Electronic Supplementary Information (ESI) for the manuscript titled **"Heteroatom functionalized activated porous biocarbons and their excellent performance for $CO_2$ capture at high pressure"**

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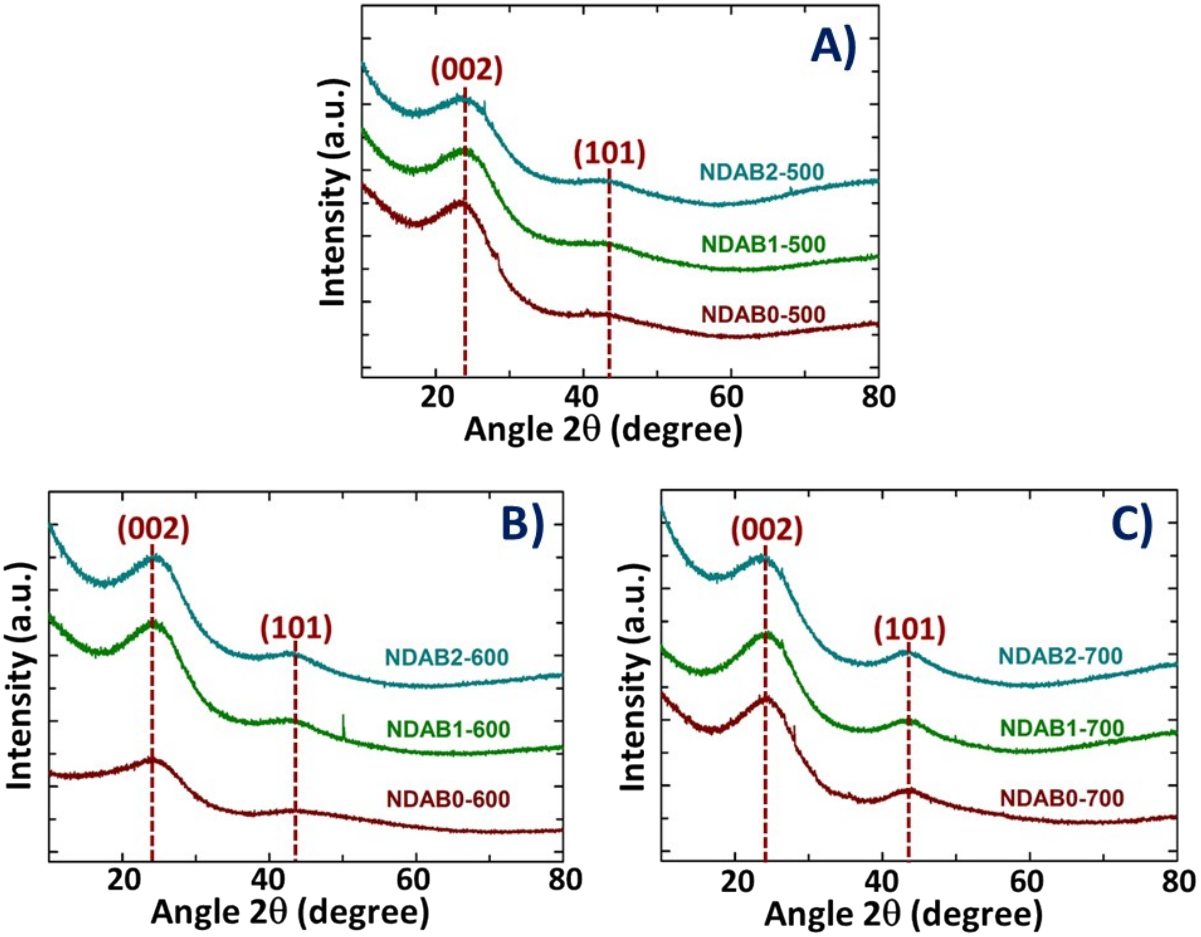


**Fig. S1.** XRD patterns of **NDABn-T** (n=0-2, T= A) 500, B) 600 and C) 700 °C).

The XRD patterns of **NDABn-T** (n=2, T=500-700 °C) show the presence of two broad peaks centered on 2θ=23° and 43°. This indicates that the samples are graphitic in nature. At any given temperature, the intensity of both the peaks increase slightly with increasing ratio of $ZnCl_2$ to *Arundo donax*/chitosan, which signifies the development of higher graphitization in the carbon structure.

