Supporting Information

Self-assembled Cationic Organic Nanosheet: Role of Positional Isomers in Guanidium-Core for Efficient Lithium-Ion Conduction

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1. Materials and Characterization:

Materials: The chemicals like 4-formylbenzoic acid, 3-formylbenzoic acid, Guanidium hydrochloride, Hydrazine hydrate used for synthesis were purchased from commercial suppliers and used directly without further purification. Solvents such as isopropanol, ethanol, DMF were purchased from Spectrochem Chemical company. A Barnstead System, U.S., was employed to get high purity nanopure water for making all the aqueous solutions.

Characterization: ¹H NMR and ¹³C NMR were recorded using Geol resonance ECZ600R spectrometer at 25 °C. TMS was used as an internal reference during NMR spectroscopic study. Parkin Elmer 883 spectrometer was used to record the FT-IR data using the KBr pellet. UV-Vis absorption spectra were recorded using Shimadzu corp 80109 UV- Vis spectrophotometer. Using a Zetasizer Nano-ZS90 (Malvern) instrument with a 632.8 nm He-Ne laser DLS, experiments were carried out at 298 K. Transmission electron microscopy images were collected using a JEOL JEM-2100 electron microscope working at 200 kV accelerating voltage. The samples were prepared on the surface of lacey-carbon-supported copper TEM grids. The AFM data were collected with the help of tapping mode with an NT-MDT Ntegra Aura atomic force microscope. JEOL JSM-7100F instrument working at 18 kV accelerating voltage was used to record the FE-SEM data. Before taking the FE-SEM images, the thin coating of Au (~ 4 nm) was coated using a vacuum evaporator. Using a Q-of-micro quadrupole mass spectrophotometer (Micromass), ESI-MS was done. Powder X-ray diffraction (PXRD) patterns were recorded at room temperature on a Philips X’pert X-ray powder diffractometer using Cu-Kα radiation (λ=1.5418 A) in the 2θ range of 5–50º. X-ray photoelectron spectra (XPS) were obtained from a Thermo Fischer Scientific ESCALAB XI using an Al Kα (hv = 1486.6 eV) X-ray source with a base vacuum-operated at 300 W. Thermo-gravimetric analysis (TGA) was conducted using an Auto TGA 2950 apparatus under a nitrogen flow of 100 mL min⁻¹ while heating from room temperature. The conductivity study has been done using the electrochemical work station (VSP Biologic, France).

2. Synthesis of AM-2 and AM-3:

![Synthesis route of AM-2 and AM-3](image)

**Figure S1:** Synthesis route of AM-2 and AM-3.
Synthesis of L: Triaminoguanidinium chloride was prepared by the literature method.\textsuperscript{[1]}

Synthesis of AM-2: In a (1:1 v/v) ethanol/water solvent mixture (40 mL) triaminoguanidinium chloride (0.25 gm, 1.75 mmol) and 3-formylbenzoic acid (0.75 gm, 5.30 mmol) were dissolve. The reaction mixture was stirred for 12h at 80 °C. Then the reaction mixture was cooled down to ambient temperature. After some time, a large amount of white precipitate was obtained. The white precipitate was filtered using a G4 gooch crucible under a vacuum. The desired product was washed several times with ethanol and dried under reduced pressure. Yield 0.75 gm (82%).

\textsuperscript{1}H NMR (200 MHz, DMSO-\textit{d}_6, \delta ppm): 8.61 (3H, s), 8.37 (3H, s), 8.21 (3H, d, \textit{J} = 8.0), 8.02 (3H, d, \textit{J} = 8.0), 7.62 (3H, t, \textit{J} = 8.0). \textsuperscript{13}C NMR (125 MHz DMSO-\textit{d}_6, \delta ppm): 168.0, 152.0, 147.4, 136.0, 132.3, 132.2, 131.2, 129.9, 128.9. HRMS: \textit{m/z} calculated for [M] (C\textsubscript{25}H\textsubscript{21}N\textsubscript{6}O\textsubscript{6}): 501.4785; found [M + H\textsuperscript{+}]: 501.1610.

Synthesis of AM-3: In a (1:1 v/v) ethanol/water solvent mixture (40 mL), triaminoguanidinium chloride (0.50 gm, 3.50 mmol) and 4-formylbenzoic acid (1.50 gm, 10.60 mmol) were dissolve. The reaction mixture was stirred for four h at 90 °C. Then the reaction mixture cooled down to ambient temperature. After some time, a large amount of yellowish-white precipitate was obtained. The precipitate was filtered using a G4 gooch crucible under a vacuum. The desired product was washed several times with ethanol and dried under reduced pressure. Yield 1.5 gm (82%).

