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ORIGINAL ARTICLE

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# Reactive extrusion process to obtain a multifunctional fiber-rich ingredient from coffee hull

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**ABSTRACT:** Brazil, known for its robust agricultural sector, generates substantial amounts of lignocellulosic residues annually. The valorization of these residues as ingredients for the food industry aligns with environmental, social, and economic sustainability goals. Coffee hull, a by-product of the coffee industry, holds potential as a source for creating high-value food ingredients. This study aimed to develop a multifunctional, fiber-rich ingredient from the coffee hull using a one-step reactive extrusion process with either alkaline hydrogen peroxide or sulfuric acid. The coffee hull was subjected to extrusion with sulfuric acid (1% and 3%) or alkaline hydrogen peroxide (1% and 3%) in these one-step processes. The resulting materials were then analyzed for their physicochemical and techno-functional properties. All treated samples demonstrated an increased cellulose and insoluble dietary fiber content, along with enhanced porosity, as observed through scanning electron microscopy. The sample treated with 3% alkaline hydrogen peroxide (AHP3) exhibited the highest levels of cellulose (45.17%), insoluble dietary fiber (78.20%), density (3.54), water absorption capacity (1.96 g/g), and oil absorption capacity (0.86 g/g). Furthermore, all samples displayed thermal stability from room temperature up to 300 °C, and a reduction in crystallinity indexes was noted after treatment. The reactive extrusion method proved effective in modifying the coffee hull, offering a green approach to producing high-value products from lignocellulosic waste. This method presents several advantages, including short reaction times, low reagent concentrations, minimal or no effluent generation, and potential scalability for industrial scale.

**Keywords:** alkaline hydrogen peroxide; coffee hull; insoluble fibers; sulfuric acid.

# *Processo de extrusão reativa para obter um ingrediente multifuncional rico em fibras a partir da casca de café*

**RESUMO:** *O Brasil, conhecido por seu robusto setor agrícola, gera quantidades substanciais de resíduos lignocelulósicos anualmente. A valorização desses*

*resíduos como ingredientes para a indústria alimentícia está alinhada com as metas de sustentabilidade ambiental, social e econômica. A casca de café, um subproduto da indústria cafeeira, tem potencial como fonte para a criação de ingredientes alimentícios de alto valor agregado. Este estudo teve como objetivo desenvolver um ingrediente multifuncional e rico em fibras a partir da casca de café, usando um processo de extrusão reativa de uma etapa com peróxido de hidrogênio alcalino ou ácido sulfúrico. A casca de café foi submetida à extrusão com ácido sulfúrico (1% e 3%) ou peróxido de hidrogênio alcalino (1% e 3%) em processos de uma etapa. Os materiais resultantes foram então analisados quanto às suas propriedades físico-químicas e tecnofuncionais. Todas as amostras tratadas demonstraram um aumento no teor de celulose e fibra alimentar insolúvel, juntamente com maior porosidade, conforme observado por microscopia eletrônica de varredura. A amostra tratada com 3% de peróxido de hidrogênio alcalino (AHP3) exibiu os maiores teores de celulose (45,17%), fibra alimentar insolúvel (78,20%), densidade (3,54), capacidade de absorção de água (1,96 g/g) e capacidade de absorção de óleo (0,86 g/g). Além disso, todas as amostras apresentaram estabilidade térmica da temperatura ambiente até 300 °C, e uma redução no índice de cristalinidade foi observada após o tratamento. O método de extrusão reativa provou ser eficaz na modificação da casca de café, oferecendo uma abordagem verde para produzir produtos de maior valor agregado a partir de resíduos lignocelulósicos. Esse método apresenta várias vantagens, incluindo tempos de reação curtos, baixas concentrações de reagentes, geração mínima ou nenhuma de efluentes e possibilidade de escalonamento para escala industrial.*

**Palavras-chave:** *ácido sulfúrico; casca de café; fibras insolúveis; peróxido de hidrogênio alcalino.*

## 1 Introduction

Brazil is the largest coffee producer in the world. In the 2022/2023 period, Brazil was expected to produce 62.6 million bags (60 kg), accounting for about one-third to half of the world's coffee production, which was estimated to reach approximately 167.5 million bags (60 kg) (Periyasamy *et al*., 2022; USDA, 2022). Additionally, Brazil is the largest coffee exporter globally and ranks second among coffee-consuming countries (ABIC, 2021).

During coffee bean processing, the residues generated constitute approximately 30%-50% of the production. This implies that the volume of processed coffee can be similar to the amount of residues produced (Peshev *et al*., 2018). Coffee hulls are lignocellulosic materials obtained from the dry processing of coffee that retains the outer peel, coffee berry's pulp, and parchment of the coffee berry (Arya *et al*., 2022). Their chemical composition includes lignin (38%), cellulose (28%), hemicellulose (25%), proteins (8%-11%), ash (3%-7%), lipids (1%-3%), and caffeine (1%) (Ávila; Martins; Goldbeck, 2021; Oliveira *et al*., 2021; Pandey *et al*., 2000; Periyasamy *et al*., 2022).

According to Hassan, Williams and Jaiswal (2018), coffee hulls are mainly used as compost, vermicompost, livestock feed, and household fuel. In some instances, they are discarded into the environment. However, they represent a valuable raw material for obtaining fiber-rich ingredients for the food industry.

Traditionally, the physiological effects of coffee and coffee processing residues have focused primarily on their caffeine content, often overlooking other components present in coffee, such as polysaccharides and dietary fibers. These components have several beneficial physiological effects on health, including reducing the risk of coronary heart disease, and diabetes, and aiding in weight control (Santos; Silva; Pintado, 2022).

Introducing fiber-rich products into the food industry, in the form of flours or additives, can be considered a promising strategy. They offer multifunctional roles, enhancing techno-functional properties, such as being used as thickeners, gelling agents, fillers, and water-retaining agents. Moreover, incorporating these residues into a food products can contribute to the eco-sustainability of the coffee industry within a circular economy, minimizing negative environmental and economic impacts resulting from the improper disposal of large amounts of residues and by-products (Difonzo *et al*., 2022; Iriondo-Dehond; Iriondo-Dehond; Castillo, 2020).

In recent years, several studies have reported the modification of agro-industrial residues using chemical, physical, and enzymatic methods, or their combination, aimed at improving their nutritional and techno-functional properties. This approach has been particularly effective in increasing dietary fiber content and enhancing technological characteristics such as water absorption capacity (Galdeanol; Grossmann, 2006; Van Buggenhout *et al*., 2015; Yoshida; Prudencio, 2020).

