



Screening and identification of bioorganic light-emitting substances from marine macrophytes

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Organic light-emitting materials in marine macrophytes from various coastal environments were identified. Twenty-eight species from the solvent fractions were examined and identified as candidates for bioorganic light-emitting materials using photoluminescence (PL) spectra and gas chromatography-mass spectrometry. We selected 16 solvent fractions from a total of 1,221 prepared from *Ishige okamurae*, *Sargassum confusum*, *Grateloupia elliptica*, *Chondracanthus intermedius*, *Porphyra yezoensis*, *Meristotheca papulosa*, *Gelidium amansii*, and *Scytosiphon lomentaria*. The maximum light-emitting PL spectra appeared at various colors, mainly between blue and green, based on chromaticity coordinates, from solvent fractions of *M. papulosa*, *G. amansii*, *G. elliptica*, *P. yezoensis*, *S. lomentaria*, *I. okamurae*, and *C. intermedius*. These results will contribute to the development of novel organic light-emitting materials.

Key Words: bioorganic light-emitting material; gas chromatography-mass spectrometry; marine macrophytes; photoluminescence spectrum

INTRODUCTION

The history of displays begins with early cathode-ray tubes, moving on to the first and second generation of liquid crystal displays (LCDs), and finally reaching the current generation of organic light-emitting diode (OLED) devices (Wong and Ho 2009). The OLED is a self-emitting form, and improvements have resulted in an improved viewing angle and outstanding performance with thin layers. The device is a simple one, and power consumption has been greatly improved over time. However, shortages of materials and problems relating to device life expectancy have yet to be solved satisfactorily. South Korea is a current leader, alongside the United States and Japan, in terms of the number of patent registrations relat-

ing to OLEDs. However, with regard to the materials used to produce OLEDs (a core technology), South Korea has only 14% of all such patents, and so room for expansion exists with respect to OLED display technology (Ministry of Science and Technology 2006).

The limitations of present OLED technology lie in the emission of light. The light-emitting materials used may affect the lighting field as well as the display field. For example, the green-colored light emitted by Alq₃ is the most commonly used light-emitting material (Hall and Schlegel 2001). However, although blue light-emitting materials with chemical stability, low color purity, and high flexibility exist, these tend to show whitening. Red light-



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emitting materials can reach a wavelength of 650 nm, but their emitting capabilities are less than ideal for industrial use (Seo and Moon 2014). Previous studies have investigated the optical and semiconducting properties of OLED materials (Karakawa et al. 2007, Sasabe et al. 2008) and were undertaken to find a way to avoid the dependence on the current range of OLED materials. In the present study, we used substances derived from living organisms to solve this problem. We required the light-emitting substance to be solid at room temperature, contain a benzene ring within its structure, have a range of molecular weights from 100 to 500 and a range of melting points above 100°C, and show little or no reaction with moisture (Karakawa et al. 2007, Sasabe et al. 2008). To examine the efficiency of LEDs, photoluminescence (PL) spectra were used to measure the bandgap from the full valence band to the empty conduction band, and to examine the basic characteristics and optical properties of the derivatives (Karakawa et al. 2007).

Few studies have reported on OLED materials derived from natural products / living organisms, and most OLED materials available at present are synthetic or a combination of organic and inorganic materials for increased functionality (Tajima et al. 2006). As “natural” OLED materials, chlorophyll *a*, chitin, cytochrome *c*, other proteins, and DNA have been studied (Steckl 2007, Singh et al. 2010), and researchers have continued to search for novel light-emitting materials, including biological products (Sanderson et al. 1987, Gomez et al. 2014). Bio-light-emitting materials derived from natural sources have been reported, including chitosan (D-glucosamine [2-amino-2-deoxy-D-glucose]), myoglobin, bovine serum albumin, deoxyribonucleic acid, cytochrome *c*, and chlorophyll (Hagen et al. 2006, Tajima et al. 2006, Steckl 2007, Singh et al. 2010, Rau et al. 2012). Adenine is a purine derivative that shows blue light emission in the 380-560 nm range, while cytosine can show high luminance at 380-560 nm. Emissions increase with purity at a reported maximum brightness of 2.16 cd m⁻² at a wavelength of 386 nm (Lewis 2006, Lee et al. 2007, Kim et al. 2010, Shin 2013). PL studies on the photosynthetic pigment chlorophyll *a* have also been reported (Choi et al. 2001), with the maximum peak wavelengths reported as 373 and 668 nm, respectively, and with respective high emitting ranges at 380-400 and 660-800 nm with a luminance of 1.16 cd m⁻² (Shin 2013).

