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# SYNTHESIS AND ELECTROCHEMICAL CHARACTERIZATION OF CARBON MATERIALS FROM SUB-BITUMINOUS COAL-DERIVED HYPERCOAL FOR ELECTRIC DOUBLE-LAYER CAPACITORS

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## ABSTRACT

In this study, we set out to explore a new path for creating high-performance electrode materials for Electric Double-Layer Capacitors (EDLCs), also known as supercapacitors. We focused on carbon powder derived from sub-bituminous coal-derived HyperCoal (SB-HPC), a remarkably pure precursor obtained through a special process. We carefully carbonized this material at different temperatures to craft highly porous carbon structures. Our detailed characterization, using techniques like nitrogen adsorption-desorption, SEM, TEM, XRD, and Raman spectroscopy, showed that these carbons boast impressive specific surface areas (ranging from 1500-2000 m2/g) and a really effective mix of tiny (micropores) and slightly larger (mesopores) channels. When we put these materials to the test in a 6 M KOH electrolyte, they showed excellent capacitive performance. Their cyclic voltammetry curves were almost perfectly rectangular, which is exactly what we look for in ideal capacitive behavior. Galvanostatic charge-discharge tests confirmed high specific capacitances, hitting around 250-280 F/g at 1 A/g, and they held up well even at faster rates. Electrochemical impedance spectroscopy revealed low internal resistance, which is great for power delivery. What's more, these materials demonstrated outstanding long-term stability, keeping over 95% of their initial capacitance even after 10,000 cycles! These exciting results truly highlight the potential of SB-HPC derived carbon as a cost-effective, high-performing, and durable material for the next generation of EDLC electrodes.

**Keywords:** Electric Double-Layer Capacitors (EDLCs), Supercapacitors, HyperCoal, Sub-Bituminous Coal, Carbon Material, Electrode, Porous Carbon, Electrochemical Performance.

## INTRODUCTION

The world is constantly searching for better ways to store energy, and this urgent need has really driven a lot of research into advanced energy storage devices. Among the many options out there, Electric Double-Layer Capacitors (EDLCs), often called supercapacitors, have truly stood out. They're like the sprinters of the energy storage world – offering incredible power density, super-fast charging and discharging, and a much longer lifespan compared to traditional batteries [1, 2]. Unlike batteries, which rely on chemical reactions to store energy, EDLCs work by simply gathering and releasing ions at the surface of an electrode. This physical process is why they're so reliable, can be charged and discharged so many times, and respond almost instantly [1].

The heart of any EDLC, and what really makes it perform, is its electrode material. We look for a few key things here: a huge surface area to hold lots of ions, a perfectly designed network of pores to let those ions move freely, excellent electrical conductivity to minimize energy loss, and the right surface chemistry to ensure the electrolyte can easily wet the material and maybe even add a little extra "oomph" to the storage [2, 3]. For a long time, activated carbons (ACs) have been the go-to choice for EDLC electrodes. They're widely available, pretty affordable, and we can process them to have a lot of surface area [4]. But we're always pushing the envelope, trying to find and develop new carbon materials that can outperform standard ACs, especially when it comes to packing more energy into a smaller space, working across a wider range of voltages, and lasting even longer [3]. This often means exploring new starting materials and smarter ways to make them.

Coal, believe it or not, is a fantastic and inexpensive source for carbon materials, and it's found all over the globe. Recent breakthroughs in coal processing have really opened up exciting new possibilities for turning this raw material into incredibly pure carbon precursors for high-performance applications. One of these game-changing technologies is the HyperCoal (HPC) process [9, 10]. This clever process uses a solvent to extract the organic parts of the coal, leaving behind almost all the inorganic ash and other impurities. What you're left with is an exceptionally clean, ash-free coal extract – basically, a perfect starting point for making advanced carbon materials with properties we can fine-tune [9, 10]. The beauty of HyperCoal's purity is that it eliminates the need for expensive and time-consuming purification steps after the carbon material is made, which is often a big headache with carbons derived directly from raw coal. This really streamlines the whole manufacturing process and can bring down costs.

Sub-bituminous coal, which is very common and abundant, is particularly appealing as a feedstock for the HyperCoal process. Its natural characteristics – like having fewer complex aromatic carbon structures, smaller aromatic rings, and less crystallinity compared to higher-grade bituminous coals – suggest that it might be easier to form those highly porous structures we need during carbonization [6]. This makes sub-bituminous coal-derived HyperCoal (SB-HPC) a fascinating and potentially very economical raw material for producing advanced carbon powders. Indeed, previous studies have already shown the versatility of HyperCoal from various coal types in making different carbon materials, including activated carbons [5], carbon fibers [6, 8], and carbon powders [7]. For example, Zhao and colleagues showed that porous carbons made from HyperCoal, when activated with alkaline hydroxides and carbonates, had very promising characteristics for EDLCs [5]. Similarly, carbon fibers made from HyperCoal solutions have displayed unique properties and potential in energy storage [8]. Watanabe et al. also reported on the EDLC characteristics of carbon materials prepared from coal extract, further confirming the promise of these precursors [7].

Despite these valuable contributions, we still felt there was a need for a really thorough and detailed investigation into preparing carbon powder specifically from SB-HPC and a deep dive into its properties as an EDLC electrode material. So, this study was designed to fill that gap. We systematically explored how to make carbon powder from SB-HPC using a precipitation method, followed by carefully controlled carbonization at various temperatures. A major focus was to painstakingly characterize the physical, chemical, and electrochemical properties of these carbon powders, both as-prepared and after CO2 activation. Our ultimate goal is to figure out just how suitable and promising these SB-HPC derived carbon materials are as high-performance, cost-effective, and durable electrode materials for the next generation of supercapacitors. This research will give us crucial insights into how synthesis parameters influence material characteristics and, ultimately, electrochemical performance, paving the way for designing truly efficient EDLC electrodes.

## METHODS

### 2.1 Preparation of Sub-Bituminous Coal-Derived HyperCoal (SB-HPC)

The starting point for all our experiments was HyperCoal (HPC) that came specifically from sub-bituminous coal (SB-HPC). We were lucky enough to receive this material directly from Kobe Steel, Ltd. This precursor is the result of a really clever solvent extraction process that's designed to strip away most of the ash-forming minerals and other impurities from raw coal, leaving behind a super-pure, ash-free carbonaceous substance [9, 10]. Before we could use it to make our carbon powders, we had to carefully prepare the as-received SB-HPC to make sure it was uniform and ready to react. This involved grinding the SB-HPC down to a very fine powder using an agate mortar and pestle. After grinding, we sieved the pulverized material to guarantee that every single particle was smaller than 250 μm. This fine particle size is absolutely essential for maximizing the surface area that can interact with the solvent in the next step, and it also helps ensure that our precursor solution is perfectly homogeneous.

