

荧光粉/SiO₂ 荧光膜用于高显指激光照明武华君¹, 潘国徽¹, 吴 昊¹, 张亮亮¹, 张家骅^{1,2}

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摘 要: 利用“蓝光激光+无机荧光体”的方式产生白光当前激光照明的主流技术, 具有结构简单、成本低等优势。但现有的无机荧光体光谱中缺乏青绿光和红光成分, 导致激光照明的显色指数偏低。本工作通过引入电荷补偿剂 Na⁺的方式, 制备出高效的蓝绿色 Ca₃Sc₂Si₃O₁₂:Ce³⁺, Na⁺荧光粉, 其内/外量子效率分别达到 86%和 55%; 然后结合商用 CaAlSiN₃:Eu²⁺红粉, 以二氧化硅溶胶为无机黏结剂, 在蓝宝石衬底上低温制备出高效的复合荧光膜。该荧光膜具有良好的热稳定性, 200 °C 时其荧光强度仍保持在常温下的 84%; 荧光膜中绿光、黄光、红光成分均匀分布, 其半高宽达 176 nm; 在蓝光激光激发下产生 647 lm 的白光, 光效 122 lm/W, 光源的显色指数 88。这些结果表明荧光粉/SiO₂ 复合荧光膜在高显色激光照明中有广阔的应用前景。

关键词: 激光照明; 荧光转换体; 高显色; 石榴石; 硅酸钪钙

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在过去的 20 年里, 基于半导体芯片的固态照明技术由于其发光效率高、使用寿命长、环保等优势, 已经逐步取代了传统的照明光源, 被称为是 21 世纪的绿色照明光源^[1]。根据激发芯片种类的不同, 固态照明可以分为发光二极管照明和激光照明。其中发光二极管照明已经广泛用于室内、公路、交通信号灯和显示器背光等低亮度照明和显示领域^[2-3]。但由于 LED 芯片在高电流密度下存在难以解决的“效率骤降”现象, 这严重限制了 LED 照明在高亮度照明领域的应用^[4]。与发光二极管照明相比, 激光照明具有亮度高和方向性好等优势, 在汽车大灯、激光投影和海上搜救等领域有广泛的应用前景^[5-6]。

激光照明当前主要是用蓝光激光激发远程荧光体的方式来实现。传统的“荧光粉+硅胶”的封装方式, 由于其热导率低(0.1~0.4 W·m⁻¹·K⁻¹)、耐热性差, 在高功率-密度的激光激发下, 会出现“黄化”问题, 不适用于激光照明。因此, 耐热性好、热导率高、抗激光照射的全无机荧光体应运而生^[7-10]。当前, 常见的全无机荧光体有单晶、荧光陶瓷、玻璃陶瓷、荧光膜等。陶瓷和单晶具有热导率高(5~

13 W·m⁻¹·K⁻¹)、热稳定性好和荧光效率高的优势, 但存在制备周期长、制备成本高和品类单一的问题^[11-12]; 荧光玻璃具有工艺简单、制备成本低和易于批量化生产的优势, 但玻璃的热导率(约 1 W·m⁻¹·K⁻¹)和饱和功率密度(1 W·m⁻²)较低, 难以用于大功率激光照明^[13-15]。荧光膜具有易制备、成本低和热导率高的优势, 在激光照明中有广阔的应用前景^[16-17]。现有的荧光膜主要是通过荧光粉和玻璃粉低温共烧的方式制备, 烧结温度一般在 600~1 000 °C, 所用的荧光粉也多是氧化物荧光粉, 以黄色 Y₃Al₅O₁₂:Ce³⁺(YAG), 绿黄色 Lu₃Al₅O₁₂:Ce³⁺(LuAG)为主^[18-19]。而白光照明中最常用的红色氮化物荧光粉容易被玻璃粉腐蚀, 导致现有的 YAG, LuAG 荧光膜中缺乏红光成分^[19-21]。此外, 与蓝光 LED (半高宽约为 20 nm)相比, 蓝光激光的半高宽只有大约 2 nm, 激光照明的光谱中缺少蓝绿光组分, 光谱中存在很宽的“蓝绿光凹陷”^[22-23]。现有的荧光膜光谱中因缺少蓝绿光和红光成分, 导致激光照明光源的显色指数偏低, 不能实现高显色激光照明。

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在 LED 照明中, 石榴石结构的 $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Ce}^{3+}$ (CSS) 荧光粉具有良好的物理化学性质、高效的发光效率和出色的热稳定性, 引起了人们的广泛关^[24-26]。首先, 该荧光粉可以被蓝光有效激发, 高效发射 505 nm 的宽带蓝绿光^[27]; 与常用的黄绿色 LuAG 荧光粉(峰位位于 530 nm)相比, CSS 荧光粉有更加蓝移的发射光谱, 蓝绿光成分更多, 可以有效填充光谱中的“蓝绿光凹陷”, 实现高显色激光照明。其次, 蓝色激发光和蓝绿色发射光谱之间的斯托克斯偏移较小, 有利于减少下转换过程中产生的热量。最后, CSS 荧光粉具有出色的热稳定性, 其荧光强度在 600 K 时仍能保持在 80 K 时荧光强度的 60%, 而常用的 YAG 荧光粉的荧光强度仅为 30%^[28]。CSS 荧光粉出色的发光特性表明, 如果能够将 CSS 用于激光照明, 它将是一种有前途的蓝绿色荧光体^[29-30]。

