

## 초임계 기반 전단 탈황 및 탄산칼슘 개질을 통한 재생 고무특성 향상 연구

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## Enhancing Properties of Reclaimed Rubber *via* Supercritical Fluid-Assisted Shear Desulfurization and Calcium Carbonate Modification

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**Abstract:** Using inorganic filler to replace waste rubber powder is an important way to improve the performance of recycled rubber. However, the poor “inorganic-organic” interface is one of the bottlenecks limiting the application of inorganic fillers in the field of waste rubber regeneration. This paper innovatively proposed using stearic acid modified calcium carbonate powder (CCP), combined with supercritical fluid-assisted shear desulfurization technology to improve the interface bonding between waste rubber and calcite powder. The vulcanizable properties and mechanical properties of the reclaimed rubber prepared with unmodified CCP, KH570 modified CCP and stearic acid modified CCP were tested. The results showed that the reclaimed rubber prepared from CCP modified by stearic acid exhibited good mechanical properties. The tensile strength of the regenerated rubber was 14.2%, 36.2%, and 11% higher than those of the regenerated rubber without CCP, unmodified CCP and KH570 modified CCP, respectively. Tear strength was higher by 7%, 16.2%, and 3.6%, respectively. The elongation at break was 13.4%, 26.8%, and 6.9% higher, respectively. This research is of great significance for improving the recycling properties of waste rubber and promoting the reuse of waste resources.

**Keywords:** stearic acid, calcium carbonate powder, recycled rubber.

### Introduction

The main components of waste tire rubber are natural rubber, synthetic rubber, carbon black, metals, nylon fibers, additives, etc.<sup>1</sup> It is difficult to degrade under natural conditions, and improper disposal can lead to serious “black pollution”.<sup>2</sup> One of the main methods for handling these waste tire rubbers is the preparation of recycled rubber.<sup>3</sup> However, after multiple uses and processing, the rubber molecules in recycled rubber undergo breakage and degradation. This results in a shortened

length and reduced molecular weight of the rubber chains in the recycled rubber, leading to decreased material strength and durability. Additionally, the processing methods for preparing recycled rubber often involve low-temperature dispersion, shear, and mixing, which limit the recovery and orderly arrangement of rubber molecular chains and consequently affect the material's mechanical properties.<sup>4,6</sup>

Introducing suitable fillers into waste rubber powder can enhance the mechanical performance of recycled rubber to some extent. calcium carbonate powder (CCP) is widely used in rubber, plastic, paper, coating, and other fields due to its advantages of resource richness, low price, chemical stability, and environmental friendliness. However, during the mechanical grinding process of CCP, unsaturated particles such as Ca<sub>2</sub><sup>+</sup>

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and  $\text{CO}_3^-$  easily react with adsorbed water molecules, resulting in the formation of hydroxyl groups and hydrophilicity in heavy calcium carbonate. However, most rubber composite materials are oleophilic and hydrophobic, and their surface properties are opposite to those of heavy calcium carbonate. This leads to poor compatibility, uneven dispersion, and weak interfacial adhesion between CCP and the rubber matrix, causing interface defects and decreased performance of rubber composite materials. Therefore, surface organic modification of CCP is necessary.<sup>7-10</sup>

Currently, common modifiers and methods are used for the surface organic modification of CCP. These include Coupling agent modification: Using coupling agents such as titanium ester and aluminum ester to react with heavy calcium carbonate, forming bonding interactions to improve its dispersibility and compatibility,<sup>11-12</sup> Oligomer modification: Adding organic oligomers such as polyethylene wax and polyethylene glycol to enhance the strength and durability of heavy calcium carbonate through physical and chemical cross-linking; unsaturated organic acid modification: Using unsaturated organic acids such as oleic acid to react with heavy calcium carbonate to improve its surface activity and plasticization; Water-soluble polymer modification: Introducing water-soluble polymers such as polyacrylic acid (salt) to form hydrogen or ionic bonds with the surface of heavy calcium carbonate, improving its dispersibility and stability.<sup>13-14</sup>

