



Upgrading the Characterization of Spent Fluid Catalytic Cracking Catalyst into Functional Adsorbent via Hybrid Ultrasonic-Chemical Approach

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Abstract: Fluid catalytic cracking catalysts (FCCCs) are essential in petroleum refining but become hazardous spent materials after deactivation due to heavy metal and coke deposition. Regenerating spent FCC catalysts (SFCCCs) is necessary to reduce hazardous waste and restore physicochemical properties. Conventional regeneration methods face limitations, such as high energy demand or potential structural damage, leaving a gap for more efficient and sustainable approaches. This study shows that a hybrid ultrasonic-chemical method can effectively regenerate SFCCCs, improving surface morphology, pore structure, and crystallinity, as evidenced by characterization using XRD, FTIR, BET, and SEM-EDS. The untreated SFCCCs exhibited a surface area of 27.255 m²/g, which increased to 33.508 m²/g after hybrid treatment and 34.344 m²/g after chemical treatment, indicating pore reopening and enhanced surface activity. FTIR analysis showed clearer Si-O-Si and -OH bands in hybrid-treated samples, suggesting improved recovery of the zeolite structure and reduced coke formation. XRD patterns showed sharper peaks in the 2 θ = 22–30° range, indicating greater crystallinity in hybrid regeneration than in untreated samples. SEM-EDS showed cleaner, more porous morphologies with reduced Fe (3.25%) and exposure of hidden Ni (9.49%) after hybrid treatment. These findings underscore the effectiveness of ultrasonic-assisted chemical regeneration, which offers a promising pathway for sustainable catalyst waste management and potential future reuse applications.

Keywords: Chemical regeneration; hybrid regeneration; petroleum refining; spent catalyst; ultrasonic

1. Introduction

Fluid Catalytic Cracking Catalysts (FCCCs) play a crucial role in various industries, especially in chemical processes such as petroleum refining [1]. Annually, it is estimated that 170.000-200.000 tons of spent catalysts are produced [2]. Once these catalysts reach the end of their operational lifespan, spent fluid catalytic cracking catalysts (SFCCCs) are categorized as hazardous and toxic waste due to their substantial heavy metal content, including iron, vanadium, nickel, magnesium, and sulfur, which poses a potential environmental risk [2-3]. Therefore, spent catalyst waste cannot be disposed of

without proper treatment, necessitating effective methods to recycle or regenerate SFCCCs while recovering their economic value.

Although considered hazardous waste, the spent catalysts have a significant potential for reuse. Typically, spent catalysts are used to enhance the pyrolysis of activated sludge in refinery waste [3], increase reactivity in cement production [4], and serve as an adsorption medium in wastewater treatment [5]. However, the treatment of spent catalysts poses challenges in identifying appropriate regeneration methods [6]. Several conventional methods have been developed to regenerate spent catalysts, including physical [7], chemical [8], and thermal methods [9]. Physical methods are known for their simple procedures and low operational costs. However, their effectiveness is limited, especially in removing strongly adsorbed contaminants from the catalyst surface [7]. Meanwhile, chemical methods are effective at removing various types of contaminants, but using large quantities of chemicals can lead to secondary pollution [10]. Thermal methods are also effective in removing organic contaminants, but require high energy consumption and pose the risk of damaging the catalyst structure, as well as incurring higher costs [11].

Several previous studies have evaluated the regeneration of spent catalysts using acid-based chemical treatments. Amalia et al. [11] reported that treatment of SFCCCs with sulfuric acid increased surface area by 110%, pore volume by 29.25%, and reduced the content of impurity metals such as Ni, V, and Fe. However, this method has limitations: it is only effective at dissolving surface metals and can lead to excessive dealumination, potentially compromising the stability of the zeolite structure. On the other hand, ultrasonically assisted regeneration, as reported by Pu et al. [12] It demonstrates cavitation's ability to remove coke deposits and material trapped in the catalyst's pores. However, these methods are not free of drawbacks, such as relatively high energy requirements and the risk of structural damage from cavitation. Based on the drawbacks of each approach, the application of an ultrasonic-chemical hybrid method becomes more relevant, as it combines the advantages of both: the effectiveness of ultrasonication in cleaning organic deposits and opening pores, and the ability of sulfuric acid to dissolve metal impurities.

Recent research has shown that combining chemical and ultrasonic methods is a promising approach [9]. Wang et al. [9] reported that ultrasonic waves can enhance the efficiency of chemical leaching processes through cavitation phenomena, resulting in more effective catalyst surface cleaning and shorter regeneration times. The combination of these two methods not only increases regeneration efficiency but also reduces chemical usage and minimizes the risk of damage to the catalyst structure, thereby creating a more environmentally friendly and sustainable solution for treating waste containing synthetic dyes and ammonia. Previous research has also shown that the application of ultrasonic-chemical hybrid methods can increase the effectiveness of adsorbent material regeneration. Gao et al. [13] reported that the combination of ultrasonic with persulfate successfully regenerated dye-saturated activated carbon with higher efficiency than the single method, while reducing chemical requirements. This reinforces that similar hybrid methods can be applied to other materials, including RFCC-based catalysts, to produce more environmentally-friendly and sustainable solutions. This research focuses on the effect of the regeneration method on the physicochemical characteristics of the spent catalyst. The results are expected to provide a fundamental understanding of how chemical and ultrasonic (hybrid) treatments can improve the material's surface morphology, pore structure, and functional groups, thereby laying the foundation for further applications in wastewater treatment. Furthermore, the reutilization of SFCCCs aligns with circular economy principles, creating added value from hazardous waste while promoting environmental sustainability.

2. Materials and Methods

2.1 Material

The SFCCCs used in this study were obtained from the RU IV Balongan Oil Refinery, Indonesia. The catalyst primarily consists of Y-type zeolite (faujasite structure) and contains several hazardous metals. Its composition is as follows: 8.17% C, 10.57% O, 0.71% Na, 26.64% Al, 41.23% Si, 0.33% K, 1.63% Ca, 4.83% Fe, and 5.88% Ni. The chemical and hybrid treatment method employed 98% sulfuric acid (H_2SO_4) (p.a. Merck, Germany).

