**Electronic Supplementary Information** 

## Binder-Free Heat Dissipation Films Assembled with Reduced Graphene Oxide and Alumina Nanoparticles for Simultaneously High In-Plane and Cross-Plane Thermal Conductivities

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S1. TEM observation of alumina NPs and nanoscale-voids of alumina NPs



**Figure S1.** (a) TEM image of 80 nm-sized alumina NPs (purchased from Korea Nanomaterials, image is available at the company's website: <u>https://koreanano.co.kr/shop/item.php?it\_id=KRU3008</u>), (b) Cross-sectional TEM observation of rGO/alumina complexed film.

**S2.** Thickness and size information of the Cu, rGO, and rGO/alumina films.

**Table S2.** Thickness and diameter of Cu, rGO and rGO/alumina films.

Sample	Thickness	Size
Cu	10 µm	Diameter 4 cm
rGO	9.3 µm	Diameter 4 cm
rGO/alumina	9.8 µm	Diameter 4 cm

S3. Electrical insulation between LED and heat dissipation film for thermal management tests.

Table S3. Measured electrical resistivities of heat dissipation films before and after applying silicon-based grease

	Electrical resistivity without grease (Ω.m)	Electrical resistivity with grease (Ω.m)
Cu	$2.2 \times 10^{-8}$	$5.49  imes 10^4$
rGO	$1.6 \times 10$	$6.71  imes 10^4$
rGO/alumina	4.2 × 10	$7.24  imes 10^4$