Alkaline peroxide has been identified as an efficient reagent for the modification of lignocellulosic residues, acting as an effective agent for delignification and hemicellulose solubilization, thereby increasing the cellulose content of samples. This effectiveness is due to the formation of the hydroperoxide anion (HOO–) – in alkaline pH, which is the main active species in peroxide. In contrast, hydrogen peroxide is unstable under alkaline conditions and decomposes into hydroxyl (OH–) and superoxide (O2–) radicals. These radicals are responsible for oxidizing the lignin structure, attacking hydrophilic (carboxyl) groups, breaking some bonds, and eventually dissolving lignin and hemicellulose (Yoshida; Prudencio, 2020). Schmitz *et al*. (2021) emphasized that alkaline hydrogen peroxide is a promising bleaching agent for retaining the composition of lignocellulosic materials, making them suitable as food ingredients. Xia *et al*. (2022) reported on the effect of alkaline hydrogen peroxide treatment on the delignification of corn stalks, indicating that oxidative treatments could be considered promising strategies for biomass utilization due to their excellent performance and cost-effectiveness.

Similarly, acid treatments can be used to derive new materials from lignocellulosic residues, as they act by hydrolyzing hemicelluloses and removing soluble lignin, resulting in increased cellulose content (Pereira; Marim; Mali, 2022; Tu; Hallett, 2019). Acidic pretreatment can be performed using either dilute acid at elevated temperature or concentrated acid at lower temperatures. Typically, dilute acid treatment involves adding an acid solution (0.2%-2.5%, w/w) to the biomass with regular mixing at temperatures ranging from 120 ºC to 210 °C (Periyasamy *et al*., 2022).

Extrusion cooking combined with chemical treatment can promote the delignification of the material and decrease the crystallinity of cellulose by breaking the hydrogen bonds that maintain its supramolecular structure. This results in materials with higher water absorption capacity and improved sensory properties (Cardoso *et al*., 2016; Vilela *et al*., 2016; Yoshida; Prudencio, 2020).

Coffee hulls are a raw material that have been underutilized, considering their promising nature due to their structural composition and high availability. This highlights the need for further studies and favorable conditions for biotechnological

applications to optimize the utilization of this biomass. Therefore, this study aimed to develop a multifunctional fiber-rich ingredient from coffee hulls using a one-step process based on reactive extrusion with either alkaline hydrogen peroxide or sulfuric acid.

The rest of this article is structured as follows: Section 2 presents the materials and methods used to conduct the study; Section 3 presents the results obtained, as along with a discussion based on relevant literature; and finally, Section 4 provides the conclusions.

## 2 Materials and methods

This section outlines the materials and methods utilized in the study. Section 2.1 details the process of obtaining and conditioning the waste (coffee hull) and the primary reagents used for its modification. Subsection 2.2 describes the method employed to modify the waste through extrusion. Subsequent subsections provide the methodologies used to characterize the materials obtained.

### 2.1 Materials

Coffee hull (CH) (*Coffea arabica*) was supplied by the Institute of Rural Development of Paraná – IDR (Londrina, Paraná State, Brazil). The residue was dried in an air-circulating oven (Marconi MA 035, São Paulo, Brazil) for 12 hours at 45 °C, followed by grinding and sieving to obtain particles between 180 µm and 300 µm. During the analysis period, the material was stored at room temperature. The reagents used in this study, including sulfuric acid, hydrogen peroxide, and sodium hydroxide, were of analytical grade (Synthlab, Diadema, SP, Brazil).

### 2.2 Modification of coffee hull by reactive extrusion

Modification of the CH was performed using reactive extrusion in a single-screw extruder (AX Plastics, Diadema, SP, Brazil), which featured a screw diameter of 1.6 cm and a screw length-to-diameter ratio (L/D) of 40. The extruder had four heating zones and a matrix diameter of 0.8 cm. The temperature across all zones was maintained 100 ºC, with a screw speed of 60 rpm. Sulfuric acid (SA) and alkaline hydrogen peroxide (AHP) were utilized as modifying agents at concentrations of 1.0% and 3.0% (g acid/100 g residue). These agents were dissolved in distilled water and mixed with the residue (100 g), resulting in a final moisture content of 32% (g water/100 g residue). Each mixture was sealed in plastic bags and equilibrated for one hour before extrusion. A control sample, prepared without any reagent other than water, was also extruded to achieve the same moisture content.

To determine the initial moisture content, samples weighing approximately 2 g to 10 g were placed in pre-weighed metal capsules and heated at 105 °C until a constant weight was achieved. Moisture content (%) was calculated based on the difference between the initial and final masses of the samples. The extruded samples underwent three washes with distilled water for achieve neutralization, and the final pH was measured using 1 g of each sample mixed with 10 mL of water. After 10 minutes, pH levels were measured with a digital potentiometer (HI 3221, HANNA, Romania), which had been calibrated with buffer solutions of pH 4.0 and 7.0. Subsequently, samples were dried in

an air-circulating oven (Marconi MA 035, São Paulo, Brazil) for 12-24 hours at 60 °C, and ground to obtain particles between 150 µm and 220 µm. The reactive extrusion parameters were adapted from a study by Pereira, Marim and Mali (2022). Samples modified with 1.0% and 3.0% SA were labeled SA1 and SA3, respectively, while those modified with 1.0% and 3.0% AHP were labeled AHP1 and AHP3.

## 2.3 Determination of chemical composition

Proteins, ash, and moisture contents were analyzed following official AOAC methodologies (AOAC, 2012). Nitrogen content was determined using the Kjeldahl method, with protein content calculated using a conversion factor of 6.25. Lipid extraction was conducted as per the method recommended by the IAL (2008). For this, 2,000 g of each sample was utilized in a Soxhlet extractor, using petroleum ether as the extraction solvent for 8 hours. Carbohydrate content was calculated by difference, subtracting the combined values of moisture, ash, proteins, and lipids from 100. Cellulose and hemicellulose were quantified using the Van Soest (1965) method, while lignin content was determined according to the Technical Association of the Pulp and Paper Industry (TAPPI, 1999) method.

## 2.4 Bulk density and pH

Bulk density (BD) was assessed following the method by Benítez *et al*. (2011), utilizing a 10 mL graduated cylinder filled with the samples to a volume of 10 mL by consistent tapping. Bulk density was calculated as grams per cubic centimeter ($g/cm^3$). pH was measured using 1 g of each sample mixed with 10 mL of water. After 10 minutes, pH readings were taken with a digital potentiometer (HI 3221, HANNA, Romania), and calibrated using buffer solutions of pH 4.0 and 7.0.

## 2.5 Scanning electron microscopy (SEM)

The microstructure of the fibers was analyzed using scanning electron microscopy (SEM) with a microscope (FEI Quanta 200, Oregon, USA). Samples were ground to particle sizes between 15 nm and 20 nm, dried at 60 °C for 24 hours, and stored in a desiccator with silica for another 24 hours before analysis. Each sample was mounted on bronze stubs, coated with a gold layer (40 nm-50 nm), and observed at an accelerating voltage of 20 kV.