2.2 Experimental apparatus

Figure 1 illustrates the experimental apparatus, which used an Ultrasonic Bath (50AL-AC220-240V, China) combined with a condenser to prevent material loss through evaporation. The ultrasonic bath generated ultrasonic waves that disrupted cell walls, thereby facilitating the efficient release of active compounds. The stand and clamps provided structural stability by securing the equipment in place. The condenser enabled repeated solvent circulation through solid samples, while the Erlenmeyer flask functioned as the final container for the extract solution.

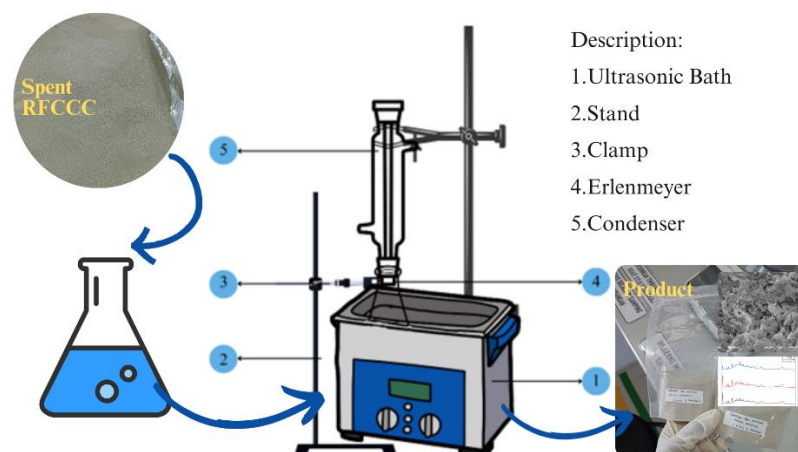


Figure 1. Ultrasonic bath apparatus

2.3 Method

2.3.1 Regeneration method of SFCCCs

The regeneration methods used for Spent Fluid Catalytic Cracking Catalysts (SFCCCs) were chemical methods and hybrid ultrasonic–chemical methods. Both treatments were carried out using H_2SO_4 as the solvent, with a solid-to-liquid ratio of 1:10 w/w. Before the SFCCCs treatment, the catalyst is first washed with deionized water to remove impurities, then dried in an electric oven (Memmert UN-30, Germany) at 105 °C for 7 hours. In the chemical method, the dried catalyst was immersed in a 0.7 M sulfuric acid solution and stirred using an orbital shaker (Biobase, China) at room temperature (30 ± 2 °C) for 8 hours. Meanwhile, in the hybrid ultrasonic–chemical method, the catalyst was immersed in a 0.7 M sulfuric acid solution and treated using an ultrasonic bath (Walfront, China) at a temperature of 70 °C for 30 minutes and power of 250 W. After the regeneration process using either the chemical or hybrid ultrasonic–chemical method, the catalyst was filtered and washed repeatedly using demineralized water until it reached a neutral pH (pH 7), then dried again in an electric oven at 105 °C for 7 hours, and finally sieved to a particle size of 150 mesh (100 μm). The regenerated catalyst was then calcined in a furnace (Thermo Fisher, USA) at 550 °C for 4 hours.

2.3.2 Characterization method

The non-treated spent FCC catalysts were denoted as SFCCCs, SFCCCs-K was defined as the chemically treated SFCC catalysts, and SFCCCs-H was defined as the ultrasound bath-treated SFCC catalysts. After the regeneration process, a series of characterization methods was used to comprehensively characterize the material's properties. X-ray Diffraction (XRD; Rigaku Miniflex-600, Japan) was used to determine the crystal structure. By analyzing the most intense peak broadening using the Scherrer equation, the crystal size was determined, which served as the basis for the subsequent characterization stage [14]. Fourier further analyzed the identified crystal structure using transform infrared spectroscopy (FTIR; Agilent Cary 630, USA). This method enabled exploration of chemical bonds and molecular structures, providing deeper insights into chemical interactions within the material [15]. To understand the surface characteristics in more detail, the Brunauer–Emmett–Teller (BET) method (Quadrascorb-evo, US) was employed. This technique enabled the determination of the surface area of nanostructures based on the principle of physical gas adsorption, which was directly related to the material's adsorption capacity [16]. The final stage of characterization involved a

combination of scanning electron microscopy and energy-dispersive spectroscopy (SEM & EDS; JEOL JSM-6510LA, Japan). This method visualized the surface morphology and semi-quantitatively analyzed the elemental composition of the material. The SEM-EDS results not only complemented the previous characterization but also provided a comprehensive visual and elemental understanding of the regeneration mechanism.

3. Results and Discussion

3.1 Identification of SFCCCs surface structure recovery based on FTIR Spectra

Infrared spectroscopy (FTIR) was used to evaluate changes in the surface chemical structure of SFCCCs before and after regeneration. Three sample conditions were analyzed, with the results shown in **Figure 2**.