Among marine organisms, algae are a well-known source of novel substances, such as proteins, fatty acids, vitamins, and pigments (Volk 2008). They also produce a wide variety of secondary metabolites with different bio-activities (e.g., antifungal, antiviral, antibiotic). Algae also

have potential for pharmaceutical research. In addition, those marine organisms that produce novel organic compounds are of particular interest.

This study was undertaken to identify bio-light-emitting materials and to estimate the efficiency of the light emission using the PL spectra. We derived the diverse functional organic light-emitting substances from marine macrophytes.

MATERIALS AND METHODS

Samples

Marine macrophytes were collected around the coastal areas of Ganjelgot in Ulsan City, Nameri in Gangneung, and Aewol in Jeju, South Korea. Materials were identified morphologically based on the “*Marine Algae of Jeju*” by Lee (2008) and Silva et al. (1996), Yoshida (1998) and Schneider and Wynne (2007) were used for further identification. The species of marine macrophytes examined comprised 5 Chlorophyta, 13 Phaeophyta, and 10 Rhodophyta (Table 1).

Extraction and separation

The collected marine macrophytes were rinsed, dried at room temperature, and then powdered. Each of the 100-g samples was powdered and extracted using 1,000 mL of 100% methanol (MeOH) over 25 h at room temperature. After filtration, the crude extract was concentrated to give a dark-green tar-like sticky residue (10 g) under reduced pressure in an evaporator, and then the MeOH extraction was purified by column chromatography using a Pyrex glass column (diameter : length = 24 : 40 cm) charged with a silica gel 60 (70 g). Elution was performed with a toluene and ethyl acetate in the following ratio: 7 : 3 in sequence. Fractionations were monitored by thin layer chromatography. Fractions of candidate organic compound materials were used to examine the samples in terms of their PL spectra, and promising candidates were selected based on their light-emitting properties.

Identification

Gas chromatography–mass spectrometry (GC-MS) analysis was performed using a Shimadzu GC MS-QP 2010-Plus (Shimadzu, Kyoto, Japan) and a 30 m × 0.25 mm × 0.25-μm Rtx-5MS capillary column (Shimadzu). The helium carrier gas was supplied at a head pressure of 35

psi to provide an initial flow rate of 1 mL min⁻¹. The temperature of the split injector was 260°C and the split ratio was 1 : 50. The injection volume for the fraction samples was 1 mL. The GC oven was maintained at 60°C for 3 min and then programmed at 100°C (2 min hold), followed by 200°C (5 min hold) and finally 235°C (10-min hold). Full-scan mass spectra were collected from *m/z* 85 to 550 at a data acquisition rate of 10 spectra s⁻¹. The MS transfer line was held at 250°C and the ion source temperature was 200°C. The electron energy was 70 eV. Isolated peaks were identified by comparison with the National Institute of Standards and Technology Library (Gaithersburg, MD, USA).

PL spectra

The selected purified substances were examined in terms of their PL spectra using the apparatus shown in Fig. 1. The substances were exposed to UV radiation at

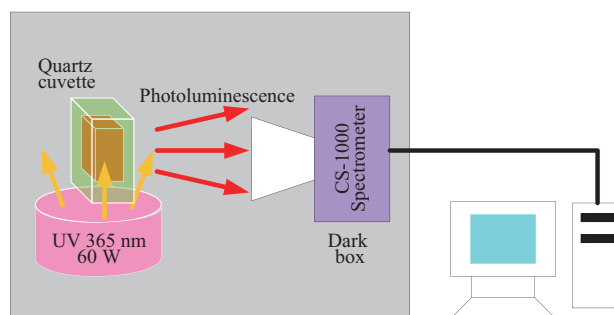


Fig. 1. Schematic diagram of photoluminescence analysis.

