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Electronic Supplementary Information for

# Cartilage-inspired superelastic ultradurable graphene aerogels prepared by selective gluing of intersheet joints

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## 1. Formation of GA@PDMS nanocomposite



**Fig. S1** (a) Schematic illustration for fabricating the GA and GA@PDMS nanocomposite. (b–c) proposed mechanism of rGO wet–gel formation under acidic conditions.

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#### 2. Electrical resistance measurements



**Fig. S2** Electrical resistance of the pristine GA and the GA@PDMS nanocomposites as a function of the PDMS amounts. All electrical measurements were conducted with Agilent Semiconductor Parameter Analyzer (4155C).

As shown in the **Fig. S2**, electrical resistance of the GA@PDMS nanocomposite were investigated as a function of PDMS amounts (0.1, 0.5, 1.0, 3.0 and 5.0 wt.%). There has not been much change in the electrical resistance up to 0.5 wt.% of PDMS contents. However, excessive insertion of PDMS can inherently produce detrimental effects on electrical conductivity because of its electrical insulating properties. Therefore, further increase in PDMS amount resulted in an increase in electrical resistance and then, reached the maximum value of 128.4 k $\Omega$  at 5.0 wt% of PDMS. The electrical resistance value of GA@PDMS nanocomposite (5.0 wt.%) was 5 times higher than that of pristine GA (25.6 k $\Omega$ ). Interestingly, this tendency becomes more pronounced when the loading pressure was applied, presumably because the excessively introduced PDMS onto graphene layer hamper the formation or redistribution of conducting path. Under 3.0 kPa of loading pressure, the The electrical resistance value of GA@PDMS nanocomposite (5.0 wt.%) was 25.0 k $\Omega$ , which is about 7-fold greater compared to the pristine GA at same loading pressure (3.6 k $\Omega$ ). Electronic Supplementary Material (ESI) for Chemical Communications This journal is © The Royal Society of Chemistry 2016

#### 3. Pore characterizations of the GA@PDMS nanocomposites



**Fig. S3** Pore characterization of the pristine GA and the GA@PDMS nanoconposite. Nitrogen adsorption-desorption isotherms and BJH pore distribution curves (inset) of (a) the pristine GA and (b) GA@PDMS nanocomposite. The BET surface areas and nitrogen adsorption-desorption isotherms were measured at 78 K using BELSORP-mini II analysis program.

	Surface area and pore size analyzer [a]			
Sample	Specific surface area	Total pore volume (cm <sup>3</sup> g <sup>-1</sup> )		
	$(m^2 g^{-1})$			
Pristine GA	302.4	1.48		
GA@PDMS nanocomposite	293.2	1.45		

Table S1. Pore characterization results of the pristine GA and GA@PDMS nanocomposite.

<sup>[a]</sup> These values were obtined by high precision surface area and pore size analyzer (BELSORP-mini II).

The nitrogen adsorption-desorption isotherms and the Barrett-Joyner-Halenda pore distribution curves of the pristine GA and GA@PDMS nanocomposite are displayed in **Fig. S3**, and the results are summarized in **Table S1**.

Most importantly, there is no significant difference between the pristine GA and GA@PDMS nanocomposite. They exhibit similar pore features (*e.g.* type of adsorption/desorption isotherms and pore volume). For both pristine GA and GA@PDMS nanocomposite, the type IV adsorption-desorption isotherm curves were demonstrated along

with a type H3 hysteresis loop. The surface areas of the pristine GA and GA@PDMS nanocomposite, which were calculated by BET equation using N<sub>2</sub> adsorption/desorption isotherms, were also estimated to be 302.4 m<sup>2</sup> g<sup>-1</sup> and 293.2 m<sup>2</sup> g<sup>-1</sup>, respectively. From the BJH pore size distribution, it was also observed that the GA@PDMS nanocomposite has a pore volume of 1.45 cm<sup>3</sup> g<sup>-1</sup> with pore diameter in the range of about 10 to 200 nm. This result is almost the same as the value of pristine GA before PDMS introduction (pore volume of 1.48 cm<sup>3</sup> g<sup>-1</sup>).

Taking these results into account, it is concluded that there was no or little effect on the pore characteristics of the pristine GA, which indicating that most pore structures of pristine GA remained after selective insertion of PDMS.

# 4. Comparison between the pristine GA and GA@PDMS nanocomposite

The influence of the selective insertion of PDMS on the physical, mechanical and electrical properties of the pristine GA was demonstrated as below:

Table 52. Comparison between the pristine Grand Grand Birls handcomposite								
	Physical, mechanical and electrical properties							
Sample	Bulk	Elastic	Electrical					
	density	modulus <sup>[a]</sup>	conductivity <sup>[b]</sup>					
	$(mg  cm^{-3})$	(kPa)	$(S m^{-1})$					
Pristine GA	17.3	42.7	$0.38 \times 10^{-2}$					
GA@PDMS nanocomposite (0.5 wt%)	18.5	87.7	$0.39 \times 10^{-2}$					

 Table S2. Comparison between the pristine GA and GA@PDMS nanocomposite

<sup>[*a*]</sup> The mechanical properties were characterized by a TA Instruments DMA (RSAIII). <sup>[*b*]</sup> These values were calculated using Agilent Semiconductor Parameter Analyzer (4155C).

### 5. Mechanical properties of the GA@PDMS nanocomposites



Fig. S4 Compressive stress-strain curves of the pristine GA and GA@PDMS nanocomposites (0.1, 0.5, 1.0, 3.0 and 5.0 wt.%) measured by a dynamic mechanical analyzer with 90% of compressive strain.

Table S3. Mechanical properties comparison as a function of PDMS amount <sup>(a)</sup>								
	Pristine GA	0.1 wt% PDMS	0.5 wt% PDMS	1.0 wt% PDMS	3.0 wt% PDMS	5.0 wt% PDMS		
Elastic modulus (kPa)	42.7	48.2	87.7	55.4	43.1	17.5		
Max. compressive strength (kPa)	174.0	352.9	448.2	315.5	188.4	194.6		

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<sup>[a]</sup> The mechanical properties were characterized by a TA Instruments DMA (RSAIII).