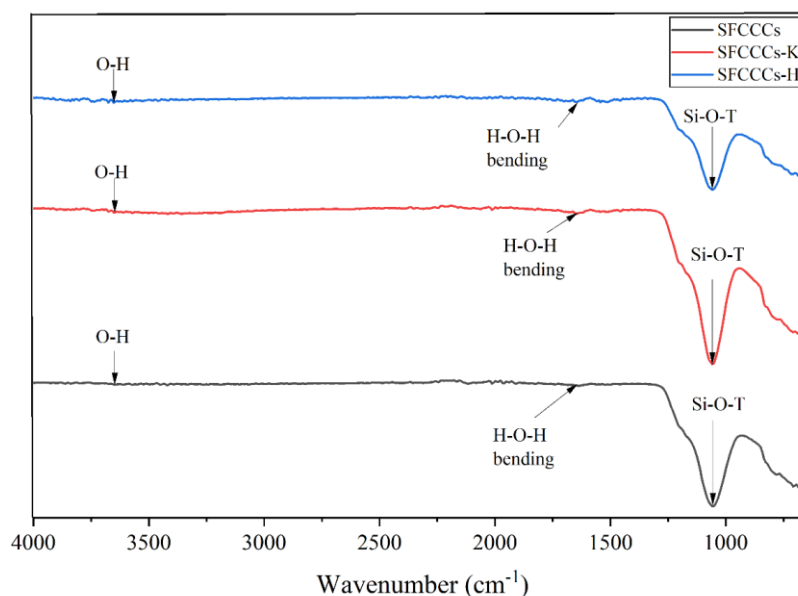


Figure 2. FTIR Spectra of SFCCCs

The FTIR spectra of the three samples in **Figure 2** show differences that reflect the effectiveness of each regeneration method. In the SFCCCs sample without regeneration, the absorption peak in the 1040–1100 cm^{-1} region (Si-O-T group) appears to be weakened, indicating damage to the zeolite structure due to heavy-metal contamination and carbon impurities (coke). This is consistent with EDS data showing carbon (C) content of 8.17%, as well as Fe and Ni metals at 4.83% and 5.88%, respectively, indicating a high level of fouling. The maximum transmittance value (%T) in SFCCCs is 100.31%, indicating high IR absorption by surface contaminants. This phenomenon is consistent with the results of Wang et al. [7], who stated that the accumulation of carbon and heavy metals on the catalyst surface causes a decrease in catalytic activity and the closure of active pores, thereby reducing the efficiency of absorption and distribution of infrared (IR) in the zeolite structure [6].

After chemical regeneration (SFCCCs-K), the intensity of the Si-O-T and Si-O-Si bands increased, indicating the beginning of zeolite structure recovery. The -OH band was also more clearly visible at ($\sim 3400 \text{ cm}^{-1}$) and H_2O ($\sim 1600 \text{ cm}^{-1}$) with moderate intensity. The appearance of a band at 1380 cm^{-1} indicated the presence of residual acid solvent from the leaching process. The EDS results show a decrease in Fe and Ni content to 2.02% and 4.88%, respectively, indicating the successful heavy-metal leaching. The increase in carbon (C) to 18.49% indicates that the remaining coke has not been completely removed. The intensity value (100.87%) indicates a very clean surface, while the low IR absorption suggests that the zeolite structure has not been fully restored. This is supported by the research of Lu et al. [18], which states that acid leaching can improve the zeolite structure, increase crystallinity, and open pores, although it does not completely remove carbon from the surface [17].

The most optimal results were shown by the SFCCCs-H sample, whose FTIR spectrum showed the sharpest and strongest peaks, particularly in the Si-O-T and Si-O-Si bands, indicating that the crystalline structure of the zeolite had been successfully restored to the maximum extent. In addition, an increase in the intensity of the -OH and H₂O bands was also clearly observed, indicating that the catalyst pores had reopened and were able to adsorb water more effectively. This demonstrates that the cavitation effect of ultrasonic waves can break down impurities in the pores, improve diffusion, and expand access to active sites within the catalyst structure [19]. The carbon content, as determined by EDS, decreased from 18.49% (SFCCCs-K) to 10.05% in SFCCCs-H, indicating that coke and organic contaminants were removed more effectively. Although the Ni content increased slightly to 9.49%, this is likely due to the opening of internal pores, making previously hidden metals easier to detect. The presence of minor elements such as Ti (0.42%) and Mg (0.16%) also suggests that the cleaning process reached the deepest pores of the catalyst surface. The maximum transmittance value in SFCCCs-H (100.84%) was slightly lower than that in SFCCCs-K (100.87%), but this may indicate that the intensity of the FTIR absorption band increased due to the presence of active groups that absorb IR more strongly. This means that the surface of the SFCCCs-H catalyst is clean but structurally active. This is consistent with Dewes et al. [20] who explain that ultrasonic waves produce cavitation effects, shear forces (mass transfer), and microjets capable of releasing heavy metals from the surface, opening pores, and increasing diffusion and adsorption into the zeolite structure [18]. Considering the FTIR data, %T value, zeolite structure recovery, and surface cleaning effectiveness, SFCCCs-H is declared the best regeneration method. The regenerated catalyst has an active, clean structure and has high potential for reuse in wastewater treatment or other applications.

3.2 Surface area of adsorbent

BET characterization was performed to determine changes in the physical properties of SFCCCs after the regeneration process. According to Table 1, the surface area of SFCCCs increased after regeneration. Untreated SFCCCs had a lower surface area of 27.255 m²/g, while after hybrid treatment (SFCCCs-H), it increased to 33.508 m²/g, and after chemical treatment, it reached 34.344 m²/g. This indicates that the regeneration process was able to open previously clogged pores and expand the active surface area [19]. The difference between the adsorption and desorption pore diameters occurred due to the hysteresis effect, which is the mismatch between the path when the gas is absorbed (adsorption) and when the gas is released again (desorption). This hysteresis mainly occurs in mesoporous materials, such as zeolite Y or silica, due to capillary condensation within the pores, which occurs when small pores begin to fill with liquid nitrogen under capillary pressure [20].

Table 1. Characteristics of SFCCCs Pore Structure

Catalyst	Adsorption		Desorption		Reff
	Surface area (m ² /g)	Pore diameter dV(d) (Å)	Surface area (m ² /g)	Pore diameter dV(d) (Å)	
SFCCCs	27.255	48.514	47.730	38.340	This study
SFCCCs-K	34.344	33.910	57.133	38.508	This study
SFCCCs-H	33.508	33.880	55.296	38.040	This study
SFCCCs with 5% C ₂ H ₂ O ₄	91.02	0.1248 cm ³ /g	82.48	0.1383 cm ³ /g	[12]
SFCCCs with H ₂ SO ₄	74.98	-	113.75	-	[9]
SFCCCs-M with 0.8–1.6 mol/L hydrochloric acid	91.9	197.1	-	-	[21]

The initial SFCCCs sample (untreated) showed the lowest surface area, namely adsorption (27.255 m²/g) and desorption (47.730 m²/g), with the largest adsorption pore diameter (48.514 Å) and desorption pore diameter (38.340 Å). This indicates that most pores remain open, but the surface area is low due to blockages in the fine pores (micro- and mesopores) caused by the accumulation of impurity metals such as Fe and Ni.