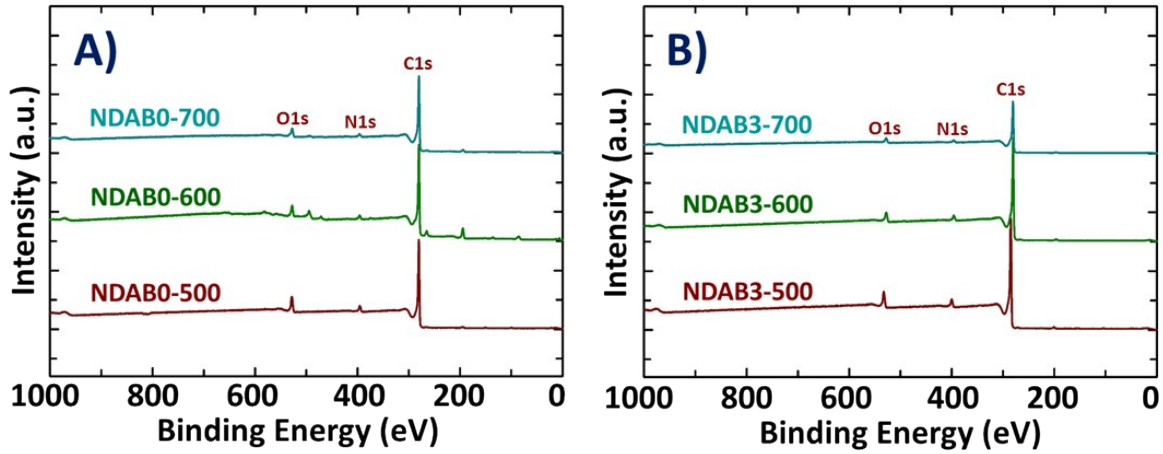


**Fig. S2.** XPS survey spectra of A) **NDAB0-T** (T = 500-700 °C) and B) **NDAB3-T** (T = 500-700 °C).

The XPS survey spectra show three main peaks corresponding to the elements carbon, nitrogen and oxygen. The XPS results prove that the presence of chitosan during the carbonization can effectively incorporate the nitrogen in the carbon structure.