以自制的蓝绿色 CSS 荧光粉和商用的红色 $\text{CaAlSiN}_3:\text{Eu}^{2+}$ (CASN) 荧光粉为荧光原料, 二氧化硅溶胶为二氧化硅的来源, 二氧化硅溶胶起到了无机黏结剂、荧光粉间隙填充物质和荧光粉保护剂的作用, 通过低温烧结制备出荧光粉和二氧化硅的复合荧光膜。本工作系统研究了荧光粉的物相性质及形貌, 荧光膜的形貌、荧光性质以及在蓝光激光激发下的荧光性能。

1 实验

1.1 样品制备

通过高温固相法制备 CSS 荧光粉, 以高纯度的 CaCO_3 , Sc_2O_3 , SiO_2 , CeO_2 和 NaHCO_3 为原料, 按照化学式 $(\text{Ca}_{0.98-x}\text{Na}_x\text{Ce}_{0.02})_3\text{Sc}_2\text{Si}_3\text{O}_{12}$ ($x=0, 0.02, 0.04, 0.06, 0.08$) 精确称量后置于玛瑙研钵中, 充分研磨均匀后装入刚玉坩埚中, 随后置于 10% H_2 +90% N_2 的管式炉中, 在 1400 °C 保温 5 h, 随炉冷却至室温, 取出后将样品粉碎研磨均匀即得到 CSS 荧光粉。

以自制的蓝绿色 CSS 荧光粉和商用的红色 CASN 荧光粉作为荧光材料, 其中红色荧光粉和蓝绿色荧光粉的质量比(R/G)是 1:8、1:12、1:20, 以二氧化硅溶胶作为无机黏结剂、聚乙烯吡咯烷酮(PVP)为有机黏结剂。荧光膜的工艺流程如下, 首先将荧光粉、二氧化硅溶胶和 PVP 按照质量比 150:50:1 混合均匀, PVP 黏结剂可以调节浆料的粘稠度, 然后将浆料通过刮涂法涂敷在蓝宝石基板上, 最后将干燥后的荧光膜置于马弗炉中, 在 600 °C 加热 20 min 以排除有机物, 即可得到荧光膜。

1.2 样品表征

用 D8-Advance 型 X 射线粉末衍射仪分析样品的晶相, 测试条件: Cu 靶, 管电压 40 kV, 管电流 30 mA。利用 Model S-4800 型扫描电子显微镜分析了荧光粉及荧光膜的显微结构。利用 F920 型荧光光谱仪分析样品的发射和激发光谱, 在 F920 光谱仪上使用时间相关单光子计数法测量了样品的荧光衰减光谱, 该光谱仪配备了 R928 型光子计数探测器光电倍增管和氢灯。利用 THMS-600 型冷热台对样品进行加热, 升温速率是 25 °C/min, 每 25 °C 为 1 个温度点, 每个温度点保温 2 min; 同时利用 QEPro 型光纤光谱仪测量变温光谱。利用 PM100D 型激光功率计测量蓝光激光器输出功率和电流的关系。利用 Quantaaurus-QY Plus C13534-12 型绝对量子效率测试系统测量荧光粉的吸收效率和内/外量子效率。利用装配有积分球和蓝光激光的 HAAS-2000 型光电测试系统测量样品的激光照明性能。本工作中的所有测试如无特殊说明均在室温下完成。

2 结果与讨论

2.1 CSS 荧光粉的物相分析及荧光性能

图 1 为不同 Na^+ 含量荧光粉的 XRD 谱。从图 2 可以看出, 当 CSS 荧光粉中不掺杂 Na^+ 时, 得到的荧光粉中含有 Sc_2O_3 和 $\text{Ca}_3(\text{Si}_3\text{O}_9)$ 杂相。当在 CSS 中引入 Na^+ 后, 随着 Na^+ 含量的增加, 杂相的含量逐渐变小, 同时 XRD 衍射峰的强度逐渐增强, 荧光粉的结晶度增加, Na^+ 起到助熔剂的作用。 $x \geq 0.06$ 时, 得到了纯相的 CSS 荧光粉, 可以和 PDF 72-1 969 卡片的衍射峰完全重合。此外, 随着 Na^+ 含量的增加, 56.2°附近的衍射峰位逐渐向大角度方向移动, 说明 Na^+ 已经固溶入 CSS 晶格中。

