

Investigating Two Activation Strategies for Sustainable Carbon Synthesis: A Characterization Approach

Mohamed Beshir

Beni-Suef University

Nabila Shehata

`nabila.shehata@psas.bsu.edu.eg`

Beni-Suef University

Safaa El-Nahas

Qena university

Research Article

Keywords: Activated carbon, sludge waste valorization, H_2SO_4 activation, KOH activation, sustainable materials, circular economy

Posted Date: September 7th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10820928/v1>

License:   This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Additional Declarations: No competing interests reported.

Investigating Two Activation Strategies for Sustainable Carbon Synthesis: A Characterization Approach

Mohamed A. Beshir¹, Nabila Shehata^{1*}, Safaa El-Nahas^{2*}

¹Environmental Science and Industrial Development Department, Faculty of Postgraduate Studies for Advanced Sciences, Beni-Suef University, Beni-Suef 62511, Egypt

²Chemistry Department, Faculty of Science, Qena University, Qena 83523, Egypt

Correspondence e-mails: Nabila Shehata nabila.shehata@psas.bsu.edu.eg; Safaa El-Nahas safaa.elnahas@sci.svu.edu.eg, Orcid: 0000-0002-8185-1338

Abstract

The conversion of waste sludge into high value activated carbon materials represents a promising approach for waste valorization and sustainable materials development. This comprehensive study investigates the morphological transformations and elemental composition changes in sludge-derived carbon materials subjected to different chemical activation strategies. Initially, controlled pyrolysis at 800 °C in a nitrogen environment was used to develop raw carbon (RC), which was then chemically activated using H₂SO₄ at 450 °C and KOH at 800 °C to produce materials with fundamentally distinct properties. Comprehensive characterization analysis revealed fundamental differences between activation methods and their resulting material properties. KOH activation successfully developed hierarchical porous structures with distinctive rod-like and plate-like morphologies, developing extensive mesopores and macropores while retaining significant potassium content (31.31 wt%) that provides beneficial basic surface functionality. In contrast, H₂SO₄ activation produced fine spherical particles with extensive agglomeration patterns, successfully incorporating sulfur-containing functional groups (18.13 wt% S) that significantly enhance surface acidity and hydrophilicity. Advanced EDX elemental mapping techniques demonstrated uniform spatial distribution of activating agents

throughout the entire carbon matrix, confirming successful penetration and chemical reaction processes. Both activation processes effectively removed ash components while simultaneously developing enhanced porosity and introducing valuable heteroatom functionalities. This research demonstrates the technical feasibility of producing task-specific carbon materials from waste streams, contributing to circular economy principles and sustainable synthesis.

Keywords: Activated carbon; sludge waste valorization; H₂SO₄ activation; KOH activation; sustainable materials; circular economy

1. Introduction

The exponential growth of global population and rapid industrialization have resulted in unprecedented generation of organic waste streams, with sewage sludge represents one of the world's most difficult waste management issues [1]. Municipal wastewater treatment plants generate millions of tons of sludge annually, containing complex mixtures of organic matter, nutrients, and potentially harmful contaminants that require careful handling and disposal [2]. Traditional sludge management approaches including landfilling, incineration, and anaerobic digestion. However, agricultural application face increasing regulatory restrictions and environmental concerns, driving the urgent need for sustainable valorization strategies that can transform this waste burden into valuable materials [3].

Activated carbon materials have emerged as versatile adsorbents with exceptional surface area, diverse surface functions and adjustable pore structures making them indispensable for numerous applications from energy storage, water remediation and catalysis to air filtration [4]. The global activated carbon market, valued at several billion dollars annually, relies heavily on non-renewable precursors such as coal, petroleum coke, and coconut shells, raising concerns about resource sustainability and supply security [5]. This economic and environmental context has stimulated intensive research into alternative

precursor materials derived from renewable biomass and organic waste streams, with sewage sludge representing a particularly attractive feedstock due to its abundance, consistent availability, and inherent carbon content [6].

The synthesis of activated carbon from waste materials involves two critical stages: initial carbonization through pyrolysis to develop a carbon framework, followed by activation processes that develop porosity and introduce functional groups essential for specific applications [7]. Chemical activation has proven superior to physical activation methods due to its ability to achieve higher surface areas, better pore structure control, and enhanced functional group incorporation at relatively lower temperatures [8]. Among chemical activation agents, potassium hydroxide (KOH) and sulfuric acid (H_2SO_4) represent two fundamentally different approaches that produce materials with contrasting properties and application potentials [9].

Potassium hydroxide activation operates through complex mechanisms involving potassium intercalation, selective carbon gasification, and metallic potassium reduction reactions that develop hierarchical porous structures with predominantly basic surface characteristics [10]. The process typically requires high activation temperatures (700-900 °C) and results in materials with exceptional surface areas which in some cases can be more than 2000 m^2/g , rendering them particularly appropriate for gas adsorption applications such as CO_2 capture and hydrogen storage [11]. The retention of potassium species within the carbon framework can provide additional functionality, particularly in electrochemical applications where potassium acts as an internal electrolyte enhancing ionic conductivity [12].

In contrast, sulfuric acid activation proceeds through dehydration, sulfonation, and controlled oxidation mechanisms. This introduces extensive oxygen and sulfur-containing functional groups while developing primarily microporous structures [13]. This activation route operates at moderate temperatures (400-500 °C) and produces materials with strong acidic surface characteristics, high hydrophilicity, and superior ion exchanging capacity,

which makes them perfect for heavy metal removal [14]. The incorporation of sulfonic acid groups ($-\text{SO}_3\text{H}$) significantly enhances the material's affinity for polar molecules and provides active sites for catalytic applications [15].

Comprehensive characterization of activated carbon materials requires advanced analytical techniques to understand the complex relationships between synthesis conditions, structural properties, and performance characteristics [16]. X-ray diffraction (XRD) analysis provides crucial information about crystallographic structure, degree of graphitization, and interlayer spacing modifications induced by different activation treatments [17]. Combining energy-dispersive X-ray spectroscopy (EDX) with scanning electron microscopy (SEM) allows for a thorough examination of morphological changes and elemental distribution patterns that are directly related to material properties [18]. XPS -technique (X-ray photoelectron spectroscopy) provides a unique understanding of surface chemical states and functional group distributions, while differential scanning calorimetry (DSC) reveals thermal stability characteristics essential for high-temperature applications [19].

Despite extensive research on activated carbon synthesis, significant knowledge gaps remain regarding the specific mechanisms governing morphological evolution and elemental redistribution during chemical activation of sludge-derived precursors [20]. Most previous studies have focused on traditional precursor materials or single activation methods, with limited systematic comparison of how different chemical activation strategies influence the final material properties [21]. Furthermore, the complex composition of sewage sludge, containing both organic and inorganic components, introduces additional variables that can significantly affect activation outcomes and require thorough investigation [22]. The relationship between activation conditions, resulting material characteristics, and performance in specific applications remains poorly understood, particularly for waste-derived precursors with heterogeneous compositions [23]. Current literature lacks comprehensive studies that systematically correlate

morphological transformations, elemental composition changes, and surface chemistry modifications with activation mechanisms, limiting the ability to design optimal synthesis protocols for specific applications [24]. Additionally, the environmental implications and sustainability metrics of different activation approaches require thorough evaluation to support the development of truly sustainable carbon materials [25].

Hence, this comprehensive study addresses these knowledge gaps by conducting detailed morphological and elemental characterization of chemically activated carbon materials derived from sewage sludge using two distinct activation strategies. The research employs advanced analytical techniques including SEM-EDX, XRD, XPS, and DSC analysis to establish clear process-structure-property relationships and provide fundamental insights into activation mechanisms. The findings contribute to the scientific understanding of waste valorization processes while supporting the development of sustainable carbon materials with tailored properties for specific environmental and energy applications.

2. Materials and Methods

2.1 Carbon Synthesis via Raw Material Processing

The solid sludge samples were initially collected, homogenized, and it was heated for 24 h at 105 °C to eliminate moisture and produce uniform feedstock characteristics. The dried sludge was then ground and sieved through a 200-mesh screen (with an opening size of 75 µm, using a standard test sieve from Retsch GmbH, Germany) to obtain uniform particle size distribution, which ensures consistent heat transfer and pyrolysis kinetics during the thermal treatment process. The homogenized sludge served as the precursor material for carbon synthesis through controlled pyrolysis under inert atmosphere. The carbon synthesis was conducted through high-temperature pyrolysis in a horizontal tubular furnace (Carbolite STF 16/180, United Kingdom) equipped with precise temperature control and

gas flow regulation systems. Approximately 50 g of the prepared sludge was placed in alumina crucibles and subjected to thermal decomposition under continuous nitrogen flow (99.99% purity, 150 mL/min) to maintain an oxygen-free environment and prevent combustion. The samples were heated from room temperature to 800 °C at a controllable rate of 5 °C per minute during the pyrolysis process to minimize thermal shock and ensure uniform carbonization throughout the material matrix. For three hours, the temperature kept at 800 °C to achieve complete devolatilization of organic compounds, removal of heteroatoms, and formation of stable carbon structures through dehydration, decarboxylation, and aromatization reactions. After the pyrolysis period, samples were cooled naturally to room temperature under continuous nitrogen purging to prevent oxidation of the newly formed carbon material.

2.2 Chemical Activation Procedures

Two distinct chemical activation methods were employed to alter the RC material's pore shape, surface chemistry, and adsorption characteristics. Chemical activation involves the introduction of activating agents that develop additional porosity through selective gasification reactions and chemical modifications at elevated temperatures, resulting in materials with enhanced surface area and tailored surface functionality.

2.2.1 Potassium Hydroxide (KOH) Activation

For the chemical activation procedure, potassium hydroxide (KOH) was chosen due to its high efficiency in enhancing the surface properties of activated carbon during pyrolysis. Raw sewage sludge was collected from Beni Suef Wastewater Treatment Plant and heated to eliminate water content in an oven that is set between 100 and 105 °C. Smaller particles with a consistent diameter of 0.75 mm were produced by grinding the dried sewage sludge to ensure consistent activation throughout the material. Known weights of the dried sewage sludge were systematically mixed with ground potassium hydroxide in a mass ratio of sewage sludge to KOH (1:1) for insight into how the final carbon characteristics are affected by the concentration of the activating agent. To facilitate intimate contact between

the precursor material and the activating agent, distilled water (10 mL) was added to each mixture to form a uniform paste with homogeneous distribution of KOH throughout the sludge matrix. The resulting paste was dried at room temperature for 24 h to allow gradual removal of excess moisture while maintaining the uniform distribution of the activating agent. Following the drying period, pyrolysis was conducted in a tube furnace under continuous nitrogen flow maintained at 100 mL/min to develop an inert atmosphere and prevent oxidation during the thermal treatment. The adsorbents produced through this activation process were then collected and stored in sealed containers for subsequent characterization studies. The washing of KAC with hydrochloric acid offers several significant advantages that enhance the quality and performance of the final product. The acid treatment ensures complete removal of residual KOH and potassium salts formed during the activation process.