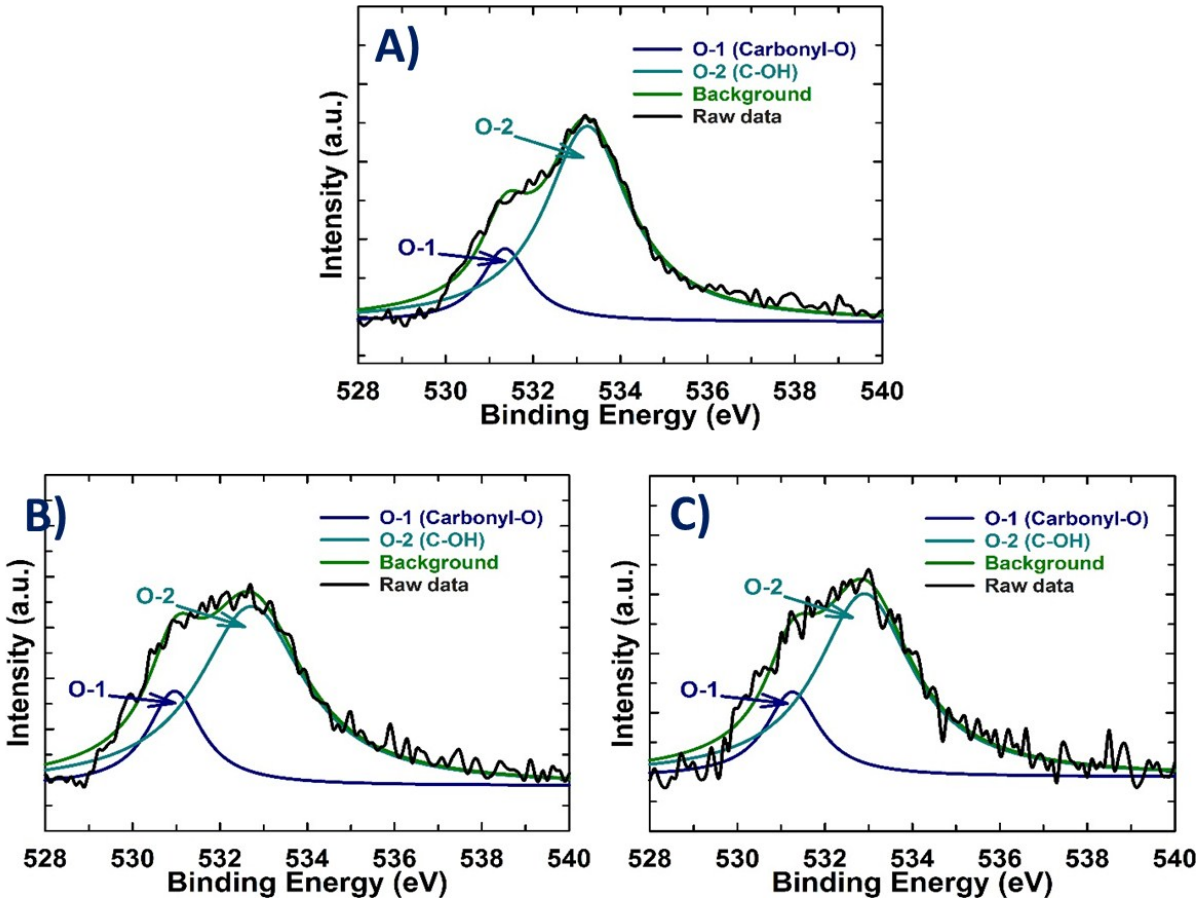


**Fig. S3.** High resolution XPS O1s spectra of **NDAB3-T** (T = A) 500, B) 600 and C) 700 °C).

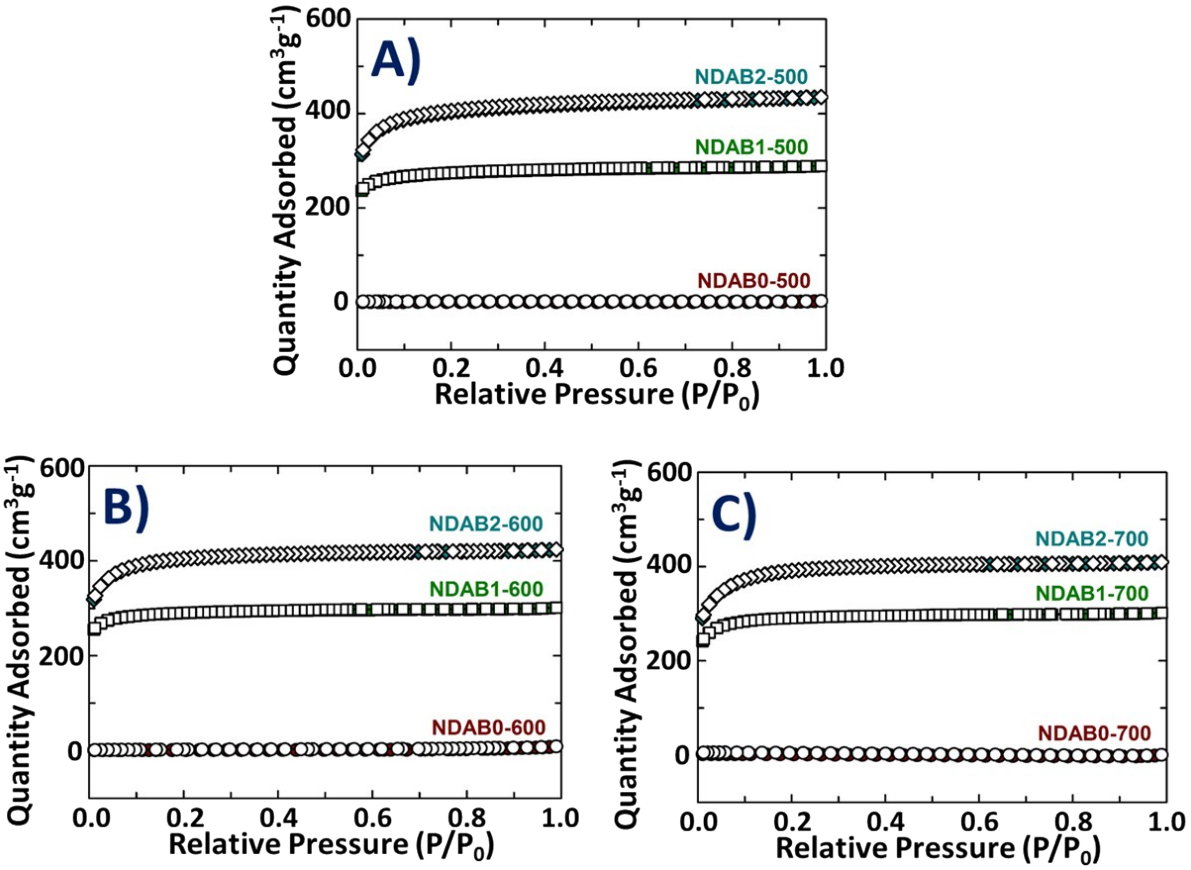


**Fig. S4.** $N_2$ adsorption-desorption isotherms of **NDABn-T** (n=0-2, T = A) 500, B) 600 and C) 700 °C).

The isotherm curves correspond to the shape which belongs to type I as per the IUPAC classification of adsorption isotherms. In other words, the porous structure of these activated biocarbons is dominated by micropores followed by a lesser amount of meso and macropores in the structure. It can also be seen that when the impregnation amount of the activating agent is increased, there is