



# Hybrid downconverters with green perovskite-polymer composite films for wide color gamut displays

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**Abstract:** We propose to use a hybrid downconverter system comprising low-cost green perovskite-polymer composite films for liquid crystal display (LCD) backlight unit (BLU) to realize wide color gamut. Recently, ultrastable, highly luminescent  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  ( $\text{MAPbBr}_3$ ) organic-inorganic perovskite-polymer composite films have been developed. These films exhibit outstanding color quality with a full-width-at-half-maximum (FWHM) of only 18 nm and a peak wavelength of 530 nm, which makes them promising candidates as green downconverters. Two configurations to hybridize these green films with state-of-the-art red emitting downconverters, including CdSe-based quantum dots (QDs) and narrow peak phosphors, are proposed. Color and efficiency analyses indicate that the hybridization of green perovskite-polymer films with red  $\text{K}_2\text{SiF}_6:\text{Mn}^{4+}$  (KSF) phosphor could lead to wide color gamut coverage of nearly 90% Rec. 2020 and high total light efficiency (TLE) of around 20 lm/W while maintaining low cost.

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## 1. Introduction

Since the International Telecommunication Union (ITU) recommendation (ITU-R BT.2020) for ultra-high definition (UHD) television came out [1], the display industry is moving toward

higher color performance, which would represent the color truthfully. To achieve this goal, quantum dots (QDs) have been widely used in liquid crystal display (LCD) backlight systems, thanks to their high photoluminescence (PL) efficiency, easily tunable wavelength and narrow emission, which helps realize greater than 90% coverage of ITU's recommended color gamut standard (commonly referred to as Rec. 2020) [2–5]. However, the well-developed narrow emitting (20–30 nm) QDs are mostly cadmium selenide (CdSe) based, while Cd is one of the regulated materials in the Restriction of Hazardous Substances (RoHS) Directive by European Union. The maximum allowable concentration for Cd in consumable electronics is 100 ppm, which is difficult to meet. Thus, Nanosys proposed a “greener” QD system by combining modified narrow green CdSe-QDs and red InP-QDs to be compliant with RoHS regulations [6]. Samsung has been using Cd-free QDs in their flagship TVs. Because of the limited stability of QDs, they are most widely used in thin film “on surface” configuration, which provides mild working temperature environment, but requires large amount of QD materials. The “on surface” configuration has been a cost limiting factor of QDs, which are usually synthesized in sophisticated hot-injection process.

On the other hand, the development of narrow linewidth phosphors which can be used directly on LED chips provides a low-cost alternative for realizing wide color gamut [7]. Among the several phosphor systems that are under active developments,  $\text{K}_2\text{SiF}_6:\text{Mn}^{4+}$  (KSF or PFS) is a narrow-band red phosphor with five sharp emission lines and the most intensified one peaks at 631 nm, and it has been widely used in LCD backlight with high efficiency and stability since the successful commercialization by GE [8–10]. Another red phosphor  $\text{Sr}[\text{LiAl}_3\text{N}_4]:\text{Eu}^{2+}$  (SLA) also shows high efficiency with longer, yet broader emission peak [11]. In comparison to red ones, green emitting phosphors normally have broader peaks [7, 12–15], thus limiting the overall color gamut this “on chip” phosphor configuration can possibly achieve.

Recently, metal halide perovskites are emerging as promising light emitting materials with high efficiency and superb color purity, particularly in green color [16–21]. However, their instability issue must be solved before widespread application can be realized [22]. A microencapsulation strategy has recently been developed by the authors' team to achieve organic-inorganic perovskite-polymer composite films with high luminescent efficiency, color purity and ultrahigh stability against heat and water exposure [23, 24]. Meanwhile, perovskite-polymer composite films with similar color purity and enhanced PL efficiency have been reported [25]. With unprecedented color quality and steadily-increasing stability and PL efficiency, these perovskite-polymer composite films represent a promising green downconverter candidate for the LCD backlight unit (BLU).

In this paper, we propose the hybridization of green perovskite-polymer emitter films with state-of-the art Cd-based red QDs, KSF/PFS red phosphor, or SLA red phosphor in two possible BLU configurations. We will provide a generalized analysis on the color gamut and efficiency performance of these hybrid downconverters with the goal to obtain low-cadmium or cadmium-free BLU with high color gamut coverage in Rec. 2020 while maintaining high efficiency for display applications.