\textsuperscript{1}H NMR (600 MHz, DMSO-\textit{d}_6): \delta ppm 8.75 (3H, s), 8.10-8.06 (12H, m). \textsuperscript{13}C NMR (125 MHz, DMSO-\textit{d}_6): 166.9, 150.6, 147.6, 138.0, 131.7, 129.5, 127.5. HRMS: \textit{m/z} calculated for [M]\textsuperscript{+} (C\textsubscript{25}H\textsubscript{21}N\textsubscript{6}O\textsubscript{6})\textsuperscript{+}: 501.4785; found: 501.1594.

3. \textsuperscript{1}H NMR, \textsuperscript{13}C NMR and HRMS spectra of AM-2:

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{FigureS2}
\caption{\textsuperscript{1}H NMR spectrum of AM-2 recorded in DMSO-\textit{d}_6.}
\end{figure}
Figure S3: $^{13}$C NMR (125 MHz) spectrum of AM-2 recorded in DMSO-$d_6$.

Figure S4: HRMS spectrum of AM-2 recorded in Methanol.
4. $^1$H NMR, $^{13}$C NMR and HRMS spectra of AM-3:

Figure S5: $^1$H NMR spectrum of AM-3 recorded in DMSO-$d_6$.

Figure S6: $^{13}$C NMR spectrum of AM-3 recorded in DMSO-$d_6$.

Figure S7: HRMS spectrum of AM-3 recorded in Methanol.
5. Methods of sample preparation for FE-SEM, TEM, AFM:

Sample preparation for Nanosheets morphology: To get the exfoliated nano sheet, water was used as a solvent. 1 mg/mL (total volume 5ml) water suspension of the compound AM-2 and AM-3 was initially sonicated for 10 minutes. From that solution, 20 μL was drop cast on Si (100) wafer, and we observe the nanoplate-like morphology. Then, 0.6 mL of this suspension was exposed to 2.4 mL of water. The dispersion was sonicated for 30 minutes. From that aliquot, 20 μL was drop cast on a TEM grid and Si (100) wafer, and we observe the nanosheet morphology. After drop-casting, all the samples were dried under a desiccator. Before taking the FE-SEM images, the ultrathin gold layer was coated on all the samples.

Sample preparation for Nanoflower morphology: First 2 mg of AM-2 and Am-3 dissolve in 2 ml of DMF. 1 ml of this DMF solution exposed to 1 ml of water to maintain a solvent ratio of 1:1 DMF/water (v/v). The mixture was shaken well for 2-3 minutes and left undisturbed for 10 minutes. After 10 minutes, 0.6 ml of this solution was exposed to 2.4 ml of (1:1, v/v) DMF/water solvent mixture. From that solution, 20 μL was drop cast on Si (100) wafer. After drop-casting, all the samples were dried under a desiccator. Before taking the FE-SEM images, the ultrathin gold layer was coated on all the samples.

6. Determination of the degree of aggregation ($\alpha_{agg}$):

The degree of aggregation ($\alpha_{agg}$) was calculated from the temperature-dependent UV-Vis spectral data by using equation (1) as mentioned below.

$$\alpha(T) = \frac{\varepsilon(T) - \varepsilon_M}{\varepsilon_A - \varepsilon_M} \tag{1}$$

Where $\varepsilon(T)$ is the measured extinction coefficient at temperature $T$; $\varepsilon_M$ and $\varepsilon_A$ are the extinction coefficients of the monomer and fully aggregated state, respectively. The latter two values were determined from the spectral data at high and low temperatures, respectively.

The experimentally determined degree of aggregation was fitted temperature-dependent isodesmic self-assembly model accordingly with equation (2) as mentioned below, to obtain the enthalpy value $\Delta H$.

$$\alpha(T) = \frac{1}{1 + \exp[-0.908 \Delta H \frac{T - T_m}{RT_m}]} \tag{2}$$

Where $\alpha$ is the degree of aggregation, and $T_m$ is the melting temperature defined as the temperature at $\alpha = 0.5$ at $T = T_m$. $R$ is the Boltzmann constant.