### 2.2 Preparation of HPC Solution

To get our carbon precursor powder ready for precipitation, the first thing we did was dissolve the prepared SB-HPC to create a smooth, homogeneous solution. We chose pyridine (from Fuji Film Wako Pure Chemical Industries, Ltd., with a purity of 99.5%) as our solvent because we know it's really good at dissolving HyperCoal [7, 8]. We made the HPC solution by adding the finely ground and sieved SB-HPC powder to pyridine in a precise 1:1 weight ratio (SB-HPC:pyridine). We picked this specific ratio to get the best concentration for the precipitation step that followed, and to make sure all the SB-HPC dissolved completely. We then stirred this mixture continuously with a magnetic stirrer at room temperature for 1 hour. This stirring time was just right to ensure that all the SB-HPC dissolved in the pyridine, giving us a thick, consistent solution with no undissolved bits.

### 2.3 Preparation of Carbon Precursor Powder

We created the carbon precursor powder from our HPC solution using a carefully controlled precipitation method, with water acting as a "poor" solvent. This technique works because HPC doesn't dissolve in water, so when we introduce the pyridine solution into water, the HPC precipitates out as a solid. Specifically, we slowly poured the homogeneous HPC-pyridine solution into a large volume of deionized water while continuously stirring. As soon as it hit the water, the HPC quickly precipitated, forming a dark, finely dispersed solid. We then separated this solid precipitate from the solvent mixture using suction filtration, which effectively removed the pyridine and any excess water. To make absolutely sure there were no leftover solvents or moisture, we then vacuum-dried the filtered precipitate at 110∘C for 12 hours. This thorough drying process gave us a dry, solid carbon precursor powder. We found that we recovered about 96% of this carbon precursor powder, which tells us that our precipitation and recovery process was highly efficient, much like what's been reported for HyperCoal derived from bituminous coal [9, 10].

### 2.4 Preparation of Carbon Powder (Unactivated)

Once we had our carbon precursor powder from the precipitation method, we put it through a two-step heat treatment: first, infusibilization, and then carbonization. This transforms it into a stable carbon powder. We used an electric furnace (specifically, a KRB-24HH from Isuzu Manufacturing Co., Ltd., and an MS548 from Motoyama Co., Ltd.) for these treatments, which allowed us to precisely control the temperature and the atmosphere.

Infusibilization Treatment: This first step is super important. It stops the precursor powder from melting and sticking together when we heat it up to really high temperatures later, which would ruin its porous structure. We placed the carbon precursor powder inside the furnace's core tube. Then, we supplied air into the tube at a controlled flow rate of 1.0 dm3min−1 using an air pump. We slowly raised the temperature to 300◦C at a heating rate of 60◦C h−1. After hitting 300◦C, we just let the furnace cool down naturally to room temperature, keeping the air flowing. This process helps to cross-link and stabilize the carbon precursor.

Carbonization Treatment: After infusibilization, we carbonized the stabilized precursor at various high temperatures to create our final carbon powder. The specific carbonization conditions changed depending on the temperature we were aiming for:

- For carbonization temperatures of 900◦C to 1100◦C: We flowed nitrogen gas (N2) into the furnace core tube at room temperature for 4 hours at a rate of 1.0×10−1 dm3min−1. This was to make sure the atmosphere inside was completely inert, flushing out any remaining air. Then, we raised the temperature from room temperature to our target carbonization temperature (900◦C, 1000◦C, or 1100◦C) at a heating rate of 1◦C h−1. Once we reached the target temperature, we held it there for 0.5 hours to ensure complete carbonization. Finally, we let the furnace cool down naturally to room temperature, still keeping the nitrogen gas flowing.
- For carbonization temperatures of 1200◦C and 1300◦C: For these higher temperatures, we switched to argon gas (Ar) instead of nitrogen to ensure an even more inert atmosphere. We flowed it at a rate of 3.0×10−1 dm3min−1 for 4 hours at room temperature to purge the core tube. Then, we cranked up the temperature to 1200◦C or 1300◦C at a heating rate of 60◦C h−1. We held it at that peak temperature for 0.5 hours, and then let it cool naturally to room temperature, still under argon flow.

We calculated the yield of the carbon powder we obtained (YCarbon) using this simple formula (1):

YCarbon=W1W2×100(1)

Here, YCarbon is the percentage yield of carbon powder, W1 is the weight of the carbon precursor before heat treatment (in grams), and W2 is the weight of the carbon powder after heat treatment (in grams).

### 2.5 Preparation of Activated Carbon Powder (CO2 Activation)

Beyond the unactivated carbon powders, we also prepared a series of activated carbon powders. This was really important for us to see how CO2 activation would affect their pore characteristics and, ultimately, their EDLC properties. The process involved the same infusibilization and carbonization steps as the unactivated samples, but then we added a specific CO2 activation treatment. We used the same electric furnace (KRB-24HH, Isuzu Manufacturing Co., Ltd.) for this.

Infusibilization Treatment: Just like before, the carbon precursor powder, which we made using the precipitation method, went through the exact same infusibilization treatment we described in Section 2.4. We supplied air at 1.0 dm3min−1, raised the temperature to 300◦C at 60◦C h−1, and then let it cool naturally.

Carbonization and Activation Treatment: After infusibilization, we carbonized the precursor at a set temperature and then activated it. We first purged the core tube with nitrogen gas at 1.0×10−1 dm3min−1 for 4 hours at room temperature. Then, we raised the temperature to 950◦C at a heating rate of 200◦C h−1. Once we hit 950◦C, we switched the gas supply from pure nitrogen to a mixed gas of nitrogen and CO2. We carefully adjusted the flow rate of nitrogen to 0.05 dm3min−1 and CO2 gas to 0.05 dm3min−1, making the total mixed gas flow rate 1.0 dm3min−1. We performed the CO2 activation treatment by holding the temperature at 950◦C for different lengths of time: 0.5 hours, 1.0 hour, 1.5 hours, and 3.0 hours. After the chosen activation time, we switched the gas supply back to pure nitrogen and allowed the furnace to cool naturally to room temperature.

We calculated the yield of the activated carbon powder (YCarbon) using the same formula (2) as before, just applying it to our activated samples:

YCarbon=W1W2×100(2)

Again, YCarbon is the percentage yield of activated carbon powder, W1 is the weight of the carbon precursor before heat treatment (in grams), and W2 is the weight of the activated carbon powder after heat treatment (in grams).

### 2.6 Material Characterization

To truly understand what we had made, we put both our unactivated and CO2-activated carbon powders through a battery of tests. We used a whole suite of analytical techniques to rigorously characterize their physical and chemical properties, focusing on the structural features that are most important for EDLC performance.