## 2. Methods

### 2.1 Perovskite-polymer composite films

Perovskite-polymer composite films have recently emerged as promising green emitting downconverters with high PL efficiency, color purity and high stability against heat and water exposure [23–25].

As a prominent example, we have reported a swelling-deswelling microencapsulation strategy to achieve well dispersed, intimately passivated perovskite nanoparticles inside polymer matrixes (Fig. 1) [23].

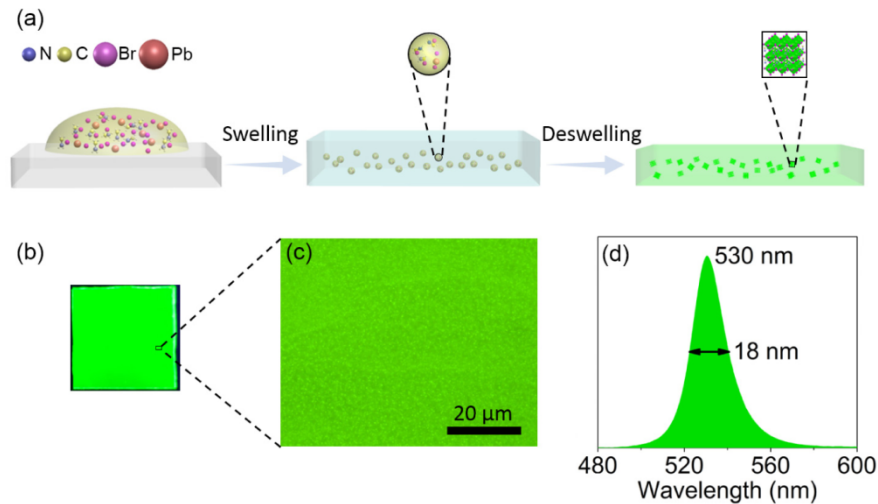


Fig. 1. (a) Scheme of MAPbBr<sub>3</sub> - polymer composite film formation process through swelling-deswelling; (b) Image of MAPbBr<sub>3</sub> - PS film under UV excitation; (c) Fluorescent optical microscope image of MAPbBr<sub>3</sub>-PS film with focal plane  $\approx 4 \mu\text{m}$  underneath the top surface; (d) Typical emission spectrum of as-prepared MAPbBr<sub>3</sub>-PS films.

CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> (MAPbBr<sub>3</sub>) perovskite precursor solutions prepared by mixing PbBr<sub>2</sub> and CH<sub>3</sub>NH<sub>3</sub>Br at a 1:3 molar ratio in dimethylformamide (DMF) solvent were spin-coated on polymer substrate, during which perovskite precursors can be introduced into polymer matrices as solute through the solvent-induced polymer swelling process. When the solvent is driven out of the polymer matrix (for example, by baking), the perovskite precursors will be left within to react and form high quality, well dispersed perovskite nanoparticles. Meanwhile the polymer matrix will deswell, shrink back and form a coherent barrier layer around the perovskite nanoparticles, protecting them from water, oxygen or heat of the surrounding environment (Fig. 1(a)).

A series of technically important polymer substrates, including polystyrene (PS), polycarbonate (PC), acrylonitrile butadiene styrene (ABS) etc., work well with this microencapsulation strategy, indicating the generality of the strategy. Figure 1(b) shows an image of a typical perovskite -PS film excited by UV light (365nm). The microencapsulation process provides intimate passivation of crystalline perovskite nanoparticles within polymer matrix, as seen from the fluorescent microscopic image (Fig. 1(c)), and yields composite films with high PL efficiency and ultrahigh stability, without any further encapsulation. Perovskite -PC and perovskite -PS composite films can survive boiling water treatment for 30min without obvious PL decay, and three kinds of perovskite-polymer composite films can be immersed in water for two months maintaining the PL intensity [23]. The emission peaks of these films lie in 528-533 nm range which is highly suitable for display application and the FWHM can be as narrow as 18 nm for perovskite -PS film, as shown in Fig. 1(d).