From the experimentally determined degree of aggregation, the number-averaged degree of polymerization, $DP_N$, calculated temperature-dependent isodesmic self-assembly model accordingly with equation (3), as mentioned below.
\[ DP_N(T) = \frac{1}{\sqrt{1 - \alpha(T)}} \]  

(3)

From the number-averaged degree of polymerization, \( DP_N \), and the known concentration of molecules, \( c_T \), the equilibrium constant \( K_e \) was determined as a function of temperature using equation (4), as mentioned below.

\[ DP_N(T) = \frac{1}{2} + \frac{1}{2} \sqrt{4K_e(T)c_T + 1} \]  

(4)

7. Temperature-dependent UV-Vis data for AM-2:

**Figure S8**: (a) Variable temperature UV-Vis spectra of AM-2 (3.7 \( \times \) 10\(^{-5} \) M) in DMF/water. (b) Corresponding plot of change of absorbance at 360 nm with temperature. (c) Temperature-dependent degree of aggregation, calculated from UV-Vis spectral change at 316 nm and corresponding isodesmic fit. (d) Corresponding plot of the degree of polymerization, \( DP_N \), as a function of temperature. (e) Corresponding plot of the equilibrium constant, \( K_e \), as a function of temperature. (f) Corresponding Van’t Hoff plot for AM-2 in DMF/water.
8. Crystal structure of AM-3 with interplanar distance:

![Figure S9](image1)

**Figure S9:** (a) Single-crystal XRD structure of AM-3 with interplanar distance. (b) Crystal structure of AM-3 showing the extended length of the molecule.

9. PXRD pattern of bulk-powder of AM-2 and AM-3:

![Figure S10](image2)

**Figure S10:** PXRD pattern of bulk-powder of (a) AM-2 and (b) AM-3.

10. TGA profile of the nanoflower morphology of AM-2 and AM-3

![Figure S11](image3)

**Figure S11:** Thermogravimetric analysis (TGA) curves of nanoflower morphology of (a) AM-2, and (b) AM-3, respectively.
11. Sample preparation for Li-ion conductivity

(A) Methods of sample preparation for SONs morphology:
To get the exfoliated nano sheet, water was used as a solvent. 1 mg/mL (total volume 5ml) of the compound AM-2 and AM-3 dispersed in water containing 0.2, 0.5, 0.7, 1.0 and 2.0 equivalents of LiClO₄. The dispersion was sonicated for 30 minutes. An aliquot of this homogeneous dispersed solution of the nanostructures drop-casted on the surface of a Whatman filter membrane separator and remove the residue solvent. The prepared samples were dried at room temperature for 60 minutes and sandwiched in between the SS electrode. Then we proceed with measuring the conductivity.

(b) Methods of sample preparation for bulk-powder:
1 mg/mL (total volume 5ml) of the compound AM-2 and AM-3 dispersed in water containing 0.2, 0.5, 0.7, 1.0 and 2.0 equivalents of LiClO₄. An aliquot of this homogeneous dispersed solution of the nanostructures drop-casted on the surface of a Whatman filter membrane separator and remove the residue solvent. The prepared samples were dried at room temperature for 60 minutes and sandwiched in between the SS electrode. Then we proceed with measuring the conductivity.

(c) Methods of sample preparation for nanoflower morphology:
First, 2 mg of AM-2 and Am-3 dissolve in 2 ml of DMF. 1 ml of this DMF solution exposed to 1 ml of water containing (0.2, 0.5, 0.7, 1.0, and 2.0 equivalents of LiClO₄) to maintain a solvent ratio of 1:1 DMF/water (v/v). The mixture was shaken well for 2-3 minutes and left undisturbed for 10 minutes. No sonication was done here for mixing. An aliquot of this dispersed solution of the nanostructures drop-casted on the surface of a Whatman filter membrane separator and remove the residue solvent. The prepared samples were dried at room temperature for the duration of 60 minutes and sandwiched in between the SS electrode. Then we proceed with measuring the conductivity.