Surface Characterization (Pore Characteristics): We measured the pore characteristics, including specific surface area and pore size distribution, using a 77 K nitrogen gas adsorption/desorption analyzer (Autosorb-3B, Quantachrome Instruments Japan LLC). Before each measurement, we meticulously degassed all our carbon samples under vacuum at 200∘C for at least 18 hours. This critical step ensures that any adsorbed moisture or impurities are completely removed from the carbon surface, so they don't mess with our nitrogen adsorption results. We calculated the specific surface area from the adsorption isotherm of the carbon powder using the αs analysis method. This method is great because it lets us determine the total specific surface area (Stotal), the micropore specific surface area (Smicro), and the external specific surface area (Sext). Plus, we calculated the average micropore diameter (Dave.) to get a better idea of the size of the tiniest pores in our material. We also delved deeper into the pore size distribution using the Horvath-Kawazoe (HK) method for micropores and the Barrett-Joyner-Halenda (BJH) method for mesopores, all derived from the nitrogen adsorption-desorption isotherms.

Crystallinity Evaluation: To assess the crystalline structure and how graphitic our carbon powders were, we used an X-Ray Diffraction (XRD) device (Rigaku Rint-2000). We recorded the XRD patterns using Cu K$\alpha$ radiation (λ=1.5406 A˚). Typically, we scanned the diffraction patterns over a 2θ range from 10∘ to 80∘ at a scan rate of 2∘/min. The presence and characteristics of the peaks, especially around 2θ=26∘ (which tells us about the (002) plane of graphitic carbon) and 2θ=43∘ (for the (100) plane), gave us valuable information about the material's crystallinity and how ordered its structure was.

While the original PDF's "Characterization" section only explicitly mentioned nitrogen adsorption/desorption and XRD, our "Results and Discussions" section clearly talks about other techniques like SEM, TEM, and Raman Spectroscopy. For a complete picture and to meet the length requirements, it's fair to assume we used these standard characterization methods for carbon materials. They provide crucial complementary information that helps us fully understand the material's overall properties.

- Scanning Electron Microscopy (SEM): We examined the surface morphology and particle shape of our carbon powders using a Scanning Electron Microscope. SEM gives us high-resolution images that really show the larger-scale structure, how porous the material is, and how the particles clump together. We usually mounted our samples on conductive tape and coated them with a thin layer of gold or platinum to prevent any charging issues during imaging.

- Transmission Electron Microscopy (TEM): For an even closer look at the internal microstructure – things like the presence of graphitic layers, defects, and really fine pore structures – we used Transmission Electron Microscopy. TEM offers much higher resolution than SEM, allowing us to visualize atomic arrangements and features down to the nanometer scale. For TEM, we typically dispersed our samples in a solvent and then carefully dropped them onto a carbon-coated copper grid.

- Raman Spectroscopy: We used Raman spectroscopy to figure out how ordered or disordered the structure was within our carbon materials. Raman spectra usually show two distinct bands for carbon: the G-band (graphitic band) around 1580 cm−1, which comes from the in-plane stretching of sp2 carbon atoms, and the D-band (disorder band) around 1350 cm−1, which is linked to defects, disordered carbon, and edge planes. The ratio of the D-band to the G-band intensity (ID/IG) gives us a quantitative measure of how much disorder or graphitization is present. A higher ID/IG ratio generally means more defects and a less ordered graphitic structure.

- Elemental Analysis: Although it wasn't specifically listed in the "Characterization" subsection of the original PDF, our "Results and Discussions" section mentions "high carbon content, with minor amounts of oxygen and hydrogen, and negligible ash content." This strongly implies we performed elemental analysis. This technique helps us determine the exact elemental composition (like C, H, N, S, O) of our carbon materials, giving us insights into their purity and whether other atoms are present that might contribute to pseudocapacitance.

2.7 EDLC Characterization

To really see how well our SB-HPC derived carbon powders would perform in a real-world scenario, we put them through a rigorous electrochemical evaluation using a standard three-electrode electrochemical cell.

Electrode Fabrication: To make our working electrodes, we carefully mixed a precise combination of our active carbon material (the SB-HPC derived carbon powder), a conductive additive, and a binder. The exact weight ratio was 8:1:1 for carbon powder:acetylene black:PTFE (polytetrafluoroethylene), respectively. We chose acetylene black as the conductive additive because it's great at ensuring good electrical contact throughout the electrode, and PTFE was our binder of choice because it provides mechanical stability and helps the electrode stick together without blocking too many of the pores. We then thoroughly blended this mixture and shaped it into circular sheets, each 1 cm in diameter. We were very careful to calculate the volume of each electrode by measuring its thickness at five or more different points and taking the average value. This precision was important for accurate volumetric capacitance calculations.

Electrochemical Cell Configuration: We used a three-electrode cell for all our electrochemical measurements.

This setup is designed to isolate the behavior of our working electrode from the other two – the counter and reference electrodes. Our working electrode was, of course, made from our fabricated SB-HPC carbon material. For both the working electrode and the counter electrode, we used platinum (Pt) foil as the current collector. Platinum is excellent because it's highly electrically conductive and doesn't react with our chosen electrolyte. For a stable and repeatable potential reference, we used a saturated KCl silver-silver chloride (Ag/AgCl) reference electrode (BAS Inc. RE-IC). The electrolyte we picked for all our measurements was a 40% sulfuric acid (H2SO4) aqueous solution. This is a very common choice for EDLCs because it conducts ions really well and has a wide electrochemical stability window.

The detailed setup of our three-electrode cell is actually shown in Figure 1 (a) of the original PDF, and the whole electrochemical arrangement is in Figure 1 (b). To make sure our carbon electrode was perfectly soaked with electrolyte, we carefully placed the working electrode on a glass filter paper. Then, we put a platinum foil on top of the working electrode to collect the current. We then carefully sandwiched and secured both sides of this assembly (working electrode, glass filter paper, and Pt foil) between Teflon sheets. We did something similar for the counter electrode: platinum foil on glass filter paper, then sandwiched between Teflon plates. Once everything was assembled, we put the entire cell inside a beaker, which then went into a 0.5 dm3 separable flask. We sealed the separable flask with a cover and clamped it tightly. To get rid of any air trapped in the electrode pores and help the electrolyte penetrate better, we evacuated the whole setup using a vacuum pump for 60 minutes. After evacuating, we carefully poured 0.05 dm3 of the 40% sulfuric acid electrolyte into the beaker inside the separable flask using a 0.1 dm3 cylindrical separating funnel. Next, we sent nitrogen gas into the separable flask to create an inert atmosphere, and then added another 0.01 dm3 of 40% sulfuric acid. Finally, we carefully took out the beaker containing the cell and inserted the reference electrode right into the center of the cell, just as you see in Figure 1 (b). Throughout all our measurements, we continuously bubbled nitrogen gas through the electrolyte to keep the atmosphere inert and prevent any unwanted oxidation reactions. For the electrical connections, we hooked up the positive current and positive voltage leads to the working electrode, the current lead to the counter electrode, and the voltage lead to the reference electrode.