These heavy metal contaminants can clog the pore surface, thereby accelerating the FCC catalyst's deactivation. According to Thommes et al. [21], in the International Union of Pure and Applied Chemistry (IUPAC) standard, the large difference in surface area and pore diameter in adsorption and desorption is known as a hysteresis isotherm, which occurs because the mechanism of filling and emptying nitrogen in the pores is not symmetrical [19]. As a result, the average pore size measured in desorption tends to be smaller. Hybrid regeneration resulted in an increase in surface area to 33,508 m²/g and a decrease in pore size to 33,880 Å. The primary mechanism for this increase was the cavitation effect of ultrasonic waves, which caused implosions of cavitation bubbles in the solution, thereby accelerating the dissolution of impurity metals from the catalyst's surface and pores [10]. The combination of ultrasound and acid can enhance heavy metal leaching efficiency and significantly reduce regeneration time compared to conventional methods, thanks to improved mass transfer and cavitation effects. Similar results were found in a study by Marafi et al. [24], which showed that an increase in convective diffusivity can occur under these conditions [22]. Although the pore diameter decreases, this is not an indication of damage, but rather the opening of previously closed small pores (micropores) that were previously blocked by metal and carbon.

Chemical treatment without ultrasonic assistance provides an adsorption surface area (34,344 m²/g) and desorption (57,133 m²/g) with chemical SFCCCs diameter almost similar to hybrid SFCCCs, adsorption pore diameter (33,910 Å), and desorption pore diameter (38,508 Å). With results that are almost identical to those of SFCCCs-H, this indicates that the difference in effectiveness between the chemical and hybrid methods is not quantitatively significant. However, in terms of regeneration time, the hybrid method is much more efficient and avoids the risk of zeolite structural damage from prolonged exposure to strong acids. Based on the analysis results, this study is consistent with Amalia et al. [11], indicating that the dissolution of impurity metals proceeds effectively. However, the use of acid solutions at high concentrations or extreme reaction temperatures, combined with longer process times, has the potential to damage the zeolite structure [10].

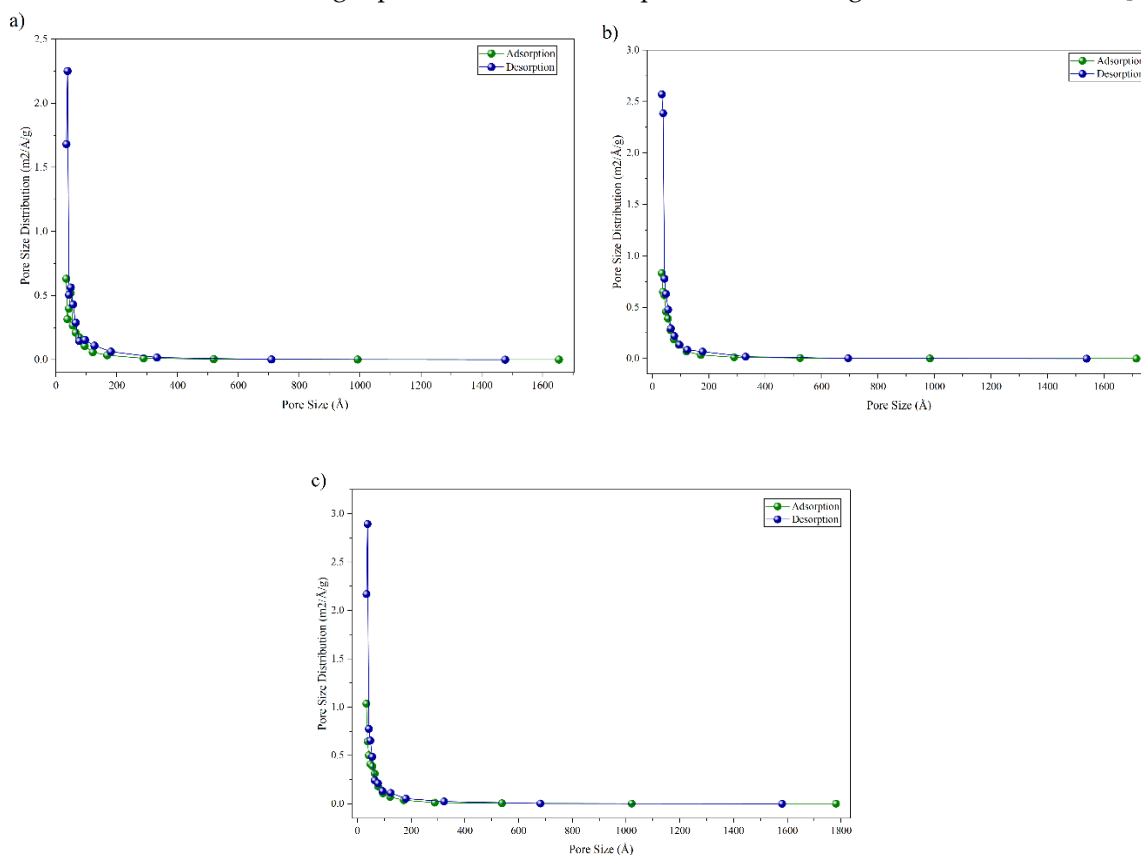


Figure 3. Pore Size Distribution of Modified SFCCCs Based on BET Analysis: (a) SFCCCs, (b) SFCCCs-K, (c) SFCCCs-H