## 2.2 Hybrid backlight system configurations

Because of their process simplicity, low cost, high stability, high luminescence efficiency and great color purity, the perovskite -polymer composite films have great potential to be used as green downconverters for the BLU of wide color gamut LCDs, particularly when being hybridized with other state-of-the-art red emitters. Three commonly used backlight downconverter geometries are “on-chip”, “on-edge” and “on-surface” [26]. A hybrid backlight system can be comprised of a blue LED, a green perovskite -polymer film and red downconverter (QD or phosphor) in two configurations as described below:

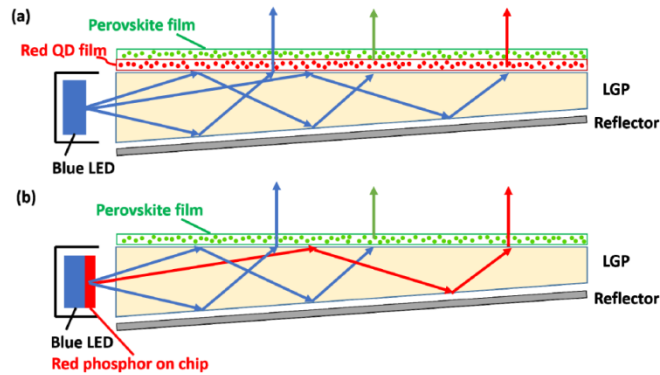


Fig. 2. Backlight system configuration for (a) On-surface green perovskite and red QD films excited by blue LEDs, and (b) On-surface green perovskite and on-chip red phosphor excited by blue LEDs. LGP stands for light guide plate.

1. On-surface green perovskite-polymer and red QD films excited by blue LEDs: Since at current stage QDs would mostly suffer from lifetime and stability decrease in “on-chip” design due to high flux and high LED junction temperature ( $\sim 150^\circ\text{C}$ ) [27], it is better to use “on-surface” configuration. To reduce reabsorption and enhance the photoluminescence, layered structures were found to be more effective with green emitters on top of red ones [28]. Figure 2(a) shows the configuration of “back-to-back” green perovskite-polymer on red QD films pumped by blue LEDs.
2. On-surface green perovskite and on-chip red KSF/PFS Phosphor or SLA Phosphor excited by blue LEDs: With phosphors’ high quantum efficiency and good stability especially under high temperature, the best configuration for this combination is “on-chip” packaged red phosphors on blue LED and remote “on-surface” green perovskite film, as shown in Fig. 2(b). The choice of phosphors and their respective characteristics and advantages will be discussed in the following section.

### 3. Results and discussions

#### 3.1 Analysis of color gamut coverage and total light efficiency (TLE)

The optical performance of a BLU needs to be evaluated with consideration of their integration into the whole LCD system. In a typical LCD panel configuration [2] white backlight passes through the thin-film-transistor (TFT) backplane, liquid crystal layer, and color filters successively. Finally, output light with different colors from three RGB light channels together forms one display pixel.

Color gamut coverage and total light efficiency (TLE) are two main parameters commonly used to evaluate color and efficiency performance of a BLU system. Within a preset LCD system, these parameters can be derived from spectral analysis of the primary colors [2–5]. As pointed out in [29], the color gamut coverage ratio in CIE 1931 is more consistent to the Rec. 2020 volume coverage ratio in color appearance model CIELAB, CIELUV and CIECAM02, compared to that in CIE 1976, so we use CIE 1931 as the metric in this paper. The TLE of the display system is calculated by

$$TLE = \frac{683 \frac{\text{lm}}{\text{W}} \int P_{out}(\lambda) V(\lambda) d\lambda}{\int P_{in}(\lambda) d\lambda} = LER * TE, \quad (1)$$

where LER and TE are defined as



$$LER = \frac{683 \frac{lm}{W} \int P_{out}(\lambda) V(\lambda) d\lambda}{\int P_{out}(\lambda) d\lambda} \quad (2)$$

$$TE = \frac{\int P_{out}(\lambda) d\lambda}{\int P_{in}(\lambda) d\lambda}, \quad (3)$$

