

## Electronic Supplementary Information

Stretchable, self-healing and adhesive sodium alginate-based composite hydrogels as  
wearable strain sensors for expansion-contraction motion monitoring

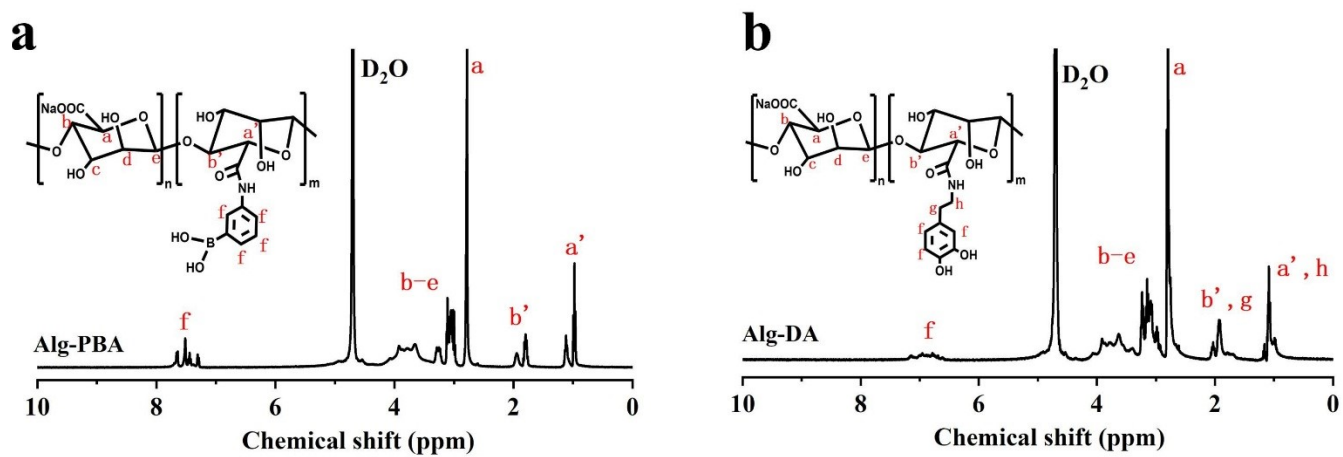
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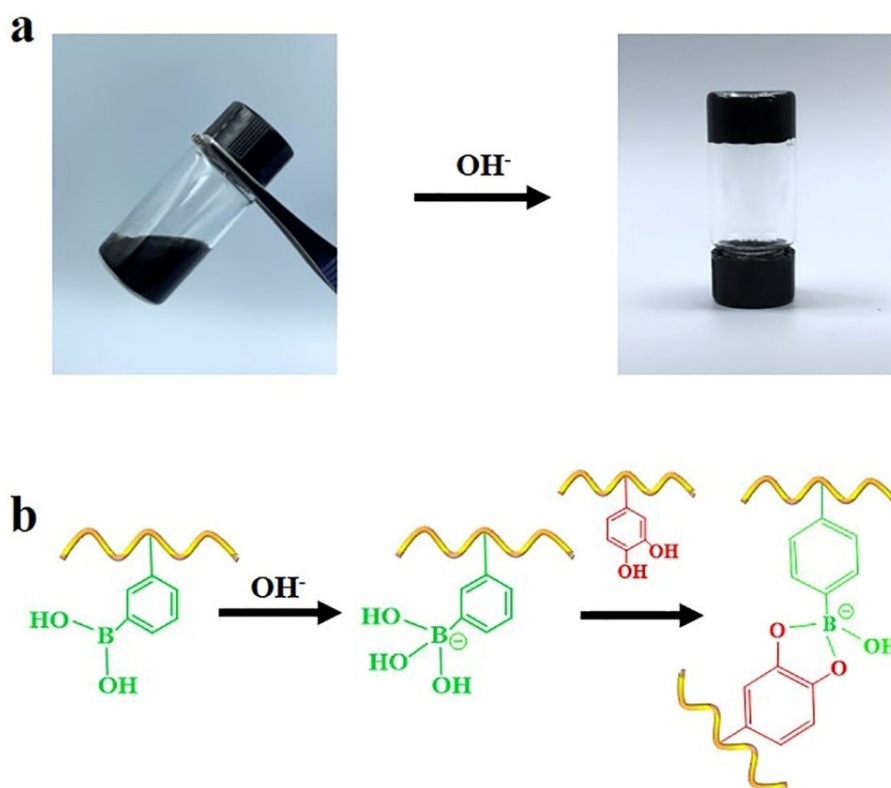
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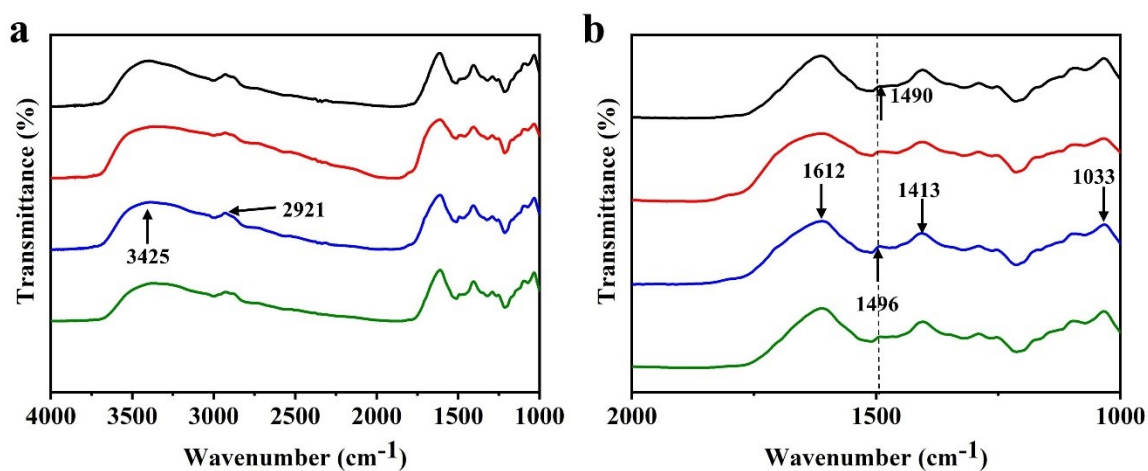
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**Fig. S1.**  $^1\text{H}$  NMR spectra of (a) Alg-PBA, and (b) Alg-DA in  $\text{D}_2\text{O}$ .