Although there have been some research achievements in the surface modification of CCP, there are still limitations in terms of the variety of surface modifiers and the development of composite modifiers. For example, titanium esters, aluminum esters, and organic phosphates are expensive, leading to high costs. Water-soluble polymer modification is complex and not conducive to industrialization. Therefore, further research and application of new modifiers are needed to achieve efficient surface modification of CCP.<sup>15</sup>

In this study, KH570( $\text{C}_{10}\text{H}_{20}\text{O}_5\text{Si}$ ) and stearic acid were used for the surface modification of CCP. The modified CCP was characterized using infrared spectroscopy and scanning electron microscopy. The study investigated the effects of CCP modification on the vulcanization characteristics and mechanical properties of reclaimed rubber. The interface interaction mechanism of modified CCP in the preparation of recycled rubber was also discussed.

## Experimental

**Materials and Methods.** Materials: Waste rubber powder

(WRP) with a particle size of 20 mesh, produced by Guangrao Changguan Renewable Resources Co., Ltd. CCP, manufactured by Insuda (Shandong) New Materials Co., Ltd, KH570, a slightly yellow liquid with a boiling point of 190 °C and a molecular formula of ( $\text{C}_{10}\text{H}_{20}\text{O}_5\text{Si}$ ), was purchased from Dongying Rich-Tech Science and Technology Co., Ltd. Anhydrous ethanol (EtOH) and others are commercially available. All of the above manufacturers are located in China.

**Instruments and Equipment:** Supercritical auxiliary shearing twin-screw machine: Dongying Jiexuan Environmental Protection Co., Ltd. XK-160 open mill: Dalian Huahan Rubber & Plastic Machinery Co., Ltd. MG2000GAN non-rotor rheometer, M-3000AU Mooney viscosity meter, Tensometer 2000 tensile testing machine: China High-Speed Railway Testing Instruments Co., Ltd. Scanning Electron Microscope (SEM): Japanese Electronics Corporation. Infrared Spectrometer: Bruker GmbH, Germany, Vector70 Fourier Transform Infrared Spectrometer.

**Preparation of KH570@CCP and SA@CCP:** As shown in Figure 1(a), 10 g of CCP and 2 g of KH570 were added to a reaction solution of 300 mL anhydrous ethanol. The mixture was stirred for 6 hours at 60 °C in a water bath, then removed and washed several times with anhydrous ethanol. The resulting product was dried in a 60 °C oven to obtain KH570@CCP.

As shown in Figure 1(b), 2 g of stearic acid was dissolved in 300 mL of anhydrous ethanol. The solution was stirred for 1 hour at 60 °C in a water bath. Then, 10 g of CCP was added to the solution and stirred for 6 hours at 60 °C in the water bath. The product was washed several times with anhydrous ethanol and dried in a 60 °C oven to obtain SA@CCP.

**Preparation of Recycled Rubber:** The process of preparing recycled rubber is divided into two stages: Stage 1: Firstly, mix rubber powder, rubber oil and CCP in a mixer for 3 min; After that, the mixture was added to the supercritical auxiliary shearing twin-screw machine for use, the temperature was set at 200 °C, the speed was 50 r/min, the time was 15 min, the mass ratio of supercritical fluid  $\text{CO}_2$  to the mixture was 0.02, and the material was taken out. In the supercritical state, carbon dioxide exhibits high solubility, which enables it to swell the rubber matrix. This swelling action aids in breaking down the cross-linking structures within the rubber. Under the influence of shear forces, the swollen rubber particles are more easily fragmented and dispersed, which contributes to the improved uniformity and production efficiency of the reclaimed rubber. The supercritical assisted shear desulfurization process is shown in Figure 2.