respectively. In Eqs. (1)-(3),  $P_{in}(\lambda)$  and  $P_{out}(\lambda)$  represent the radiation spectral power distribution (SPD) of the input and output light,  $V(\lambda)$  is the human eye sensitivity function which is centered at  $\lambda = 550$  nm. From Eq. (1) we can see that TLE contains two parts: the luminous efficacy of radiation (LER) and the transfer efficiency (TE). LER reflects how efficient the output light can be converted to the brightness feeling of human eye and TE stands for the power ratio of transmitted light through the LCD panel over the input light. Together TLE represents how much input light transmits through the LCD panel and finally gets converted to the brightness perceived by human eye. TLE is a measure of BLU's total efficiency considering most of the factors including the efficacy of light source, the transmittance of color filters, LC layer and polarizers, and the aperture ratio of each light channel. Only one factor of the wall plug efficiency of the light source itself is excluded for the following reasons: First, even among the same type of light source, the wall plug efficiency can be quite different [30], not to say different types of emitters. Secondly, different configurations of BLU have different working temperature environment so that the emitters' performance vary. Moreover, the experimental temperature dependent PLQY of many kinds of emitters are not openly available, which makes it hard to take the intrinsic wall plug efficiency comparison into evaluation. Last but not least, perovskite-polymer composite films are still emerging material systems, and their PLQY or other performance still have plenty of room to improve, with lots of researchers' endeavor [23–25]. As a result, in this work we do not consider the emitters' own PLQY or downconversion efficiency and mainly focus on the effect of different backlight spectra on LCD color performance and system efficiency, using color gamut and TLE as sufficient criteria. This meanwhile makes the analysis more general and not limited to certain material/film process or even kinds of material, as long as the emission properties are similar (peak wavelength, FWHM).

For the calculation of color gamut and TLE, we use same method as [3, 5] and choose D65 ( $x = 0.312$ ,  $y = 0.329$  in CIE 1931 diagram) as the white light reference point, which is very close to sunlight and prevalent in most display products and can be used as a representative white point for display performance evaluations. The green spectrum used in all calculation was measured from our perovskite-PS film, i.e. peak wavelength  $\lambda = 530$  nm and FWHM = 18 nm. The blue spectrum was measured from a commercial 550 mW high power LED (Royal-Blue Cree XLamp® XT-E from LEDsupply), with  $\lambda = 450$  nm and FWHM = 18.5 nm. The red QD spectrum was measured from CdSe-based colloidal QDs synthesized in-house using reported method [31], with  $\lambda = 630$  nm and FWHM = 22.7 nm. The KSF/PFS phosphor spectrum was taken from [8], and SLA phosphor spectrum from [11]. All the primary colors are marked in CIE 1931 diagram without passing through the LC layer or color filters so that we can get an idea of the color purity of each source (Fig. 3, "Pero-" stands for "Perovskite" in all figures and tables). Because the peak wavelength of QDs can be easily tuned through particle size control, the "Red QD" circle markers represent original QD spectrum centered at 630 nm, redshifted spectra centered at 635 nm, 640 nm, 650 nm, from top-left to down-right, respectively. The CIE coordinates of the RGB components in BLU are listed in Table 1.

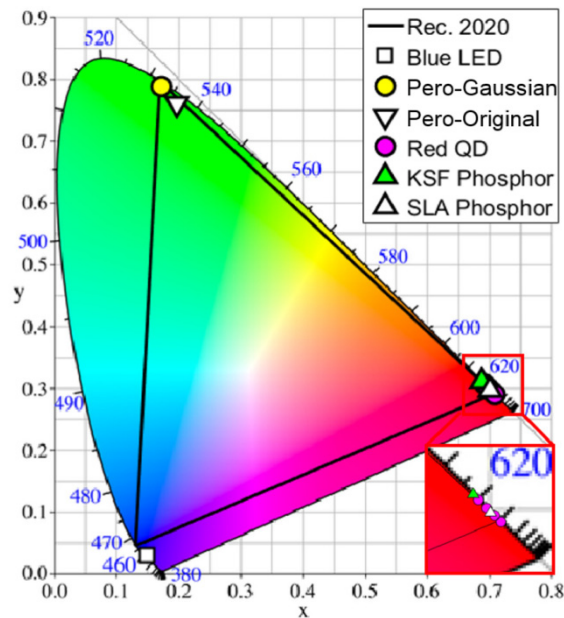


Fig. 3. RGB color primaries in CIE 1931. Rec. 2020 is plotted for reference. Inset shows details of different red emitters.