a corresponding increasing the amount of $N_2$ being adsorbed. This happens due to the fact that higher porosity is generated by the chemical reaction with activating agent.

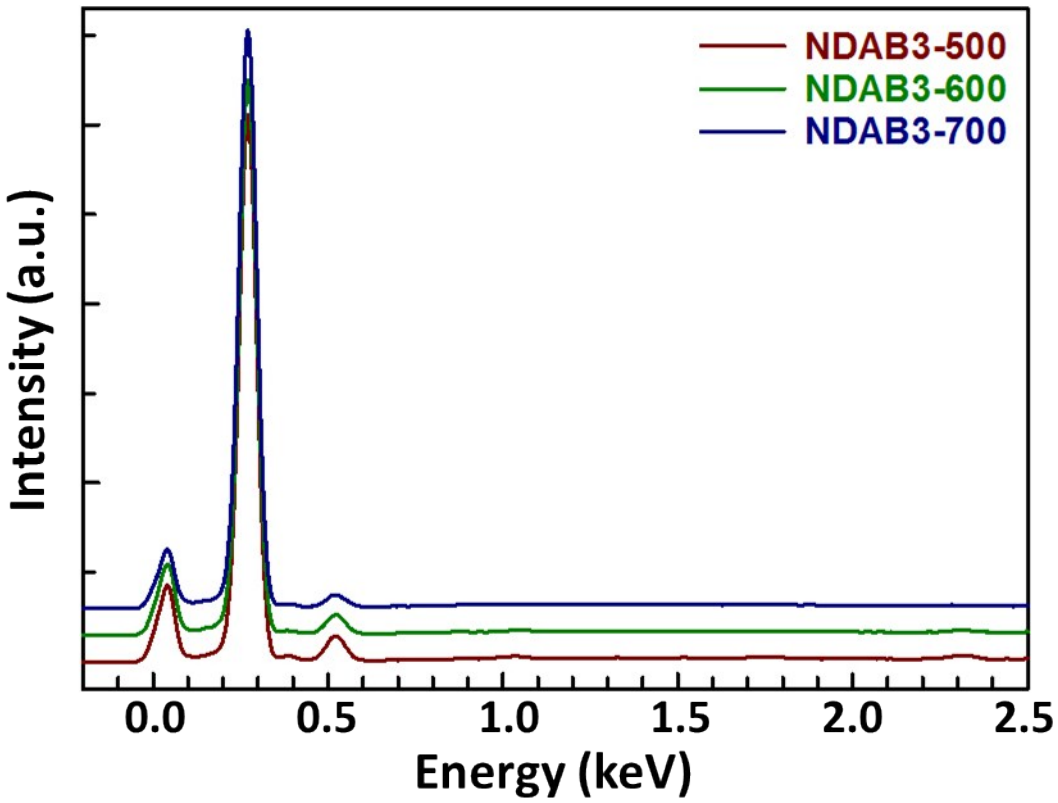


**Fig. S5.** EDS analysis of **NDAB3-T** (T = 500-700 °C).

The result of EDS analysis further confirms the heteroatom functionalization of the porous activated biocarbons. The sample NDAB3-500 shows the highest amount of nitrogen content which is in consistency with the CNS and XPS analysis elemental composition. From these results, it can be concluded that the one step strategy of synthesizing heteroatom doped porous activated biocarbons has been successful.