## 2.6 Fourier transform-infrared spectroscopy (FTIR)

Samples were dried at 60 °C for 24 hours and stored in a desiccator with silica for an additional 24 hours. FTIR spectra were obtained using a spectrophotometer (Shimadzu FTIR-8300, Kyoto, Japan), with analysis conducted over a range of 4000 $cm^{-1}$ to 500 $cm^{-1}$ at a resolution of 4 $cm^{-1}$. A total of 64 scans were performed for each sample.

## 2.7 X-ray diffraction (XRD)

XRD analysis was conducted using a Panalytical X'PERT PRO MPD diffractometer with copper k radiation (λ = 1.5418 Å) operating at 40 kV and 30 mA. The scanning range was from 2θ = 5° to 2θ = 45°, with a step size of 0.1 and a scanning speed of 1°/min. A secondary graphite beam monochromator was employed. The crystallinity index (CI) of the cellulose was calculated using Equation 1, based on the method by Segal *et al*. (1959):

$$CI = \left( \frac{I_{200} - I_{am}}{I_{200}} \right) \times 100 \quad (1)$$

where, CI is the crystallinity index of the cellulose, $I_{200}$ represents the peak intensity at 2θ = 20° – 22°, and $I_{am}$ is the peak intensity corresponding to the peak at 2θ = 18°.

## 2.8 Differential scanning calorimetry (DSC)

DSC analysis was performed using a Shimadzu DSC 60 calorimeter (Kyoto, Japan). Samples were placed in aluminum containers and heated from room temperature to 300 °C at a rate of 10 °C/min under a nitrogen atmosphere (flow rate: 50 mL/min).

## 2.9 Water absorption capacity (WAC)

WAC was determined according to the methodology described by Lu, Liu and Li (2013). Precisely 2,000 g of each sample was mixed with 25 mL of distilled water in pre-weighed Falcon tubes. The mixture was agitated at 150 × g for 30 minutes at room temperature using a Quimis orbital shaking incubator (Diadema, Brazil), followed by centrifugation at 3500 × g for 10 minutes (Hettich Centrifuge, Universal model 320R, Darmstadt, Germany). The supernatant was discarded, and the tube containing the wet sediment was weighed. WAC was calculated as the ratio of the weight of the hydrated fiber to the weight of the dehydrated fiber, expressed in grams of water absorbed per gram of sample.

## 2.10 Oil absorption capacity (OAC)

OAC was determined following the method by Lu, Liu and Li (2013). Exactly 2,000 g of each sample was combined with 25 mL of commercial soybean oil in pre-weighed Falcon tubes. The mixture was subjected to constant agitation at 150 × g for 30 minutes using a Quimis orbital shaking incubator, followed by centrifugation at 3500 × g for 10 minutes. The supernatant was discarded, and the remaining sample was weighed. OAC was calculated as the ratio of the final weight of the sediment to the weight of the dry matter, expressed in grams of oil absorbed per gram of sample.

## 2.11 Swelling capacity (SC)

Approximately 1000 g of each sample was placed in a 25 mL test tube, and 20 mL of distilled water was added. The mixture was agitated continuously for 2 hours using a shaker incubator (CT-712 Cientec, Brazil). After agitation, the suspensions were allowed to rest for 18 hours to ensure complete decantation. The swelling capacity was calculated as the ratio of the volume occupied by the sample (mL) to the sample weight (g).

## 2.12 Water adsorption isotherms

The samples (approximately 0.500 g) were conditioned in a desiccator containing calcium chloride (relative humidity ~ 0%) for 48 hours before being analyzed using Aqua Sorp Isotherm Generator equipment (Decagon Devices, Pullman, WA, USA). The moisture content at equilibrium was expressed as grams of water per gram of dry matter. Each analysis was performed in triplicate, and the resulting adsorption curves were fitted to the Guggenheim, Anderson and De Bôer (GAB) model. The GAB isotherm model (Bizot, 1984) is represented by Equation 2:

$$M = \frac{m_0 CK a_w}{1 - K a_w}\left(1 - K a_w + CK a_w\right) \quad (2)$$

where $M$ represents the equilibrium moisture content (g water/100 g solids), $a_w$ denotes water activity, $m_0$ is the monolayer moisture content (g water/100 g solids), and $C$ and $K$ are GAB model constants. All assays were conducted in triplicate.

## 2.13 Glucose adsorption (GA)

The glucose adsorption capacity was assessed according to the method described by Ou *et al*. (2001), with minor modifications. One gram of each sample was mixed with 100 mL of a glucose solution (500 mmol/L) and incubated at 37 °C for one hour. Following the incubation period, the samples were centrifuged at 3500 × g for 15 minutes. The glucose concentration in the supernatant was then determined using the dinitrosalicylic acid (DNS) method (Miller, 1959).

## 2.14 Statistical analysis

Statistical analysis was conducted using analysis of variance (ANOVA) and Tukey's mean comparison test, with a significance level at $p \leq 0.05$. These analyses were performed using R software (R Foundation for Statistical Computing, Vienna, Austria).

## 3 Results and discussions

This section presents the results and discussions. The chemical composition of the raw and modified waste samples is detailed in Subsection 3.1. The characterization results of the materials are organized in Subsections 3.2 to 3.8. Each result is accompanied by a relevant discussion, supported by the data obtained in this study and corroborated by findings in the literature.

### 3.1 Chemical composition

Table 1 ▼
Chemical composition of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

Table 1 provides the chemical composition of coffee hulls. The samples did not exhibit statistically significant differences in terms of moisture, lipid, and carbohydrate content. However, ash and protein contents differed significantly, as determined by Tukey's test ($p < 0.05$) with a reduction observed in the samples subjected to the treatments involving reactive extrusion.

| Samples | Moisture (%) | Ash (%) | Protein (%) | Lipids (%) | Carbohydrates (%) |
|---|---|---|---|---|---|
| **Raw CH** | 6.60$^{a}$ | 5.37$^{b}$ | 11.24$^{a}$ | 2.50$^{a}$ | 74.29$^{a}$ |
| **Control sample** | 6.25$^{a}$ | 6.16$^{a}$ | 11.10$^{a}$ | 1.29$^{a}$ | 75.20$^{a}$ |
| **SA1** | 5.67$^{a}$ | 3.20$^{d}$ | 10.14$^{b}$ | 1.06$^{a}$ | 79.93$^{a}$ |
| **SA3** | 5.61$^{a}$ | 2.81$^{d}$ | 9.45$^{b}$ | 1.06$^{a}$ | 81.07$^{a}$ |
| **AHP1** | 3.89$^{a}$ | 2.86$^{d}$ | 9.55$^{b}$ | 1.15$^{a}$ | 82.55$^{a}$ |
| **AHP3** | 4.31$^{a}$ | 3.60$^{c}$ | 10.53$^{ab}$ | 0.89$^{a}$ | 80.67$^{a}$ |

Means followed by different letters in the same column differ according to Tukey's Test ($p \leq 0.05$).