2.2.2 Sulfuric Acid (H₂SO₄) Activation

A 2 L glass beaker with heating and stirring capabilities was used for the sulphuric acid activation process. To guarantee full impregnation of the precursor material with the activating agent, 200 g of dried sludge and 200 mL of concentrated sulphuric acid were completely combined. After that, the mixture was heated to 200 °C for an hour while being constantly stirred to promote uniform activation throughout the material. During this heating period, water present in the mixture gradually evaporated, causing the initially liquid mixture to transform into a slurry-like consistency and eventually begin to solidify as carbonization progressed. At the point when the mixture exhibited significant solidification, mechanical agitation was discontinued to prevent damage to the forming carbon structure, while heating continued to drive the activation reactions to completion. The thermal treatment proceeded until the material transformed into a carbon-like substance with characteristic black color and porous texture. To further promote the activation process and enhance pore development, a 10 mL syringe was used to gently inject tiny quantities of distilled water into the carbonized substance which generated steam

and facilitated additional gasification reactions within the carbon matrix. The resultant carbon material was allowed to gradually cool to room temperature under ambient circumstances once the activation process was finished. After cooling, the carbon received an extensive washing process that started with several rinses with distilled water to get rid of excess acid and water-soluble contaminants. The carbon was steeped in a diluted sodium hydroxide solution for half an hour to eliminate any residual acidity and acid-soluble mineral components. After the neutralization step, the material underwent a second thorough washing with distilled water to remove residual sodium hydroxide and achieve a neutral pH. The washed carbon was then dried to constant weight and stored in a tightly closed container to prevent moisture absorption and maintain material properties prior to characterization studies.

2.3 Methods of Characterization

2.3.1 Scanning electron microscopy equipped with EDX

Scanning electron microscopy analysis was performed for morphological and elemental Analysis using a ZEISS EVO 18 instrument (Germany) operated at accelerating voltages of 15-20 kV. Energy dispersive X-ray spectroscopy (EDX) analysis was conducted using an Oxford Instruments X-MaxN 80 EDS detector (United Kingdom). Peak identification and quantitative analysis were performed using the Oxford AZtec software package.

2.3.2 X-ray diffraction (XRD) analysis

X-ray diffraction (XRD) analysis was performed to investigate the crystallographic structure, degree of graphitization, and interlayer spacing of the tested materials using a Rigaku MiniFlex 600 diffractometer (Japan) equipped with Cu K α radiation ($\lambda = 1.5406$ Å) operating at 40 kV and 15 mA. Diffraction patterns were recorded over a 2θ range of $10-80^\circ$ with a scan rate of $2^\circ/\text{min}$, step size of 0.02° , and count time of 1 second per step to ensure adequate signal-to-noise ratio. The interlayer spacing (d_{002}) was calculated using

Bragg's equation ($n\lambda = 2d \sin\theta$), while crystallite dimensions including stack height (Lc) and crystallite diameter (La) were determined using the Scherrer equation with appropriate shape factors ($K = 0.89$ for Lc and $K = 1.84$ for La). These structural parameters provide quantitative information about the degree of structural ordering, and the effect of different activation treatments on the carbon crystalline structure.

2.3.4 X-ray photoelectron spectroscopy (XPS)

X-ray photoelectron spectroscopy (XPS) was performed using a Thermo Scientific K-Alpha+ instrument (United Kingdom) equipped with a monochromatic Al K α X-ray source (1486.6 eV, 400 μ m spot size) under ultra-high vacuum conditions (base pressure $<5 \times 10^{-9}$ Torr) to determine surface elemental composition, chemical oxidation states, and functional group distribution. Survey spectra were acquired over a binding energy range of 0-1350 eV with pass energy of 200 eV and energy step of 1 eV to identify all elements present on the surface.

2.3.5 Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) was conducted using a TA Instruments Q2000 calorimeter (USA) to investigate thermal transitions, oxidation behavior, and thermal stability of the carbon materials under controlled atmospheric conditions. Samples (5-10 mg) were sealed in aluminium pans and analyzed over a temperature range of 25-600 °C at heating rates of 10 °C/min under both nitrogen (50 mL/min) and air (50 mL/min) atmospheres.

3. Results and Discussion

Total Organic Carbon (TOC) measurement is a method widely used to evaluate the organic matter in wastewater sludge. Besides, the morphological and elemental characterization of the three carbon materials reveals distinct structural and compositional changes induced by

different activation treatments. The comprehensive SEM-EDX analysis demonstrates how activation chemistry directly influences the final properties of sludge-derived carbon materials.

3.1 Total Organic Carbon (TOC) measurement and activation yield

Soluble TOC concentrations can be used as a reliable indication of sludge organic matter discharge. Table 1 demonstrates that while treating the sludge with H₂SO₄ the TOC values reduced from 2.91 to 2.14 g/L, KOH reduced the TOC value from 2.91 to 1.69 g/L.

Table 1. TOC data for Raw and modified samples

Sample No.	RC (mg/L)	KOH (mg/L)	H ₂ SO ₄ (mg/L)
1	2850	1680	2120
2	3120	1890	2340
3	2760	1520	1980
Mean ± SD	2910 ± 189	1697 ± 186	2147 ± 181

Activation yield was defined as:

$$\text{Yield (\%)} = (\text{mass of activated carbon} / \text{mass of raw precursor}) \times 100 \quad (1)$$

The results showed that the yield (%) of SAC (70.61%) was higher than that of KAC (64.52%).

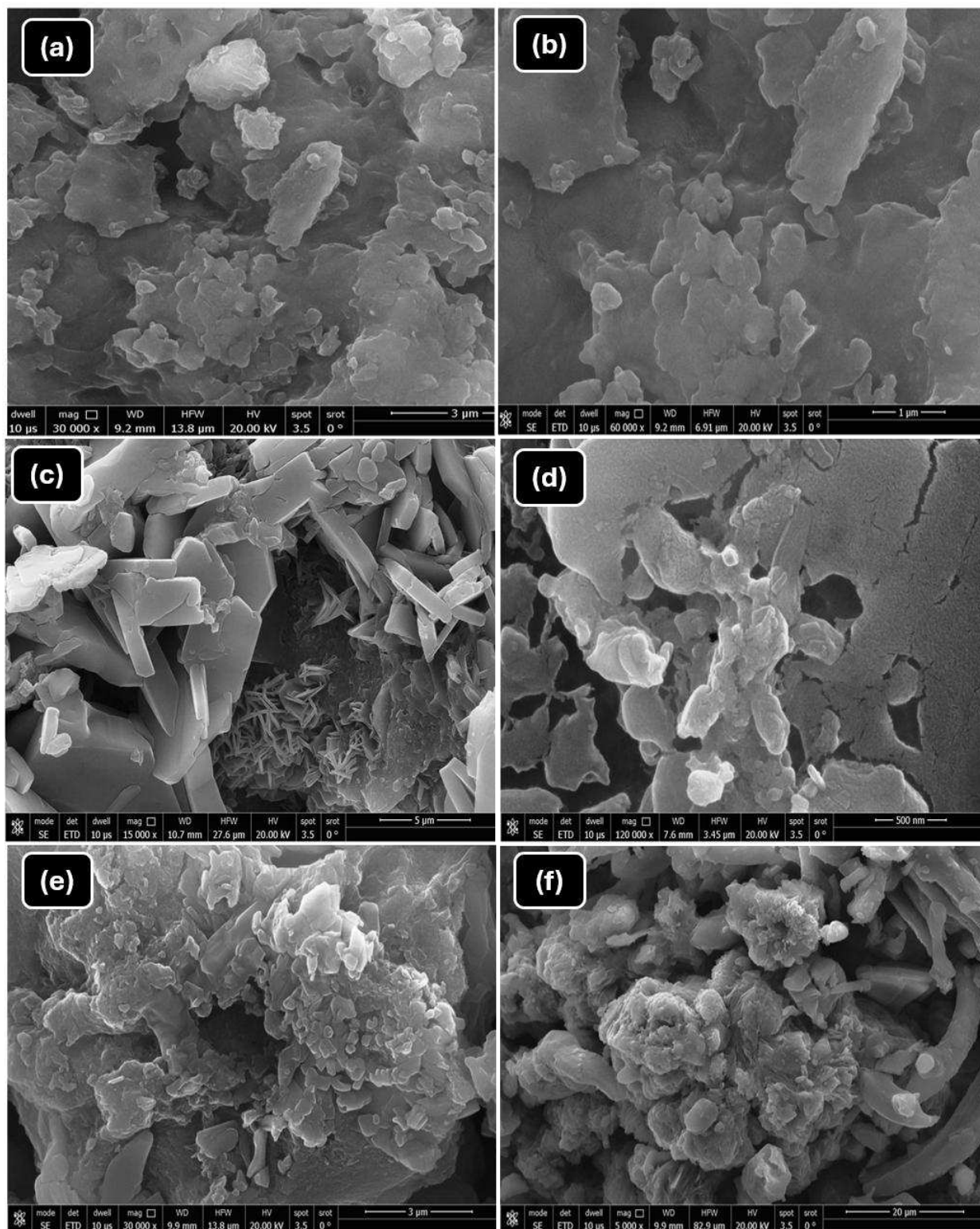
3.1 Morphological Transformations and Activation Mechanisms

The RC sample exhibits a highly heterogeneous granular morphology with irregular particle shapes and sizes ranging from approximately 500 nm to 5 µm, as shown in Figure (1a). The particles display angular, fragmented characteristics typical of pyrolyzed organic matter, with evident surface roughness and textural variations. At higher magnification (Fig. 1b), the surface topography reveals a moderately porous structure with visible micro-cracks and surface irregularities, indicating incomplete structural development during the pyrolysis process at 800 °C under nitrogen atmosphere. The particle agglomeration is

moderate, with individual particles maintaining distinct boundaries while forming loose clusters. This morphology reflects the thermal decomposition of organic components in the sludge during pyrolysis, where volatile matter removal develops initial porosity without significant structural reorganization.

The KOH activation process fundamentally transforms the carbon structure through intercalation and gasification mechanisms, as demonstrated in Figure 1c. The activated sample shows dramatically different morphology compared to RC, developing well-defined rod-like and plate-like structures with dimensions ranging from 1-3 μm in length and 200-500 nm in width. At higher magnification (Fig. 1d), the surface reveals an extensively developed porous network with visible mesopores and macropores, indicating successful structural reorganization during the alkaline activation process. The particles exhibit sharp edges and well-defined crystalline facets, suggesting partial graphitization promoted by the catalytic effect of potassium species. This morphological transformation results from potassium intercalation between carbon layers, followed by selective carbon gasification at high temperature, developing the hierarchical porous architecture crucial for applications requiring high surface area such as supercapacitors or gas adsorption.

In contrast, the H_2SO_4 activation produces a distinctly different morphological pattern characterized by fine particulate morphology with extensive agglomeration, as illustrated in Figure 1e. The particles range in size from 100 to 800 nm and are primarily spherical to sub-spherical, forming dense clusters and aggregates. The surface appears smoother compared to both RC and KAC samples, with less pronounced surface roughness, reflecting a different activation mechanism involving dehydration and carbonization at lower temperature (450 $^\circ\text{C}$). Sulfuric acid acts as both a dehydrating agent and a sulfonating reagent, developing microporous structures and introducing sulfur-containing functional groups. The observed particle agglomeration results from hydrogen bonding and electrostatic interactions between sulfonic acid groups (Fig. 1f), which is characteristic of sulfonated carbon materials and indicates successful surface functionalization.