365 nm in a 1-mL crystal cuvette, and PL reactions were initiated. The CS-1000 spectra were used to measure the chromaticity, brightness, and light emission of the samples (Fig. 1).

Table 1. List of marine macrophytes collected in South Korean coastal areas

Phylum	Order	Family	Species			
Chlorophyta	Ulvales	Ulveaceae	<i>Ulva pertusa</i> <i>Enteromorpha intestinalis</i>			
		Cladophorales	Cladophoraceae	<i>Cladophora wrightiana</i>		
	Siphonocladales	Siphonocladaceae	<i>Cladophoropsis herpestica</i>			
		Codiolales	Codiaceae	<i>Codium fragile</i>		
Phaeophyta	Dictyotales	Dictyotaceae	<i>Dictyota okamurae</i> <i>Padina arborrescens</i>			
			Scytosiphonales	Scytosiphonaceae	<i>Scytosiphon lomentaria</i>	
	Ishigeales	Ishigeaceae	<i>Ishige okamurae</i> <i>Ishige sinicola</i> <i>Ecklonia cava</i> <i>Ecklonia kurome</i> <i>Undaria pinnatifida</i>			
			Laminariales	Alariaceae	<i>Laminaria japonica</i>	
					Fucales	Sargassaceae
	Rhodophyta	Bangiiales	Bangiaceae	<i>Porphyra yzoensis</i>		
		Gelidiales	Gelidiaceae	<i>Gelidium amansii</i> <i>Chondrus ocellatus</i>		
				Gigartinales	Gigartinaceae	<i>Chondracanthus intermedius</i> <i>Meristotheca papulose</i> <i>Delisea pulchra</i>
		Bonnemaisoniales	Bonnemaisoniaceae			<i>Grateloupia elliprica</i> <i>Grateloupia kurogii</i> <i>Grateloupia angusta</i> <i>Grateloupia okamurai</i>
						Halymeniales

RESULTS

Purification

In total, 1,221 solvent fractions were isolated from 28 marine macrophyte species. Sixteen solvent fractions with no pigments were selected and verified by PL reactions. The structures and material properties of the candidate substances were analyzed by GC-MS. In total, 14 bioorganic light-emitting materials identified from the 16 solvent fractions were selected for potential use as OLED materials (Tables 2 & 3).

Identification

1,3,5-Cycloheptatriene and benzyl isopentyl ether

were found in *Ishige okamurae* and *Sargassum confusum*; *S. confusum* also contained isopropyl phenylacetate. 1,1-Diphenyl-3-[methoxymethoxy]-1-butene was identified from *Grateloupia elliptica*. Benzene acetic acid, 3-methoxy-4-[(trimethylsilyloxy)-ethyl ester, trimethylsilyl ergometrine, 9H-fluorene-2-carboxylic acid, 9-oxo-[2-hydroxyethyl] [methyl]amide, and benzamide, 2-chloro-4-trifluoromethyl were found in *Chondracanthus intermedius*, *Porphyra yezoensis*, *Meristotheca papulosa*, and *Gelidium amansii*. Carbamic acid and 1-(4-acetamidoanilino)-3,7-dimethylbenzo[4,5]imidazo[1,2-a]pyridine-4-carbonitrile were identified from *G. elliptica*. 1-[4-Acetamidoanilino]-3,7-dimethylbenzo[4,5]imidazo[1,2-a]pyridine-4-carbonitrile and 8,9,9,10,10,11-hexafluoro-4,4-dimethyl-3,5-dioxatetracyclo [5.4.1.0[2,6],0[8,11]]dodecane were found in