In general, the three graphs in **Figure 3** are dominated by pores smaller than 200 Å, indicating mesoporous characteristics. In untreated SFCCCs, the pore distribution appears low and narrow, indicating pore blockage due to metal accumulation during use, which affects diffusion capacity and catalyst activity. The hysteresis between the adsorption and desorption curves is also large, indicating asymmetrical or blocked pores[23]. After regeneration with the hybrid method, pore distribution increased sharply, especially at sizes ≤ 100 Å. The adsorption and desorption curves became more parallel, indicating more open and regular pores. The synergistic effect of ultrasonication and sulfuric acid accelerated metal release via dissolution and complexation, thereby efficiently removing pore blockers, increasing surface area, pore openness, and a more uniform pore distribution [24]. Chemical regeneration also improved pore distribution, although the curve was slightly more dispersed, suggesting a risk of changes in pore structure under the strong acidic conditions. Overall, hybrid regeneration provides the most optimal results because it can open pores efficiently, maintain structural stability, and minimize the risk of material damage, making it a superior method for restoring the characteristics of SFCCCs.

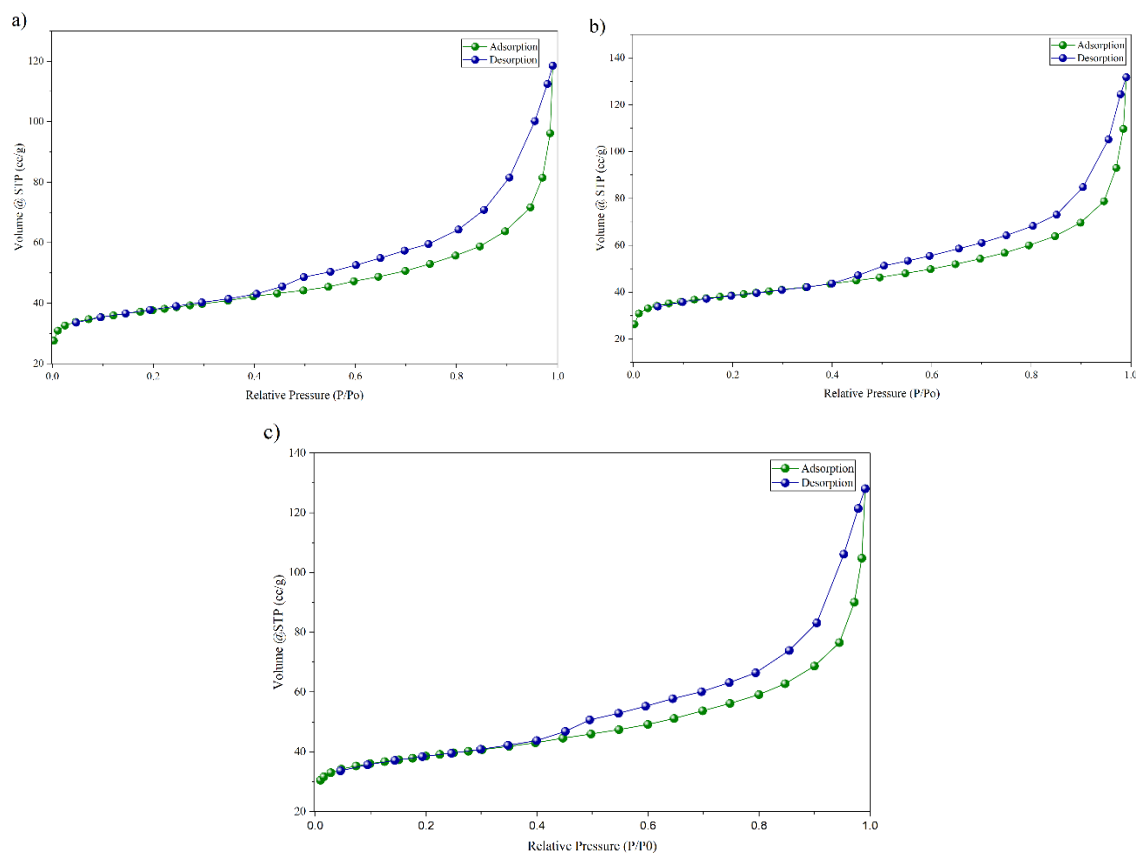


Figure 4. Adsorption-Desorption Isotherm Curves for SFCCCs Samples: (a) SFCCCs, (b) SFCCCs-K, (c) SFCCCs-H

The three curves in **Figure 4** show type IV isotherms with hysteresis loops, which are characteristic of mesoporous materials. The presence of pores ranging in size from 2 to 50 nm, a size commonly used as an adsorbent. The hysteresis loop formed between the adsorption and desorption curves indicates the presence of capillary condensation within the mesopores, which is an indicator of open pore structure and inter-pore connectivity. In the three **Figures 4. (a), (b), and (c)**, it can be seen that the adsorption volume increases sharply at a relative pressure approaching 1 (P/P_0), which indicates the formation of multilayers or significant pore filling[6]. At **Figure 4. (a)**, the adsorption and desorption volumes reached approximately 120 cc/g with wide and non-parallel hysteresis, especially at relative pressures ($P/P_0 > 0.4$). This indicates that SFCCCs experience pore-blocking or bottleneck effects due to the formation of coke and metal deposits, solid residues from the heating of organic materials that clog and coat the catalyst's pores, making it difficult for the catalyst to decompose and inhibiting access to the pores. This is consistent with the adsorption surface area obtained (27.255 m²/g). The hysteresis loop area indicates limited pore connectivity, and the total adsorption volume