287

288 *Figure 1. Combined SEM micrographs of RC (a& b) , KAC (c& d), and SAC (e& f),*
 289 *showing the contrasting morphologies and pore development observed after activation*

3.2 Elemental Composition and Chemical Modifications

The elemental composition analysis reveals the effectiveness of different activation strategies in modifying the carbon's chemical nature. The RC sample, analyzed over a bulk area, shows major elements including oxygen (57.8 wt%, 71.69 at%), silicon (28.73 wt%, 20.3 at%), and aluminum (3.55 wt%, 2.61 at%), as presented in Table 2. Minor elements comprise sulfur (2.59 wt%, 1.6 at%), calcium (2.93 wt%, 1.45 at%), phosphorus (2.25 wt%, 1.44 at%), potassium (0.9 wt%, 0.46 at%), and iron (1.25 wt%, 0.44 at%). The high oxygen content reflects the presence of oxygen-containing functional groups and possible oxidation during pyrolysis, while the significant silicon and aluminum concentrations indicate the presence of inorganic ash components originating from the sludge feedstock. These ash components, while potentially detrimental to electrochemical performance, may contribute to the mechanical stability of the carbon framework.

KOH activation dramatically alters the elemental composition, as shown in Table 2, with oxygen remaining the dominant element (60.62 wt%, 78.34 at%) but accompanied by an exceptionally high potassium content (31.31 wt%, 16.56 at%). Other detected elements include calcium (2.69 wt%, 1.39 at%), silicon (1.5 wt%, 1.11 at%), magnesium (1.41 wt%, 1.2 at%), aluminum (1.25 wt%, 0.96 at%), and iron (1.22 wt%, 0.45 at%). The substantial potassium concentration indicates either incomplete removal during washing or strong chemical interactions between potassium species and the carbon framework. This retention could be advantageous for certain applications, as potassium can serve as an internal electrolyte in supercapacitor applications. The reduced concentrations of silicon and aluminum compared to the RC confirm that alkaline activation effectively removes ash components through dissolution mechanisms, simultaneously purifying and activating the carbon material.

The sulfuric acid-activated carbon exhibits a unique elemental profile, as detailed in Table 2, with oxygen (53.27 wt%, 65.81 at%) as the major component, accompanied by significant sodium (23.63 wt%, 20.31 at%) and sulfur (18.13 wt%, 11.17 at%)

concentrations. Minor elements include silicon (1.13 wt%, 0.8 at%), aluminum (1.16 wt%, 0.85 at%), calcium (0.76 wt%, 0.38 at%), and iron (1.91 wt%, 0.68 at%). The high sulfur content confirms successful incorporation of sulfur-containing functional groups during acid activation, significantly enhancing the material's hydrophilicity and ion-exchange capacity. The unexpected presence of sodium likely originates from the neutralization process during washing, where sodium hydroxide may have been used to adjust pH, resulting in ion exchange with protons from sulfonic acid groups. The relatively low ash content compared to RC indicates effective acid leaching of inorganic impurities during the activation process.

Table 2. EDX elemental composition of RC, KAC, and SAC

Element	RC wt.%	RC at.%	KAC wt.%	KAC at.%	SAC wt.%	SAC at.%
O K	57.80	71.69	60.62	78.34	53.27	65.81
Si K	28.73	20.30	1.50	1.11	1.13	0.80
Al K	3.55	2.61	1.25	0.96	1.16	0.85
Ca K	2.93	1.45	2.69	1.39	0.76	0.38
S K	2.59	1.60	-	-	18.13	11.17
P K	2.25	1.44	-	-	-	-
K K	0.90	0.46	31.31	16.56	-	-
Mg K	-	-	1.41	1.20	-	-
Fe K	1.25	0.44	1.22	0.45	1.91	0.68
Na K	-	-	-	-	23.63	20.31
Total	100.0	100.0	100.0	100.0	100.0	100.0

3.3 Spatial Elemental Distribution and Activation Homogeneity

330 The EDX elemental mapping provides crucial insights into the homogeneity and
331 effectiveness of the activation processes. EDX spectrum of the RC in Figure 2a showing
332 an elemental composition analysis conducted at 30 kV accelerating voltage. The spectrum
333 reveals silicon (Si) as the dominant element with the highest intensity peak at 1.74 keV,
334 followed by oxygen (O) at 0.53 keV. Minor elements detected include aluminum (Al) at
335 1.49 keV, sulfur (S) at 2.31 keV, phosphorus (P) at 2.01 keV, potassium (K) at 3.31 keV,
336 calcium (Ca) with characteristic peaks at 3.69 and 4.01 keV, and iron (Fe) showing $K\alpha$ and
337 $K\beta$ peaks at 6.40 and 7.06 keV respectively. The analysis was performed over a full area
338 with 10 seconds live time, demonstrating the heterogeneous inorganic composition typical
339 of sludge-derived carbon materials. Figure 2a (inset) illustrates the spatial distribution of
340 major elements across the RC sample, demonstrating relatively uniform distribution of
341 oxygen (22% coverage) and silicon (56% coverage) throughout the analyzed area, with
342 aluminum (6% coverage) showing more localized concentrations. Phosphorus, sulfur, and
343 potassium exhibit scattered distribution patterns with concentrations ranging from 2-5%
344 coverage each, while calcium shows discrete localization (6% coverage) in specific
345 regions, possibly associated with calcium-containing mineral phases. The uniform
346 distribution of oxygen and silicon suggests homogeneous incorporation of these elements
347 during the pyrolysis process, while the heterogeneous distribution of other elements
348 reflects the complex mineralogy of the original sludge feedstock.

349 EDX spectrum of the KAC sample (Fig. 2b) shows the elemental composition analysis
350 conducted at 30 kV accelerating voltage. The spectrum is dominated by potassium (K) with
351 an exceptionally high intensity peak at 3.31 keV, demonstrating significant retention of the
352 KOH activating agent. Oxygen (O) appears as the second most prominent element at 0.53
353 keV, while silicon (Si) at 1.74 keV shows dramatically reduced intensity compared to the
354 RC, indicating effective ash removal during alkaline activation. Minor elements detected
355 include magnesium (Mg) at 1.25 keV, aluminum (Al) at 1.49 keV, calcium (Ca) with peaks
356 at 3.69 and 4.01 keV, and iron (Fe) showing characteristic $K\alpha$ and $K\beta$ peaks at 6.40 and

7.06 keV respectively. The analysis was performed over a full area with 10 seconds live time, confirming successful KOH activation and substantial elemental composition modification compared to the precursor material.

The elemental mapping of the KAC, presented in Figure 2b (inset), reveals remarkably uniform distribution of potassium (72% coverage) across the entire mapped area, indicating successful penetration and interaction of the KOH activating agent throughout the carbon matrix. This uniform distribution confirms the effectiveness of the activation process and suggests that the hierarchical porous structure facilitates deep penetration of the activating agent. Oxygen distribution (13% coverage) shows good correlation with potassium-rich regions, supporting the formation of potassium-oxygen complexes or hydroxide species during activation. Other elements including aluminum (3% coverage), silicon (4% coverage), calcium (7% coverage), and iron (1% coverage) exhibit more localized distributions, indicating selective concentration or preferential retention in specific structural domains during the activation process.

EDX spectrum of the SAC sample showing elemental composition analysis conducted at 30 kV accelerating voltage (Fig. 2c). The spectrum is dominated by oxygen (O) with the highest intensity peak at 0.53 keV, indicating extensive oxidation during acid activation. Sulfur (S) appears as a highly prominent peak at 2.31 keV, confirming successful incorporation of sulfur-containing functional groups from H₂SO₄ activation. Sodium (Na) shows significant presence at 1.04 keV, likely resulting from neutralization procedures during sample processing. Silicon (Si) at 1.74 keV exhibits substantially reduced intensity compared to the RC, demonstrating effective acid leaching of silicate components. Minor elements detected include aluminum (Al) at 1.49 keV, calcium (Ca) with characteristic peaks at 3.69 and 4.01 keV, and iron (Fe) showing K α and K β peaks at 6.40 and 7.06 keV respectively. The analysis confirmed successful sulfuric acid activation and extensive surface functionalization with heteroatoms. Figure 2c (inset) demonstrates the elemental distribution in the sulfuric acid-activated carbon, showing highly uniform sulfur

384 distribution (38% coverage) throughout the sample, confirming successful sulfonation
385 reactions across the carbon surface. This uniform distribution indicates that the acid
386 activation process effectively introduces sulfonic acid groups throughout the material, not
387 just on external surfaces. Oxygen distribution (30% coverage) correlates well with sulfur-
388 rich regions, consistent with the formation of sulfonic acid functional groups ($-\text{SO}_3\text{H}$) and
389 other oxygen-containing species. Sodium distribution (13% coverage) appears relatively
390 uniform but with some localized enrichment zones, supporting the hypothesis of ion
391 exchange processes during neutralization. The minor elements (aluminum, silicon, iron)
392 show reduced and more dispersed distributions compared to the raw carbon, confirming
393 effective acid leaching during the activation process.

