Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information for

Long-term stability improvement of light-emitting diode using highly transparent graphene oxide paste

Seungae Lee,[b] Yun Ki Kim[a] and Jyongsik Jang*[a]

[a]School of Chemical and Biological Engineering, College of Engineering, Seoul National University, Gwanak 1, Gwanak-ro, Gwanak-gu, Seoul 151-742, Korea

[b]Department of Materials Science and Engineering, Northwestern University, 2220 Campus Drive, Evanston, IL 60208, USA

1. Reliability test results of white light-emitting diodes with various-sized graphene oxide-embedded paste

Various-sized graphene oxide (GO) sheets were fabricated through a ball-milling process and a Hummers' method. The prepared GO sheets were incorporated in a die attach paste adhesive of white light-emitting diode (LED) using the solvent-exchange method. To confirm an influence of GO size on the LED performance, the reliability test was carried out on the LEDs comprised with the GO-embedded paste (Table S1). In case of using 50 nm-sized GO sheets, the long-term stability of the LEDs was improved under the condition of 85 °C compared to that of the LEDs with conventional white paste. On the contrary, under a condition of 85 °C and 85% relative humidity (RH), the relative luminous flux of the LEDs with the GO paste was lower than that of the LEDs with white paste after 1000 h. In case of using the GO sheets with a size range from 100 to 150 nm, the reliability of LEDs was enhanced under the condition of 85 °C. Under the condition of 85 °C and 85% RH, the relative luminous flux of the LEDs was similar to that of the LEDs composed of white pate. When the micrometer-sized GO sheets (1–2 μm) were utilized, the reliability of LEDs was slightly improved compared to that of the conventional LEDs under the condition of 85 °C. Interestingly, under the condition of 85 °C and 85% RH, the long-term stability of the LEDs was significantly enhanced compared with the LEDs with white paste. The results of the reliability test depending on the size of GO sheets were explained by the dispersibility of GO sheets and moisture barrier property. As the size of GO sheets decreased, the GO sheets were evenly dispersed in silicone paste, resulting in the effective heat dissipation and high transmittance. On the other hand, the protection ability against external moisture, *i.e.* moisture barrier property, was reduced as the size of GO sheets decreased. Accordingly, the reliability of LEDs was declined under the high-moisture condition (85% RH) when the small-

sized GO sheets were used. When the 50 nm-sized GO sheets were embedded in silicone paste, the effect of GO sheets on the moisture barrier property declined due to the shorter mean-free path. In case of the conventional white pate, several fillers, such as zinc oxide (ZnO) and aluminum oxide (Al_2O_3), are added to silicone paste in order to enhance the heat dissipation. The size of ZnO which is included in white paste in large quantities is from 0.1 to 5 μm with an average size of 1 μm. It is considered that the white paste exhibited slightly better moisture barrier property compared to the GO paste composed of the 50 nm-sized GO sheets because of the effect of relatively large-sized ZnO. On the contrary, the LEDs comprised with the large-sized GO sheets exhibited outstanding moisture barrier property. However, the large-sized GO sheets were not evenly dispersed in silicone paste and formed some aggregation, reducing transparency. Consequently, the GO sheets having a size range from 300 to 400 nm were used in this research because it represented high dispersibility and excellent moisture barrier property.

Table S1. Reliability test results of white light-emitting diodes comprised of various-sized graphene oxide (GO)-embedded paste adhesive.[a]

Test condition	Properties	White paste	Graphene oxide paste[b]			
			50 nm	100–150 nm	300–400 nm	1–2 μm
85 °C	Forward voltage variation [V][c]	0.56	0.84	0.5	0.52	0.74
	CIE x coordinate variation[d]	0.0011	–0.0017	–0.0022	0.0008	–0.0021
	CIE y coordinate variation[d]	0.0036	0.0011	0.0003	0.0027	0.0009
	Relative luminous flux [%][e]	99.47	99.85	99.82	99.81	99.56
85 °C, 85% RH	Forward voltage variation [V][c]	0.10	0.40	0.79	0.10	0.23
	CIE x coordinate variation[d]	–0.0010	–0.0043	–0.0036	–0.0006	–0.0008
	CIE y coordinate variation[d]	–0.0032	–0.0043	–0.0024	–0.0026	–0.0027
	Relative luminous flux [%][e]	88.92	88.86	88.94	90.71	90.85

[a] Total operating time was 1000 h and applied current was 400 mA.