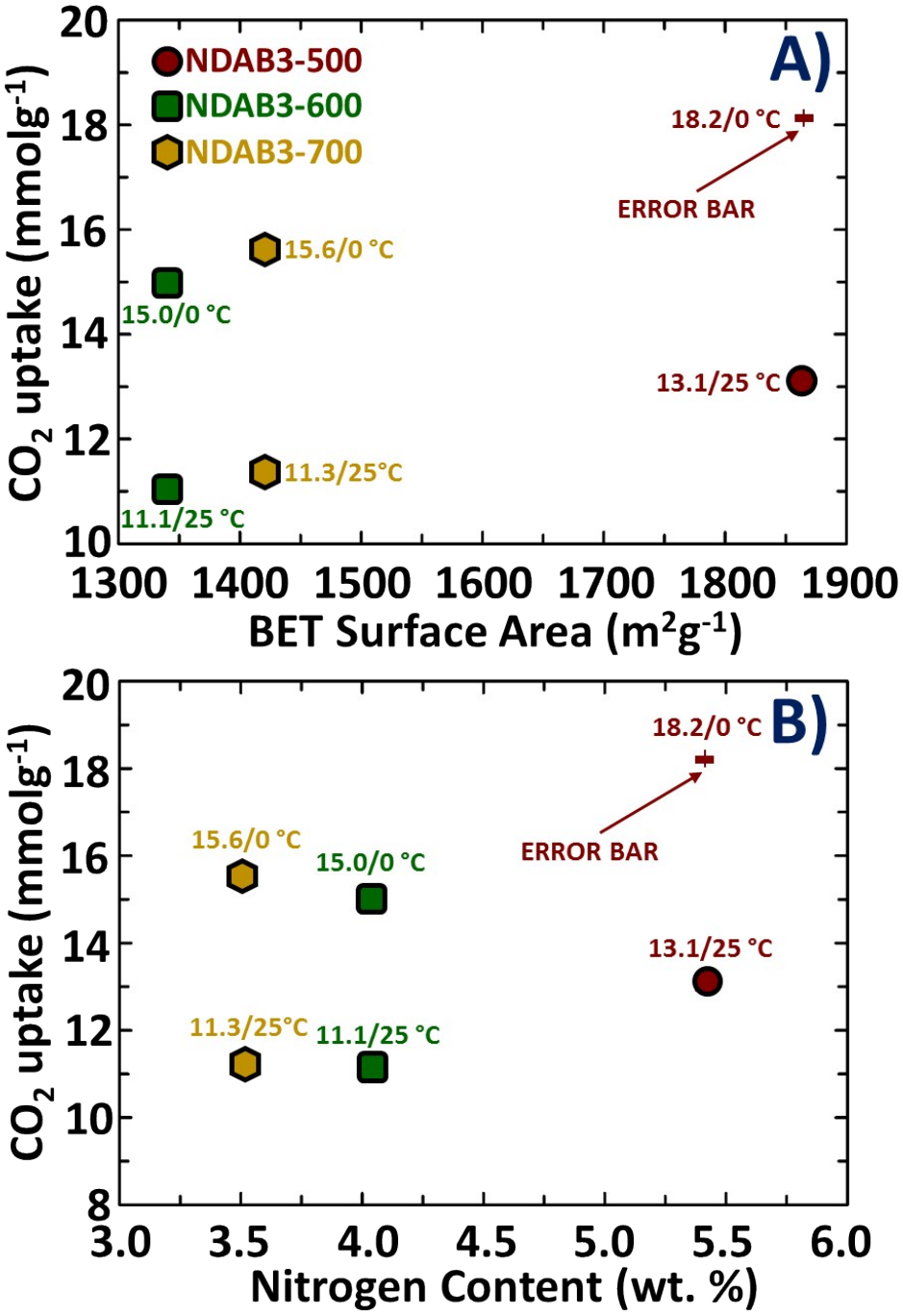


**Fig. S6.** The relation between $CO_2$ adsorption measured at different conditions with A) BET surface area and B) nitrogen content of the **NDAB3-T** materials. (Note: Error bars included for $CO_2$ measurements recorded at 0°C)