Previous studies on raw coffee hulls reported protein values ranging from 8% to 11%, lipid content between 0.5% and 3%, and carbohydrate content ranging from 58% to 85% (Gouvea *et al.*, 2009), which aligns with the findings of the present study. Variations in the chemical composition of coffee hulls may result from differences in processing, storage, transport, and processing conditions.

Table 2 presents the results for hemicellulose, cellulose, and lignin content. No significant differences were observed between the samples for hemicellulose and lignin before and after reactive extrusion treatments. However, a significant increase in cellulose content was noted in the modified samples compared to the raw coffee hull, with the AHP3 sample showing the highest cellulose value. Similar results were reported by Vilela *et al.* (2016), who found that chemical treatment with hydrogen peroxide alters the physical properties of fibers, leading to solubilizing of lignin and reducing in cellulose crystallinity, thereby improving certain techno-functional properties of the fibers.

Table 2 ▶
Composition of fiber in raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

| Samples | Cellulose (%) | Hemicellulose (%) | Lignin (%) | Insoluble dietary fibers (%)* |
|---|---|---|---|---|
| **Raw CH** | 27.92 ± 4.86$^{c}$ | 9.33 ± 2.02$^{a}$ | 28.72 ± 2.21$^{a}$ | 65.97 ± 3.04$^{b}$ |
| **Control sample** | 30.85 ± 1.87$^{c}$ | 11.06 ± 5.05$^{a}$ | 27.01 ± 1.59$^{a}$ | 68.92 ± 2.95$^{b}$ |
| **SA1** | 39.84 ± 1.41$^{b}$ | 11.27 ± 4.26$^{a}$ | 27.75 ± 2.13$^{a}$ | 78.86 ± 2.53$^{a}$ |
| **SA3** | 40.96 ± 0.54$^{ab}$ | 8.19 ± 0.66$^{a}$ | 28.02 ± 0.79$^{a}$ | 77.17 ± 0.88$^{a}$ |
| **AHP1** | 42.18 ± 0.09$^{ab}$ | 9.55 ± 1.19$^{a}$ | 25.58 ± 2.04$^{a}$ | 77.31 ± 0.97$^{a}$ |
| **AHP3** | 45.17 ± 1.55$^{a}$ | 8.46 ± 0.25$^{a}$ | 24.57 ± 1.95$^{a}$ | 78.20 ± 1.05$^{a}$ |

Means followed by different letters in the same column differ according to Tukey's Test ($p \leq 0.05$).
* Values obtained from the sum of cellulose, hemicellulose, and lignin contents.

Other studies have suggested that chemical treatments in an alkaline medium preserve cellulose structure (Chen; Zhao; Xia, 2009) and that the addition of hydrogen peroxide can facilitate the removal of lignin and hemicellulose (Alvira *et al*., 2010). However, this was not observed in the present study. The reactive extrusion process used here was relatively short, with samples remaining in the extruder for only 2 to 3 minutes, which may not have been sufficient to remove lignin or hemicellulose. Additionally, the concentrations of SA and AHP used were lower, and reaction times shorter, compared to those in other studies. Dutra *et al*. (2018) reported AHP concentrations between 0.5% and 7.4%, with reaction times ranging from 1 to 24 hours for various lignocellulosic materials.

All treated samples exhibited higher insoluble fiber content (Table 2), likely due to the removal of proteins, ashes, and lipids during the reactive extrusion process. Insoluble fibers are known to accelerate intestinal transit, increase fecal bulk, and slow glucose hydrolysis, thereby contributing to the prevention of certain colon diseases (Santos; Silva; Pintado, 2022).

## 3.2 Bulk density and pH

The bulk density of raw coffee hulls was measured at 6.11, and a decrease was observed in all treated samples, including the control sample, with values ranging from 3.54 to 3.38 (Table 3). This suggests that the extrusion process altered the physical structure of the materials, potentially increasing their porosity, as observed in SEM images. Yoshida and Prudencio (2020) reported that a decrease in bulk density could be beneficial for bakery applications, noting that fiber modification leading to increased porosity corresponds to lower density values.

The pH values ranged from 5.47 and 6.20 (Table 3), as expected since all samples were washed to remove unreacted SA or AHP and subjected to a neutralization step.

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Table 3 ▶
Bulk density and pH of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

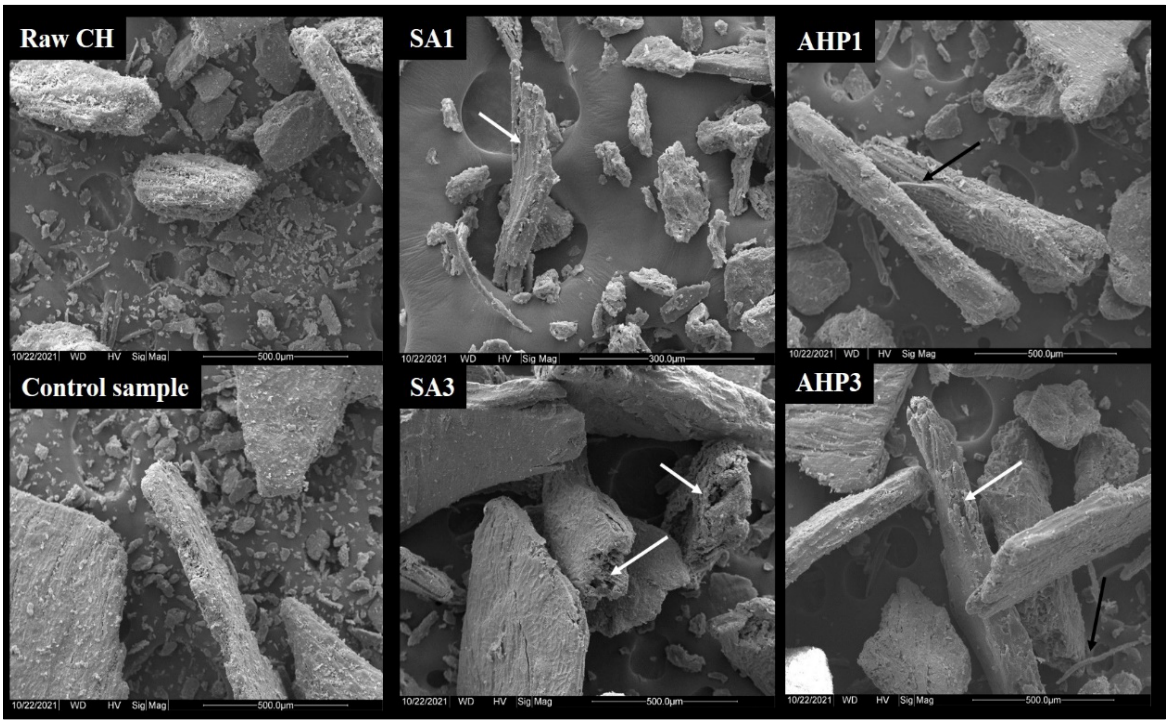
| Samples | Bulk density (g/cm$^3$) | pH |
|---|---|---|
| **Raw CH** | 6.1 ± 0.2$^a$ | 5.47 ± 0.02$^b$ |
| **Control sample** | 3.7 ± 0.1$^c$ | 5.58 ± 0.01$^b$ |
| **SA1** | 4.1 ± 0.1$^b$ | 5.19 ± 0.04$^b$ |
| **SA3** | 4.1 ± 0.1$^b$ | 5.20 ± 0.04$^b$ |
| **AHP1** | 4.3 ± 0.1$^b$ | 6.08 ± 0.12$^a$ |
| **AHP3** | 3.6 ± 0.1$^c$ | 6.20 ± 0.10$^a$ |