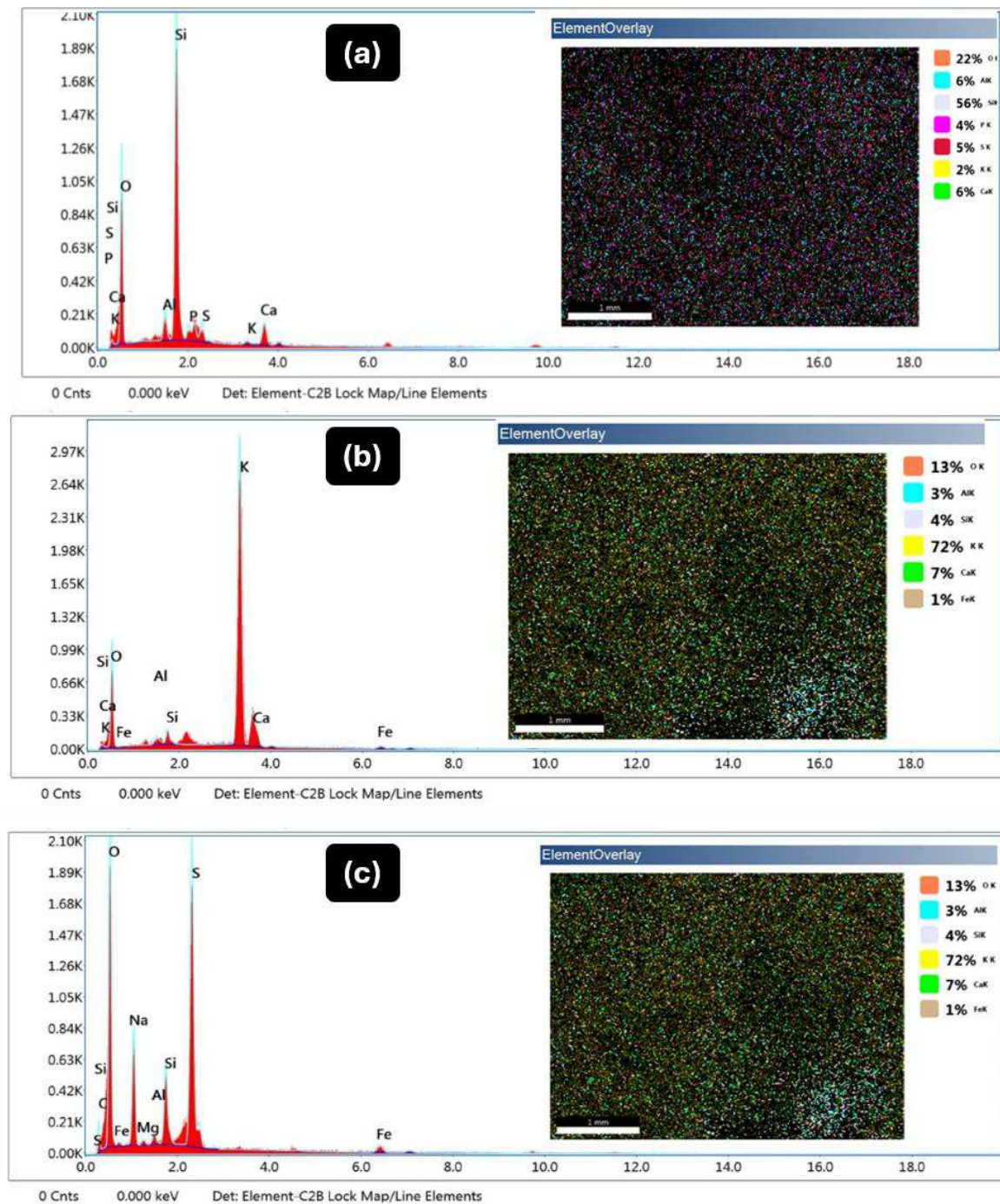


Figure 2. EDX spectra and elemental maps for RC (a), KAC (b), and SAC (c), highlighting the altered distribution of the major inorganic and activating elements

3.4 Structural and Crystallographic Analysis

The diffraction pattern shown in Figure (3a) exhibits well-defined peaks with varying intensities, indicating good crystallinity of the constituent phases. Phase identification was accomplished through pattern matching with the Crystallography Open Database (COD), while quantitative analysis was performed using Rietveld refinement methodology. X-ray diffraction pattern of the RC in Figure 3a showing major crystalline phases identified: Cristobalite II (COD 9016225), Calcite (COD 9016706), Aragonite (COD 9014838), Vuagnatite (COD 9000514), and Gismondine (COD 9012418). The quantitative phase analysis revealed that calcite (CaCO_3) constitutes the most abundant phase at 34.8%, followed by vuagnatite ($\text{CaAlSi}(\text{OH})\text{O}_4$) at 27.7%, and cristobalite II (SiO_2) at 23.3%. Minor phases include gismondine ($\text{CaAl}_2\text{Si}_2\text{O}_8 \cdot 4\text{H}_2\text{O}$) at 10.6% and aragonite (CaCO_3) at 3.6%. This mineralogical composition is particularly noteworthy as it combines common carbonate minerals with rare aluminosilicate phases, suggesting a complex formation history involving multiple geological processes.

The crystallographic parameters of the identified phases, summarized in Table 3, reveal diverse crystal systems ranging from hexagonal calcite to monoclinic cristobalite II. The predominance of orthorhombic structures (vuagnatite, gismondine, and aragonite) indicates formation under specific stress conditions, while the variety of crystal systems suggests multiple formation episodes or varying local conditions during crystallization. The unit cell parameters are consistent with literature values for these mineral phases, confirming the accuracy of the phase identification. Semi-quantitative elemental analysis complemented the phase identification, revealing oxygen as the dominant element at 48.8%, followed by calcium at 23.0%, silicon at 17.2%, aluminum at 6.1%, carbon at 4.6%, and hydrogen at 0.3%. This elemental distribution is entirely consistent with the identified mineral phases and provides additional validation of the quantitative phase analysis. The high oxygen content reflects the abundance of oxides, carbonates, and hydroxides, while the significant calcium content correlates directly with the presence of calcite, aragonite,

vuagnatite, and gismondine. The identification of cristobalite II is particularly significant from a mineralogical perspective, as this high-temperature SiO₂ polymorph typically forms above 1470 °C, indicating either volcanic origin or high-temperature industrial processing. The monoclinic crystal structure represents the β-cristobalite form, which suggests cooling from elevated temperatures under specific thermal conditions. The coexistence of both calcite and aragonite polymorphs of CaCO₃ provides important insights into the formation environment, as aragonite represents the higher-pressure polymorph and typically forms under different pressure-temperature conditions compared to calcite. Vuagnatite, representing over a quarter of the sample composition, is a relatively rare calcium aluminum silicate hydroxide mineral whose presence, along with the zeolite mineral gismondine, strongly suggests hydrothermal alteration processes or formation in alkaline conditions with readily available calcium and aluminum sources. The abundance of these aluminosilicate phases indicates that the raw material experienced significant chemical alteration, possibly in a hydrothermal environment where aluminum and silicon could be mobilized and recrystallized into these complex framework structures. The overall mineral assemblage provides compelling evidence for a complex formation environment involving multiple processes. The presence of high-temperature cristobalite suggests initial formation or processing at elevated temperatures, while the abundance of carbonates indicates significant carbonation reactions, possibly through interaction with CO₂-rich fluids. The hydrothermal alteration minerals (vuagnatite and gismondine) suggest subsequent low-to-moderate temperature aqueous alteration processes that modified the original mineral assemblage.

X-ray diffraction analysis of SAC reveals a complex assemblage of feldspar minerals with subordinate quartz and trace radioactive phases. The diffraction pattern shown in Figure (3b) exhibits characteristic peaks of alkali and plagioclase feldspars, with the most prominent reflections occurring around 27-28° 2θ, corresponding to the major feldspar phases. X-ray diffraction pattern of SAC in Figure (3b) showing major crystalline phases

identified: Anorthoclase (COD 9013308), Sanidine (COD 9004246), Quartz low (COD 1011176), Labradorite (COD 9000748), Albite (COD 9000525), Microcline (COD 9000943), Uranium (COD 4124713), and Thorium (COD 1523048). The quantitative phase analysis revealed a feldspar-dominated mineralogy with anorthoclase ((Na,K)AlSi₃O₈) constituting the most abundant phase at 30.4%, followed by albite (NaAlSi₃O₈) at 20.5%, labradorite ((Ca,Na)(Al,Si)₄O₈) at 20.1%, microcline (KAlSi₃O₈) at 16.4%, and sanidine (KAlSi₃O₈) at 10.6%. Minor phases include quartz (SiO₂) at 1.9%, thorium at 0.1%, and trace amounts of uranium. This mineralogical assemblage is characteristic of felsic igneous rocks or feldspar-rich pegmatitic materials, indicating formation under conditions favorable for alkali feldspar crystallization. The H₂SO₄ activation process appears to have preserved the original feldspar mineralogy while potentially modifying surface properties and developing enhanced porosity within the carbon matrix.

The crystallographic parameters of the identified phases, summarized in Table 3, reveal a predominance of triclinic crystal systems among the feldspar phases, with the exception of hexagonal quartz and the higher-symmetry radioactive phases. The coexistence of multiple feldspar phases suggests either primary magmatic crystallization under varying conditions or subsequent exsolution processes that led to the separation of initially homogeneous alkali feldspars into distinct compositional domains. The preservation of these crystalline structures in the SAC indicates that the H₂SO₄ activation process operated under conditions that maintained structural integrity while enhancing surface area. Semi-quantitative elemental analysis revealed oxygen as the dominant element at 47.4%, followed by silicon at 29.8% and aluminum at 11.3%, which is entirely consistent with the feldspar-dominated mineralogy. The presence of sodium (4.2%) and potassium (4.8%) reflects the abundance of alkali feldspars, while calcium (2.3%) corresponds to the plagioclase feldspar component. The detection of trace uranium (0.1%) indicates the presence of an accessory radioactive phase, which is not uncommon in granitic or pegmatitic environments. The

480 H₂SO₄ activation process appears to have concentrated these mineral phases while
481 developing the carbon structure through controlled carbonization and chemical activation.
482 The mineral assemblage provides strong evidence for formation in a felsic igneous
483 environment, likely representing either a granitic rock or pegmatite that served as the
484 precursor material for activated carbon production. The abundance of alkali feldspars
485 (anorthoclase, sanidine, microcline) suggests crystallization from a K-Na-rich magma
486 under relatively low-pressure conditions. The presence of both high-temperature sanidine
487 and low-temperature microcline indicates either a complex cooling history or multiple
488 episodes of crystallization and recrystallization. The successful preservation of these
489 phases during H₂SO₄ activation suggests that the treatment conditions were optimized to
490 develop porosity while maintaining the beneficial properties of the feldspar minerals for
491 potential applications in adsorption or catalysis.

492 X-ray diffraction analysis of KAC was conducted under identical conditions to the SAC
493 sample, revealing a simplified but related feldspar assemblage with enhanced
494 concentrations of specific phases. The diffraction pattern shown in Figure 3c exhibits
495 similar characteristic feldspar peaks but with different relative intensities, indicating a
496 modified phase distribution compared to the SAC. The pattern quality remains excellent
497 with well-resolved peaks enabling accurate phase identification and quantification. The
498 KOH activation process appears to have selectively modified the phase assemblage,
499 possibly through preferential dissolution or transformation of certain mineral components.
500 Figure 3c shows the major crystalline phases identified: Anorthoclase (COD 9013308),
501 Quartz low (COD 1011176), Albite (COD 9000525), Microcline (COD 9000943),
502 Uranium (COD 4124713, COD 7222830), and Thorium (COD 9008485). The quantitative
503 phase analysis revealed a more concentrated feldspar assemblage with anorthoclase
504 dramatically increased to 43.7%, representing the dominant phase. Albite constitutes
505 29.5% and microcline 23.6%, while quartz remains minor at 2.7%. Notably, this sample
506 shows slightly elevated concentrations of radioactive phases, with uranium present in two

different structural forms (COD 4124713 and COD 7222830) at 0.1% each, and thorium at 0.1%. The absence of sanidine and labradorite compared to the SAC sample suggests that the KOH activation process either preferentially dissolved these phases or promoted their transformation into the more stable anorthoclase and albite phases. This selective phase modification is consistent with the alkaline activation mechanism, which typically involves dissolution-reprecipitation processes under high pH conditions. The crystallographic parameters for the major phases remain consistent with those reported for the SAC sample, as shown in Table 3, confirming the structural integrity of the feldspar minerals. The simplified phase assemblage with higher concentrations of anorthoclase and albite suggests that KOH activation not only developed porosity but also promoted phase concentration and purification. This could be advantageous for applications requiring specific feldspar compositions or enhanced uniformity in mineral properties.

The elemental composition analysis for the KAC sample was not provided in the available data, but based on the mineralogical composition, it would be expected to show similar proportions to the SAC sample, with potentially enhanced silicon and aluminum contents due to the increased feldspar concentration and reduced accessory phase content. The KOH activation process may have also introduced additional potassium into the system, which could be incorporated into the feldspar structures or present as residual activation agent. Both activated carbon samples demonstrate the dominance of feldspathic minerals characteristic of felsic igneous rocks that served as precursor materials for activated carbon production. The progression from SAC to KAC shows increasing concentration of anorthoclase and albite, with elimination of the calcium-rich labradorite and high-temperature sanidine phases. This evolution suggests that different activation chemistries (H_2SO_4 vs. KOH) produce distinct effects on the mineral assemblage, with KOH promoting more selective phase dissolution and concentration. Alkaline activation appears to preferentially remove less stable phases while concentrating the more chemically resistant anorthoclase and albite components.