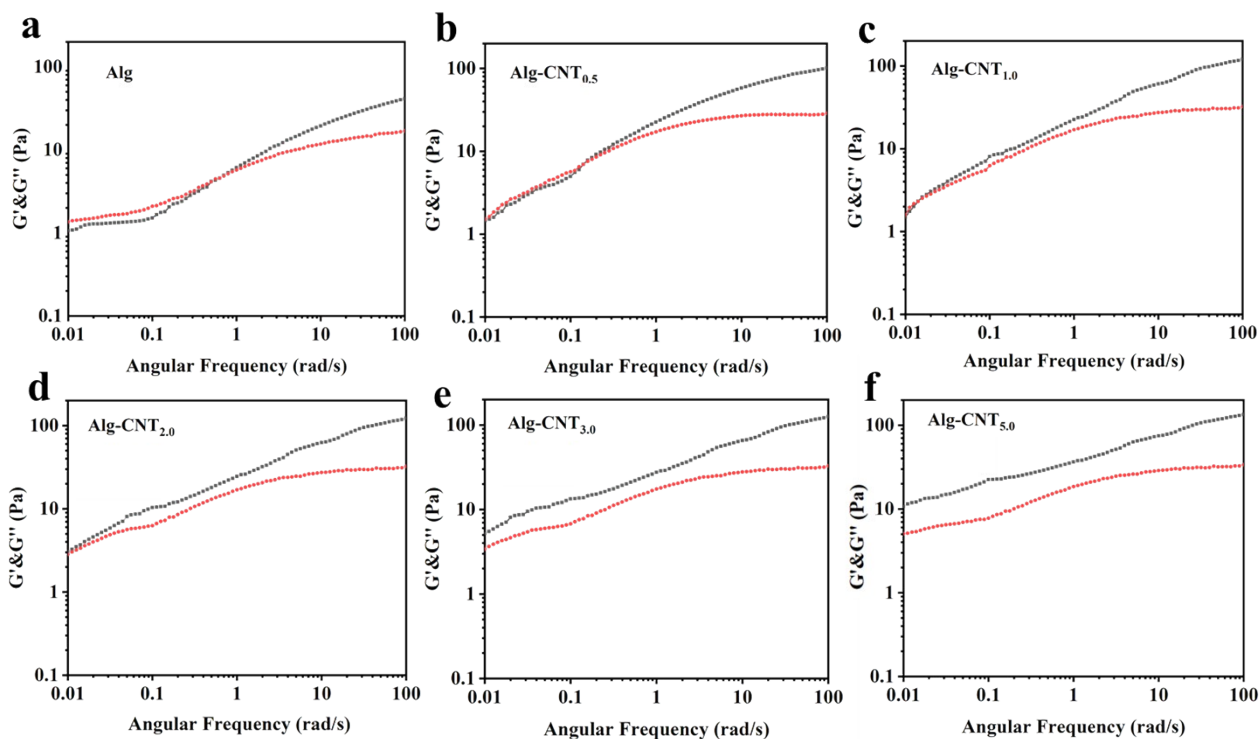
**Fig. S2.** (a) Photograph and (b) schematic of Alg-CNT hydrogel formation.