Means followed by different letters in the same column differ according to Tukey's Test ($p \leq 0.05$).

## 3.3 Scanning electron microscopy (SEM)

Figure 1 ▼
SEM images of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

Figure 1 presents scanning electron microscopy (SEM) images of raw coffee hulls and samples modified by extrusion and chemical treatments. The raw CH and control sample exhibit a compact structure due to the presence of non-cellulosic materials and waxes on their surface.



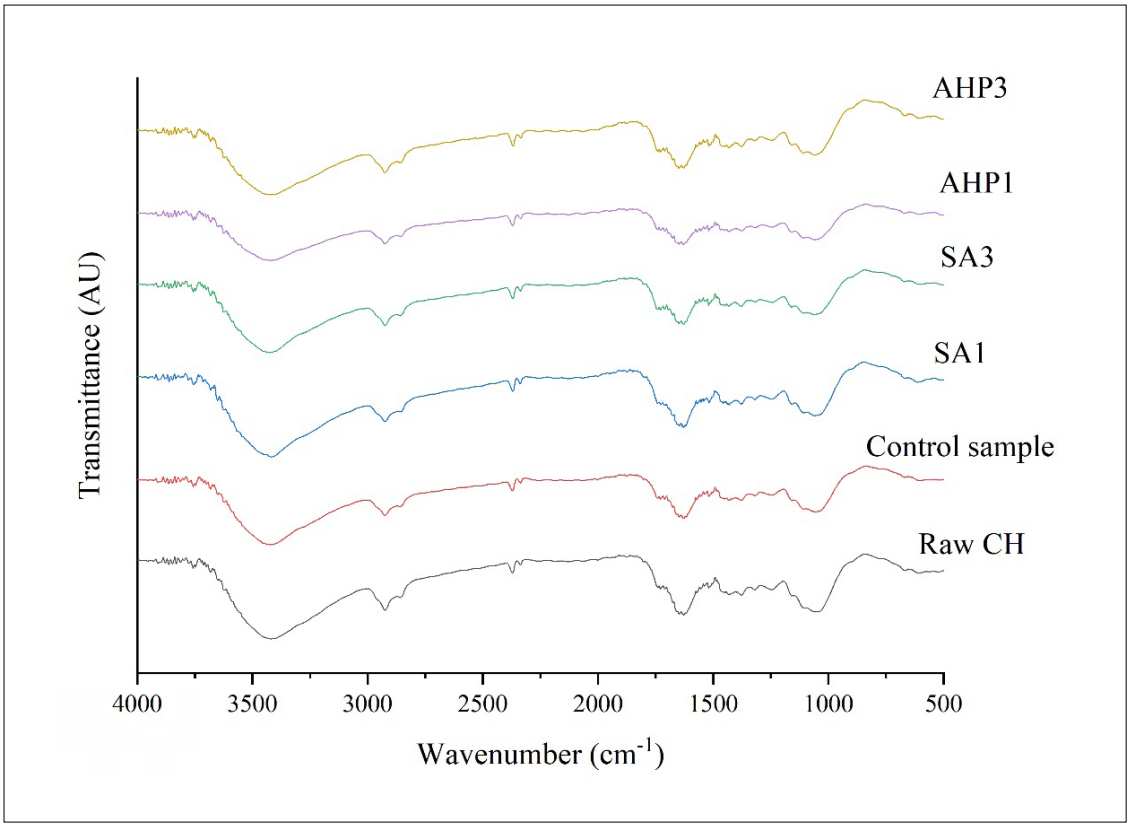

All treated samples displayed surfaces pores, likely due to the combined effects of extrusion and chemical treatments, which contributed to the removal of waxes and superficial fatty acids from the fibers, consistent with bulk density results. Meng *et al*. (2022) observed similar outcomes for bamboo residues treated with AHP. Other studies have reported that acid and/or alkaline hydrolysis effectively remove fatty acids and waxes from the surface, exposing the fibers (Brígida *et al*., 2010).

Samples treated with acid exhibited a rougher surface, attributed to the chemical treatment and the shear force, high temperatures, and pressure during the extrusion process (Figure 1). The samples treated with AHP showed more fiber bundles and deeper valleys.

## 3.4 Fourier transform-infrared spectroscopy (FTIR)

Figure 2 ▼
FTIR spectra of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

Figure 2 shows the FTIR spectra of the samples, indicating no significant differences between raw CH and treated samples. In all samples, bands at 3400 $cm^{-1}$ were attributed to the stretching of OH groups from cellulose, hemicellulose, lignin, proteins, and other components. Bands at 2920 $cm^{-1}$ and 2840 $cm^{-1}$ were observed across all samples, corresponding to the CH stretching vibration of alkyl groups.