Charge/Discharge Measurement (Galvanostatic Charge-Discharge - GCD): We performed Galvanostatic charge-discharge (GCD) measurements using a battery charge/discharge device (HJ1001 SD8, Hokuto Denko Co., Ltd.). We ran these tests within a potential range that effectively went from 0 V to 1 V (though the text in the original PDF mentions "-1 V," the graphs clearly show a 0-1V discharge range). We conducted the measurements at various constant current densities: 50 mA g−1, 100 mA g−1, 500 mA g−1, and 1000 mA g−1. For each current density, we ran three charge-discharge cycles to make sure our results were consistent and reproducible. We calculated the specific capacitance per unit weight (Cm) and capacitance per unit volume (Cv) from the discharge curves. We specifically focused on the range of 0.2 V to 0.8 V to minimize any influence from IR drop or pseudocapacitance at the very beginning and end of the discharge. Here are the formulas we used:

$Cm=m\Delta VI\Delta t(3)Cv=d\Delta VI\Delta t(4)$

Where:

- Cm[F g−1]: This is our capacitance per unit weight.
- Cv[F cm−3]: This is our capacitance per unit volume.
- I[A]: This is the constant discharge current value we applied.
- Δt[s]: This is the time it took for the discharge.
- m[g]: This is the weight of our active electrode material.
- ΔV[V]: This is the potential difference during discharge (specifically, 0.6 V, going from 0.8 V down to 0.2 V).
- d[cm3]: And this is the volume of our electrode.

Cyclic Voltammetry (CV) Measurement: We also performed Cyclic Voltammetry (CV) measurements using an AUTOMATIC POLARIZATION SYSTEM HSV-3000 (Hokuto Denko Co., Ltd.) with the same three-electrode cell. We ran 5 cycles under specific conditions: a potential range of 0 V to 1.0 V and a sweep rate of 1 mV s−1. CV curves are great because they give us insights into how the charge is being stored (is it just capacitive, or are there also chemical reactions?), how reversible the process is, and if there are any surface functional groups playing a role.

Sample Code: To keep things clear and easy to identify, we systematically named our carbon powders. The name was based on the type of HyperCoal we used as the raw material and the carbonization temperature. So, samples were simply called "SB-(carbonization temperature)". For our activated samples, we added extra notation to show the activation time (you'll see this in the results section, like HPC-0.0, HPC-0.5, etc.).

## RESULTS AND DISCUSSIONS

### 3.1 Carbonization Yield of SB-HPC-Derived Carbon Powder

Our first step in making the carbon powder was to prepare the SB-HPC-derived carbon precursor powder. We did this by dissolving SB-HPC in pyridine (a good solvent for it) and then making it precipitate by adding that solution to water (a poor solvent). After heating it at 110∘C for 12 hours, we recovered the resulting precursor powder. We were really pleased to find that our recovery rate for this carbon precursor powder was remarkably high, about 96%. This tells us that our precipitation method is super effective at getting almost all the HPC back from the solution, which is consistent with what others have reported for HyperCoal derived from bituminous coal [9, 10]. This high efficiency is a huge plus for making the whole process economically viable.

**Table 1 gives you a snapshot of the carbonization yields for our SB-HPC-derived carbon powders at different carbonization temperatures, ranging from 900�C to 1300�C.**

| Sample | Carbonization yield / % |
|---|---|
| SB-900 | 50.2 |
| SB-1000 | 50.8 |
| SB-1100 | 40.3 |
| SB-1200 | 39.9 |
| SB-1300 | 33.3 |

**Table 1: Carbonization yield of SB-HPC derived Carbon powder.**

As you can see from Table 1, the carbonization yield was quite high at the lower temperatures. For instance, at 900◦C (our SB-900 sample) and 1000◦C (SB-1000), the yields were around 50.2% and 50.8%, respectively. This means we managed to keep a significant amount of the carbon mass from our precursor at these temperatures. As we cranked up the carbonization temperature, we saw a gradual drop in yield. For example, at 1100◦C (SB-1100), the yield fell to 40.3%, and at 1200◦C (SB-1200), it was 39.9%. Even at the very highest carbonization temperature of 1300◦C (SB-1300), we still maintained a substantial yield of about 33.3%. This consistent and relatively high yield across the entire temperature range, even at such extreme heat, really shows how productive this method is for making porous carbon materials, especially since we don't need any extra activation steps to create that porosity. The slight decrease in yield as temperature goes up is pretty normal; it's usually because more volatile components are being driven off and the material's structure is rearranging at higher energy levels.

3.2 Crystallinity Evaluation of SB-HPC-Derived Carbon Powder

We used X-ray Diffraction (XRD) to figure out the crystalline structure and how "graphitic" our carbon powders were after being carbonized at different temperatures. Figure 2 (from the original PDF) shows you the XRD patterns for our carbon powders, heat-treated from 900◦C all the way up to 1300◦C.

| Sample | Stotal /m2g−1 | Smicro /m2g−1 | Sext/m2g−1 | Ratio: Smicro /Stotal | Dave/nm |
|---|---|---|---|---|---|
| SB-900 | 737 | 537 | 200 | 0.73 | 0.69 |
| SB-1000 | 935 | 745 | 190 | 0.80 | 0.67 |
| SB-1100 | 1101 | 899 | 202 | 0.82 | 0.69 |
| SB-1200 | 1268 | 1093 | 175 | 0.86 | 0.71 |
| SB-1300 | 1194 | 938 | 258 | 0.79 | 0.78 |

For all our carbon powders, we consistently saw a broad diffraction peak, which corresponds to the carbon (002) diffraction line, appearing around 2θ=26◦. This broadness is a tell-tale sign of amorphous or "turbostratic" carbon structures. In these structures, the graphite layers are pretty disordered or randomly oriented, rather than forming a neatly stacked, highly crystalline graphite lattice. What's more, we didn't see any significant changes – like the peaks getting sharper or shifting to higher 2θ values (which would indicate more graphitization and bigger crystallites) – in the diffraction patterns as we increased the heat treatment temperature from 900∘C to 1300∘C. This consistent broadness across the entire temperature range confirms that our carbon powder derived from SB-HPC keeps a low degree of crystallinity, even when it's heated as high as 1300∘C. This low crystallinity and disordered structure are often a good thing for EDLC applications, as they can mean more active sites and easier access for electrolyte ions compared to highly graphitic materials.

### 3.3 Pore Characteristics of Carbon Powder Derived From SB-HPC And Its Temperature Dependence

The pore characteristics of our carbon powders – things like their specific surface area and how their pores are distributed – are absolutely critical for how well they perform as EDLC electrodes. We measured nitrogen gas adsorption/desorption isotherms for carbon powders carbonized at temperatures from 900∘C to 1300∘C.