**Table 1. CIE 1931 chromaticity coordinates of RGB primaries in BLU**

Chromaticity coordinates	x	y
630 nm	0.692	0.308
635 nm	0.698	0.302
640 nm	0.704	0.296
650 nm	0.709	0.291
KSF/PFS Phosphor	0.688	0.312
SLA Phosphor	0.701	0.291
Green Pero-Original	0.197	0.761
Green Pero-Gaussian	0.172	0.789
Blue LED	0.151	0.030

When carrying out spectra analysis for color gamut, a common practice is to use Gaussian spectrum based on the peak wavelength and FWHM of the emission, since it is convenient to tune the characters of the spectrum and analyze the wavelength/FWHM effect on the performance. However, when it comes to a specific backlight, it is better to do analysis based on experimental spectral data without Gaussian fitting, to provide more realistic and accurate performance of the backlight. To simply show the discrepancy of Gaussian fitting here [2], we also mark the position of Gaussian fitted perovskite spectrum in Fig. 4. We can see that

even with the same FWHM, the fitted spectrum “appears” much purer because the tail is cut off. To ensure the reliability of the results, all the spectra used for calculation in this paper are original without Gaussian fitting or other processing. During the calculation, commercial color filters are employed, whose transmission spectra are shown in Fig. 5. Color filter-1 (CF-1 from AUO of Taiwan) which is commonly used in TV [3] has higher transmittance but larger overlap between adjacent colors, while Color filter-2 (CF-2), developed and used by LG [32] has less color crosstalk, yet the transmittance for blue and green is reduced by ~20%. So, there is always tradeoff between color purity and total light efficiency, which will be discussed in detail for each BLU combination. The BLU color gamut without passing through LC/color filter is also calculated for reference.

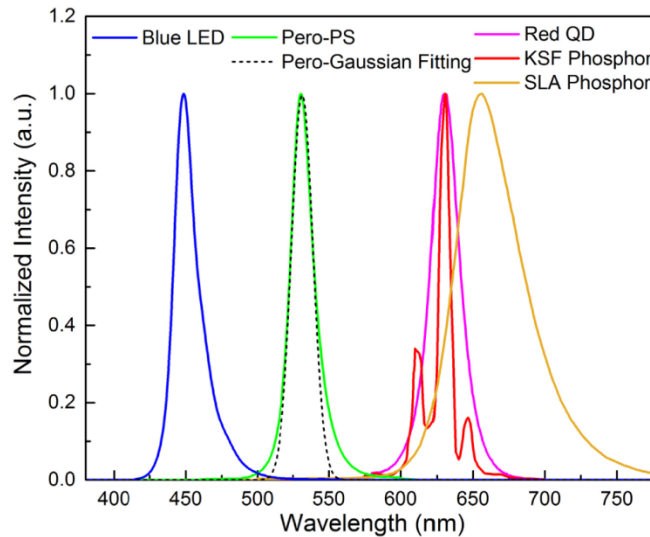


Fig. 4. Spectra of RGB primary colors used for calculation.

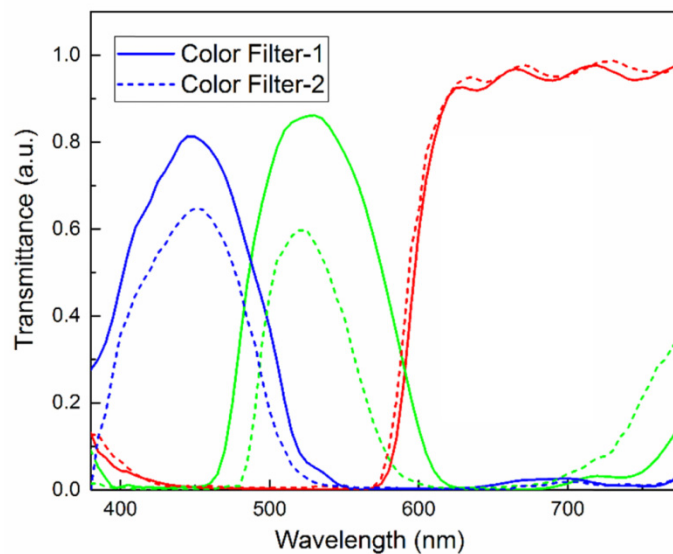


Fig. 5. Transmission spectra of two commercial color filters.