534 The persistent presence of radioactive phases (uranium and thorium) in both activated
535 carbon samples, albeit in trace quantities, require careful consideration for any potential
536 applications. While the concentrations are low, appropriate handling protocols should be
537 implemented to ensure occupational safety during production and use. The detection of
538 two different uranium phases in the KAC sample suggests that the KOH activation process
539 may have promoted structural reorganization of the radioactive components, potentially
540 affecting their leaching behavior or stability. The feldspar-rich nature of these activated
541 carbon materials suggests unique properties that combine the high surface area and porosity
542 typical of activated carbons with the chemical stability and ion-exchange capabilities of
543 feldspar minerals. This hybrid character could be advantageous in applications such as
544 selective adsorption, catalysis support, or environmental remediation where both high
545 surface area and specific mineral-solution interactions are desired. The excellent
546 crystallinity evidenced by the sharp, well-defined XRD peaks indicates that both activation
547 processes successfully developed porosity while preserving the beneficial structural
548 characteristics of the feldspar components, developing materials with potentially superior
549 performance compared to conventional activated carbons derived from purely organic
550 precursors.

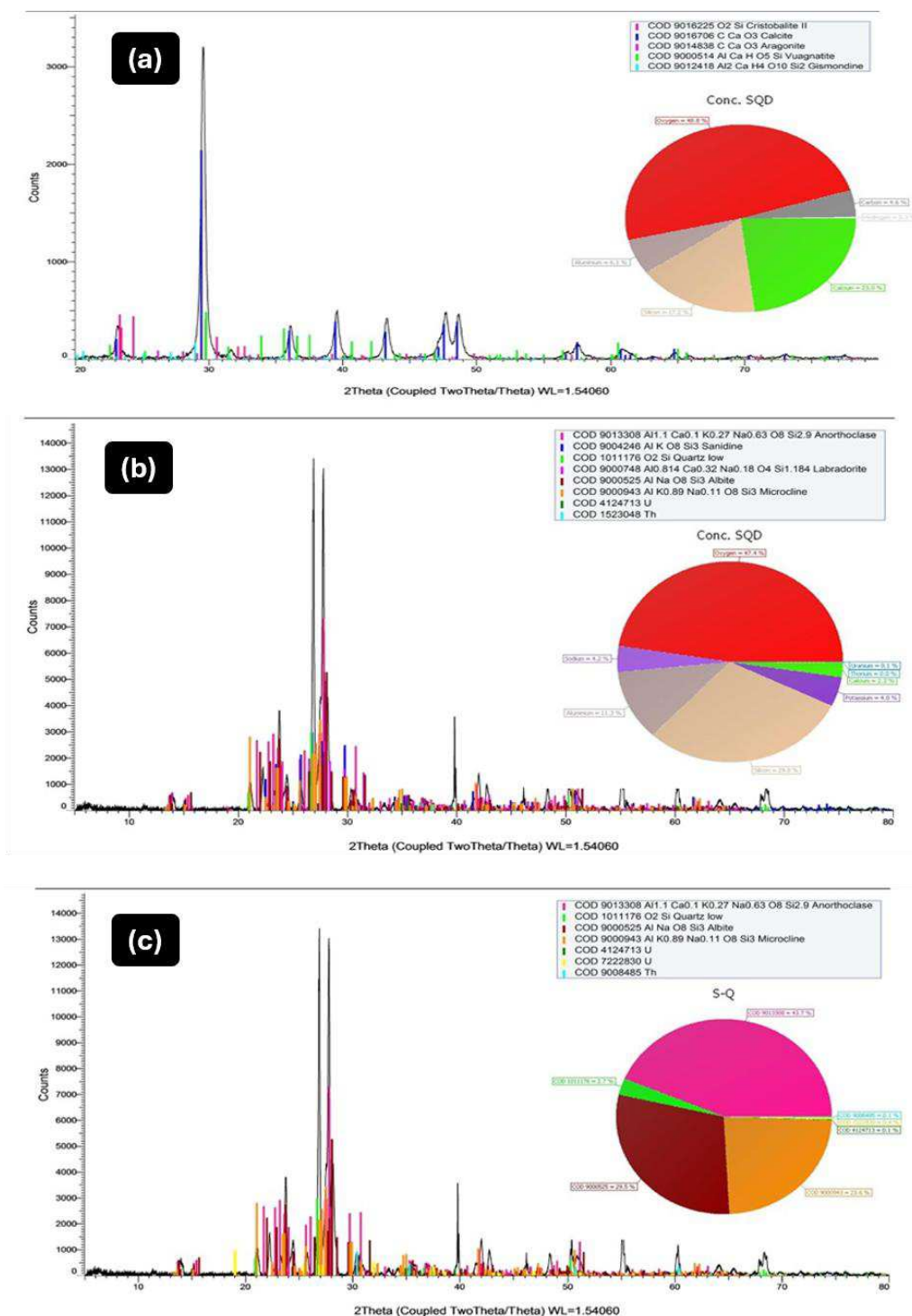


Figure 3. XRD patterns of RC, SAC, and KAC, allowing direct comparison of the phase assemblages before and after activation

554 *Table 3. Unified crystallographic parameters for RC, SAC, and KAC*

Sample	Phase	Formula	Crystal system	Space group	Unit-cell parameters	Volume (Å ³)
RC	Calcite	CaCO ₃	Hexagonal	R -3 c (167)	a = 4.9844 Å; c = 17.0376 Å	366.58
	Vuagnatite	CaAlSi(OH)O ₄	Orthorhombic	P 21 21 21 (19)	a = 7.0550 Å; b = 8.5420 Å; c = 5.6830 Å	342.48
	Cristobalite II	SiO ₂	Monoclinic	P 1 21/c 1 (14)	a = 8.0110 Å; b = 4.5440 Å; c = 8.8900 Å; β = 121.00°	277.39
	Gismondine	CaAl ₂ Si ₂ O ₈ ·4H ₂ O	Orthorhombic	P 21 21 21 (19)	a = 13.9020 Å; b = 8.8920 Å; c = 13.9520 Å	1724.70
	Aragonite	CaCO ₃	Orthorhombic	P m c n (62)	a = 4.9586 Å; b = 7.9867 Å; c = 5.7547 Å	227.90
SAC	Anorthoclase	Al _{1.1} Ca _{0.1} K _{0.27} Na _{0.63} O ₈ Si _{2.9} A _{11.1} Ca _{0.1} K _{0.27} Na _{0.63} O ₈ Si _{2.9}	Triclinic	C -1 (2)	a = 8.2780 Å; b = 12.9490 Å; c = 7.1450 Å; α = 91.790°; β = 116.160°; γ = 90.190°	686.97

Sanidine	AlKSi_3O_8	Monoclinic	$C 1 2/m 1 (12)$	$a = 8.6110 \text{ \AA}; b = 13.0460 \text{ \AA}; c = 7.1774 \text{ \AA}; \beta = 116.020^\circ$	-
Quartz low	SiO_2	Hexagonal	$P 3 2 2 1 S (154)$	$a = 4.9000 \text{ \AA}; c = 5.4000 \text{ \AA}$	112.28
Labradorite	$\text{Al}_{0.814}\text{Ca}_{0.32}\text{Na}_{0.18}\text{O}_4\text{Si}_{1.184}$	Triclinic	$C -1 (2)$	$a = 8.1510 \text{ \AA}; b = 12.8290 \text{ \AA}; c = 7.1044 \text{ \AA}; \alpha = 93.170^\circ; \beta = 116.300^\circ; \gamma = 90.230^\circ$	-
Albite	$\text{AlNaSi}_3\text{O}_8$	Triclinic	$-1 (2)$	$a = 8.1530 \text{ \AA}; b = 12.8694 \text{ \AA}; c = 7.1070 \text{ \AA}; \alpha = 93.521^\circ; \beta = 116.458^\circ; \gamma = 90.257^\circ$	665.91
Microcline	$\text{AlK}_{0.89}\text{Na}_{0.11}\text{O}_8\text{Si}_3$	Triclinic	$C -1 (2)$	$a = 8.5520 \text{ \AA}; b = 12.9750 \text{ \AA}; c = 7.2050 \text{ \AA}; \alpha = 90.090^\circ; \beta = 115.920^\circ; \gamma = 89.510^\circ$	719.03
Uranium	U	Orthorhombic	$C m c m (63)$	$a = 2.8520 \text{ \AA}; b = 5.8650 \text{ \AA}; c = 4.9450 \text{ \AA}$	82.71
Thorium	Th	Tetragonal	$I 4/m m m$	$a = 2.8220 \text{ \AA}; c = -$	-

				(139)		
KAC	Anorthoclase	$\text{Al}_{1.1}\text{Ca}_{0.1}\text{K}_{0.27}\text{Na}_{0.63}\text{O}_8\text{Si}_{2.9}$	Triclinic	C -1 (2)	$a = 8.2780 \text{ \AA}; b = 12.9490 \text{ \AA}; c = 7.1450 \text{ \AA}; \alpha = 91.790^\circ; \beta = 116.160^\circ; \gamma = 90.190^\circ$	686.97
	Quartz low	SiO_2	Hexagonal	P 32 2 1 S (154)	$a = 4.9000 \text{ \AA}; c = 5.4000 \text{ \AA}$	112.28
	Albite	$\text{AlNaSi}_3\text{O}_8$	Triclinic	C -1 (2)	$a = 8.1530 \text{ \AA}; b = 12.8694 \text{ \AA}; c = 7.1070 \text{ \AA}; \alpha = 93.521^\circ; \beta = 116.458^\circ; \gamma = 90.257^\circ$	665.91
	Microcline	$\text{AlK}_{0.89}\text{Na}_{0.11}\text{O}_8\text{Si}_3$	Triclinic	C -1 (2)	$a = 8.5520 \text{ \AA}; b = 12.9750 \text{ \AA}; c = 7.2050 \text{ \AA}; \alpha = 90.090^\circ; \beta = 115.920^\circ; \gamma = 89.510^\circ$	719.03
	Uranium (form 1)	U	Orthorhombic	C m c m (63)	$a = 2.8520 \text{ \AA}; b = 5.8650 \text{ \AA}; c = 4.9450 \text{ \AA}$	82.71
	Uranium (form 2)	U	Monoclinic	C 1 2/c 1 (15)	$a = 9.3400 \text{ \AA}; b = 7.0000 \text{ \AA}; c = 9.3000 \text{ \AA};$	607.99

					$\beta = 89.290^\circ$	
	Thorium	Th	Cubic	F m -3 m (225)	$a = 5.0843 \text{ \AA}$	131.43

555

3.5 XPS Analysis

The XPS survey spectrum of the RC, shown in Figure 4, exhibits characteristic photoelectron peaks corresponding to the major surface elements, with carbon and oxygen representing the dominant species. High-resolution spectra of C 1s (~285 eV), O 1s (~533 eV), N 1s (~400 eV), and S 2p (~165 eV) core levels were recorded with pass energy of 50 eV and energy step of 0.1 eV to analyze detailed chemical environments and bonding configurations. Peak deconvolution analysis was performed using Casa XPS software with Gaussian-Lorentzian peak shapes and Shirley background subtraction to quantify the relative concentrations of different functional groups including C-C/C-H (graphitic carbon), C-O (hydroxyl, ether), C=O (carbonyl), O-C=O (carboxyl), C-SO₃H (sulfonic groups in SAC), and other oxygen and sulfur-containing functionalities.