appears lower than that of the two chemically and hybridly regenerated samples. The same findings in the study by Ihli et al. [27] explain that this phenomenon occurs due to pore closure and nitrogen inaccessibility resulting from carbon deposition or accumulation of impurity metals [27]. The adsorption volume increased to 130 cc/g in **Figure 4. (b)**, the adsorption and desorption curves appear more parallel with narrower hysteresis compared to SFCCCs, indicating that hybrid treatment can increase pore accessibility and total pore volume in the catalyst. Ultrasound plays an important role by producing a cavitation effect that can reopen clogged pores and disperse particle agglomerates. As a result, gas diffusion pathways such as nitrogen become more open, thereby significantly increasing adsorption capacity. This increase also indicates that the mesoporous structure, which was previously clogged by metal or coke deposition, can be restored, supporting the recovery of catalyst structural properties after hybrid regeneration treatment [25]. The highest adsorption volume increase was close to 135 cc/g in **Figure 4. (c)** SFCCC-K with a value not much different from SFCCCs-U but wider, indicating the effectiveness of chemical regeneration in cleaning some of the pores, but without the mechanical assistance of ultrasound, this process was less than optimal in removing carbon residues trapped in the deepest pores[9]. This is also supported by the high surface area of Chemical SFCCCs (57,133 m²/g). These differences indicate that hybrid regeneration can significantly unclog pores and restore the catalyst's internal structure, a process that cannot be achieved with conventional chemical methods alone.

3.3 XRD Diffraction Patterns as an Indicator of SFCCCs Regeneration Success

From the diffraction pattern, it can be determined to what extent the regeneration method restores or enhances the active crystal structure, particularly the Zeolite Y phase, which plays a crucial role in catalytic activity. The XRD results are shown in **Figure 5**.

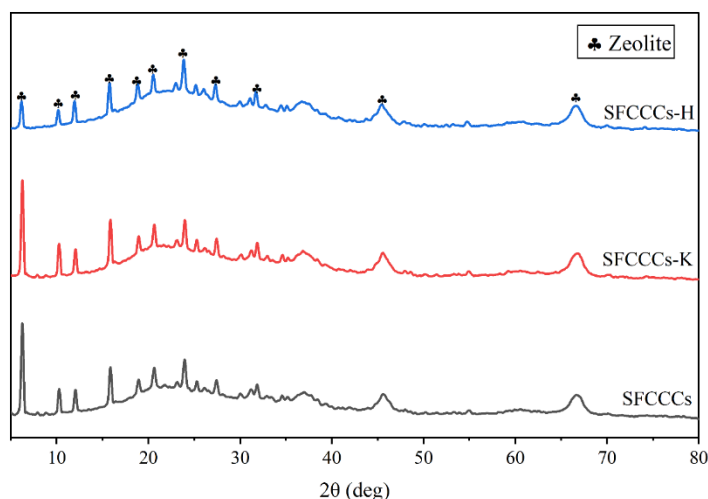


Figure 5. Crystal Structure of SFCCCs Based on XRD Analysis: (a) SFCCCs, (b) SFCCCs-H, (c) SFCCCs-K

Based on the results of X-Ray Diffraction (XRD) analysis in **Figure 5**, the untreated sample (SFCCCs) shows relatively strong diffraction peaks, which may be attributed to the presence of residual crystalline phases and deposited metals, both of which contribute to peak intensity. The identified phases were mostly amorphous silica, kaolinite, and residual zeolite phases that were no longer dominant, indicating that the zeolite crystal structure had been severely damaged and was no longer effective for use in catalytic applications[17]. In the ultrasonically regenerated sample, **Figure 5. (b)** SFCCCs-H, slight variations in peak intensity and peak broadening were observed. These changes suggest structural modification and partial removal of impurities rather than a straightforward increase in crystallinity. According to Dewes et al. (2022), ultrasound induces cavitation, improving mixing and accelerating crystallization, thereby accelerating zeolite crystal formation. In addition, ultrasonic waves accelerate mass transfer and crystal nucleation during zeolite synthesis. This shortens zeolite formation time by 30-40% and produces zeolite with a more uniform particle size and higher crystallinity [20]. The main phases are indicated by the reappearance of active phases, such as silica and aluminosilicate, identified in this sample, demonstrating the success of the hybrid method in

partially restoring the active structure of SFCCCs. Meanwhile, in regeneration using the chemical method (Figure 5). (c) SFCCCs-K, the diffraction pattern in this sample shows sharp and well-defined peaks, but the intensity of some diffraction peaks is lower than that of the SFCCCs-H sample. This may be due to an overly aggressive chemical leaching process that can dissolve part of the aluminosilicate zeolite framework, especially under extreme pH or high-acid conditions. This is in line with Christopher et al. (2019), who stated that although chemical regeneration effectively removes contaminants, inappropriate selection of leaching concentration and duration can cause excessive dissolution of the zeolite framework and reduce its structural stability [29]. The intense peaks of the zeolite and SiO phases appearing in the XRD pattern indicate that the catalyst's crystallinity has increased compared to the initial conditions. Overall, hybrid regeneration treatment (SFCCCs-H) is the most effective method for optimally restoring the crystal structure of SFCCCs. This method is not only capable of cleaning contaminant deposits but also of maintaining the integrity of the zeolite structure.

3.4 Morphology of SFCCCs Microstructure After Regeneration through SEM-EDS

To observe the effect of regeneration on SFCCCs, SEM was performed on samples treated with ultrasonic and chemical treatments. The results of the analysis are shown in Figure 6.