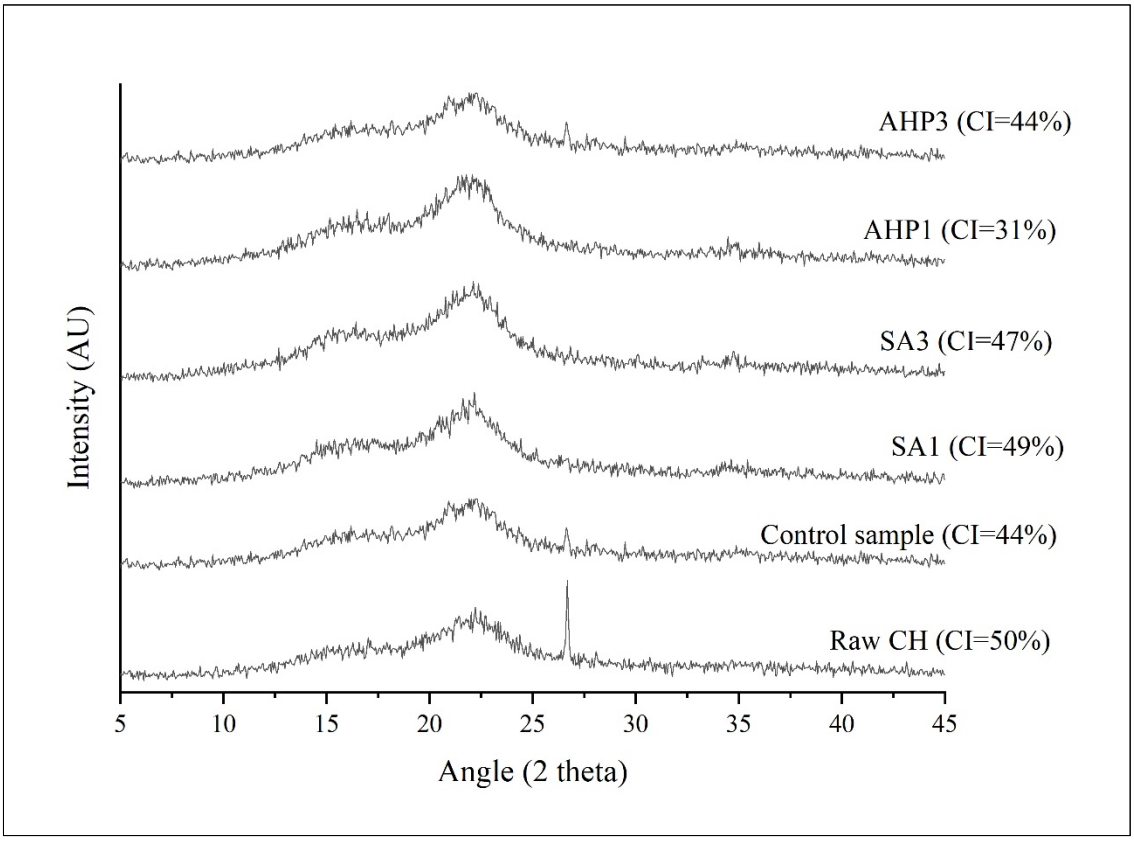

A band at 2900 $cm^{-1}$ was observed, which can be attributed to the C=O group in hemicellulose and/or the ester bonds of carboxyl groups present in hemicellulose and lignin. According to Cardoso *et al*. (2016), AHP may induce changes in hemicellulose and lignin. The C=O group in the fiber can be enhanced by oxidation reactions or reduced by cyclization reactions, both of which are promoted by hydrogen peroxide. The bands between 1400 $cm^{-1}$ and 1000 $cm^{-1}$ are characteristic of fiber spectra (lignin, hemicellulose, and cellulose).

Meng *et al*. (2022) reported higher-intensity cellulose bands after chemically treating bamboo residues with acid and alkaline reagents, a result not observed in this study.

## 3.5 X-ray diffraction (XRD)

The XRD patterns of raw CH, control sample, and samples modified with SA and AHP are presented in Figure 3, along with their respective relative crystallinity indexes. During the reactive extrusion process, the fibers were subjected to high temperature and shear forces. However, these conditions did not alter the crystallinity pattern of the modified samples, all of which exhibited a peak around $2\theta = 22^{o}$, indicative of cellulose I, as reported by Gabriel *et al*. (2020). Similar studies on cellulose extracted from oat hulls subject to reactive extrusion also found that this process did not affect the inherent crystalline structure of the sample likely due to the resistance of chemical reagents to penetrate the stable crystalline region of cellulose (Oliveira *et al*., 2017).

Figure 3 ▼
XDR patterns and relative crystallinity indexes (CI) of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*




The raw coffee hull exhibited a crystallinity index of 50% (Figure 3), consistent with Gabriel *et al*. (2020), who reported a CI of 55% for raw coffee hulls. Despite the high temperature and shear forces applied during the reactive extrusion, the crystallinity pattern of the modified samples remained unchanged. Larrea *et al*. (1997) similarly observed no significant alterations in XRD patterns for rice hulls subjected to reactive extrusion with hydrogen peroxide.

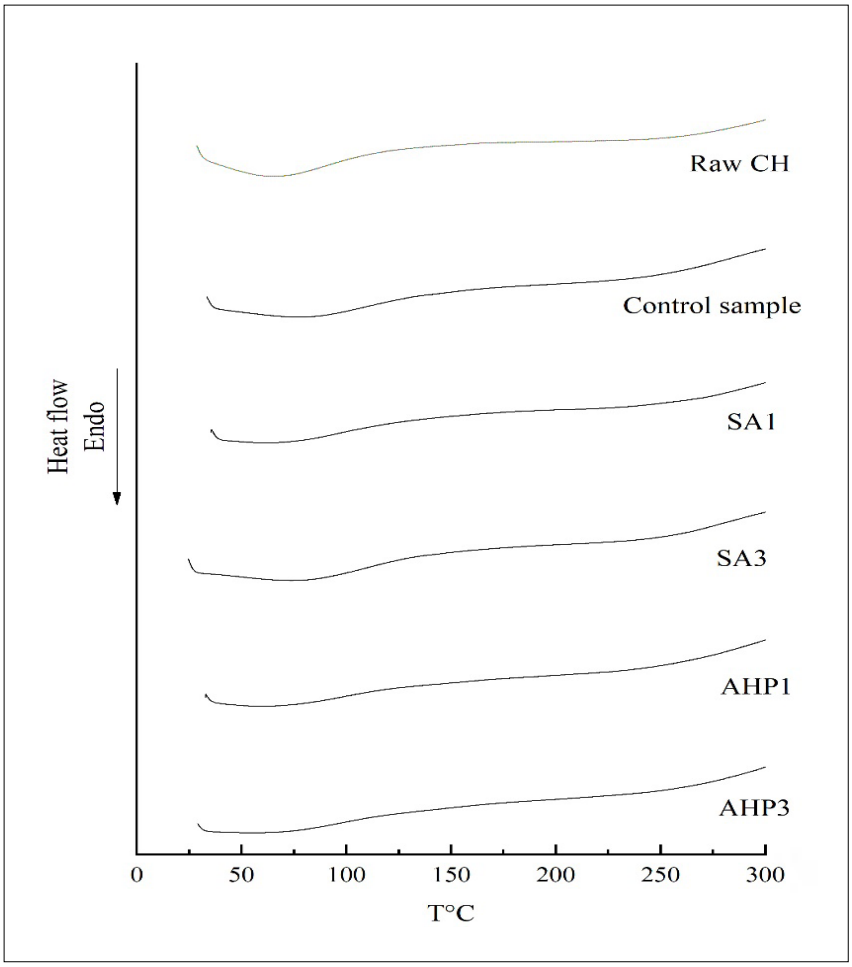
The crystallinity indexes (CI) of treated samples showed a decrease compared to the raw coffee hull, and the AHP1 sample displaying the lowest CI value. Vandenbossche *et al*. (2014) reported a reduction in CI for blue agave bagasse after undergoing alkaline treatment via reactive extrusion. Oliva *et al*. (2017) noted that extrusion, a highly versatile process for lignocellulosic residues, facilitates mixing, rapid heat transfer, and high shear stress, leading to defibrillation and a reduction in both the crystallinity index and the degree of polymerization of cellulose.