The quantitative surface elemental analysis of the RC revealed a carbon-dominated surface composition with C1s representing 77.56 atomic %, followed by oxygen (O 1s) at 15.96 atomic %. Minor surface constituents include silicon (Si 2p) at 2.30 atomic %, nitrogen (N 1s) at 2.25 atomic %, calcium (Ca 2p) at 1.89 atomic %, and trace amounts of iron (Fe 2p) at 0.04 atomic %. The high carbon concentration is consistent with the organic nature of the precursor material, while the presence of inorganic elements (Si, Ca, Fe) indicates the incorporation of mineral components during the carbonization process. The C1s peak binding energy of 285.83 eV is characteristic of sp²-hybridized carbon typically found in graphitic structures, confirming the formation of aromatic carbon frameworks during thermal treatment.

The detailed binding energy analysis, summarized in Table 4, provides insight into the chemical states of the surface elements. The O1s peak at 532.74 eV suggests the presence of various oxygen-containing functional groups including C-O, C=O, and possibly hydroxyl groups, which are commonly formed during the carbonization of organic materials. The Si 2p binding energy of 103.71 eV is consistent with silicate minerals or

silica phases, while the Ca 2p peak at 348.51 eV indicates calcium in oxide or carbonate form, correlating well with the calcite phases identified in the XRD analysis.

Table 4. XPS surface elemental composition and binding energies for all samples

Element	RC		KAC		SAC	
	BE (eV)	At.%	BE (eV)	At.%	BE (eV)	At.%
C 1s	285.83	77.56	286.09	43.46	285.98	25.10
O 1s	532.74	15.96	532.98	32.55	533.13	47.80
Si 2p	103.71	2.30	103.92	4.13	103.92	0.68
Ca 2p	348.51	1.89	-	-	349.31	1.01
N 1s	400.91	2.25	400.46	2.04	402.45	4.12
Fe 2p	712.81	0.04	716.22	0.63	716.08	0.27
K 2p	-	-	294.95	11.97	-	-
S 2p	-	-	169.87	1.81	170.14	11.81
Cl 2p	-	-	199.93	3.41	-	-
Na 1s	-	-	-	-	1072.35	9.21

The XPS analysis of the KAC revealed significant compositional changes compared to RC, as illustrated in Figure 4. The surface carbon content decreased substantially to 43.46 atomic %, while oxygen increased dramatically to 32.55 atomic %, indicating extensive oxidation and the introduction of oxygen-containing functional groups during the KOH activation process. The appearance of potassium (K 2p) at 11.97 atomic % with a binding energy of 294.95 eV confirms the incorporation of the KOH activating agent, likely present as potassium compounds or intercalated potassium species within the carbon structure. The presence of chlorine (Cl 2p) at 3.41 atomic % with a binding energy of 199.93 eV in the KAC sample suggests either contamination from the activation process or the formation of potassium chloride phases. The sulfur content (S 2p) at 1.81 atomic % with a binding

energy of 169.87 eV indicates the presence of sulfur-containing functional groups or compounds, possibly derived from the precursor material or introduced during processing. The silicon content increased to 4.13 atomic % compared to RC, while iron content increased significantly to 0.63 atomic %, suggesting concentration effects during the activation process. The shift in C 1s binding energy to 286.09 eV indicates a change in the carbon chemical environment, likely due to the formation of carbon-oxygen bonds and the development of more oxidized carbon species.

SAC exhibited the most dramatic compositional changes among the three samples, with oxygen becoming the dominant surface species at 47.80 atomic %, as shown in Figure 4. The carbon content decreased further to 25.10 atomic %, indicating extensive oxidation and the introduction of numerous oxygen-containing functional groups during the H₂SO₄ activation process. The appearance of sodium (Na 1s) at 9.21 atomic % with a binding energy of 1072.35 eV is unexpected and may indicate contamination or the use of sodium-containing reagents during sample preparation or washing procedures.

The high sulfur content (S 2p) at 11.81 atomic % with a binding energy of 170.14 eV in the SAC sample confirms the extensive incorporation of sulfur-containing species, likely in the form of sulfonate groups (-SO₃H), sulfate esters, or other organosulfur functionalities developed during the H₂SO₄ activation process. This sulfur incorporation is significant as it can provide additional surface acidity and potentially enhance adsorption properties for specific applications. The nitrogen content increased to 4.12 atomic % with a binding energy of 402.45 eV, suggesting the formation of various nitrogen-containing functional groups or the concentration of nitrogen species during activation. The silicon content decreased dramatically to 0.68 atomic %, while calcium content remained low at 1.01 atomic %, indicating possible leaching of mineral components during the acidic activation treatment.

However, the progression from raw material to activated carbons demonstrates the profound effects of different activation chemistries on surface composition and

functionalization. The KOH activation resulted in moderate oxidation while introducing basic potassium species, developing a material with enhanced oxygen functionality and potential basic character. In contrast, the H₂SO₄ activation produced extensive oxidation and significant sulfur incorporation, likely developing a highly functionalized material with acidic surface properties. The C 1s binding energy shifts (285.83 eV → 286.09 eV → 285.98 eV) reflect the changing chemical environments and the extent of functionalization, with both activated samples showing evidence of carbon-heteroatom bonding.

The iron content changes across the samples (0.04% → 0.63% → 0.27%) suggest that activation processes may concentrate or redistribute metallic species, potentially developing catalytically active sites. The binding energies for Fe2p (712.81-716.22 eV) are consistent with iron oxide phases, which could contribute to additional functionality in adsorption or catalytic applications. The preservation of silicon in both activated carbons, albeit at different concentrations, indicates that some mineral components survive the activation treatments, potentially contributing to structural stability and unique surface properties.

From a surface chemistry perspective, these results indicate that both activation processes successfully introduced substantial heteroatom functionalization, with KOH developing a more basic, potassium-rich surface and H₂SO₄ producing a highly oxidized, sulfur-rich, and potentially acidic surface. The high oxygen contents in both activated carbons (32.55% and 47.80%) suggest excellent potential for applications requiring high surface polarity, such as water treatment, gas separation, or as supports for catalytic applications. The distinct chemical signatures of each activation route provide opportunities for tailored surface properties depending on the intended application, with the SAC sample showing promise for acid-catalyzed processes and the KAC sample potentially beneficial for applications requiring basic surface sites.

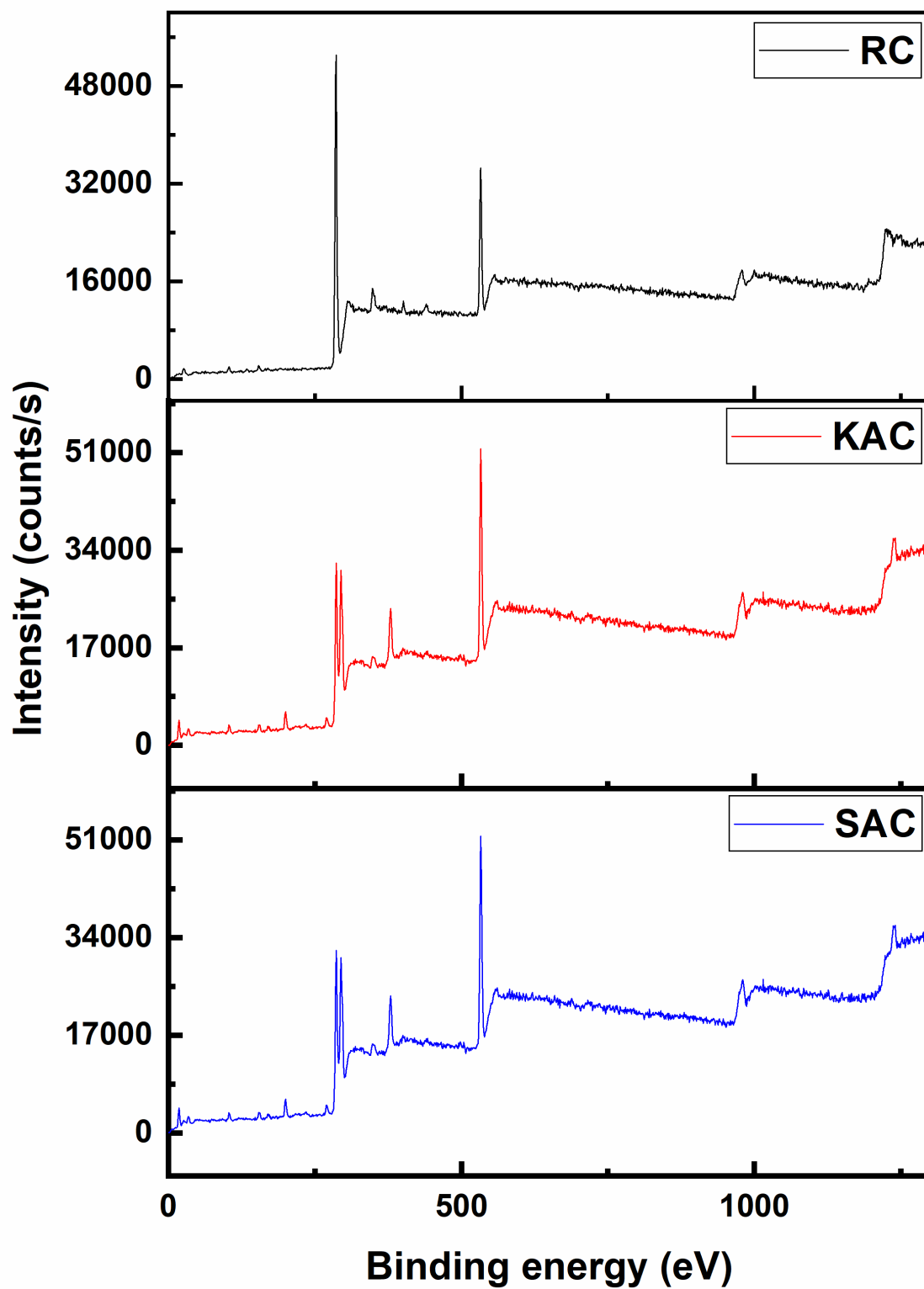


Figure 4. XPS survey spectrum of RC showing major photoelectron peaks and their corresponding binding energies

3.6 Differential Scanning Calorimetry (DSC) analysis

Differential Scanning Calorimetry (DSC) analysis was performed on RC, SAC, and KAC to investigate their thermal behavior and identify characteristic thermal transitions. The DSC thermogram of RC, shown in Fig. 5, exhibits distinct thermal events including endothermic moisture loss at lower temperatures and exothermic combustion processes at elevated temperatures.