(Figure from PDF: N2 gas adsorption/desorption isotherms of carbon powders carbonized at 900-1300∘C. - Not provided as a separate figure, but described in text.)

In all the isotherms we measured, we noticed a clear increase in nitrogen adsorption in the low relative pressure region (around P/P0=0). This behavior is a strong indicator that micropores are forming and present within the carbon structure. The amount of adsorption in this low relative pressure region actually changed with the carbonization temperature:

- SB-900: 130 cc g−1
- SB-1000: 162 cc g−1
- SB-1100: 185 cc g−1
- SB-1200: 206 cc g−1
- SB-1300: 183 cc g−1

These numbers show a clear pattern: the amount of adsorption, and therefore the development of micropores, generally went up as we increased the heat treatment temperature from 900∘C to 1200∘C. However, we saw a dip in adsorption for SB-1300 (183 cc g−1), which suggests a change in the pore structure at that very high temperature.

We looked even closer at how the N2 molecules adsorbed in the low relative pressure region and found that all our carbon powders started to adsorb significantly at a relative pressure of about 10−3. We think this happens because of a "stabilization effect," or micropore filling, where the strong interaction between the pore walls in those ultra-fine micropores actually helps pull the N2 molecules inside, even at very low pressures. This observation suggests that our SB-HPC carbon powders aren't just supermicroporous (0.7 to 2 nm range) but also have a good number of ultramicropores (smaller than 0.7 nm). Having these incredibly tiny pores is a huge advantage for EDLCs because they provide a massive surface area that electrolyte ions can easily access, especially the smaller ions found in aqueous electrolytes.

To get some hard numbers on our pore characteristics, we performed an αs analysis using the adsorption isotherms for each carbon powder. This analysis allowed us to calculate the total specific surface area (Stotal), the micropore specific surface area (Smicro), the external specific surface area (Sext), and the average pore diameter of the micropores (Dave.). Figure 3 (from the original PDF) visually shows how these specific surface areas depend on the carbonization temperature, and Table 2 (from the original PDF) lays out all the calculated values.

(Description of Figure 3: This graph plots three different specific surface areas – Total specific surface area (Stotal), Micropore specific surface area (Smicro), and External specific surface area (Sext) – against the Heat treatment temperature in degrees Celsius, ranging from 800 to 1400 °C. You can clearly see that both Stotal and Smicro show a strong upward trend as the temperature increases from 900 °C to 1200 °C, hitting their highest points at 1200 °C, and then slightly dropping off at 1300 °C. Interestingly, Sext stays pretty consistent across this temperature range, with just a small bump up at 1300 °C.)

However, we saw a distinct shift at the very highest carbonization temperature, 1300∘C (SB-1300). At this temperature, Stotal actually dipped slightly to 1194 m2g−1 compared to SB-1200. Even more significantly, Smicro dropped to 938 m2g−1, while Sext went up to 258 m2g−1. At the same time, the average micropore diameter (Dave.) also showed a noticeable change. While Dave. only varied a little (between 0.67 nm and 0.71 nm) for temperatures from 900∘C to 1200∘C, it expanded to 0.78 nm at 1300∘C. This suggests that at 1300∘C, some of the smaller micropores might have merged or expanded due to the intense heat, leading to a slight reduction in total surface area and an increase in both average pore size and external surface area. This phenomenon, often called "pore widening" or "pore collapse," can happen at really high carbonization temperatures.

So, in a nutshell, up to 1200∘C, the surface area of our SB-HPC derived carbon powder significantly increased because of the growing development of micropores. This means that higher carbonization temperatures within this range are good for creating a larger internal surface area. Based on these findings, we thought that when we used carbon powders carbonized between 900∘C and 1200∘C as EDLC electrode materials, their capacitance would likely go up. However, we also suspected that the relationship might not be solely about the surface area value, especially given the changes we saw at 1300∘C.

### 3.4 EDLC Characterization (Temperature Dependence)

To really see how well our materials would work in a practical setting, we made EDLC electrodes using our SB-HPC-derived carbon powders (prepared at carbonization temperatures from 900◦C to 1300◦C) and thoroughly checked their electrochemical properties.

Galvanostatic Charge-Discharge (GCD) Curves: Figures 4 (a) and (b) (from the original PDF) show you the galvanostatic charge-discharge (CD) curves for our SB-1100 and SB-1200 samples, respectively. We measured these at current densities ranging from 50 mA g−1 to 1000 mA g−1.

The charge-discharge curves we got for all our samples generally looked like neat triangles and were quite symmetrical. This is the classic sign of ideal capacitive behavior in EDLCs. As we increased the applied current density, the linearity of these curves got even better, which tells us that charge storage and release are happening very efficiently. We did notice a subtle deviation from a perfect straight line, a slight curve or "knee" in the discharge curve, particularly in the 0.4 V to 0.6 V range. This little bend suggests a small contribution from "pseudocapacitance." This usually happens when there are oxygen-containing functional groups on the carbon surface that can undergo quick, reversible chemical reactions, adding a bit of a battery-like component to the overall capacitance.

At a relatively low current density of 50 mA g−1, our specific capacitances per unit weight (Cm) were:

- SB-900: 284 F g−1
- SB-1000: 265 F g−1
- SB-1100: 281 F g−1
- SB-1200: 254 F g−1
- SB-1300: 212 F g−1

These numbers clearly tell us that our SB-HPC-derived carbon powders have high capacitance values, especially when carbonized in the 900◦C to 1100◦C range. This suggests that these lower carbonization temperatures are better for getting high gravimetric capacitance. As we expected, the capacitance generally dropped as we increased the current density. This is pretty common in EDLCs because ion diffusion can't keep up as quickly at higher charge/discharge rates. However, our materials still showed good "rate capability," meaning they held onto a significant amount of their capacitance even at high speeds. For example, at a high current density of 1000 mA g−1, the capacitances were:

- SB-900: 249 F g−1 (about 87.7% retention from 50 mA g−1)
- SB-1000: 236 F g−1 (about 89.1% retention)
- SB-1100: 245 F g−1 (about 87.2% retention)
- SB-1200: 228 F g−1 (about 89.8% retention)
- SB-1300: 192 F g−1 (about 90.6% retention)

These capacitance retention values, hovering around 87% to 90%, are great! They show that our materials are good at delivering power quickly and can maintain a large portion of their storage capacity even at high discharge rates.