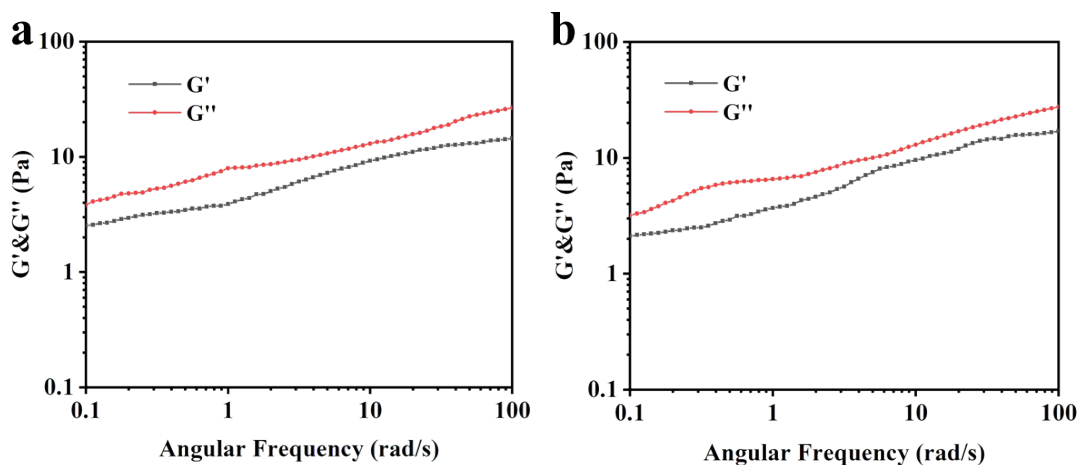
**Fig. S3.** (a) FT-IR spectra of Alg, Alg-CNT<sub>0.5</sub>, Alg-CNT<sub>2.0</sub>, and Alg-CNT<sub>5.0</sub> hydrogels, and (b) their corresponding magnified images in the wavenumber range of 1000–2000.

**Table S1.** The assignment of the characteristic vibrational bands of hydrogels.

| Alg  | Alg-CNT <sub>0.5</sub> | Alg-CNT <sub>2.0</sub> | Alg-CNT <sub>5.0</sub> | Assignment                     |
|------|------------------------|------------------------|------------------------|--------------------------------|
| 1033 | 1033                   | 1033                   | 1033                   | $\nu_s$ (C–O–C)                |
| 1413 | 1413                   | 1413                   | 1413                   | $\nu_s$ (COO <sup>-</sup> )    |
| 1490 | 1494                   | 1496                   | 1496                   | Amide II                       |
| 1612 | 1612                   | 1612                   | 1612                   | $\nu_{as}$ (COO <sup>-</sup> ) |
| 2921 | 2921                   | 2921                   | 2921                   | $\nu_s$ (C–H)                  |
| 3425 | 3425                   | 3425                   | 3425                   | $\nu_s$ (O–H)                  |



**Fig. S4.** Storage modulus  $G'$  (black dotted line) and loss modulus  $G''$  (red dotted line) of (a) Alg, (b) Alg-CNT<sub>0.5</sub>, (c) Alg-CNT<sub>1.0</sub>, (d) Alg-CNT<sub>2.0</sub>, (e) Alg-CNT<sub>3.0</sub>, and (f) Alg-CNT<sub>5.0</sub> hydrogels in oscillatory frequency sweep measurements.