The thermal analysis of RC (initial mass: 27.9 mg) revealed several distinct thermal events across the temperature range examined. A prominent endothermic peak observed around 200 °C corresponds to the removal of physically and chemically bound water, as well as the decomposition of low-stability organic components. This dehydration process is characteristic of natural organic materials and indicates the presence of hygroscopic components and weakly bound moisture. The heat flow magnitude during this endothermic transition suggests significant mass loss, consistent with the removal of volatile components and structural water from the carbonaceous matrix and associated mineral phases.

The thermal behavior becomes markedly exothermic starting around 400 °C, with a broad exothermic region extending to approximately 650 °C. This temperature range corresponds to the oxidative combustion of the organic carbon framework, representing the primary decomposition of the carbonaceous structure. The exothermic nature of this process indicates energy release during carbon oxidation, and the broad temperature range suggests the presence of carbon species with varying thermal stabilities. A second exothermic peak observed around 550-650 °C may correspond to the combustion of more thermally stable

675 aromatic carbon structures or the oxidation of residual char formed during the initial
676 decomposition stages.

677 The experimental parameters and key thermal characteristics for all samples are
678 summarized in Table 5, providing a comprehensive comparison of their thermal behaviors.
679 RC exhibited the highest initial mass (27.9 mg) and demonstrated the most complex
680 thermal profile with multiple distinct transitions, reflecting the heterogeneous nature of the
681 untreated material.

682

683 **Table 5.** DSC experimental parameters and thermal characteristics of all samples

Sample	Initial Mass (mg)	Heat Range (mW)	Flow Major Endothermic Peak (°C)	Exothermic Region (°C)	Key Thermal Events
RC	27.9	6.76 - 43.48	~200	400-650	Dehydration, organic combustion
KAC	18.0	8.02 - 37.50	~250-300	400-550	Modified dehydration, altered combustion
SAC	19.7	2.26 - 32.65	~100-150	400-600	Multiple dehydration events, complex combustion

684

685

686 The DSC thermogram of the KAC, presented in Fig. 5, demonstrates significantly modified
687 thermal behavior compared to RC, reflecting the structural and chemical changes
688 introduced during the alkaline activation process. The initial mass of 18.0 mg represents a
689 lower sample quantity, which may contribute to the altered heat flow magnitudes observed
690 throughout the analysis. The endothermic transition appears shifted to higher temperatures
691 (250-300 °C) compared to RC, suggesting changes in the water binding mechanisms or the
692 formation of different hydrated species during KOH activation.

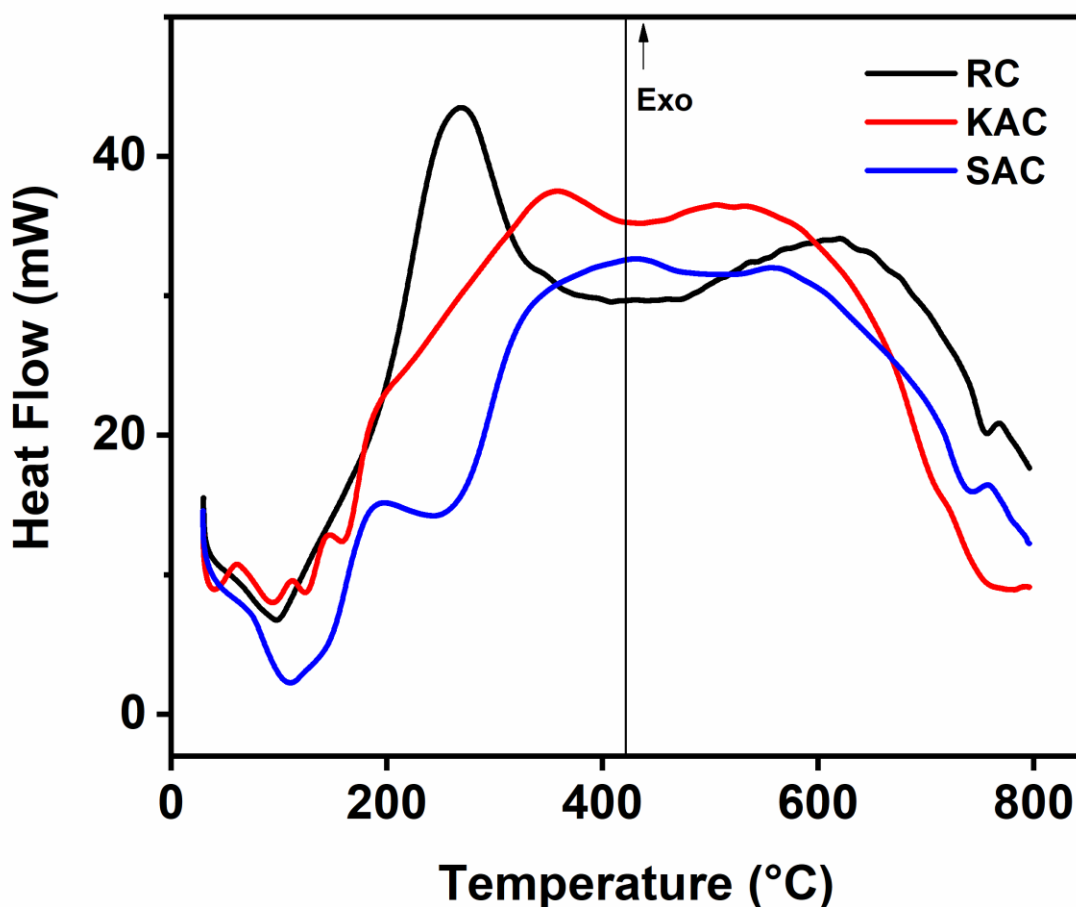
693 The thermal profile of the KAC sample shows a more compressed exothermic region
694 extending from approximately 400-550 °C, indicating that the KOH activation process has
695 developed a more thermally uniform carbon structure. This narrower temperature range for
696 combustion suggests that the activation process may have eliminated some of the thermal
697 heterogeneity present in the raw material, possibly through the removal of less stable
698 components or the creation of more uniform pore structures. The reduced intensity of the
699 exothermic processes compared to RC may reflect the lower carbon content resulting from
700 the extensive oxidation observed in the XPS analysis, where carbon decreased from
701 77.56% to 43.46%. The presence of potassium species, confirmed by XPS analysis, may
702 also influence the thermal behavior by acting as catalytic sites for carbon gasification
703 reactions or by modifying the thermal decomposition pathways. The KOH activation
704 process typically involves carbon gasification reactions that consume carbon and develop
705 porosity, which could explain the modified thermal signature and the shift toward lower
706 heat flow magnitudes in the exothermic region. SAC exhibits the most dramatically altered
707 thermal behavior among the three samples, as illustrated in Figure 5. With an initial mass
708 of 19.7 mg, the SAC sample demonstrates multiple endothermic events at relatively low
709 temperatures (100-150 °C), indicating the presence of various hydrated sulfur-containing
710 functional groups and residual activation agent. These low-temperature endothermic
711 transitions suggest the dehydration of sulfuric acid hydrates, the decomposition of

sulfonate groups, or the removal of water associated with hygroscopic sulfur-containing species.

The SAC sample shows the lowest heat flow range (2.26 - 32.65 mW), indicating significantly reduced thermal activity compared to both the RC and KAC. This suppressed thermal response likely reflects the extensive functionalization with sulfur-containing groups (11.81% S from XPS) and the high degree of oxidation (47.80% O from XPS). The exothermic combustion region extends from approximately 400-600 °C, showing a broader temperature range than the KAC but different peak characteristics compared to RC. The presence of sulfur-containing functional groups may influence the combustion mechanism, potentially acting as fire retardants or modifying the carbon oxidation pathways. The complex thermal behavior observed in the SAC sample, with multiple overlapping thermal events, reflects the heterogeneous nature of the sulfur-functionalized carbon surface. The H₂SO₄ activation process appears to have developed a material with diverse thermal stability characteristics, ranging from highly labile hydrated species that decompose at low temperatures to more stable carbonaceous structures that require higher temperatures for oxidative decomposition.

Comparative analysis of the three samples reveals distinct thermal signatures that correlate well with their chemical compositions determined by XPS analysis. The progression from raw material to activated carbon shows systematic changes in thermal behavior that reflect the extent of chemical modification achieved by different activation routes. RC exhibits the most intense thermal transitions due to its high carbon content and structural heterogeneity, while both activated carbons show moderated thermal responses consistent with their enhanced functionalization and modified surface chemistry. The thermal stability implications of these results are significant for practical applications. The KAC sample demonstrates relatively stable thermal behavior with a defined combustion range, making it potentially suitable for applications requiring moderate thermal stability. The SAC sample, with its complex low-temperature transitions and extensive functionalization,

739 may be more suitable for applications where high surface reactivity is desired, though its
 740 thermal complexity may limit high-temperature applications. The thermal analysis
 741 confirms that both activation processes successfully modified the carbon structure while
 742 introducing distinct thermal characteristics that could be exploited for specific applications
 743 in adsorption, catalysis, or energy storage systems.



744
 745 *Figure 5. DSC thermograms of RC, KAC, and SAC*
 746

747 **3.7 Process-Structure-Property Relationships and Applications Implications**

748 The combined SEM-EDX analysis reveals strong correlations between activation method,
 749 morphological development, and chemical composition, establishing clear process-

structure-property relationships. The KOH activation successfully develops a hierarchical porous structure with mesopore and macropore development while incorporating basic sites through potassium retention. This combination of high surface area and basic surface chemistry makes the material ideal for applications such as CO₂ capture, where basic sites enhance CO₂ adsorption capacity, or supercapacitors operating in basic electrolytes, where the retained potassium can improve ionic conductivity. H₂SO₄ activation produces a fundamentally different material with microporous structure and acidic surface functionality, making it suitable for ion exchange applications, heavy metal removal from aqueous solutions, or supercapacitors in acidic electrolytes. The fine particle size and high surface functionality suggest excellent dispersibility in aqueous media, which is advantageous for liquid-phase applications. The high concentration of sulfonic acid groups provides strong ion-exchange capacity and enhanced hydrophilicity. The scientific significance of these findings extends beyond materials characterization to waste valorization and circular economy principles. The successful synthesis of structurally and chemically distinct carbon materials from sludge waste demonstrates the potential for converting waste streams into high-value carbon materials. The ability to tune properties through activation chemistry opens possibilities for designing task-specific carbon materials from renewable waste sources, contributing to sustainable materials development.

3.8 Optimization Insights and Process Understanding

The incomplete removal of activating agents (K in KAC, Na in SAC) provides important insights for process optimization. The high potassium retention in KAC suggests either the need for more aggressive washing protocols or indicates strong chemical bonding between potassium species and the carbon framework. For applications where high potassium content is detrimental, optimization of washing conditions (temperature, pH, multiple cycles) would be necessary. Conversely, for applications where potassium acts as a beneficial dopant, the current process appears effective. The sodium presence in SAC

indicates that neutralization procedures require careful control to achieve the desired final composition. The correlation between oxygen and activating elements (K, S) in the mapping data supports the proposed activation mechanisms involving specific functional group formation and chemical complex development, validating the theoretical understanding of these processes. The selective removal of ash components during both activation processes demonstrates that these treatments simultaneously purify and activate the carbon materials. KOH activation is particularly effective in removing silica and alumina through alkaline dissolution, while H₂SO₄ activation achieves similar purification through acid leaching. This dual functionality of activation and purification adds significant value to the waste-to-carbon conversion process, producing materials with enhanced purity compared to simple pyrolysis.