In contrast, other studies have reported an increase in the CI following chemical treatments. Gabriel *et al*. (2020) demonstrated that treating various lignocellulosic residues, including coffee hulls, with NaOH (10%) resulted in cellulose-rich materials with higher CI, attributing this to the removal of hemicellulose and lignin from amorphous regions, thereby realigning cellulose molecules. Similarly, Meng *et al*. (2022) found that bamboo residues exhibited increased crystallinity after the removal of non-crystalline components such as lignin and hemicellulose.

## 3.6 Differential scanning calorimetry (DSC)

The DSC results (Figure 4) indicated no significant differences between the samples, with no modifications observed during the analysis. This suggests that the samples possess thermal stability when processed within this temperature range. All samples exhibited minor endothermic events between 50 ºC and 100 ºC, likely associated with water loss.

Figure 4 ▶
DSC thermograms of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*




## 3.7 Determination of techno-functional properties

Water absorption capacity (WAC) measures the amount of water a fiber can absorb, a property closely related to the soluble fibers content in the sample. Similarly, oil absorption capacity (OAC) quantifies the amount of lipids a fiber can absorb, which

correlates with the fiber's ability to bind substances in the intestine, such as acids, bile salts, and cholesterol.

In this study, both WAC and OAC were found to increase in the modified samples compared to the raw CH, with a significant difference observed for WAC e OAC (Table 4). This increase is attributed to physical and structural changes, particularly the enhanced porosity observed in all treated samples via SEM, which made them more susceptible to interactions. The AHP3 samples exhibited the highest WAC and OAC values among all samples (Table 4).

Table 4 ▶
Technological functional properties of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

| Samples | WAC (g/g) | OAC (g/g) | SC (mL/g) | GA (%) |
|---|---|---|---|---|
| **Raw CH** | 1.33 ± 0.07$^{b}$ | 0.06 ± 0.05$^{a}$ | 13.90 ± 0.25$^{a}$ | 88 ± 0.13$^{a}$ |
| **Control sample** | 1.57 ± 0.07$^{ab}$ | 0.56 ± 0.01$^{b}$ | 13.25 ± 1.23$^{a}$ | 90 ± 0.01$^{a}$ |
| **SA1** | 1.58 ± 0.06$^{ab}$ | 0.68 ± 0.01$^{ab}$ | 14.50 ± 0.78$^{a}$ | 90 ± 0.02$^{a}$ |
| **SA3** | 1.56 ± 0.06$^{ab}$ | 0.69 ± 0.03$^{ab}$ | 14.86 ± 2.37$^{a}$ | 89 ± 0.06$^{a}$ |
| **AHP1** | 1.47± 0.06$^{ab}$ | 0.52 ± 0.05$^{b}$ | 15.45 ± 1.55$^{a}$ | 91 ± 0.12$^{a}$ |
| **AHP3** | 1.96± 0.10$^{a}$ | 0.87 ± 0.10$^{a}$ | 15.83 ± 0.49$^{a}$ | 88 ± 0.13$^{a}$ |

Mean values ± standard deviation ($n = 3$). Means followed by the same letter, in the same column, do not differ from each other, according to Tukey's Test ($p \leq 0.05$).
SC = Swelling capacity; GA = Glucose adsorption.

Vilela *et al*. (2016) reported an increase in OAC from 2.6 g/g to 3.1 g/g when modifying spent coffee grounds with 5%–25% alkaline peroxide, likely due to the lower reagent concentrations employed in this study. Galdeano and Grossmann (2006) evaluated the impact of 7% hydrogen peroxide treatment combined with extrusion on the hydration properties of oat hulls, finding increases of 70% in water retention capacity and 55% in swelling volume. Moura *et al*. (2011) noted that low concentrations of hydrogen peroxide (0.3%–0.9%) did not affect the oil absorption and retention capacity of fibers.

The WAC values obtained in this study were lower than those observed by Mantovan *et al*. (2023) for raw orange bagasse and for orange bagasse subjected to alkaline treatment combined with autoclaving, where values of 4.56 g/g and 3.10 g/g were reported, respectively. Oats, soybean, and coffee hulls treated with alkaline hydrogen peroxide (2%, pH 11.5) combined with hydrothermal autoclaving showed a significant increase in WAC values; raw coffee hulls exhibited 4.5 g/g, increasing to 6.6 g/g after treatment (Silva *et al*., 2022).

A product with high OAC is highly valued in the industry, offering numerous applications such as improving the texture and shelf life of products through polymorphism control and enhancing the crystalline structure of fats to promote flavor solubilization (Vilela *et al*., 2016). Difonzo *et al*. (2022) highlighted that appropriate technologies can transform lignocellulosic residues into innovative ingredients for use in gluten-free foods, including coffee by-products.

Swelling occurs due to a spontaneous fixation of water within the fibrous matrix via intermolecular forces. This property was not affected by the treatments applied in this study, with SC ranging from 13.90 mL/g to 15.83 mL/g (Table 4). Larrea *et al*. (1997) used alkaline hydrogen peroxide (1%) pre-treatment followed by extrusion to modify rice hull fiber, observing an SC of 12.4 g/mL, while Benitez *et al*. (2019) reported

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SC of 3.8 g/mL for coffee parchment flour. The SC values obtained in this study were higher than those reported by Mantovan *et al*. (2023) for raw orange bagasse and for orange bagasse subjected to alkaline treatment combined with autoclaving, where values of 8.83 g/g and 8.43 g/g were observed, respectively.

Glucose Absorption (GA) is measured by the amount of glucose a fiber can absorb. Recent studies suggest that the consumption of insoluble fiber is associated with a reduced rate of diabetes and glucose absorption in the intestinal tract (Mello; Laaksonen, 2009). Benitez *et al*. (2019) indicated that glucose absorption depends on its concentration, with higher concentrations leading to higher absorption rates. In this study, GA values were not significantly affected by the different treatments (Table 4), with values ranging from 88% to 91% (Table 4). This indicates that the use of this residue could reduce the amount of glucose available in the intestinal lumen, thereby mitigating postprandial hyperglycemia. According to Difonzo *et al*. (2022), fiber-rich ingredients can serve as valuable additives to enhance the nutritional quality of bread, pasta, cake/muffins, biscuits, and snacks, particularly in gluten-free products, by lowering the glycemic index of foods.