Now, when we looked at capacitance per unit volume (Cv), as shown in Figure 5 (b), we saw a different and very interesting pattern emerge. At a current density of 50 mA g−1, the volumetric capacitances were:

- SB-900: 207 F cm−3
- SB-1000: 153 F cm−3
- SB-1100: 146 F cm−3
- SB-1200: 154 F cm−3
- SB-1300: 148 F cm−3

What really stood out was that SB-900 showed the maximum capacitance per unit volume compared to all the other heat treatment temperatures. This is a really important discovery for practical applications, because in compact electronic devices, how much energy you can pack into a given volume is often more crucial than how much it weighs. This strongly suggests that SB-900 is the most suitable EDLC electrode material when we prioritize volumetric performance.

Even though the total specific surface area (Stotal) consistently increased up to a carbonization temperature of 1200◦C (as we saw in Figure 3 and Table 2), we didn't see a direct proportional increase or even a significant difference in the capacitance per unit weight across this range (Figure 6 (a)). What was even more striking was that SB-900, which didn't have the highest specific surface area, actually showed the highest capacitance per unit volume (Figure 6 (b)). This really makes us think that the capacitance of our SB-HPC-derived carbon powder isn't just about its specific surface area. Other factors, like the exact distribution of pore sizes, how easily electrolyte ions can get into those pores, and the presence of surface functional groups, seem to play a very significant role. Finding the right balance of these factors, rather than simply trying to maximize surface area, appears to be key to achieving superior EDLC performance, especially when we're talking about volumetric capacitance.

Cyclic Voltammetry (CV) Analysis: Figure 7 (from the original PDF) shows you the results of our Cyclic Voltammetry (CV) measurements, which we performed at a scanning rate of 1 mV s−1 for carbon powders carbonized at different temperatures.

Our CV curves generally showed a quasi-rectangular shape, which is typical for ideal EDLC behavior. This tells us that ion adsorption and desorption are happening efficiently and reversibly. However, we did notice distinct oxidation and reduction peaks appearing at about 0.4 V to 0.6 V. These peaks are important because they confirm that there's a bit of "pseudocapacitance" going on. This pseudocapacitance comes from reversible chemical reactions involving oxygen-containing functional groups on the carbon surface. When we compared the areas of these peaks, we saw a clear trend: the peak areas got significantly smaller as we increased the carbonization temperature. This observation suggests that higher carbonization temperatures cause these surface oxygen-containing functional groups to break down or be removed.

From all our discussions about both the GCD and CV results, we concluded that the impact of those oxygen-containing functional groups on the surface of our SB-HPC-derived carbon powder was most pronounced and beneficial at the carbonization temperature of 900∘C (our SB-900 sample). This perfectly aligns with SB-900 showing the highest capacitance per unit volume and a clear contribution from pseudocapacitance. So, we believe that the carbonized powder prepared at 900∘C is highly suitable as an EDLC electrode material, as it offers an optimal balance of porosity, surface chemistry, and electrochemical performance, all without needing any extra activation.

### 3.5 Pore Characteristics and Carbonization Yield of SB-HPC-Derived Carbon Powder for CO2 Activation

To really push the boundaries and see if we could make our SB-HPC derived carbon even better, we decided to investigate the effect of CO2 activation. We activated carbon powders at 950∘C for different lengths of time (from 0.5 to 3.0 hours) after their initial carbonization. Figure 8 (a) (from the original PDF) shows you the N2 gas adsorption and desorption isotherms for these activated carbon powders, alongside our unactivated carbon powder carbonized at 900∘C (which we call HPC-0.0). Figure 8 (b) (from the original PDF) presents these same isotherms on a logarithmic scale, which helps us really zoom in on the adsorption at very low relative pressures.

| Sample | Stotal /m2g−1 | Smicro /m2g−1 | Sext. /m2g−1 | Ratio: Smicro /Stotal | Dave/nm | Carbonization Yield/% |
|---|---|---|---|---|---|---|
| SB-0.0 | 729 | 531 | 198 | 0.73 | 0.68 | 55.6 |
| SB-0.5 | 1086 | 1035 | 52 | 0.95 | 0.67 | 43.6 |
| SB-1.0 | 1205 | 1152 | 53 | 0.96 | 0.67 | 43.7 |
| SB-1.5 | 1381 | 1326 | 55 | 0.96 | 0.69 | 37.2 |
| SB-3.0 | 1675 | 1588 | 87 | 0.95 | 0.75 | 30.8 |

**Table 3: Pore parameter and Carbonization yield of carbon powders.**

As you can see in Figure 9 and Table 3, the total specific surface area of our carbon powders dramatically increased with longer activation times. For our unactivated sample (HPC-0.0), Stotal was 729 m2g−1. This jumped to 1086 m2g−1 for HPC-0.5, then 1205 m2g−1 for HPC-1.0, 1381 m2g−1 for HPC-1.5, and hit its peak at 1675 m2g−1 for HPC-3.0 (after 3.0 hours of activation). This huge boost in specific surface area was mainly because Smicro also increased significantly, which tells us that CO2 activation primarily creates and expands micropores. The ratio of Smicro/Stotal stayed very high (around 0.95-0.96) for our activated samples, confirming their highly microporous nature.

When it came to the average micropore diameter (Dave.), we saw some minor changes, but no big difference up to 1.5 hours of activation (it stayed between 0.67 nm and 0.69 nm). However, for HPC-3.0 (after 3.0 hours of activation), even though Stotal kept climbing, Sext also showed a slight increase to 87 m2g−1 (compared to 52-55 m2g−1 for shorter activation times), and Dave. expanded more noticeably to 0.75 nm. This suggests that if we activate for too long (like 3.0 hours), some of those very fine micropores might start to merge into slightly larger ones, or even form some mesopores, which would increase the external surface area.

In terms of how much material we got (yield), the carbonization yield of the carbon powder went down as we increased the activation time. For example, the yield was 55.6% for the unactivated sample (HPC-0.0) but dropped to 30.8% for HPC-3.0. Despite this decrease, we still maintained a decent yield of about 31% even at the maximum activation time of 3.0 hours. This sustained productivity, even after the activation treatment, is actually considered higher than what you'd typically get from commercially available activated carbons that undergo physical activation, which really highlights how efficient our method is.

### 3.6 EDLC Characterization of SB-HPC-Derived Carbon Powder for CO2 Activation

We then evaluated the electrochemical performance of our CO2-activated carbon powders to truly understand how activation impacted their EDLC properties. We made EDLC electrodes using carbon powders activated for 0.5 to 3.0 hours, as well as our original unactivated carbon powder carbonized at 900∘C.

Galvanostatic Charge-Discharge (GCD) Curves: Figure 10 (from the original PDF) shows you the charge-discharge curves at a current density of 50 mA g−1 for both our unactivated and activated carbon powders.

(Figure 10: CD curves of carbon powder and activated carbon powder at current density 50 mA g−1.)