图 2 为在 450 nm 蓝光激发下 Na^+ 掺杂对荧光粉荧光性能的影响。从图 2 可以看出, 在 450 nm 的蓝光激发下 CSS 荧光粉发射宽带蓝绿光, 峰值位于 507 nm, 对应 Ce^{3+} 的 5d 激发态到 $^2\text{F}_{5/2}$ 和 $^2\text{F}_{7/2}$ 基态的跃迁。该非对称发射带可分解为 2 个峰值位于 19920 cm^{-1} (约 502 nm) 和 18 414 cm^{-1} (543 nm) Gaussian 型光谱, 其能量差是 1 506 cm^{-1} , 对用于 Ce^{3+} 的 $^2\text{F}_{5/2}$ 和 $^2\text{F}_{7/2}$ 之间的能量差^[30]。此外, 随着 Na^+ 含量的增加, 发射光谱的强度逐渐增强。当 $x=0.06$ 时, 发射光谱的强度达到最大, 其对应内量子效率和外量子效率分别是 86% 和 55%。从图 2b 可以看出, 随着 Na^+ 含量的增加, 荧光粉的吸收效率逐渐提高, 主要是由于在 CSS 晶格结构中, 三价

Ce^{3+} 取代十二面体格位的二价 Ca^{2+} , 导致电荷失配, 在晶格中引入点缺陷, 这导致 Ce^{3+} 在晶格中的溶解度有限; 而一价的 Na^+ 也取代十二面体格位的二价 Ca^{2+} , Na^+ 起到电荷补偿的作用, 可以有效缓解电荷

失配, 减少晶格中的点缺陷, 进而增加 Ce^{3+} 在 CSS 晶格中的溶解度, 导致荧光粉吸收效率的增加。晶格中点缺陷的减少, 同时也会提升荧光粉的内量子效率。

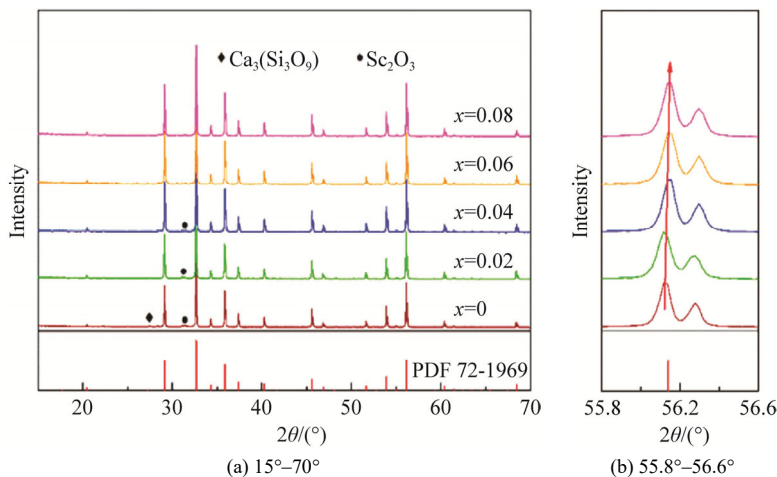
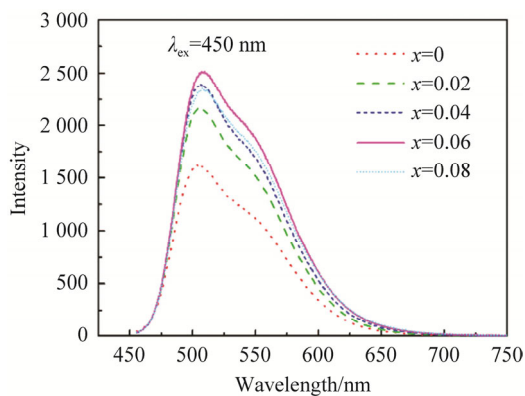
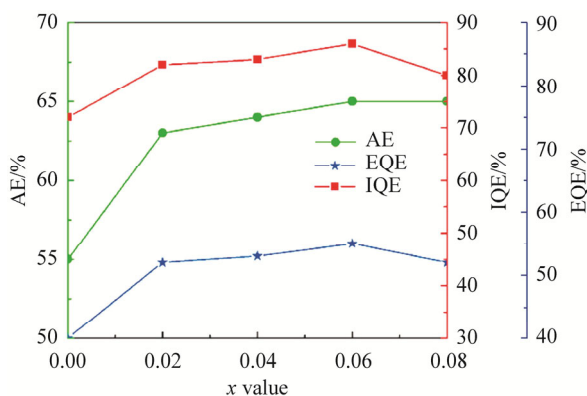


图1 荧光粉 $(\text{Ca}_{0.98-x}\text{Na}_x\text{Ce}_{0.02})_3\text{Sc}_2\text{Si}_3\text{O}_{12}$ 的XRD谱

Fig. 1 XRD patterns of $(\text{Ca}_{0.98-x}\text{Na}_x\text{Ce}_{0.02})_3\text{Sc}_2\text{Si}_3\text{O}_{12}$ phosphors



(a) Emission spectra



(b) AE, IQE and EQE

图2 不同 Na^+ 含量的CSS荧光粉的发射光谱、内/外量子效率及其吸收效率

Fig. 2 Variations of emission spectra, absorption efficiency (AE), internal quantum efficiency (IQE) and external quantum efficiency (EQE) of CSS phosphors with different Na^+ content

