbBr₃ NCs, after which the PL intensity remained constant. These results illustrate that the PDMS-urea gel greatly improves the stability of the MAPbBr₃ NCs against water.

To illustrate the versatility of our strategy to improve the stability of perovskite NCs, we further synthesised CH₃NH₃PbI₃ (MAPbI₃) NCs with red emission. The measured XRD spectrum (Figure S8) confirms the tetragonal phase of the MAPbI₃ NCs and agrees well with previous reports [31, 32]. Figure 5A gives the UV-Vis absorption and PL spectra of the obtained MAPbI₃ NCs dispersed in toluene. The MAPbI₃ NCs shows a main absorption peak at 639 nm and the emission peak at 746 nm with a FWHM of 54 nm. The MAPbI₃ NCs are more sensitive to water than the MAPbBr₃ NCs. Moreover, the PL intensity of the MAPbI₃ NCs dispersed in toluene drops very fast to nearly zero within 1 h after the addition of water, as shown in Figure 5B. Compared to the MAPbI₃ NCs dispersion in toluene, the absorption and emission peak of MAPbI₃/PDMS-urea gel red-shift to 670 and 749 nm (Figure 5C), respectively. The emission peak becomes broad and the

FWHM increases to 60 nm. Similar to the MAPbBr₃/PDMS-urea gel, the formation of the MAPbI₃/PDMS-urea gel greatly improves the PL stability of the MAPbI₃ NCs. After immersing the gel into water for 2 h, the PL intensity is only reduced by 27% of its initial value (Figure 5D). After 48 h, the PL intensity still maintains 39% of its initial value. The MAPbI₃/PDMS-urea gel also exhibits excellent self-healing capability, as illustrated in Figure 5E. The scratch made by cutting completely disappeared after 15 min, illustrating the full recovery of the gel.

The enhanced PL stability and excellent self-healing capability of the OHP NC/PDMS-urea gels with simple preparation process make them very promising materials for use in many optical systems, such as LEDs, imaging and sensing. To illustrate their capability as phosphors for LEDs, blue GaN LED chips (0.8 watt) were coated with MAPbBr₃ (0.8 wt. %)/PDMS-urea and MAPbI₃ (0.8 wt. %)/PDMS-urea gels, respectively, as shown in Figure 6A and B, respectively. The emission of the used GaN LED chip is at 458 nm with a FWHM of 21 nm. After coating with the MAPbBr₃/PDMS-urea gel, the blue emission of the LED chip is fully converted to green and

Figure 6: Photographs of (A) the MAPbBr₃/PDMS-urea gel and (B) the MAPbI₃/PDMS-urea gel-coated GaN LEDs under the operation potential of 2.7 V and (C) Emission spectra of the blue GaN LED (I), the MAPbBr₃/PDMS-urea gel coated green LED (II) and the MAPbI₃/PDMS-urea gel coated red LED (III).

the emission peak is at 549 nm with a FWHM of 23 nm (Figure 6C). The MAPbI₃/PDMS-urea gel can also completely convert the blue emission of the LED chip to red emission.

4 Conclusions

In this work, we have successfully prepared luminescent OHP NC/PDMS-urea gels by a simple process. These gels are flexible, stretchable and relatively transparent. More importantly, the prepared OHP NC/PDMS-urea gels present excellent self-healing capability. Due to the hydrophobic feature of the PDMS-urea copolymer, the stability of the OHP NCs (MAPbBr₃ NCs and MAPbI₃ NCs) against water is greatly improved after the formation of the composite gels. The use of MAPbBr₃/PDMS-urea and MAPbI₃/PDMS-urea gels as green and red phosphors for LEDs, respectively, has been successfully demonstrated. Due to the above-mentioned properties, the OHP NC/PDMS-urea polymer composite gels are very promising materials for fabricating next generation smart optical devices with self-healing property.

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