**Table S1.** Carbon and nitrogen content in all **NDABn-T** (n=0-3, T=500-700 °C) materials.

| Material | C (wt.%) | N (wt.%) | N/C (wt. ratio) | C (at.%) | N (at.%) | N/C (at. ratio) |
|---|---|---|---|---|---|---|
| NDAB0-500 | 65 | 6.5 | 0.01 | 5.4 | 0.46 | 0.085 |
| NDAB1-500 | 68 | 6.2 | 0.09 | 5.6 | 0.44 | 0.078 |
| NDAB2-500 | 70 | 5.9 | 0.08 | 5.8 | 0.42 | 0.072 |
| NDAB3-500 | 75 | 5.4 | 0.07 | 6.2 | 0.39 | 0.061 |
| NDAB0-600 | 68 | 4.2 | 0.06 | 5.7 | 0.30 | 0.053 |
| NDAB1-600 | 73 | 5.1 | 0.07 | 6.1 | 0.36 | 0.060 |
| NDAB2-600 | 75 | 5.1 | 0.07 | 6.3 | 0.36 | 0.058 |
| NDAB3-600 | 81 | 4.1 | 0.05 | 6.7 | 0.29 | 0.043 |
| NDAB0-700 | 76 | 3.3 | 0.04 | 6.4 | 0.23 | 0.037 |
| NDAB1-700 | 77 | 3.9 | 0.05 | 6.4 | 0.28 | 0.043 |
| NDAB2-700 | 76 | 4.0 | 0.05 | 6.4 | 0.28 | 0.045 |
| NDAB3-700 | 82 | 3.5 | 0.04 | 6.8 | 0.25 | 0.037 |

The concentration of carbon and nitrogen in the activated materials is dependent upon the impregnation ratio of $ZnCl_2$ used and the carbonization temperature.

**Table S2.** XPS surface elemental composition of **NDAB0-T** (T = 500-700 °C) and **NDAB3-T** (T = 500-700 °C).

| Sample | C (at.%) | N (at. %) | O (at. %) | N/C (ratio) | O/C (ratio) |
|---|---|---|---|---|---|
| **NDAB0-500** | 89 | 4.0 | 6.7 | 0.045 | 0.076 |
| **NDAB0-600** | 88 | 2.4 | 4.5 | 0.027 | 0.051 |
| **NDAB0-700** | 92 | 2.0 | 4.7 | 0.022 | 0.051 |
| **NDAB3-500** | 90 | 4.4 | 5.4 | 0.049 | 0.060 |
| **NDAB3-600** | 92 | 3.0 | 4.1 | 0.032 | 0.044 |
| **NDAB3-700** | 94 | 1.9 | 3.7 | 0.021 | 0.040 |

**Table S3. EDS** elemental composition of **NDAB3-T** (T = 500-700 °C).

| Sample | C (at.%) | N (at. %) | O (at. %) | N/C (ratio) | O/C (ratio) |
|---|---|---|---|---|---|
| **NDAB3-500** | 82 | 5.2 | 13.1 | 0.064 | 0.16 |
| **NDAB3-600** | 87 | 2.1 | 11.0 | 0.024 | 0.13 |
| **NDAB3-700** | 91 | 1.5 | 7.2 | 0.016 | 0.08 |

**Table S4.** Textural properties of N-doped activated carbons derived from *Arundo donax* and chitosan under mild activation.

| Sample | $SA_{BET}$ ($m^2g^{-1}$) | $SA_{micro}$ (%) /$SA_{meso}$ (%) | $V_{total}$ ($cm^3g^{-1}$) | $V_{micro}$ (%) /$V_{meso}$ (%) | PW (nm) |
|---|---|---|---|---|---|
| **NDAB0-500** | 3.0 | - | - | - | - |
| **NDAB1-500** | 1070 | 87/13 | 0.45 | 80/20 | 1.6 |
| **NDAB2-500** | 1559 | 80/20 | 0.67 | 72/18 | 1.6 |
| **NDAB0-600** | 4.1 | - | - | - | - |
| **NDAB1-600** | 1145 | 90/10 | 0.47 | 85/15 | 1.5 |
| **NDAB2-600** | 1328 | 80/20 | 0.66 | 76/24 | 1.6 |
| **NDAB0-700** | 3.7 | - | - | - | - |
| **NDAB1-700** | 1148 | 89/11 | 0.47 | 83/17 | 1.5 |
| **NDAB2-700** | 1335 | 75/25 | 0.63 | 71/29 | 1.5 |

**$SA_{BET}$** – BET surface area (Brunauer Emmett Teller), **$SA_{micro}$** – micropore surface area, **$SA_{meso}$** – mesopore surface area, **$PV_{total}$** – total pore volume, **$PV_{micro}$** – micropore volume, **$PV_{meso}$** – mesopore volume and **PW –** Pore Width calculated using DFT method

At any given temperature, a progressive increase in the surface area and pore volume is observed when the $ZnCl_2$ impregnation ratio is increased from 0 to 2. At any given impregnation ratio, the highest textural parameters are observed at a temperature of 500 °C.