Table 2. List of OLED candidates identified by GC-MS

Species	Name of substance	Molecular weight	Retention time (min)
<i>Ishige okamurae</i>	1,3,5-Cycloheptatriene	92	3.57
<i>Sargassum confusum</i>			14.14
<i>Sargassum fulvellum</i>			4.98
<i>Ishige okamurae</i>	Benzyl isopentyl ether	178	10.99
<i>Sargassum confusum</i>			5.12
<i>Ecklonia cava</i>			16.78
<i>Cladophoropsis wrightiana</i>			30.41
<i>Sargassum confusum</i>	Isopropyl phenylacetate	178	13.69
<i>Grateloupia elliptica</i>	1,1-Diphenyl-3-[methoxymethoxy]-1-butene	268	12.4
<i>Chondracanthus intermedius</i>	Benzeneacetic acid, 3-methoxy-4-(trimethylsilyloxy)-ethyl ester	282	11.53
<i>Grateloupia elliptica</i>	Carbamic acid	353	33.19
	1-(4-Acetamidoanilino)-3,7-dimethylbenzo[4,5]imidazo[1,2-a]pyridine-4-carbonitrile	369	8.8
<i>Porphyra yezoensis</i>	Trimethylsilylergometrine	397	11.7

OLED, organic light-emitting diode; GC-MS, gas chromatography-mass spectrometry.

Table 3. List of OLED candidates identified by GC-MS

Species	Name of substance	Molecular weight	Retention time (min)
<i>Meristotheca papulosa</i>	1-[4-Acetamidoanilino]-3,7-dimethylbenzo[4,5]imidazo[1,2-a]pyridine-4-carbonitrile	369	12.4
	8,9,9,10,10,11-Hexafluoro-4,4-dimethyl-3,5-dioxatetracyclo[5.4.1.0[2,6],0[8,11]]dodecane	302	35.64
<i>Meristotheca papulosa</i>	9H-Fluorene-2-carboxylic acid, 9-oxo-, [2-hydroxyethyl] [methyl]amide	281	22.56
<i>Scytosiphon lomentaria</i>	4-Methyl-2,4-bis[4'-trimethylsilyloxyphenyl]pentene-1	412	33.43
	Trimethyl[4-[1,1,3,3,-tetramethylbutyl]phenoxy]silane	278	30.63
<i>Gelidium amansii</i>	Benzamide, 2-chloro-4-trifluoromethyl	233	14.84
	2-[1,3-Dihydro-3,3-dimethyl-1-phenyl-3,4-benzofuran-1-yl]acetic acid	282	9.8

OLED, organic light-emitting diode; GC-MS, gas chromatography-mass spectrometry.