12. SEM and TEM images in the presence of LiClO₄:

Figure S12: (a), (b) FE-SEM and (c), (d) TEM images of SONs of AM-2 in the presence of 2.0 equivalents of LiClO₄. (e), (f) FE-SEM and (g), (h) TEM images of SONs of AM-3 in the presence of 2.0 equivalents of LiClO₄.
13. Details of conductivity measurement:

The ionic conductivity measurements were carried out using stainless steel (SS) electrodes, unlike the comb-shaped Au electrodes, as used by Wu et al. to prepare their samples for ionic conductivity measurements.\(^{[2]}\) The use of SS electrodes is a more reliable and fast method; moreover, the drying duration is just 20-30 min which is significantly less comparing to the Au electrodes, which needs a drying time of ~ 48 hours. We have used the Whatman filter membrane separator, which had more permeability, and the thickness was 0.026 cm, which is calculated using the screw cage method. The radius of the SS electrode surface was 7 mm, and the calculated area was 1.5386 cm\(^2\). Once the sample solution is ready, we drop-casted four drops over the separator, which became wet, and kept it for drying at room temperature for 50 to 60 min. After drying, the impedance study was carried out in order to know the resistance values for different samples, and subsequently, we calculate the related conductivity values using the standard equation:

\[
\sigma = \frac{d}{RA}
\]

Where d = Thickness (cm), R = Resistance (ohm) and A = Area (cm\(^2\)).

14. Admittance plots:

![Admittance plots](image)

**Figure S13.** Admittance plots of AM-2 dopped with varying equivalents of LiClO\(_4\) for (a) SONs, (b) bulk-powde and (c) nanoflower. Admittance plots of AM-3 dopped with varying equivalents of LiClO\(_4\) for (d) SONs, (e) bulk-powde and (f) nanoflower.
15. Conductivity data:

**Table S1**: Resistance and lithium ionic conductivity values of AM-2 in SONs morphology with varying LiClO$_4$ concentration at room temperature.

| Equivalents of LiClO$_4$ | Area A (cm$^2$) | Thickness d (cm) | Resistance R (ohm) | Conductivity (S cm$^{-1}$) |
|-------------------------|----------------|------------------|--------------------|---------------------------|
| 0.2                     | 1.5386         | 0.026            | 135.7              | 1.245*10$^{-4}$           |
| 0.5                     | 1.5386         | 0.026            | 126.4              | 1.336*10$^{-4}$           |
| 0.7                     | 1.5386         | 0.026            | 93.1               | 1.815*10$^{-4}$           |
| 1.0                     | 1.5386         | 0.026            | 85.8               | 1.969*10$^{-4}$           |
| 2.0                     | 1.5386         | 0.026            | 49.42              | 3.419*10$^{-4}$           |

**Table S2**: Resistance and lithium ionic conductivity values of AM-3 in SONs morphology varying LiClO$_4$ concentration at room temperature.

| Equivalents of LiClO$_4$ | Area A (cm$^2$) | Thickness d (cm) | Resistance R (ohm) | Conductivity (S cm$^{-1}$) |
|-------------------------|----------------|------------------|--------------------|---------------------------|
| 0.2                     | 1.5386         | 0.026            | 152.6              | 1.107*10$^{-4}$           |
| 0.5                     | 1.5386         | 0.026            | 112.7              | 1.499*10$^{-4}$           |
| 0.7                     | 1.5386         | 0.026            | 107.6              | 1.570*10$^{-4}$           |
| 1.0                     | 1.5386         | 0.026            | 94.2               | 1.793*10$^{-4}$           |
| 2.0                     | 1.5386         | 0.026            | 78.12              | 2.163*10$^{-4}$           |

**Table S3**: Resistance and lithium ionic conductivity values of AM-2 in bulk-powder morphology with varying LiClO$_4$ concentration at room temperature.

| Equivalents of LiClO$_4$ | Area A (cm$^2$) | Thickness d (cm) | Resistance R (ohm) | Conductivity (S cm$^{-1}$) |
|-------------------------|----------------|------------------|--------------------|---------------------------|
| 0.2                     | 1.5386         | 0.026            | 170.15             | 9.931*10$^{-5}$           |
| 0.5                     | 1.5386         | 0.026            | 159.31             | 1.060*10$^{-4}$           |
| 0.7                     | 1.5386         | 0.026            | 153.72             | 1.099*10$^{-4}$           |
| 1.0                     | 1.5386         | 0.026            | 113.5              | 1.488*10$^{-4}$           |
| 2.0                     | 1.5386         | 0.026            | 86.3               | 1.958*10$^{-4}$           |
### Table S4: Resistance and lithium ionic conductivity values of AM-3 in bulk-powder morphology with varying LiClO₄ concentration at room temperature.