**Fig. S5.** Storage modulus  $G'$  (black dotted line) and loss modulus  $G''$  (red dotted line) of polymer mixtures containing Alg-PBA and Alg-DA (a) in aqueous solution without adding NaOH and (b) in a buffer solution (pH = 7.4) in oscillatory frequency sweep measurements.

**Table S2.** The calculated self-healing efficiency of Alg-CNT<sub>2.0</sub> hydrogel.

| Sample          | Self-healing efficiency <sup>a</sup> [%] | Self-healing efficiency <sup>b</sup> [%] |
|-----------------|--|--|
| First healing   | 99.2                                     | 99.1                                     |
| Sencond healing | 96.5                                     | 98.5                                     |
| Third healing   | 95.4                                     | –  |

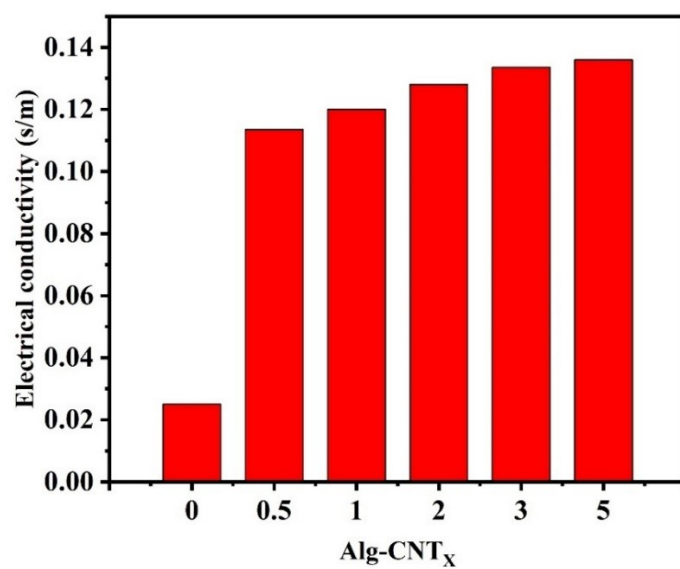
a: calculated from the tensile tests by the retained tensile strength; b: calculated from the rheological tests by the retained tensile strength.

**Table S3.** The summarized adhesion strengths of recently reported hydrogel sensors.

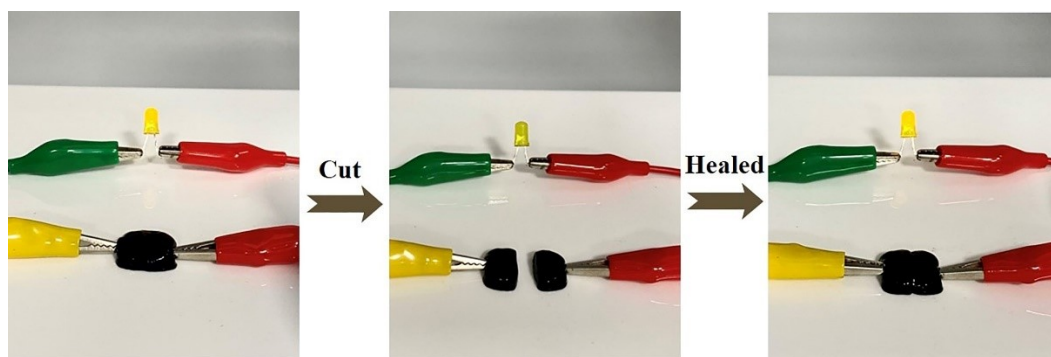
| Ref.      | Gels                | Substrates   | Adhesion strength (kPa) |
|-----------|---------------------|--------------|-------------------------|
| 4         | PAA-PANI            | glass        | 17.4                    |
| 5         | PDA-pGO-PAM         | porcine skin | 17.6                    |
| 6         | PC-CNF-GG           | porcine skin | 2.3                     |
| 9         | PVA-G-PDA-AgNPs     | porcine skin | 1.7                     |
| 23        | PVA/C-Chitosan      | porcine skin | 6.0                     |
| 24        | PDA@Ag NPs/CPHs     | porcine skin | 29                      |
| 26        | PAA-PEDOT:SL        | porcine skin | 4.6                     |
| 32        | DTPAM               | porcine skin | 15.2                    |
| 33        | PNIPAM/L/CNT        | porcine skin | 6.1                     |
| 35        | PVA-FSWCNT-PDA      | porcine skin | 5.2                     |
| 50        | MXene nanocomposite | porcine skin | 7.4                     |
| 51        | DTG                 | porcine skin | 9.8                     |
| This work | Alg-CNT             | porcine skin | 15.4                    |