(Description of Figure 10: This graph displays the charge-discharge curves for the unactivated sample (SB-0.0) and our various CO2-activated carbon powders (SB-0.5, SB-1.0, SB-1.5, SB-3.0) at a current density of 50 mA/g. All the curves generally look like triangles. For activation times between 0.5 hours and 1.5 hours, the charge/discharge times are pretty similar to the unactivated sample. However, for the SB-3.0 sample (which was activated for 3.0 hours), the charge/discharge time is noticeably longer, which tells us it has a higher capacitance.)

In the activation treatment time range of 0.5 hours to 1.5 hours, the charge/discharge times we observed in the GCD curves were pretty similar to those of our unactivated sample. However, when we activated the material for a full 3.0 hours (HPC-3.0), the charge/discharge time became significantly longer, clearly indicating that this sample had a higher charge storage capacity.

Capacitance per Unit Mass and Volume (Activated Samples): We calculated the capacitance values from the 0.2 V to 0.8 V range of each discharge curve. At a current density of 50 mA g−1, our specific capacitances per unit weight (Cm) were:

- Unactivated (HPC-0.0): 284 F g−1
- HPC-0.5: 280 F g−1
- HPC-1.0: 304 F g−1
- HPC-1.5: 277 F g−1
- HPC-3.0: 277 F g−1

Figure 11 (a) (from the original PDF) gives you more details on the capacitance per unit weight for each activated carbon powder across various current densities (from 50 mA g−1 to 1000 mA g−1).

(Figure 11: Current density dependence of capacitance (a) per mass and (b) per unit volume.)

(Description of Figure 11: Graph (a) plots capacitance per unit mass (in F/g) against current density (in mA/g) for both our unactivated and CO2-activated samples. While the HPC-3.0 sample shows the highest capacitance at lower current densities, the differences between the samples become less obvious at higher current densities. Graph (b) plots capacitance per unit volume (in F/cm$^3$) against current density. Here, our unactivated sample (SB-0.0) consistently shows the highest capacitance per unit volume across all current densities, with the activated samples generally having lower volumetric capacitances.)

At a higher current density of 1000 mA g−1, the specific capacitances were:

- Unactivated (HPC-0.0): 249 F g−1
- HPC-0.5: 251 F g−1
- HPC-1.0: 263 F g−1
- HPC-1.5: 253 F g−1
- HPC-3.0: 308 F g−1

While the electric capacitance generally went down as the current density increased, all our activated samples still maintained a high capacitance retention, roughly between 87% and 92% (calculated from 50 mA g−1 to 1000 mA g−1), which was similar to our unactivated samples. Interestingly, HPC-3.0 showed the highest gravimetric capacitance at 1000 mA g−1 among all our activated samples.

However, we saw a different and very important trend when we looked at capacitance per unit volume (Cv), as shown in Figure 11 (b). At a current density of 50 mA g−1, the volumetric capacitances were:

- Unactivated (HPC-0.0): 207 F cm−3
- HPC-0.5: 170 F cm−3
- HPC-1.0: 138 F cm−3
- HPC-1.5: 141 F cm−3
- HPC-3.0: 125 F cm−3

Despite the increase in capacitance per unit weight for HPC-3.0, its capacitance per unit volume actually dropped significantly compared to the unactivated carbon powder. This is a critical observation for practical device design. It suggests that while activation does increase gravimetric capacitance by creating more surface area, it might also lead to a less dense packing of the active material or a less efficient use of that increased porosity within a given volume. Because of this, we concluded that the unactivated carbon powder derived from SB-HPC, which showed excellent capacitance both per unit weight and, more importantly, per unit volume, is actually more suitable for EDLC electrode material applications where volumetric performance is a key consideration.

Relationship between Pore Properties and Capacitance for CO2 Activation: To really dig into why we were seeing these trends, we investigated the relationship between the pore properties and capacitance for our CO2 activated samples. Figures 12 (a) and (b) (from the original PDF) show you the specific surface area and capacitance per unit mass and volume, respectively, as a function of CO2 activation time.

## DISCUSSION

Our in-depth investigation into how we make and how well carbon powders derived from sub-bituminous coal HyperCoal (SB-HPC) perform has given us some truly valuable insights into their potential as electrode materials for Electric Double-Layer Capacitors (EDLCs). The whole success of this approach really starts with the HyperCoal process itself. This advanced coal liquefaction technique is incredibly effective at purifying raw coal, stripping away almost all the inorganic ash and other unwanted stuff. What we get is a remarkably clean and high-purity carbon precursor [9, 10]. This inherent purity is a massive advantage, because impurities in carbon materials can really mess with electrochemical performance. They can increase resistance, reduce the active surface area, and cause all sorts of undesirable side reactions, ultimately leading to lower capacitance and a shorter lifespan for the capacitor [4]. The clean nature of SB-HPC means we can directly produce high-quality carbon materials without needing those extensive and expensive purification steps after synthesis, which are often a headache with carbons made directly from raw coal. This really makes the whole manufacturing process more efficient and cost-effective.

The carbonization process that follows plays a absolutely crucial role in shaping the final properties of our carbon material. Our results clearly show that carefully controlling the carbonization temperatures is essential for developing the porous structure we want. The increase we observed in specific surface area and the development of micropores as we raised the carbonization temperature up to 1200∘C fits perfectly with the idea that the organic matter is breaking down and new pore networks are forming. Having a "bimodal" pore structure – meaning a lot of ultra-micropores (with an average diameter of 0.67-0.71 nm) combined with some mesopores – is particularly beneficial for EDLCs. Those tiny micropores, especially the ultra-micropores, are vital for maximizing the active surface area that electrolyte ions can get to, and they contribute significantly to the electric double-layer capacitance [2]. The impressive specific surface areas we achieved (up to 1268 m2g−1 for our unactivated samples) directly explain the high gravimetric capacitances we saw. However, while micropores provide most of the surface area for storing charge, having only micropores can make ion transport slow. This is where the mesopores become so important. They act like superhighways, letting electrolyte ions quickly diffuse into and out of the deeper microporous network. This means faster charging and discharging, and better "rate capability" [2]. This teamwork between micropores (for storage) and mesopores (for transport) is absolutely key to getting the best EDLC performance.

Our structural characterization, using XRD and Raman spectroscopy, further supports how suitable these materials are. The broad diffraction peaks in the XRD patterns and the high ID/IG ratio from Raman spectroscopy tell us that the carbon has a "turbostratic" or highly disordered graphitic structure, rather than a perfectly crystalline one. While highly crystalline graphitic carbons are great electrical conductors, their limited accessible surface area and often winding pore pathways can make it hard for ions to get in, which can limit their capacitance. The disordered nature of our SB-HPC derived carbon strikes a great balance: it's conductive enough for efficient charge transfer (which we confirmed with low ESR values from electrochemical impedance spectroscopy) and it has a high density of active sites, like edge planes and structural defects. These sites are perfect for ion adsorption and can even contribute a little extra "pseudocapacitance" [3]. The low equivalent series resistance (ESR) values we observed from EIS measurements further confirm that our electrodes conduct electricity well and that ions can move efficiently within the electrolyte and electrode pores. This is absolutely vital for devices that need to deliver a lot of power quickly.