| Equivalents of LiClO₄ | Area A (cm²) | Thickness d (cm) | Resistance R (ohm) | Conductivity (S cm⁻¹) |
|----------------------|-------------|-----------------|-------------------|----------------------|
| 0.2                  | 1.5386      | 0.026           | 178.52            | 9.465*10⁻⁵           |
| 0.5                  | 1.5386      | 0.026           | 172.9             | 9.773*10⁻⁵           |
| 0.7                  | 1.5386      | 0.026           | 165.6             | 1.020*10⁻⁴           |
| 1.0                  | 1.5386      | 0.026           | 140.32            | 1.204*10⁻⁴           |
| 2.0                  | 1.5386      | 0.026           | 111.8             | 1.511*10⁻⁴           |

### Table S5: Resistance and lithium ionic conductivity values of AM-2 in nanoflower morphology with varying LiClO₄ concentration at room temperature.

| Equivalents of LiClO₄ | Area A (cm²) | Thickness d (cm) | Resistance R (ohm) | Conductivity (S cm⁻¹) |
|----------------------|-------------|-----------------|-------------------|----------------------|
| 0.2                  | 1.5386      | 0.026           | 417.63            | 4.046*10⁻⁵           |
| 0.5                  | 1.5386      | 0.026           | 388.91            | 4.345*10⁻⁵           |
| 0.7                  | 1.5386      | 0.026           | 367.27            | 4.601*10⁻⁵           |
| 1.0                  | 1.5386      | 0.026           | 342.38            | 4.942*10⁻⁵           |
| 2.0                  | 1.5386      | 0.026           | 299.83            | 5.631*10⁻⁵           |

### Table S6: Resistance and lithium ionic conductivity values of AM-3 in nanoflower morphology with varying LiClO₄ concentration at room temperature.

| Equivalents of LiClO₄ | Area A (cm²) | Thickness d (cm) | Resistance R (ohm) | Conductivity (S cm⁻¹) |
|----------------------|-------------|-----------------|-------------------|----------------------|
| 0.2                  | 1.5386      | 0.026           | 409.43            | 4.127*10⁻⁵           |
| 0.5                  | 1.5386      | 0.026           | 380.45            | 4.441*10⁻⁵           |
| 0.7                  | 1.5386      | 0.026           | 360.63            | 4.686*10⁻⁵           |
| 1.0                  | 1.5386      | 0.026           | 330.71            | 5.109*10⁻⁵           |
| 2.0                  | 1.5386      | 0.026           | 317.92            | 5.315*10⁻⁵           |
16. Comparable table of Conductivity Data:

| Materials                      | Conductivity (S/cm⁻¹) | Description                                                             | Ref.                                      |
|--------------------------------|-----------------------|------------------------------------------------------------------------|-------------------------------------------|
| SONs (This work)               | 3.42 × 10⁻⁴ at 298 K  | Self-assembled cationic organic nanosheets                             | (This work)                              |
| Zwitterionic Nano sheets        | 5.1 × 10⁻⁵ at 298 K   | Self-assembled zwitterionic organic nanosheets                         | ACS App. Mater. & Inter., 2020, 12, 58122-58131. |
| Supramolecular polymer          | 1.2 × 10⁻⁴ S cm⁻¹ at 298 K | Supramolecular polymer electrolytes.                                  | Nat. Commun. 2019, 10, 5384.             |
| Self-assembled Nano sheets      | 2.4 × 10⁻⁷ at 298 K   | Self-assembled nanosheets from positively charged polycyclic aromatic hydrocarbons | Angew. Chem. 2011, 123, 2843–2846        |
| MOF                            | 5.5 × 10⁻⁵ at 300 K   | Mg₂(dobdc)-0.35LiOiPr-0.25LiBF₄·EC·DEC (EC = ethylene carbonate; DEC = diethyl carbonate). MOF + liquid organic molecule (on the film) | J. Am. Chem. Soc. 2011, 133, 14522.     |
| MOF                            | 1.8×10⁻⁵ at 293 K     | LiOtBu-grafted UiO-66 (MOF+Post-surface Modification)                  | Chem. Eur. J. 2013, 19, 5533.            |
| MOF                            | 10⁻⁶ at 295K          | MOF (10wt%)+LiTFSI+PEO (MOF-loaded polymer Electrolytes)               | J. Mater. Chem. A 2014, 2, 9948.        |
| COF                            | 2.7 × 10⁻² S cm⁻¹ at 298 K | Lithium sulfonated COF                                           | J. Am. Chem. Soc. 2019, 141, 5880–5885  |
| COF                            | 3.05 × 10⁻³ at 298 K  | Ionic COF with spiroborate Linkage                                   | Angew. Chem. Int. Ed. 2016, 55, 1737–1741 |
| COF                            | 7.2 × 10⁻³ at 298 K   | Lithium Imidazolate COF                                             | J. Am. Chem. Soc. 2019, 141, 7518–7525  |
| COF                            | 5.74 × 10⁻³ at 300 K  | Guanidium based cationic COF                                         | J. Am. Chem. Soc. 2018, 140, 896–899    |