[b] The concentration of graphene oxide in silicone paste was 0.004 wt%.

[c] The initial forward voltage was assumed as zero for evaluating the variation of forward voltage.

[d] CIE coordinates were analyzed according to CIE 127, and the variation of CIE coordinates were evaluated by regarding the initial value as origin of coordinates.

[e] Relative luminous flux was acquired by considering the initial luminous flux as *ca.* 100%.

2. Thermal properties of LEDs with the GO-embedded paste adhesive

Thermal properties of white LEDs made of GO-embedded paste adhesive were measured at 25 °C and 85 °C respectively (Table S2). The LEDs with the GO paste showed lower junction temperature (T_j) and thermal resistance (R_{th}) than the LEDs with white paste at both 25 °C and 85 °C.

Table S2. Thermal properties of white light emitting diode made of graphene oxide (GO)-embedded paste adhesive.[a]

Samples	Temperature	Junction temperature (T_j)	Thermal resistance (R_{th})
	[°C]	[°C]	[K W^{-1}]
White paste	25	67.8	29.9
	85	131.8	34.3
0.004 wt% GO paste	25	61.7	24.8
	85	125.8	28.7

[a] Drive current was 400 mA.

Judging from the variation of thermal capacitance (C_{th}) depending on thermal resistance (R_{th}), the LEDs containing the GO paste exhibited lower thermal resistance than that of the white paste (Fig. S1).

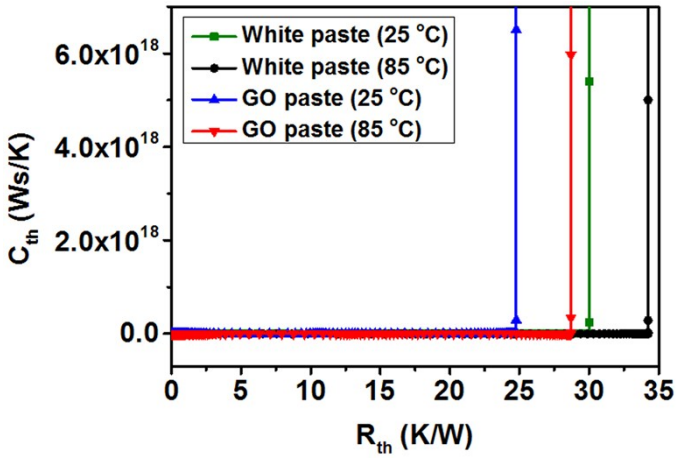

Fig S1. Variation of thermal capacitance (C_{th}) depending on thermal resistance (R_{th}) for the LEDs with white paste and GO paste.

3. Moisture barrier properties of the LEDs with various-sized GO-embedded paste

The adhesive strength was enhanced with increasing the GO contents. In case of the moisture barrier property, the moisture absorption value was reduced with increasing the concentration of GO, representing improved moisture barrier property (Fig. S2).

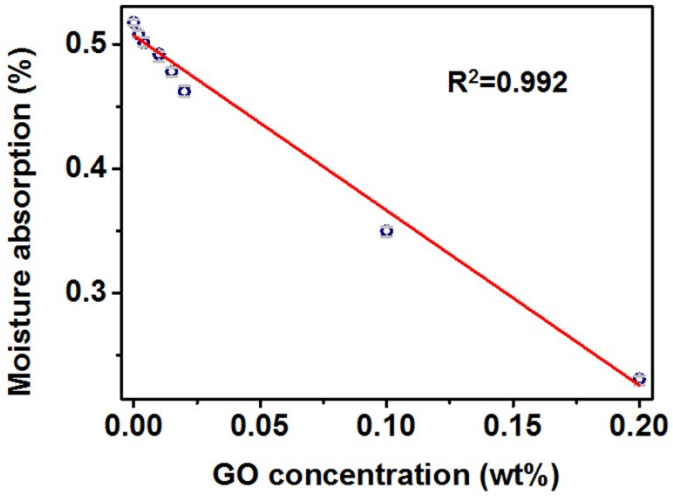

Fig. S2. Moisture absorption value of silicone resin containing various concentrations of GO.