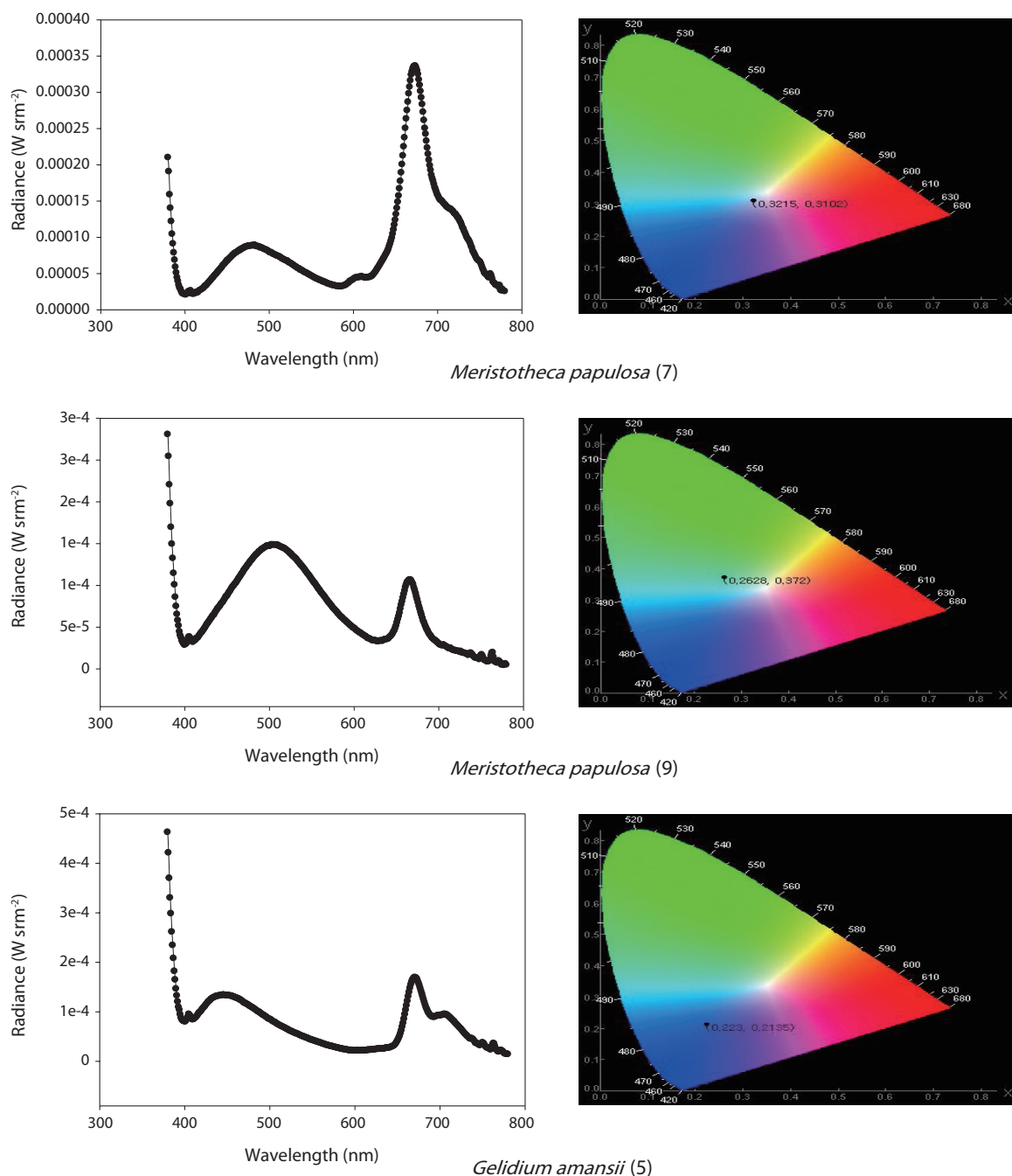


Fig. 2. Photoluminescence spectra of candidate light-emitting materials (order of extracted fraction).

M. papulosa. 4-Methyl-2,4-bis[4'-trimethylsilyloxyphenyl]pentene-1 and trimethyl[4-[1,1,3,3,-tetramethylbutyl]phenoxy]silane were identified from *Scytosiphon lomentaria*, and benzamide, 2-chloro-4-trifluoromethyl, and 2-[1,3-dihydro-3,3-dimethyl-1-phenyl-3,4-benzofuran-1-yl]acetic acid were found in *G. amansii*.

Photoluminescence spectra

Two solvent fractions of *M. papulosa* showed purple and sea-green PL reactions (Fig. 2). A solvent fraction of *G. amansii* exhibited maximum light emission peaks at 380-400 nm and 600 nm, similar to the chlorophyll *a* PL