2.2 CSS 荧光膜的微观形貌及荧光性质

选取 $x=0.06$ 的 CSS 荧光粉制作荧光膜。从图 3a 可以看出, CSS 荧光粉有良好的分散性, 颗粒尺寸比较均匀。图 3b 显示了荧光粉分布在 $5\sim 24\ \mu\text{m}$ 之间, 平均粒径约为 $13\ \mu\text{m}$ 。从图 3c 荧光膜的表面形貌可以看出, 荧光膜由荧光粉紧密堆积而成, 粉体的间隙填充有 SiO_2 颗粒。从图 3d 荧光膜的断面形貌可以看出, 荧光膜紧密的粘结在蓝宝石衬底上, 膜层表面平整, 基本没有气孔, 荧光膜的厚度均匀, 平均厚度约为 $135\ \mu\text{m}$ 。二氧化硅充当无机黏结剂的主要机理是硅溶胶含有大量的 SiO_2 胶粒, 胶粒表面有大量的硅羟基 ($-\text{SiOH}$), 在干燥过程中, 胶粒之间的 $-\text{SiOH}$ 不断地脱水缩合, 形成硅氧烷 ($-\text{SiOSi}-$), 最终形成三维网络结构, 将荧光粉固定在网络结构中, 干燥后 SiO_2 填充在荧光粉颗粒之间的间隙内; 同时 SiO_2 颗粒包覆在荧光粉表面, 对荧光粉起到保护作用, 防止荧光粉被氧化^[31]。

CSS 荧光粉和对应荧光膜的稳态光谱测试结果显示于图 4a。在 $445\ \text{nm}$ 的蓝光激发下, 二者的发射光谱基本一致, 主要来源于 Ce^{3+} 的发射。而检测 $507\ \text{nm}$ 的激发谱, 观察到二者的最佳激发峰都位于 $445\ \text{nm}$ 附近, 主要对应于 Ce^{3+} 从 $4f$ 能级跃迁到 $5d_1$ 能级, 可以被商用的蓝光激光有效激发。图 4b 是 CSS 荧光粉和 CSS 荧光膜在 $445\ \text{nm}$ 蓝光激发下, 检测 $508\ \text{nm}$ 位置的荧光衰减曲线, 二者均呈现单指数衰减, 对应的荧光寿命分别是 $70.5\ \text{ns}$ 和 $69.9\ \text{ns}$,

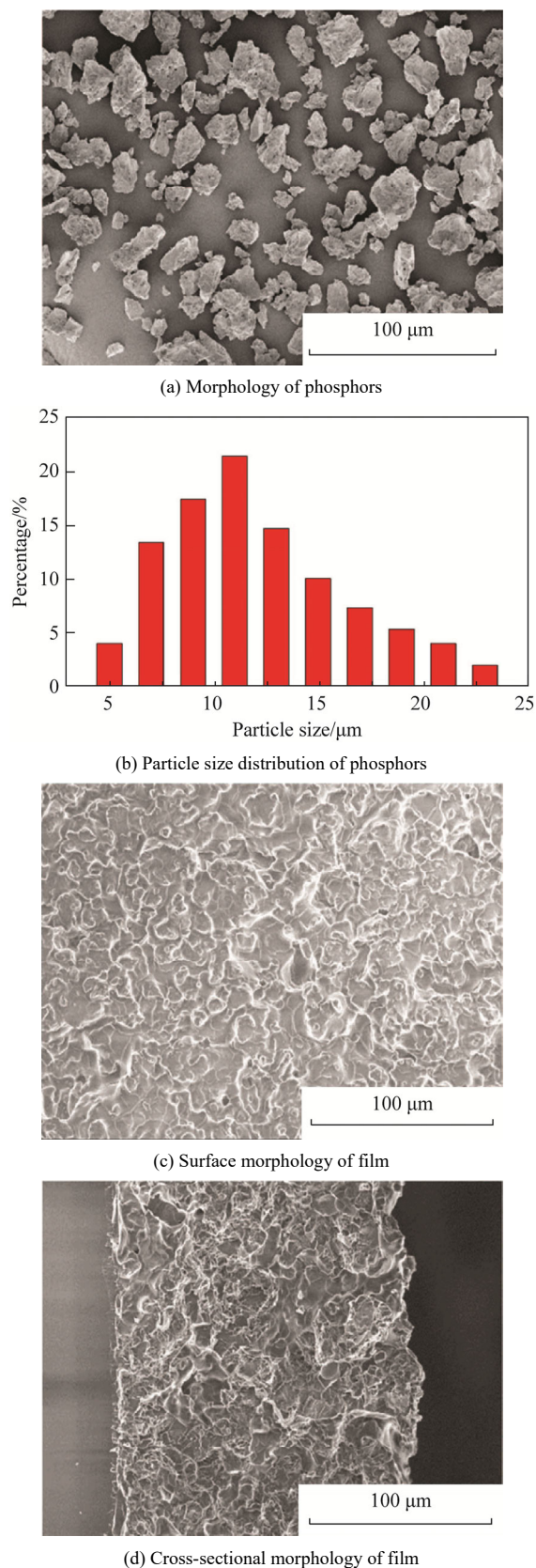


图 3 CSS 荧光粉的形貌、粒径分布以及相应荧光膜的表面形貌、断面形貌

Fig. 3 Morphology and particle size distribution of the CSS phosphors; Surface morphology and cross-sectional morphology of the corresponding phosphor film