**Table S4.** The caclulated absolute values of the resistances of Alg-CNT hydrogels.

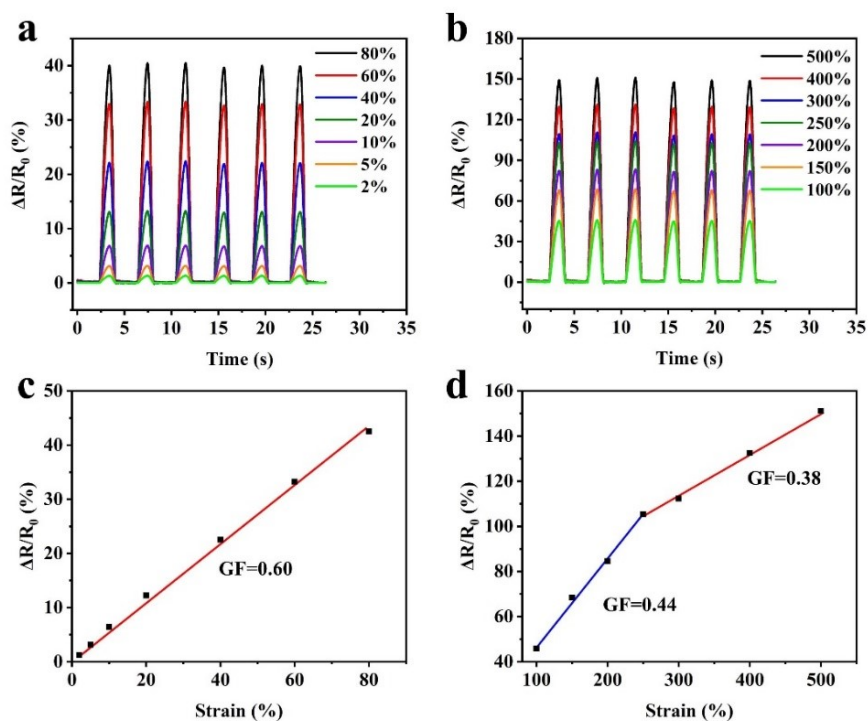
| Samples        | Alg   | Alg-CNT <sub>0.5</sub> | Alg-CNT <sub>1.0</sub> | Alg-CNT <sub>2.0</sub> | Alg-CNT <sub>3.0</sub> | Alg-CNT <sub>5.0</sub> |
|----------------|-------|------------------------|------------------------|------------------------|------------------------|------------------------|
| R ( $\Omega$ ) | 11396 | 8799                   | 8474                   | 7813                   | 6791                   | 5917                   |



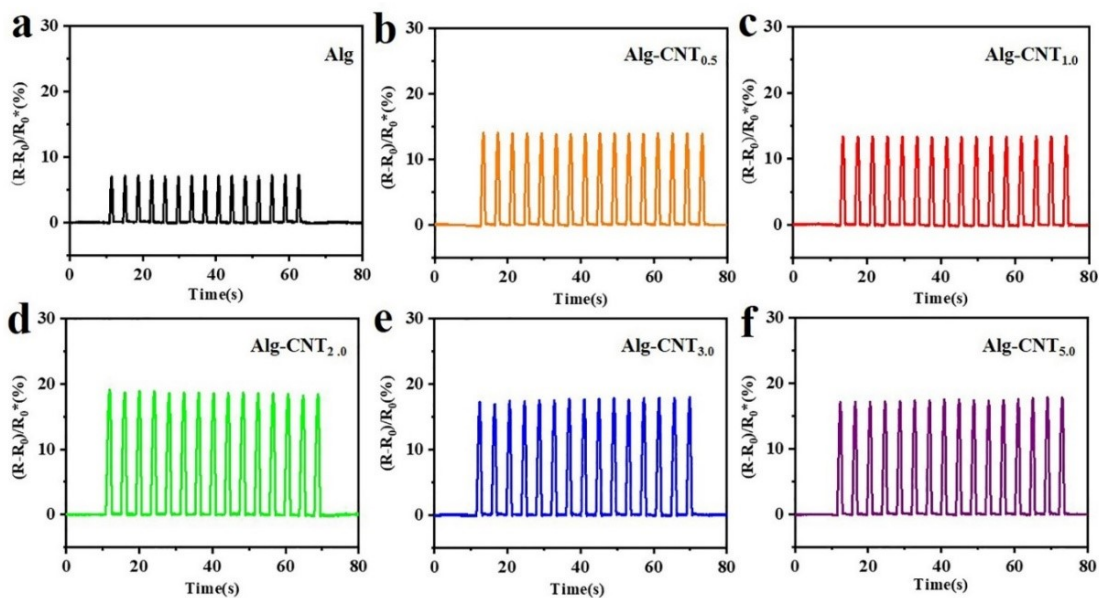
**Fig. S6.** The electrical conductivity of Alg-CNT<sub>x</sub> hydrogels doped with different contents of CNTs.