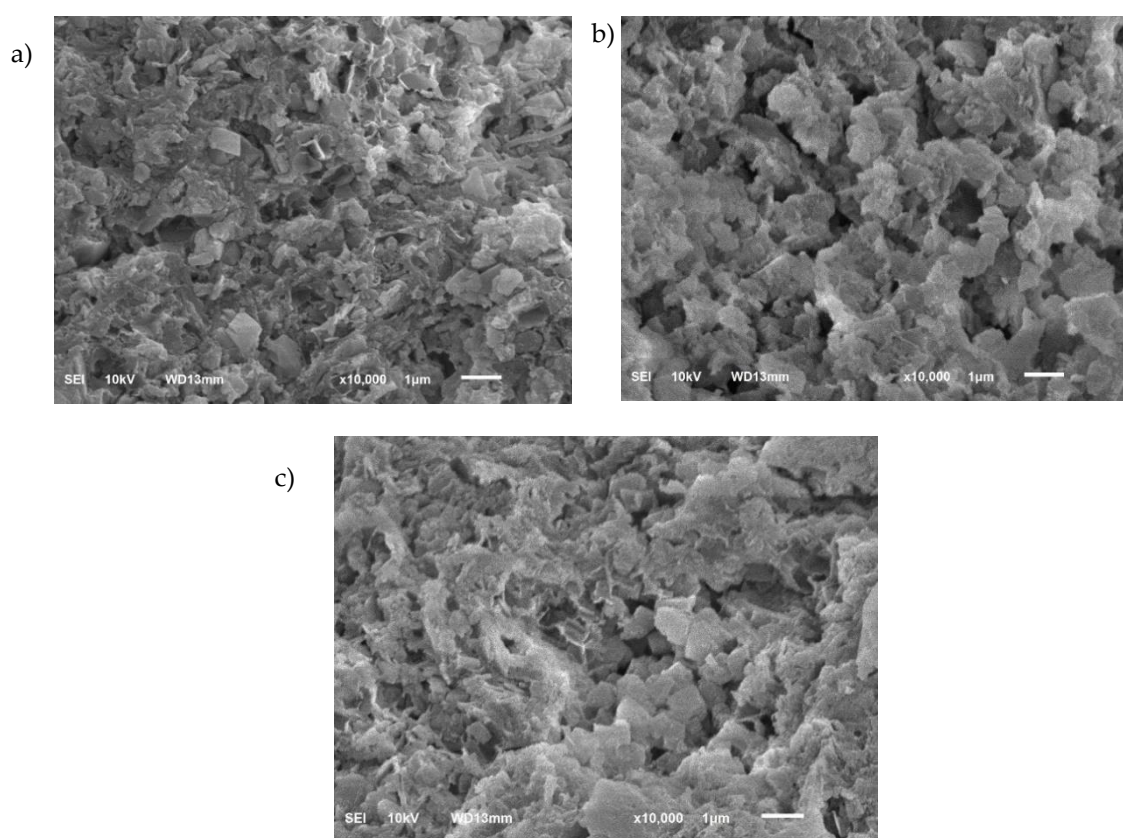


Figure 6. SEM micrographs of SFCCCs at 10,000x magnification (a)SFCCCs, (b)SFCCCs-K, (c)SFCCCs-H

The untreated SFCCCs (Figure 6. (a)) show a relatively dense surface with several pores partially covered by deposits, indicating the presence of coke and metal contaminants accumulated during catalytic operation. After regeneration (Figure 6. (b)), the surface appears relatively cleaner with some visible pores; however, the overall morphology remains similar to the untreated sample. No significant structural damage or erosion can be conclusively identified from the SEM images. Al_2O_3 in the catalyst can dissolve as acidity increases, triggering the collapse of the catalyst framework and reducing its durability [18]. In comparison, the Hybrid SFCCCs in Figure 6(c) show changes in the surface structure, with cleaner areas and less sharp grooves appearing. This is related to the cavitation phenomenon induced by ultrasonic waves, indicating a more porous structure and a much looser morphology [30]. The effectiveness of this method is listed in Table 1, which shows a desorption surface area of $55.269 \text{ m}^2/\text{g}$, only slightly lower than the chemical method, but with

a smaller pore diameter of 38.040 Å. This demonstrates that the hybrid method can reopen clogged micropores without degrading the catalyst framework, thereby increasing surface area and enhancing catalytic performance. Cavitation causes microcracks on the solid surface and increases the specific surface area, thereby directly increasing leaching efficiency and shortening reaction time.

Table 2. Elemental Analysis Before and After Regeneration

Element Mass (%)	Catalyst		
	SFCCCs	SFCCCs-H	SFCCCs-K
Si/Al	1.55	0.88	1.75
Al	26.64	36.69	23.78
Si	41.23	32.31	41.52
C	8.17	10.05	18.49
O	10.57	6.68	7.97
Na	0.71	0.46	0.37
Mg	-	0.16	-
K	0.33	0.06	0.10
Ca	1.63	0.45	0.86
Ti	-	0.42	-
Fe	4.83	3.25	2.02
Ni	5.88	9.49	4.88

During the cracking reaction, the surface of FCCC catalyst particles can be contaminated by various cations, including Ni, V, Fe, Cu, and/or Na, as well as by carbon deposits (coke) at levels of ppm or higher [31]. Based on Table 2 and Figure 7, the EDS results support the observations with quantitative elemental composition data. In SFCCCs, the C content of 8.17% and O content of 10.57% indicate the presence of organic deposits (coke) and oxide compounds, while the Ni content of 5.88% and Fe content of 4.83% indicate the presence of impurity metals from the previous process. The initial Si/Al ratio of 1.55, with 41.23% Si and 26.64% Al, indicates a silica-alumina-based catalyst framework. After ultrasonic regeneration of SFCCCs-H, the carbon content increased slightly to 10.05%, but the oxygen content decreased to 6.68%, accompanied by a decrease in Fe to 3.25% and an increase in Ni to 9.49%. This increase occurred due to the removal of impurities from the catalyst surface, exposing Ni particles that had previously been hidden and detected by EDS [27]. The Si/Al ratio decreased to 0.88 (Si 32.31%; Al 36.69%), indicating the possibility of selective Si dissolution due to ultrasonic treatment. This is in accordance with Sariman [33], who stated that the decrease in Si elements occurred due to silica leaching from the catalyst framework, caused by local pressure cavitation waves, especially in the amorphous phase or in parts that were already weak due to previous use [33]. Meanwhile, chemical regeneration (SFCCCs-K) produced the highest C content of 18.49% and O of 7.97%, indicating that carbon residue was still quite high. The Fe content decreased to 2.02% and the Ni content to 4.88%, indicating a highly effective metal-cleaning process. The Si/Al ratio increased to 1.75 (Si 41.52%; Al 23.78%), likely due to aluminum dissolution (dealuminization) by the chemical solution, resulting in reduced Al content in the framework.