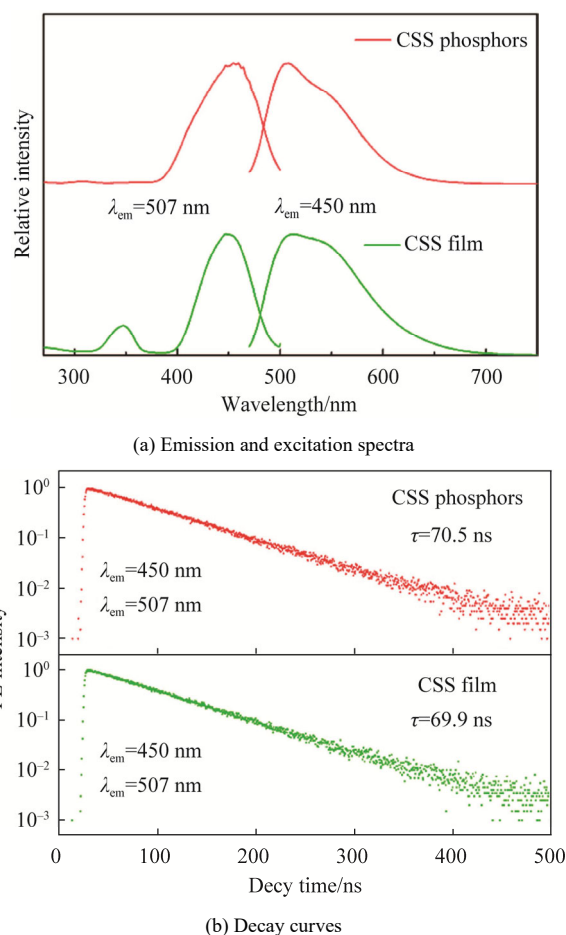


图 4 CSS 荧光粉和荧光膜的发射和激发光谱及其荧光衰减曲线

Fig. 4 Normalized emission and excitation spectra and decay curves of CSS phosphors and the corresponding phosphor film

没有明显的区别。此外, 还进一步测试了荧光膜的内量子效率和外量子效率, 分别是 84% 和 57%, 和 CSS 荧光粉得到量子效率(内、外量子效率分别是 86% 和 55%)基本一致, 在荧光膜的制备过程中, 荧光粉没有受到显著侵蚀。

对于激光照明, 荧光材料的热稳定性是其关键参数之一, 直接决定了激光照明光源的效率及其可靠性。图 5a 显示了在 452 nm 的蓝光激光激发下, CSS 荧光膜的光谱随温度的变化趋势。从图 5a 可以看出, 随着温度的升高, 荧光膜的荧光强度逐渐下降, 即发生了荧光热猝灭。其主要机理是随着温度的升高, 晶格振动增强, 激发态的电子无辐射弛豫到基态的概率增大, 荧光强度降低。从图 5b 可以看出, CSS 荧光膜和荧光粉的热稳定性基本一致, 在 200 °C 时荧光膜的荧光积分强度仍保持在常温下的 86%; 荧光膜具有优异的热稳定性, 可用于高亮度激光照明^[29]。

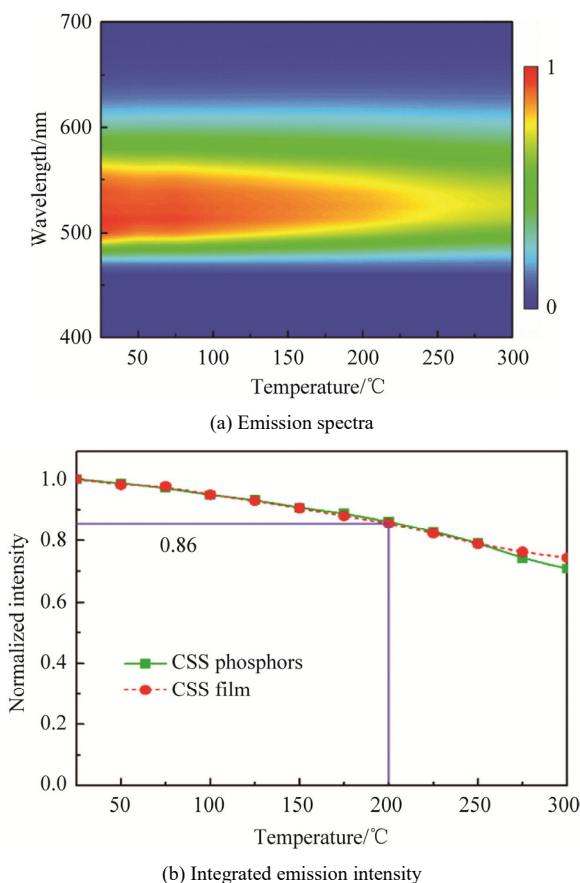


图5 CSS 荧光膜的荧光光谱及光谱积分强度随温度的变化规律

Fig. 5 Emission spectra and integrated emission intensity for CSS phosphor film as a function temperature