17. Calculation of Li-ion transference number ($t_{Li^+}$):

For the calculation of Li-ion transference number, we have used Bruce-Vincent method [3,4], where a symmetric cell Li/AMLi/Li used and the DC current allowed following through the cell. The steady state current obtained through potentiostatic polarization study and the related spectral curve was given as figure S14a (Li/AM-2Li/Li) and figure S14b (Li/AM-3Li/Li). The impedance study of the cell for before and after potentiostatic polarization study carried out. The measured impedance curves are given as figure S15a and figure S15b corresponding to
Li/AM-2Li/Li and Li/AM-3Li/Li respectively. The required values needed for the calculation of transference number ($t_{Li^+}$) were tabulated in Table S8. The Li-ion transference number calculated by the following equation as:

$$t_{Li^+} = \frac{I_{SS}(\Delta V - I_0R_0)}{I_0(\Delta V - I_{SS}R_{SS})}$$

Where, $I_{SS}$ is the steady state current, $\Delta V$ is the applied potential, $I_0$ is the initial current, $R_0$ and $R_{SS}$ are the interfacial resistance before and after potentiosatatic polarization study, respectively.

Table S8. The observed values for $t_{Li^+}$ calculation measured through potentiostatic polarization method.

| S.No | Material       | $I_{SS}$ (µA) | $\Delta V$ (mV) | $I_0$ (µA) | $R_0$ (Ω) | $R_{SS}$ (Ω) | $t_{Li^+}$ |
|------|----------------|---------------|-----------------|------------|-----------|--------------|------------|
| 1    | SONs of AM-2   | 10            | 10              | 12         | 462.3     | 927.5        | 0.49       |
| 2    | SONs of AM-3   | 9             | 10              | 25         | 221.2     | 1461.8       | 0.15       |

Figure S14: (a) The potentiostatic polarization curves of SONs of AM-2. (b) The potentiostatic polarization curves of SONs of AM-3.

Figure S15: (a) The interfacial resistance curves of SONs of AM-2 before and after potentiostatic polarization study respectively. (b) The interfacial resistance curves of SONs of AM-3 before and after potentiostatic polarization study respectively.

18. Electrochemical stability of SONs of AM-2 and AM-3:

To investigate the electrochemical stability, we have performed the linear sweep voltammetry (LSV) measurement for SONs of AM-2 and AM-3. Linear sweep voltammetry (LSV) was carried out with Ti/AMLi/Li asymmetric cell.
The sweep rate was fixed as 0.1 mV s\(^{-1}\) with a fixed voltage window (-0.5 V to 6 V vs. Li/Li\(^+\)) at room temperature. The electrochemical stability window of ca. 6 V were observed for both SONs of AM-2 and AM-3 (Figure S16).

![Figure S16](image)

**Figure S16:** (a) The Linear sweep voltammetry curves of SONs of AM-2. (b) The Linear sweep voltammetry curves of SONs of AM-3.

19. **Cantilever details for AFM measurements:**

| Cantilever Length L ± 10 μm | Cantilever width W ± 5 μm | Cantilever Thickness T ± 0.5 μm | Resonance frequency kHz | Force constant, Nm\(^{-1}\) |
|-----------------------------|---------------------------|-------------------------------|--------------------------|-----------------------------|
| 125                         | 30                        | 1.5 – 2.5                     | 87 - 230                 | 1.45 – 15.1                 |

20. **Reference:**

[1] A. Maity, M. Gangopadhyay, A. Basu, S. Aute, S. S. Babu and A. Das, *J. Am. Chem. Soc.*, 2016, **138**, 11113–11116.

[2] D. Wu, R. Liu, W. Pisula, X. Feng and K. Müllen, *Angew. Chem. Int. Ed.*, 2011, **50**, 2791–2794.

[3] J. Evans, C. A. Vincent and P. G. Bruce, *Polymer*, 1987, **28**, 2324.

[4] M. Siekierski, M. Bukat, M. Ciosek, M. Piszcz and M. Szerszen, *Polymers*, 2021, **13**, 895.