The Rec. 2020 color gamut coverages in CIE 1931 and TLE are listed in Table 2. Since we use green perovskite-PS spectrum and blue LED for all the BLUs, and the only difference among each BLU is the red emitter. Thus, we only list the red color materials in Table 2.

From Table 2, we can see that after passing through CF-1, the color gamut coverages shrink a lot, mainly due to color crosstalk [33]. After passing through the narrower CF-2 filters, the color gamut is wider than that of the original BLU. We can also observe the difference in Fig. 6, in which the BLUs with red QD630/KSF Phosphor/SLA Phosphor passing through the LC and CF-2 are plotted, and the corresponding original primary colors are also marked for reference. The green vertex slightly shifts from the “Pero-original” point and gets closer to the Rec. 2020 green vertex. This is because the color filter narrows down the green spectrum. And each of the three red sources is shifted away from Rec. 2020 edge, resulting from the color crosstalk between green and red.

**Table 2. Rec. 2020 color gamut coverage in CIE 1931 and total light efficiency (TLE) for different BLUs.**

BLU Red Emitter	No LC/CF	CF-1		CF-2		
	Rec. 2020	Rec. 2020	TLE (lm/W)	Rec. 2020	TLE (lm/W)	
QDs	630 nm	88.7%	82.8%	27.4	89.6%	19.5
	635 nm	89.5%	84.0%	26.0	90.5%	18.5
	640 nm	90.0%	85.0%	24.3	91.2%	17.2
	650 nm	90.0%	85.6%	22.3	91.6%	15.8
KSF/PFS Phosphor	88.2%	81.7%	28.3	88.9%	20.2	
SLA Phosphor	89.7%	85.2%	16.1	91.3%	11.4	

With the 630-nm red QD spectrum, we can get 89.6% Rec. 2020 coverage. Considering the easy tuning property of QDs, we also compare the results with a longer red wavelength, 635 nm, 640 nm and 650 nm. As Table 2 shows, the color gamut increases by 1-3%, but the TLE decreases accordingly from ~20 lm/W to ~16 lm/W. So, in the first configuration, Cd-based QD offers attractive features in narrow FWHM and tunable peak wavelength and the combination with green perovskite film could provide wide color gamut and high TLE. However, the cadmium-based QD is still under RoHS regulation and for the on-surface configuration where QD amount needed is proportional to screen size, the cost issue should be considered as well.

Considering the limitation of QDs, here we explore the recent advances of red phosphor materials. Conventional phosphors usually have broad emission band, which is not ideal for wide color gamut displays. However, recently several narrow-band phosphors have been developed [8–10, 34, 35]. KSF/PFS is a red phosphor with very narrow FWHM (each of the five peaks < 3 nm) and the most intensified peak occurs at 631 nm, which is suitable for display. From Table 2, we can see that BLU with KSF/PFS phosphor can reach ~89% Rec. 2020 gamut and TLE remains larger than 20 lm/W, which is comparable to our 630-nm QDs. Moreover, it can be readily implemented in “on-chip” LEDs for low cost and compact display applications, while getting rid of cadmium.

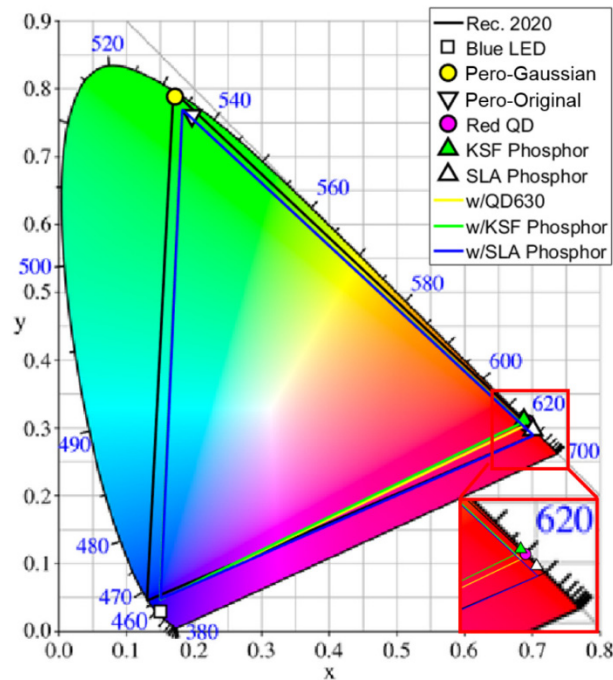


Fig. 6. Simulated color gamut in CIE 1931 for different BLUs. CF-2 is used. The color primaries before passing through LCD system are also marked for reference.