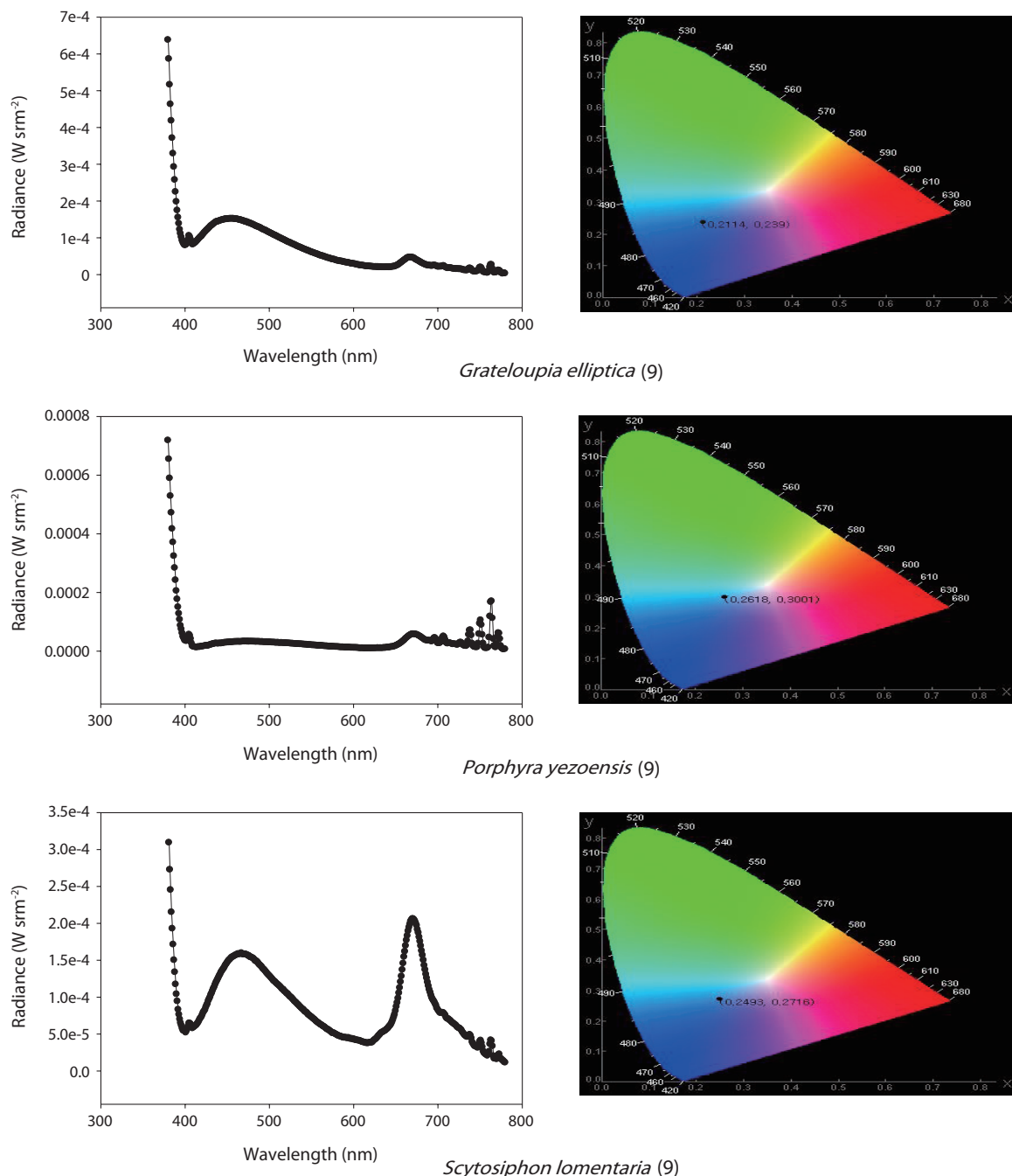


Fig. 3. Photoluminescence spectra of candidate light-emitting materials (order of extracted fraction).

reaction, but appeared dark blue (Fig. 2). Maximum light emission peaks were exhibited for *G. elliptica* at 380-400 and 430-500 nm (Fig. 3), and *P. yezoensis* at 390-400 and 680-800 nm with a blue color (Fig. 3), while *S. lomentaria* showed maximum light emission peaks at 380-400 and 680-700 nm (Fig. 4). *I. okamurae* (Rhodophyta) and *C. intermedius* showed maximum light emission peaks at 440-

520 nm, with a color between blue and green, based on chromaticity coordinates.