Table 6. Consolidated DSC experimental parameters and thermal characteristics

Sample	Initial mass (mg)	Heat flow range (mW)	Major endothermic peak (°C)	Exothermic region (°C)	Key thermal events
RC	27.9	6.76 - 43.48	~200	400-650	Dehydration; organic combustion
KAC	18.0	8.02 - 37.50	~250-300	400-550	Modified dehydration; altered combustion
SAC	19.7	2.26 - 32.65	~100-150	400-600	Multiple dehydration events; complex combustion

790

3.5 Optimization Insights and Process Understanding

791

The residual K in KAC and Na in SAC provide useful information for process optimization. Higher residual potassium may indicate the need for optimized washing when potassium retention is undesirable, whereas controlled retention may be relevant to future material-design studies. The sodium detected in SAC indicates that the neutralization step should be carefully controlled. These observations are process-optimization considerations and are not evidence of application performance. The elemental-mapping results support the proposed activation mechanisms and show that both treatments altered the distribution of inorganic components.

The selective removal of ash components during both activation processes indicates that activation also altered the inorganic fraction. KOH treatment was associated with reduced silicon-containing phases, while H₂SO₄ treatment produced acid-leached materials with substantial sulfur incorporation. These changes are consistent with simultaneous structural modification and mineral-phase redistribution during waste-to-carbon conversion.

4. Conclusions

This comprehensive study successfully demonstrates the feasibility of transforming sewage sludge waste into high value activated carbon materials with tailored properties through strategic chemical activation approaches. The systematic characterization using advanced analytical techniques has established clear process-structure-property relationships and provided fundamental insights into the mechanisms governing morphological evolution and elemental redistribution during chemical activation processes as follows. The XRD analysis revealed that both activation methods produce distinctly different crystalline assemblages compared to raw material. While the raw sample exhibited a complex mineralogy dominated by calcite, vuagnatite, and cristobalite phases, the activated carbons showed feldspar-dominated structures with KOH activation

817 developing a simplified assemblage concentrated in anorthoclase and albite phases, and
818 H₂SO₄ activation producing a more diverse feldspar distribution. The XPS surface analysis
819 shows that KOH activation achieved moderate surface oxidation while incorporating
820 significant potassium content (11.97 atomic %), developing materials with enhanced
821 oxygen functionality and potential basic character suitable for CO₂ capture and basic
822 electrolyte applications. In contrast, H₂SO₄ activation produced extensive surface oxidation
823 with oxygen becoming the dominant surface species (47.80 atomic %) alongside
824 substantial sulfur incorporation (11.81 atomic %), developing highly functionalized
825 materials with acidic surface properties ideal for ion exchange and water treatment
826 applications. The thermal analysis through DSC confirmed the distinct thermal signatures
827 of each activation route, with KOH activation producing thermally uniform materials
828 exhibiting compressed exothermic regions, while H₂SO₄ activation developed materials
829 with complex multi-stage thermal behavior reflecting extensive heteroatom
830 functionalization. The morphological investigation shows that KOH activation
831 successfully developed hierarchical porous structures with distinctive rod-like and plate-
832 like morphologies, while H₂SO₄ activation produced fine spherical particles with extensive
833 agglomeration patterns. The demonstrated ability to produce materials with contrasting
834 properties from identical precursors through different activation chemistries represents a
835 significant advancement in waste valorization technology. The KAC materials, with their
836 hierarchical porosity, basic surface functionality, and retained potassium content, show
837 excellent potential for gas adsorption applications, supercapacitors operating in basic
838 electrolytes, and CO₂ capture systems. The SAC materials, characterized by fine particle
839 morphology, extensive surface functionalization, and acidic properties, are particularly
840 suitable for water treatment applications, heavy metal removal, and ion exchange
841 processes. The scientific significance of this work extends beyond materials
842 characterization to establish fundamental process-structure-property relationships that can
843 guide future research and development efforts. The systematic comparison of two

chemically distinct activation approaches provides valuable insights into activation mechanisms and their influence on final material properties, contributing to the theoretical understanding of chemical activation processes for waste-derived carbon materials. Future research directions should focus on scaling up the most promising activation routes, conducting detailed performance evaluations in target applications, and investigating the long-term stability and recyclability of these materials. Additionally, economic and life cycle assessments would provide valuable information for commercial viability and environmental impact evaluation. The development of hybrid activation approaches combining the advantages of both methods could potentially develop materials with even more tailored properties for specific applications.

Authors contributions

M. A. B. wrote the original draft, and he is responsible for data curation, formal analysis, and Investigation; N. Sh. contributes in supervision, validation, visualization, and writing – review & editing; S. El-N. is responsible for conceptualization, methodology, and she contribute in supervision, validation, writing – review & editing

Funding

The characterization analyses for this research paper were funded by the United States Agency for International Development (USAID). The contents are the responsibility of the Authors and do not necessarily reflect the views of USAID or the United States Government.

Data Availability

The data in this paper may be obtained from the authors upon reasonable request.

Declarations

Conflict of interest: There are no conflicts to declare. Ethical approval: Not applicable.

Consent to participate: The authors declare that they consent to participate in the work. Consent to publish: The authors declare that they consent to publish this paper.

Acknowledgments

The authors acknowledge the institutional and funding support that enabled the characterization analyses reported in this study.

References

1. Rodriguez C, Alaswad A, Benyounis KY, Olabi AG. Pretreatment techniques used in biogas production from grass. *Renew Sustain energy Rev.* 2017;68:1193–204.
2. Fonts I, Gea G, Azuara M, Ábrego J, Arauzo J. Sewage sludge pyrolysis for liquid production: a review. *Renew Sustain energy Rev.* 2012;16:2781–805.
3. Raheem A, Sikarwar VS, He J, Dastyar W, Dionysiou DD, Wang W, et al. Opportunities and challenges in sustainable treatment and resource reuse of sewage sludge: A review. *Chem Eng J.* 2018;337:616–41.
4. Marsh H, Rodríguez-Reinoso F. CHAPTER 2 - Activated Carbon (Origins). In: Marsh H, Rodríguez-Reinoso FBT-AC, editors. Oxford: Elsevier Science Ltd; 2006. p. 13–86. Available from: <https://www.sciencedirect.com/science/article/pii/B9780080444635500169>
5. Bansal RC, Goyal M. Activated carbon adsorption. CRC press; 2005.
6. Hadi P, Xu M, Ning C, Lin CSK, McKay G. A critical review on preparation, characterization and utilization of sludge-derived activated carbons for wastewater treatment. *Chem Eng J.* 2015;260:895–906.
7. Yahya MA, Al-Qodah Z, Ngah CWZ. Agricultural bio-waste materials as potential

- sustainable precursors used for activated carbon production: A review. *Renew Sustain energy Rev.* 2015;46:218–35.
8. González-García P. Activated carbon from lignocellulosics precursors: A review of the synthesis methods, characterization techniques and applications. *Renew Sustain energy Rev.* 2018;82:1393–414.
9. Bedia J, Peñas-Garzón M, Gómez-Avilés A, Rodríguez JJ, Belver C. Review on activated carbons by chemical activation with FeCl_3 . *C.* 2020;6:21.
10. Lillo-Ródenas MA, Cazorla-Amorós D, Linares-Solano A. Understanding chemical reactions between carbons and NaOH and KOH: an insight into the chemical activation mechanism. *Carbon N Y.* 2003;41:267–75.
11. Wang J, Kaskel S. KOH activation of carbon-based materials for energy storage. *J Mater Chem.* 2012;22:23710–25.
12. Ismail SH, Hamdy A, Ismail TA, Mahboub HH, Mahmoud WH, Daoush WM. Synthesis and characterization of antibacterial carbopol/ZnO hybrid nanoparticles gel. *Crystals.* 2021;11:1092.
13. Proctor A, Toro-Vazquez JF. The Freundlich isotherm in studying adsorption in oil processing. *J Am Oil Chem Soc.* 1996;73:1627–33.
14. Foo KY, Hameed BH. Preparation, characterization and evaluation of adsorptive properties of orange peel based activated carbon via microwave induced K_2CO_3 activation. *Bioresour Technol.* 2012;104:679–86.
15. Youssef F, Mohamed G, Ismail S, Elzorba H, Galal A, Elbanna H. Synthesis, characterization and in vitro antimicrobial activity of florfenicol-chitosan nanocomposite. *Egypt J Chem.* 2021;64:941–8.
16. Abdel-Hamied M, Hassan RRA, Salem MZM, Ashraf T, Mohammed M, Mahmoud N, et al. Potential effects of nano-cellulose and nano-silica/polyvinyl alcohol nanocomposites in the strengthening of dyed paper manuscripts with madder: an experimental study. *Sci Rep.* 2022;12:19617.

- 921 17. El-Wakeel ST, Abdel-Karim A, Ismail SH, Mohamed GG. Development of Ag-
922 dendrites@ Cu nanostructure for removal of selenium (IV) from aqueous solution. *Water*
923 *Environ Res.* 2022;94:e10713.
- 924 18. Rahman ANA, Shakweer MS, Algharib SA, Abdelaty AI, Kamel S, Ismail TA, et al.
925 Silica nanoparticles acute toxicity alters ethology, neuro-stress indices, and physiological
926 status of African catfish (*Clarias gariepinus*). *Aquac Reports.* 2022;23:101034.
- 927 19. Hassan GK, Abdel-Karim A, Al-Shemy MT, Rojas P, Sanz JL, Ismail SH, et al.
928 Harnessing Cu@ Fe₃O₄ core shell nanostructure for biogas production from sewage
929 sludge: Experimental study and microbial community shift. *Renew Energy.*
930 2022;188:1059–71.
- 931 20. Gharib A, Maher MA, Ismail SH, Mohamed GG. Effect titanium dioxide/paraloid B.
932 72 nanocomposite coating on protection of treated Cu-Zn archaeological alloys. *Int J*
933 *Archaeol.* 2019;7:47.
- 934 21. Abdel Rahman AN, Ismail SH, Fouda MMS, Abdelwarith AA, Younis EM, Khalil
935 SS, et al. Impact of *Streptococcus agalactiae* challenge on immune response, antioxidant
936 status and hepatorenal indices of Nile tilapia: the palliative role of chitosan white poplar
937 nanocapsule. *Fishes.* 2023;8:199.
- 938 22. Liu Z, Zhang F-S, Wu J. Characterization and application of chars produced from
939 pinewood pyrolysis and hydrothermal treatment. *Fuel.* 2010;89:510–4.
- 940 23. Demirbas A. Heavy metal adsorption onto agro-based waste materials: a review. *J*
941 *Hazard Mater.* 2008;157:220–9.
- 942 24. Ahmad M, Rajapaksha AU, Lim JE, Zhang M, Bolan N, Mohan D, et al. Biochar as a
943 sorbent for contaminant management in soil and water: a review. *Chemosphere.*
944 2014;99:19–33.
- 945 25. Gao F, Qu J, Zhao Z, Wang Z, Qiu J. Nitrogen-doped activated carbon derived from
946 prawn shells for high-performance supercapacitors. *Electrochim Acta.* 2016;190:1134–
947 41.