To further broaden the color gamut, we need a red emitter with longer emission wavelengths. A good choice is the highly efficient SLA phosphor. With the peak around 650 nm and  $\text{FWHM} \approx 50$  nm, SLA phosphor enables the BLU to get 91.3% Rec. 2020 coverage, without using Cd-based QDs. However, its TLE is only 11.4 lm/W because a portion of its emission spectrum exceeds 700 nm where the eye sensitivity is low.

After all, we should reach a delicate balance between color gamut and total light efficiency. To maintain a moderate TLE and gain wide color gamut, BLUs with 630-nm QDs or KSF/PFS phosphor are ideal choices, with nearly 90% Rec. 2020 and around 20 lm/W efficiency. Especially with the combination of high brightness blue LED, green perovskite film, and on-chip red KSF/PFS phosphor, we can achieve low cost, efficient, vivid color, and cadmium-free displays.

### 3.2 Discussions on potential cost reduction benefits and hazardous concerns

Besides color and efficiency, several parameters that are of particular interests to the display industry are costs, potential hazardous concerns and film form factors (e.g. thickness etc). While at this stage, perovskite-polymer composite film is still an active research topic in academic laboratories with a lot of experimental optimization efforts pending, it is worthwhile to provide a high level analysis of these parameters with current industrial QD films for comparison.

Costs of downconverter films can be separated into two parts: chemical precursor cost and processing cost. The chemical precursors for perovskite-polymer films are simply low cost  $\text{PbBr}_2$ ,  $\text{CH}_3\text{NH}_3\text{Br}$ , DMF solvent, and commodity plastic substrates, with no expensive phosphine or surfactants involved. The fabrication can be completed within two simple steps (swelling and deswelling) under room temperature or mild annealing under  $100^\circ\text{C}$ . There are no needs for multi-steps, time consuming, high temperature growth or post growth purification processes like QDs. The procedure is very simple and can be easily scaled up. So

even if additional barrier films might be needed like QD films, significant cost reductions can be expected for perovskite-polymer composite films.

Like cadmium (Cd), the lead (Pb) component inside perovskites is also a RoHS restricted element and could be a potential hazardous concern. However, because of its lower toxicity (vs Cd), maximum allowable concentration by RoHS for Pb in consumable electronics is 1000 ppm, which is one order higher than Cd's limit (100 ppm). While systematic quantitative analysis is still needed to evaluate whether high performing perovskite-polymer films can meet this relaxed limit, we believe such concerns can be alleviated with careful perovskite precursor concentration control, film system designs, and/or developments of new perovskite material with less or without lead components.

Downconverter film form factor, particularly their thickness, has been one of the major limiting issues holding their wide applications in mobile displays where limited room is available. For perovskite-polymer composite films, what will be the ultimate thickness limit remains one open question to be explored. Given the demonstrated generality regarding to polymer substrate options (which range from ABS of  $\sim 76 \mu\text{m}$  to PS of  $\sim 600 \mu\text{m}$ ), we are optimistic that, with further polymer substrate engineering, the film thickness could be scaled down to close to  $\sim 50 \mu\text{m}$ , which might enable drop-in applications of such films as a down-converting diffusive film to replace current diffusion films even in mobile LCDs.

#### 4. Conclusion

Ultra-thin, highly luminescent perovskite-polymer composite films are fabricated with narrow green emission band. Such films can be integrated with blue LEDs and red downconverters, including red CdSe-based QDs, KSF phosphor or SLA phosphor, in two hybrid BLU configurations to obtain wide color gamut of 89%~91% Rec. 2020 coverage while maintaining high TLE, without special optical design. Specifically, cadmium free BLU with blue LED, perovskite film and on-chip KSF phosphor can achieve  $\sim 20 \text{ lm/W}$  light efficiency with  $\sim 89\%$  Rec. 2020 coverage, which is highly promising for low cost, high efficiency and RoHS compliant wide color gamut LCD applications.

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