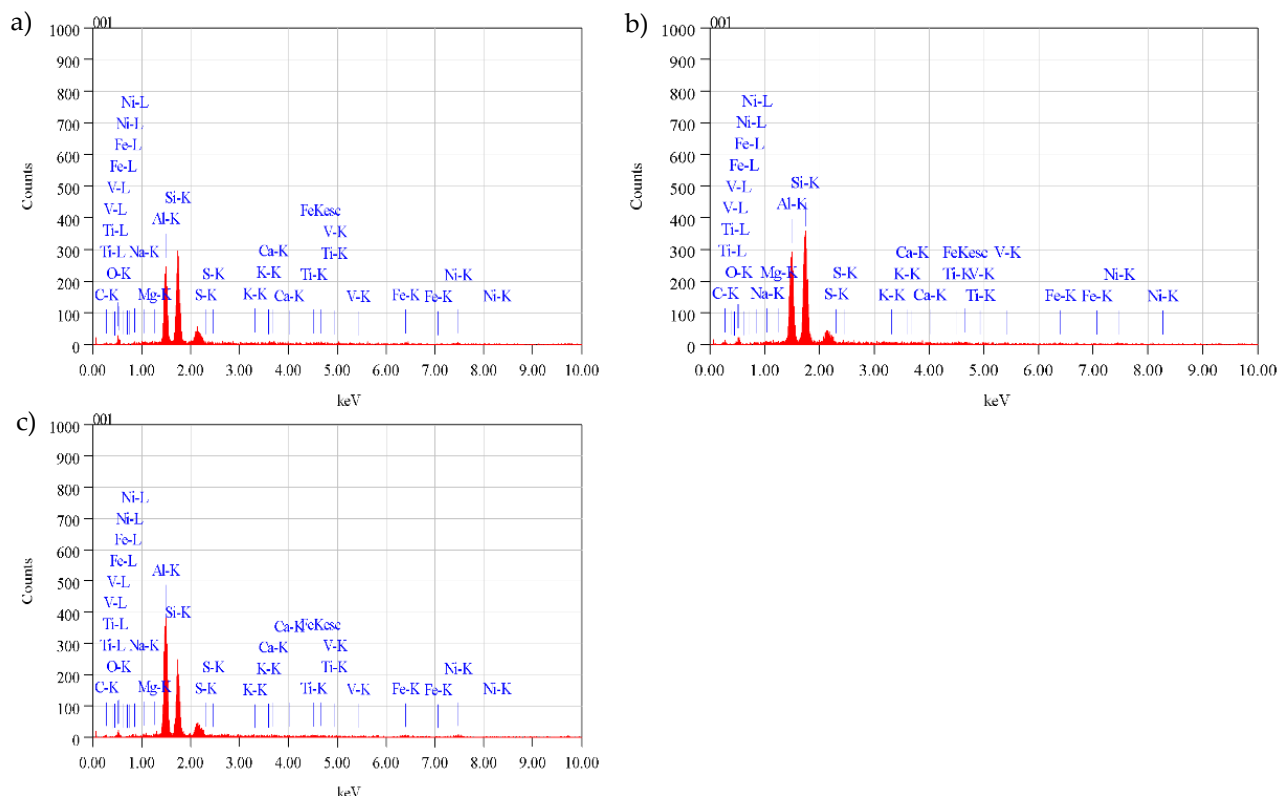


Figure 7. Elemental mapping from SEM-EDS (a)SFCCCs, (b)SFCCCs-K, (c)SFCCCs-H

Overall, SEM and EDS observations show that ultrasonic regeneration is more effective in opening pores and reducing certain impurity metals. In contrast, chemical regeneration is more efficient in removing metals but tends to leave carbon residues and alter the Si/Al ratio. These findings align with those of Sulman et al. [34], who stated that ultrasonic treatment excels at morphological restoration and surface area enhancement, while chemical treatment emphasizes chemical composition recovery, with the risk of altering the catalyst framework structure [34]. Ultrasound-assisted leaching, or this hybrid method, has great potential to improve leaching efficiency, shorten process time, and reduce chemical consumption. This is due to the advantages of ultrasonic irradiation over conventional mechanical agitation, particularly in accelerating reactions and improving contact between the solvent and solid particles.

4. Conclusions

The regeneration of spent fluid catalytic cracking catalysts (SFCCCs) using chemical and hybrid ultrasonic-chemical methods demonstrated significant improvements in physicochemical properties. BET analysis showed that the surface area increased from 27.255 m²/g for SFCCCs to 34.344 m²/g for SFCCCs-K and further to 33.508 m²/g for SFCCCs-H, indicating successful pore reopening and enhanced adsorption capacity. FTIR spectra revealed stronger Si–O–Si and –OH bands in SFCCCs-H, confirming more effective restoration of the zeolite framework and reduction of coke residues, with carbon content decreasing from 18.49% for SFCCCs-K to 10.05% for SFCCCs-H. XRD results showed sharper, more intense peaks in the 2θ = 22–30° range for SFCCCs-H, indicating improved crystallinity compared to SFCCCs and SFCCCs-K. SEM-EDS analysis further confirmed structural recovery, where Fe content decreased from 4.83% for SFCCCs to 3.25% for SFCCCs-H, while surface morphology appeared cleaner and more porous under ultrasonic-assisted regeneration. Overall, the hybrid ultrasonic-chemical method yielded the most balanced results, effectively removing impurities, reopening micropores, and restoring crystallinity while minimizing structural damage, thereby making it superior to conventional chemical regeneration for sustainable SFCCC reuse. Moreover, regenerated SFCCCs have strong potential as low-cost adsorbents for wastewater treatment.

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