2.3 CSS 荧光膜在激光照明中的应用

测试了荧光膜在激光照明应用中的荧光性能。

图6为测试装置示意图，将荧光膜固定在积分球的中央，蓝光激光发射的激光经透镜聚焦在荧光复合膜上，利用积分球上的探测器测试了在不同激发功率下，激光照明的相关参数。从图7a可以看出，激光照明的光谱由2部分组成，位于452 nm窄带的蓝光对应于蓝光激光的发光，而位于470~720 nm的宽带蓝绿光对应于CSS荧光膜的发光。随着入射蓝光激光的功率从0.32 W增加到5.30 W，光谱中的蓝光和黄光成分同步增强，光源光谱的形状基本不变；继续增加入射激光功率，光谱中的蓝光强度继续增强，但蓝绿光强度却下降，出现荧光饱和现象。当入射激光功率为5.30 W时，激光照明光源的光通量最大，达到717 lm，此时的光效是135 lm/W (图7b)。荧光膜的荧光饱和主要是由荧光热猝灭引起的，随着入射激光功率的增大，荧光膜的温度逐渐升高，而温度的升高导致荧光效率的下降，产生更多的热量，引起温度进一步升高。

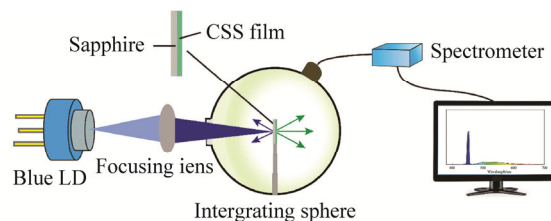


图6 激光照明测试系统的示意图

Fig. 6 Schematic of laser-driven white light and the photoelectric performance testing device for the phosphor film driven by blue laser

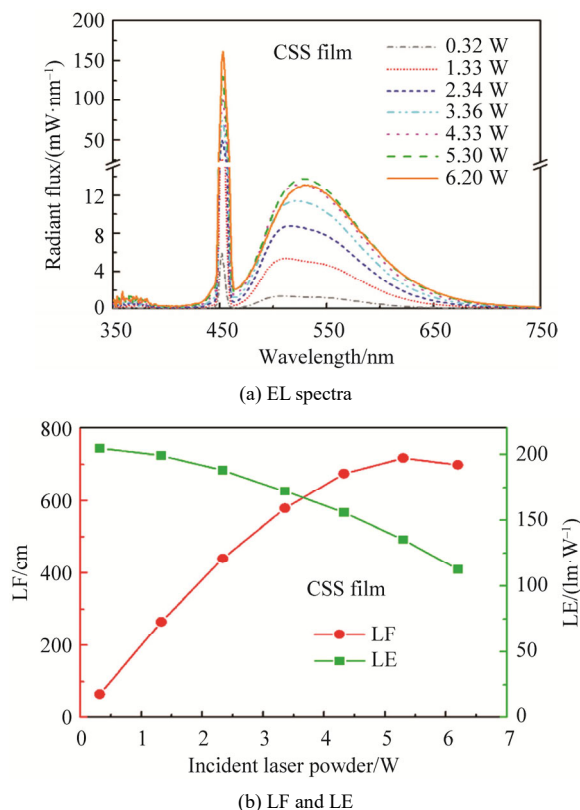


图7 CSS 荧光膜的电致发光光谱、光通量和荧光效率随入射激光功率的变化规律

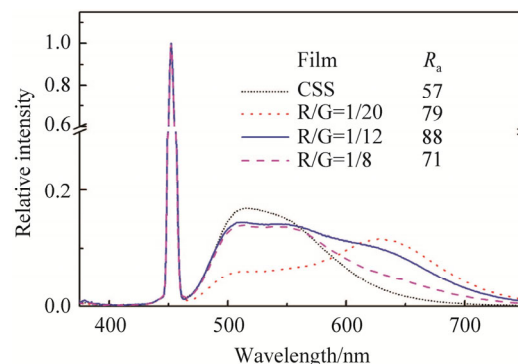
Fig. 7 Electroluminescence (EL) spectra, luminous flux (LF) and luminous efficacy (LE) of the CSS film as a function of the incident laser power

2.4 复合荧光膜的光谱调控

虽然CSS荧光膜可以实现高效的蓝绿光发射，但因光谱中缺少红光成分，导致光源的显色指数只有57，不能满足高品质照明的需求。为了提高激光照明光源的显色指数，利用商用CASN红色荧光粉来补充红光成分。通过调整不同荧光粉的比例，对荧光膜进行光谱调控。从图8可以看出，复合荧光膜的光谱分布在460~750 nm，光谱覆盖蓝绿光、黄光和红光区域。荧光膜的发射光谱主要有2个，主峰分别位于510 nm和620 nm，分别对用蓝绿色CSS荧光和CSASN红色荧光粉的发射。随着复合