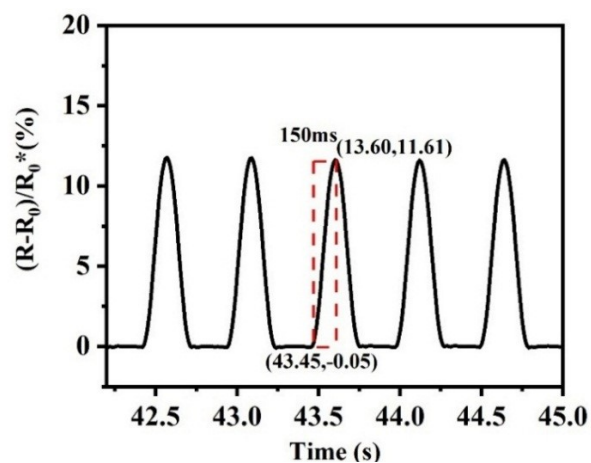
**Fig. S7.** Photographs showing the brightness change of the LED bulb in the circuit before and after the self-healing process of Alg-CNT<sub>2.0</sub> hydrogel.



**Fig. S8.** (a, b) Relative resistance changes of the Alg-CNT<sub>2.0</sub> hydrogels by stretching to different strains, and (c, d) their corresponding relative resistance change as a function of strain.



**Fig. S9.** In vitro monitoring of the relative resistance change of (a) Alg hydrogel, (b) Alg-CNT<sub>0.5</sub> hydrogel, (c) Alg-CNT<sub>1.0</sub> hydrogel, (d) Alg-CNT<sub>2.0</sub> hydrogel, (e) Alg-CNT<sub>3.0</sub> hydrogel, and (f) Alg-CNT<sub>5.0</sub> hydrogel adhered onto the surface of balloon under same expansion-contraction motion.



**Fig. S10.** The relative resistance change of Alg-CNT<sub>2.0</sub> hydrogel adhered onto the surface of balloon under expansion-contraction motion, in which the response time was marked in red frame.

**Table S5.** The summarized sensing performances of recently reported hydroge sensors.

| Ref.      | GF value  | Working range [%] | Cycles | Monitoring type         |
|-----------|-----------|-------------------|--------|-------------------------|
| 2         | 3.4       | 0-300             | 20     | Joint flexion/speaking  |
| 4         | 18.28     | 0-269             | 1000   | Touch keyboard/speaking |
| 7         | 1.25      | 0-1000            | 300    | Pressure/Joint flexion  |
| 9         | 0.13      | 0-331             | 50     | Joint flexion/speaking  |
| 12        | 1.51      | 0-1000            | 1000   | Human motion            |
| 17        | 3.44      | 0-363             | 50     | Pressure/human motion   |
| 18        | 3.15      | 0-300             | 500    | Human motion/Breathe    |
| 19        | 1.5-3.39  | 0-250             | 300    | Human motion            |
| 20        | 0.8-1.6   | 0-50              | 2000   | Pressure/human motion   |
| 25        | 0.26-0.58 | 0-550             | 2000   | Human motion            |
| 37        | 2.71      | 0-70              | 100    | Human motion            |
| 40        | 6.0       | 0-700             | 100    | Joint flexion/speaking  |
| 41        | 1.41-1.48 | 0-2000            | 300    | Human motion            |
| 43        | 0.4-2.9   | 10-4000           | 350    | Joint flexion           |
| 44        | 2.5-6.6   | 0-400             | 1000   | Human motion            |
| 53        | 0-3.12    | 0-95              | 360    | Pressure/human motion   |
| This work | 0.38-0.60 | 0-500             | 30000  | Breathe/Heartbeat       |