Through chemical composition, microbiological, aflatoxin, and acute toxicity analyses, Beltrán-Medina *et al*. (2020) demonstrated that coffee silverskin exhibits a good food safety profile and can be used in the production of extruded cereal-based food product as a ready-to-eat source of protein and fiber. The study of the functional properties of agro-industrial by-products is crucial for their viability as raw materials with higher added value for the food industry (Deepak; Jayadeep, 2022).

## 3.8 Water adsorption isotherms

Figure 5 presents the water adsorption isotherms of samples, showing an increase in equilibrium moisture content that becomes more pronounced at $a_w$ values greater than 0.65.

**Figure 5 ▶**
Water adsorption isotherms of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

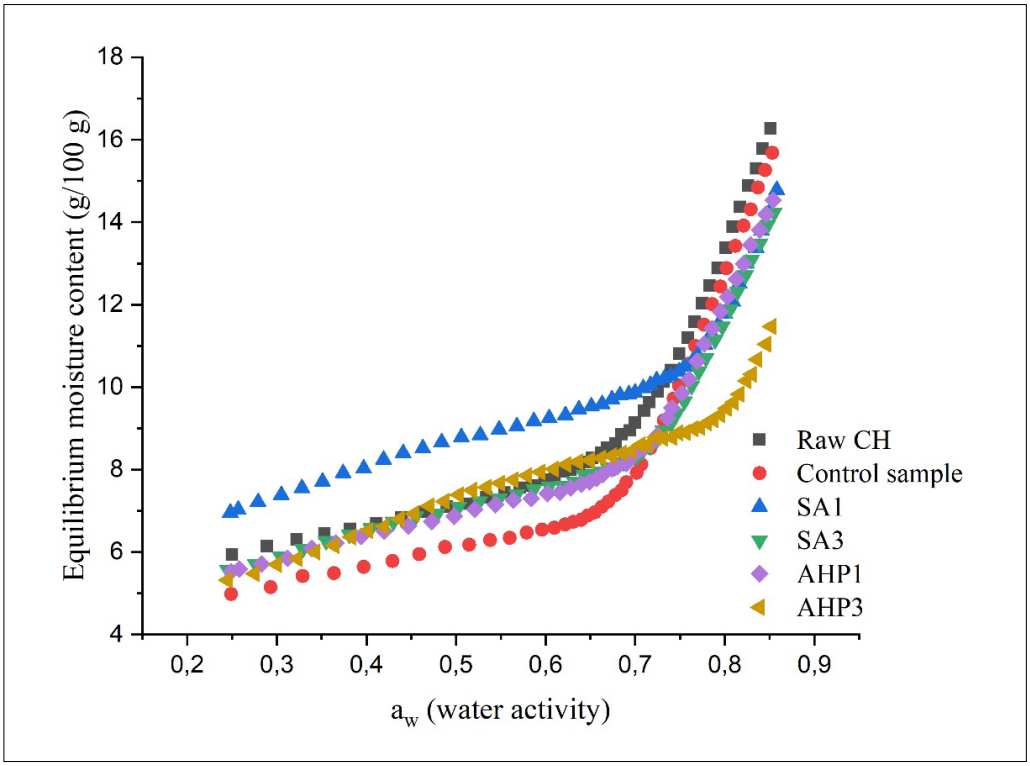

Table 5 presents the results of the adsorption isotherms, where $m_0$ represents the maximum amount of water that can be adsorbed in a single layer per gram of dry matter. Both chemical modification processes led to increased $m_0$ values. The SA1 sample exhibited the highest $m_0$ value, whereas the control sample had the lowest.

Table 5 ▶
GAB model parameters of raw CH, control sample, and samples modified with SA and AHP.
*Source: research data*

| Samples | $m_0$ | C | K | $R^2$ |
|---|---|---|---|---|
| Raw CH | 3.61 | 10000 | 0.9055 | 0.96 |
| Control sample | 2.82 | 10000 | 0.9634 | 0.96 |
| SA1 | 5.56 | 10000 | 0.6685 | 0.90 |
| SA3 | 3.80 | 10000 | 0.8353 | 0.93 |
| AHP1 | 3.58 | 10000 | 0.8691 | 0.94 |
| AHP3 | 5.16 | 56,262 | 0.6002 | 0.96 |

*GAB model: $M = (m_0CKaw)/(1-Ka_w) \times (1-Ka_w + CKa_w)$; M = equilibrium moisture (g water/100 g solids); $a_w$ = water activity; $m_0$ = monolayer value (g water/100 g solids); C and K = GAB constants.

Mihranyan *et al*. (2004) emphasized the complexity involved in understanding the moisture sorption mechanisms of cellulosic materials, due to the intricate nature of their structure. The material's composition and structural properties influence the water adsorption capacity, such as surface area, pore volume, and crystallinity. Higher porosities and surface areas typically result in greater moisture adsorption. At the same time, increased crystallinity reduces the number of hydroxyl groups available for interaction on the material's surface, thereby leading to lower water adsorption. Mantovan *et al*. (2023) corroborated this, observing that samples with higher crystallinity were less prone to water absorption. They also noted that the presence of pores can increase the values of lignocellulosic materials.

## 4 Conclusions

The sample modified with 3% alkaline hydrogen peroxide (AHP3) exhibited the highest cellulose content (45.17%), insoluble dietary fiber (78.20%), density (3.54), water absorption capacity (1.96 g/g), and oil absorption capacity (0.86 g/g), demonstrating its potential as a multifunctional ingredient. These characteristics contribute to nutritional, sensorial, and techno-functional enhancements, broadening the application possibilities of this ingredient in products such as breads, pastas, cookies, snacks, and gluten-free items, while also reducing the glycemic index of foods.

All samples exhibited thermal stability from room temperature up to 300 ºC, and a decrease in the crystallinity indexes was observed after treatment. Reactive extrusion proved to be effective in modifying the physicochemical and techno-functional properties of coffee hulls, offering advantages such as short reaction times, low reagent concentrations, minimal or no effluent generation, and scalability to an industrial level.

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## Conflicts of interest

The authors declare that there are no conflicts of interest associated with the execution of this work.

## Contributions to the article

**JACINTO, J. S.; MALI, S.:** conception or design of the study/research; data analysis and/or interpretation; final review with critical and intellectual participation in the manuscript. **MANTOVAN, J.:** conception or design of the study/research; data analysis and/or interpretation. **SILVA, J. B. M. D.:** conception or design of the study/research. All authors contributed to the writing, discussion, reading, and approval of the final version of the article.

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