膜中红色荧光粉含量的增加, 光谱中的红光成分逐渐增加。当红色荧光粉和蓝绿荧光粉质量比 $R/G=1/12$ 时, 光源呈暖白光, 光谱中蓝绿光、黄色和红色组分均衡分布, 发射光谱的半高宽达 176 nm, 光源显色指数 88, 色温 6 182 K。然后研究了 $R/G=1/12$ 的复合荧光膜在激光照明中的应用, 从图 9a 可以看出, 随着入射蓝光功率的增加, 光谱的强度逐渐增强, 其光通量逐渐增大。当蓝光功率为 5.3 W 时, 光通量达到最大 647 lm, 对应的光效是 122 lm/W。从图 9c 可以看出, 随着激光功率的增大, 光源的色坐标逐渐向蓝光区域转移, 色温逐渐升高, 其原因是随着入射功率的增大, 蓝光吸收效率和荧光转化效率均逐渐下降, 光源中的蓝光比例逐渐增多。此外, 还研究了复合荧光膜的热稳定性, 从图 9d 可以看出复合荧光膜具有良好的热稳定性, 当温

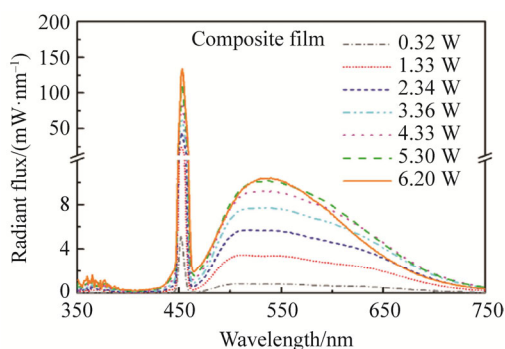
度升高至 200 °C 时其发光强度仍保持为常温下的 84%, 适用于高亮度激光照明。



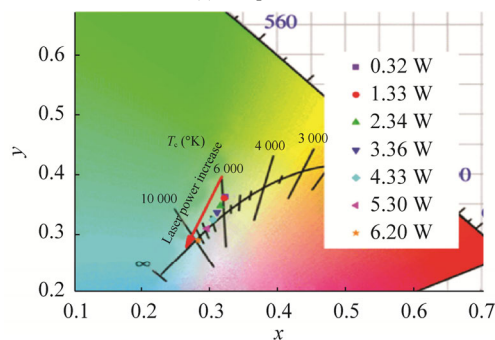
R/G is mass ratio of red to blue-green phosphor.

图 8 不同荧光粉组成的荧光膜在蓝光激光激发下的电致发光光谱

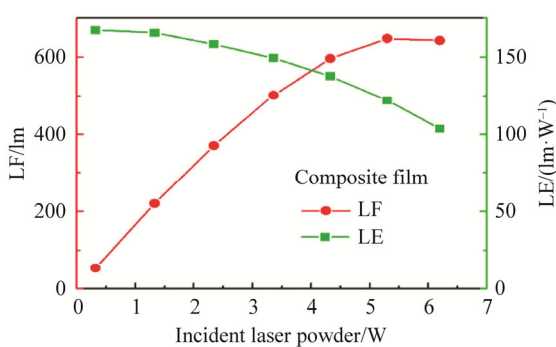
Fig. 8 Normalized EL spectra of the films with different content of phosphors driven by blue laser



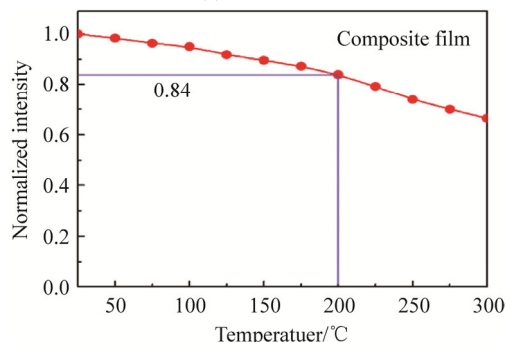
(a) EL spectra



(c) CIE color



(b) LF and LE



(d) Integrated emission intensity

图 9 复合荧光膜($R/G=1/12$)的电致发光光谱、光通量、荧光效率以及色坐标随入射激光功率的变化和荧光强度随温度的变化规律

Fig. 9 EL spectra, LF, LE and CIE color of the composite film ($R/G=1/12$) as a function of the incident laser power and integrated emission intensity as a function temperature

3 结论

利用二氧化硅溶胶作为无机黏结剂, 以自制的 CSS 蓝绿色荧光粉和商用 CSASN 红色荧光粉为荧光材料, 在高温高导率的蓝宝石衬底上, 低温制备出致密均匀的复合荧光膜。该荧光膜在 200 °C 时其荧

光强度仍保持在常温下的 84%, 其发射光谱位于 460~750 nm, 半高宽 176 nm, 光谱中蓝绿光、黄光和红光成分均衡分布。在蓝光激发下, 复合荧光膜产生 647 lm 的白光, 光效 122 lm/W, 显色指数是 88。该荧光膜制备工艺简单、性能优良在高显色激光照明中有广泛的应用前景。