DISCUSSION

Choi et al. (2001) reported on an OLED device consist-

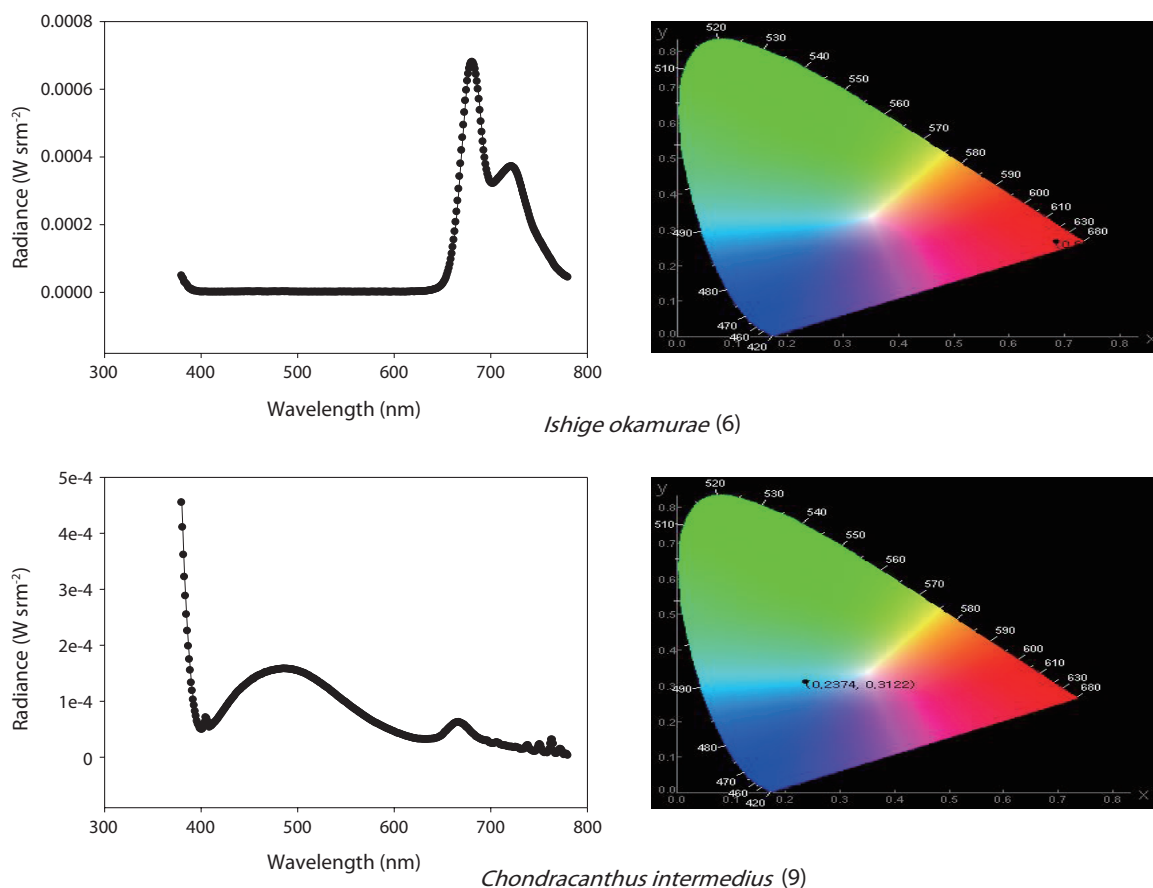


Fig. 4. Photoluminescence spectra of candidate light-emitting materials (order of extracted fraction).

ing of a single layer of chlorophyll *a* and aluminum, with a coating of chlorophyll *a* using the Langmuir-Blodgett method. The OLED efficiency and thickness of the chlorophyll coating both increased. However, chlorophyll *a* is not used in commercial OLEDs because its chemical bonds break down with heat, and also the high costs associated with its commercial use. Chlorophyll *a* has a blue irradiance of electroluminescence from 300 to 400 nm, with low red band wavelengths and a tetrapyrrole ring with magnesium atoms in the porphyrin at its center. A great deal of research has investigated its PL properties. Chlorophyll *a* emits mainly at 380-400 and 660-800 nm, and its maximum emission wavelengths have been found to show a brightness of 1.16 cd m^{-2} at 373 and 668 nm.