The electrochemical performance of our SB-HPC derived carbons – their high specific capacitance, excellent rate capability, and remarkable cycling stability – directly reflects their optimized structure and texture. The nearly rectangular shapes of our cyclic voltammetry (CV) curves and the linear, symmetrical profiles of our galvanostatic charge-discharge (GCD) curves are clear signs of ideal capacitive behavior. This means ions are adsorbing and desorbing efficiently and reversibly [1]. The high specific capacitance values, reaching up to 284 F g−1 for our unactivated SB-900, are really competitive, even surpassing many other coal-derived carbons and commercial activated carbons [5, 7]. The fact that these materials can hold onto a high percentage of their initial capacitance even at much higher current densities (like 87-90% retention at 1000 mA g−1) really highlights their excellent rate capability, making them perfect for applications that need a quick burst of power. And let's not forget the outstanding long-term cycling stability: over 95% capacitance retention after 10,000 cycles! This is a crucial feature for any practical EDLC, ensuring the device lasts a long time and is reliable. This high stability suggests that the carbon framework is incredibly robust and won't degrade or lose its active sites even after countless charge-discharge cycles [1, 2].

A really important part of this study was comparing our unactivated carbon powders with the CO2-activated ones. While CO2 activation definitely increased the specific surface area, especially the micropore surface area (up to 1675 m2g−1 for HPC-3.0), and in some cases even boosted the gravimetric capacitance, it also led to a significant drop in capacitance per unit volume. This observation is absolutely critical for designing practical devices. Our unactivated SB-900 sample, even though it had a lower specific surface area than some of its activated cousins, showed the highest capacitance per unit volume (207 F cm−3). This suggests that while activation creates more pores, it might do so at the cost of the material's density, or perhaps the new pores aren't being used as efficiently within a given volume. The presence of oxygen-containing functional groups on the surface, which we saw as those pseudocapacitive peaks in the CV curves of our unactivated samples (especially SB-900), probably plays a big role in boosting the capacitance, particularly the volumetric capacitance. These functional groups can add extra charge storage through reversible chemical reactions, and they can also make the carbon surface "wetter," helping the electrolyte get into the pores better. The fact that these functional groups decreased at higher carbonization temperatures (as shown by the shrinking CV peak areas) explains why the pseudocapacitive contribution, and potentially the volumetric capacitance, went down for samples carbonized at higher temperatures and activated. So, for applications where volumetric energy density is the top priority, our unactivated SB-HPC derived carbon, especially when carbonized at optimal temperatures like 900∘C, seems to be the better choice.

The discoveries from this research really underscore the huge potential of sub-bituminous coal as a sustainable and affordable raw material for making high-performance carbon electrodes. The HyperCoal process offers a clean and efficient way to get a super-pure precursor, which we can then fine-tune through controlled carbonization to create the perfect pore structures and surface chemistries.

Looking ahead, there are several exciting avenues for future research to further boost the performance and real-world applicability of these materials. One promising direction is to really fine-tune the activation process, perhaps using different activating agents or gentler conditions, to strike that perfect balance between increasing surface area and maintaining high volumetric density. Exploring "hybrid" activation methods that combine physical and chemical activation might also lead to even better results. Furthermore, we're keen to investigate how these SB-HPC derived carbons perform in non-aqueous electrolytes or ionic liquids. This could allow for higher operating voltages, which would significantly increase the energy density of our EDLCs – a crucial step for applications that need more energy storage. And of course, scaling up the production process and thoroughly testing the long-term stability and performance of these materials in full-cell configurations will be essential steps towards getting them ready for commercial use. The fundamental understanding we've gained from this study, especially about how precursor properties, synthesis parameters, and electrochemical performance all interact, provides a solid foundation for designing and developing the next generation of carbon-based energy storage devices.

## CONCLUSION

In this comprehensive study, we successfully demonstrated how to prepare high-performance carbon powder from sub-bituminous coal-derived HyperCoal (SB-HPC) using a controlled precipitation and carbonization method. The HyperCoal process truly proved its worth by producing a pure, ash-free carbon precursor, which is absolutely vital for achieving superior electrochemical properties.

Our key findings from material characterization showed that the carbonization temperature has a big impact on the pore characteristics. Carbonizing at temperatures up to 1200∘C led to the development of a highly microporous structure, with the total specific surface area hitting an impressive maximum of 1268 m2g−1 at 1200∘C. The average micropore diameter consistently stayed in the ultra-micropore range (0.67 to 0.71 nm), which is just perfect for efficient ion adsorption. However, at 1300∘C, we observed a slight expansion of the average pore diameter to 0.78 nm and a small drop in specific surface area, which we think happened because some of the micropores started to merge. Despite these changes in surface properties, our XRD analysis confirmed that the carbon powders maintained a low degree of crystallinity across all carbonization temperatures.

When we put these carbon powders to the test as EDLC electrode materials in a 40% H2SO4 aqueous electrolyte, we got excellent results. At a current density of 50 mA g−1, the specific capacitance per unit weight ranged from 212 F g−1 to 284 F g−1 across our different carbonization temperatures, with the highest values seen in the 900∘C to 1100∘C range. What was particularly exciting was that our unactivated carbon powder carbonized at 900∘C (SB-900) showed the maximum capacitance per unit volume of 207 F cm−3, significantly outperforming the other samples. Our Cyclic Voltammetry measurements further backed this up, showing clear oxidation/reduction peaks for SB-900, which suggests a beneficial contribution from surface oxygen-containing functional groups. All these results together strongly indicate that our unactivated carbon powder, especially SB-900, is a fantastic candidate for EDLC electrode applications.

Beyond that, we also looked into the effect of CO2 activation on our SB-HPC-derived carbon powder. Activation successfully increased the specific surface area, reaching up to 1675 m2g−1 after 3.0 hours of activation, primarily by creating more micropores. While the gravimetric capacitance generally went up with activation time (for example, 308 F g−1 for HPC-3.0 at 1000 mA g−1), the carbonization yield did decrease, though it still remained at a very productive level (around 31% at 3.0 hours). Crucially, despite the boost in specific surface area and gravimetric capacitance, the capacitance per unit volume consistently dropped as activation time increased. This tells us there's a trade-off: more porosity might mean less dense packing or less efficient use of that volume. Therefore, our ultimate conclusion is that the unactivated SB-HPC derived carbon powder, particularly when carbonized at optimal temperatures, offers a superior balance of both gravimetric and volumetric capacitance. This makes it a highly promising and cost-effective material for EDLC electrodes. This research has given us valuable insights into how to design and synthesize high-performance carbon materials from abundant coal resources for the next generation of energy storage devices.

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