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Phosphor/silica Phosphor Films as a Promising Color Converter for White Laser Lighting with Superior Color Rendering

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Extended Abstract

Introduction Laser-driven white lighting has received much attention due to its high brightness, high efficiency, and compact size. Blue laser active inorganic color converter technology is a commonly used method to produce white laser lighting due to its cost-effectiveness and simple structure. However, the existing laser lighting endures a low color rendering index (CRI) because of insufficient cyan-green and red light components in the spectra of existing color converters. Therefore, there is a demand for the development of color converters with intense cyan-green and red emission to improve the optical performance of laser lighting.

Methods (Ca_{0.98-x}Na_xCe_{0.02})₃Sc₂Si₃O₁₂ ($x=0, 0.02, 0.04, 0.06, 0.08$) (CSS) phosphors were synthesized by a solid-state reaction method. The prepared CSS phosphors were then blended with commercial CaAlSiN₃:Eu²⁺ (CASN) phosphors at different weigh ratios (abbreviated as R/G), which were chosen to be 1/8, 1/12 and 1/20. An inorganic binder used was commercially available silica colloidal particles, while an organic binder was polyvinyl pyrrolidone (PVP). Firstly, phosphors, PVP and silica colloidal particles were prepared by thoroughly mixing at a mass ratio of 150:50:1. The resulting slurries were coated on the sapphire substrates *via* blade coating. Finally, the dried samples were sintered in a muffle furnace at 600 °C for 20 min to remove the organic residues.

Results and discussion The CSS host has a cubic Garnet-type structure with the $Ia\bar{3}d$ space group. Na⁺ ions are introduced into the CSS host to function as a charge compensator. This enables the elimination of the impurity phases of phosphors. Under the excitation of blue light at 450 nm, the CSS phosphors emit a broadband cyan-green light with a peak at 507 nm. As Na⁺ content increases, the emission intensity of CSS phosphors gradually increases due to the enhanced absorption efficiency (AE) and internal quantum efficiency (IQE). As $x=0.06$, the emission intensity reaches its maximum, which is corresponding to internal/ external quantum efficiencies (IQE/EQE) of 86% and 55%, respectively.

Dense CSS films were produced using colloidal silica. Clearly, the film is composed of tightly packed phosphors and the gaps between phosphor particles are also filled with SiO₂. The film is bonded to the substrate tightly, which can be ascribed to the adhesion function of the colloidal SiO₂. The surface of the film is sufficiently smooth without polishing, and its thickness is 135 μm. The SiO₂ colloidal particles serve as an effective inorganic binder, a filler of gaps and a protective coating.

The excitation and emission spectra, decay curves and quantum efficiency of the CSS film are similar to those of the related CSS phosphors. The unimpaired luminescence properties of the film indicate that sintering does not impair the intactness of CSS phosphors. The CSS film displays a thermal quenching at evaluated temperature, and the decrease of the integrated emission intensity of CSS film aligns with that of CSS phosphors, retaining 86% of its initial value at 200 °C. When the incident laser power increases from 0.3 W to 5.30 W, the emission intensity of the film increases monotonously. However, when the laser power further increases to 6.20 W, the emission intensity of the film decreases sharply and a luminous saturation appears. The maximum luminous flux of 717 lm is achieved at an incident laser power of 5.30 W, and the corresponding luminous efficacy is 135 lm/W.

Although the CSS film exhibits a high thermal stability and a high luminous efficacy, the CRI of this phosphor-converted white light is 57 due to the deficiency of red light components. To improve the CRI, a composite film comprising CASN and CSS phosphors was fabricated. When the mass ratio of CSS and CASN phosphors is 1/12, the spectral analysis of the composite film reveals a uniform distribution of cyan-green, yellow and red light components, with a half-height width of 176 nm. Also, when excited by 5.3 W blue laser, the composite film emits a white light with a high luminous flux of 647 lm, a luminous efficacy of 122 lm/W and a CRI of 88.

Conclusions Highly efficient cyan-green CSS phosphors with an IQE/EQE of up to 86%/55% were prepared *via* introducing charge compensatory additives Na⁺. A composite phosphor film was developed on thermally conductive sapphire substrates using silica colloidal particles as an inorganic binder *via* combining the prepared cyan-green CSS and commercial red CaAlSiN₃:Eu²⁺ phosphors. The phosphor film exhibited a superior thermal stability and it maintained 84% of its room-temperature luminescence intensity even at 200 °C. The spectral analysis of the film showed an even distribution of cyan-green, yellow and red light components, with a half-height width of 176 nm. Under 5.3 W blue laser excitation, the composite film emitted a white light with a high luminous flux of 647 lm, a luminous efficacy of 122 lm/W and a CRI of 88. These results indicated that the composite phosphor film could have a promising potential for application in high-brightness laser lighting with superior color rendering.

Keywords laser lighting; color converter; high color rendering; garnet; calcium scandium silicate