Generally, higher plants and animals, fungi, and yeast have cytochrome *c* as part of the electron transport system within their mitochondria. Reductive sulfate protein absorbance bands appear at 550, 520, and 415 nm, while an oxidative protein has an absorbance band at 407 nm. Tajima et al. (2006) reported on the use of cytochrome *c* in

an OLED coating plate.

To determine the methods for OLED candidate PL measurements, Alq3 is a commercially available light-emitting material with a known spectrum and chromaticity coordinates. Its PL spectrum is in the range of 480-700 nm, its luminance is 18.66 cd m^{-2} , and the maximum emission wavelength was found to be 531 nm in primary examination (Shin 2013). Furthermore, commercial *N,N'*-bis-(1-naphthalenyl)-*N,N'*-bis-phenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) PL spectrum and color analysis chromaticity coordinates are available and can be used as a representative material with benzene rings and two amines. NPB shows a 400-700 nm PL spectrum, broad emissions with green and blue chromaticity coordinates, and color purity. The luminance is 10.15 cd m^{-2} and the maximum emission wavelength is 488 nm. Chitosan PL spectra and chromaticity coordinates (reference material) are also available. Chitosan is derived from crustacean shells and has been assessed as an OLED material, but requires a high purity. Unlike other substances, it emits

from 380 to 400 nm (blue), showing high color purity up to 0.59 cd m⁻² and a maximum light emission wavelength of 380 nm.

In this study, we described the identification and physical properties of functional bio-light-emitting materials from marine macrophytes. The compounds were first identified as functional bio-light-emitting materials. In total, 16 solvent fractions were selected and confirmed by the PL reactions, and structures and material properties were analyzed for candidate substances by GC-MS. Fourteen bio-light-emitting materials identified from the 16 solvent fractions potentially contained OLED materials from 28 strains of marine macrophytes. Many benzene derivatives were identified, including benzylisopentyl ether, isopropyl phenylacetate, 1,1-diphenyl-3-[methoxymethoxy]-1-butene, benzene acetic acid, benzamide, 4-methyl-2,4-bis[4'-trimethylsilyloxyphenyl]pentene-1, and trimethyl[4-[1,1,3,3-tetramethylbutyl]phenoxy]silane benzamide. In addition, two fluorine compounds, 9H-fluorene-2-carboxylic acid and 8,9,9,10,10,11-hexafluoro-4,4-dimethyl-3,5-dioxatetracyclo[5.4.1.0[2,6],0[8,11]]dodecane, were recognized. One amine, carbamic acid, was discovered in *G. elliptica*. Two solvent fractions of *M. papulosa* showed purple and sea-green PL reactions. The solvent fraction of *G. amansii* exhibited maximum light emissions at 380-400 and 600 nm, similar to chlorophyll *a* but with a dark-blue color. The maximum light emissions shown by *G. elliptica* were at 380-400 and 430-500 nm, while those of *P. yezoensis* were at 390-400 and 680-800 nm with a blue color. *S. lomentaria* exhibited maximum light emissions at 380-400 and 680-700 nm. *I. okamurae* (Rhodophyta) and *C. intermedius* showed maximum light emissions at 440-520 nm, the color of which was between blue and green based on chromaticity coordinates.

In conclusion, marine algae were screened to discover new OLED material candidates. As an example, we extracted the red alga *G. amansii* with an organic solvent system and confirmed the active fraction using PL methods and characterized the bio-light-emitting materials by GC-MS analysis. The OLED candidates identified in this study should be studied further with a view to improving their emission efficiencies.

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