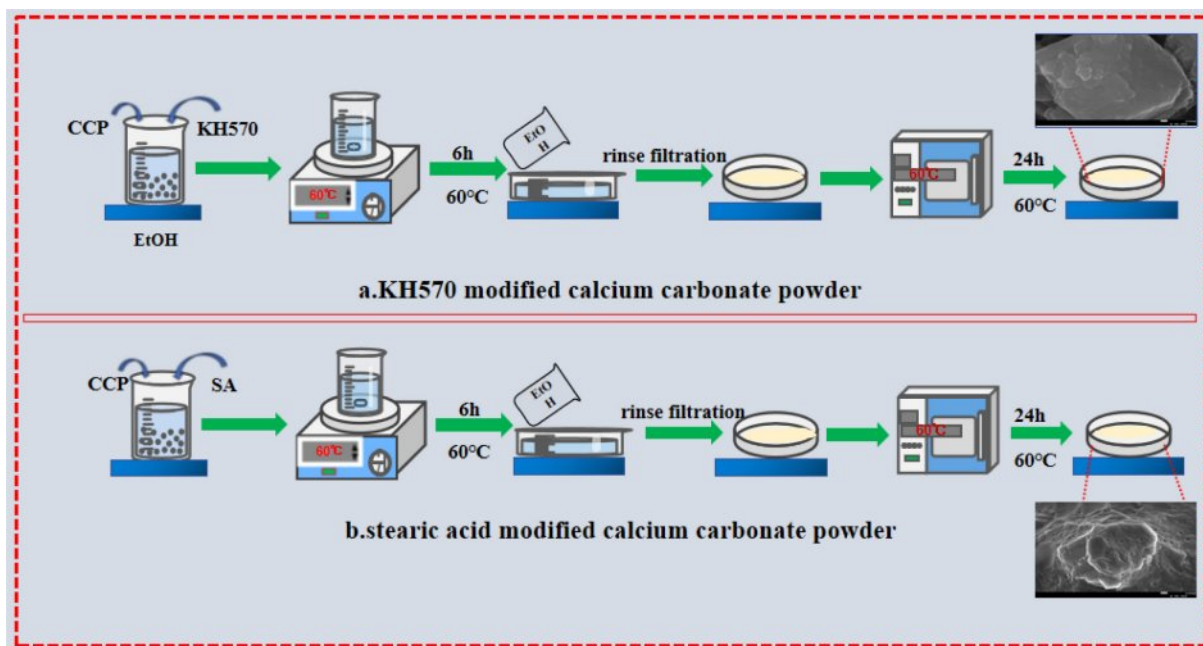


Figure 1. Process flowchart of modified calcium carbonate.

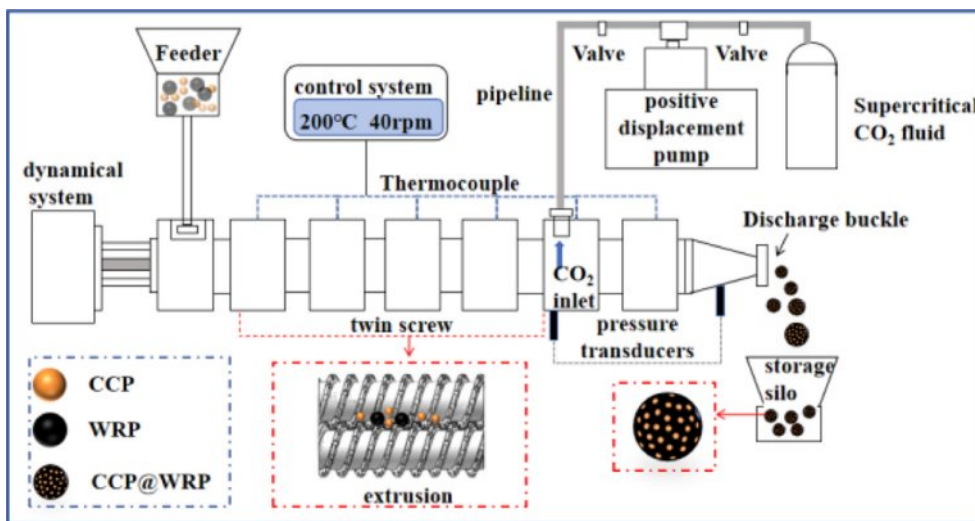


Figure 2. Flow chart of supercritical fluid-assisted shear desulfurization.

Step 2: The formulation for the preparation of vulcanized rubber is shown in Table 1. The roll spacing of the open mill was set to  $1.5 \pm 0.1$  mm and preheated to  $30$  °C. After milling for 1 minute, the accelerator TBBS was added by cutting the rubber on both sides three times. The rubber was milled for another minute and then zinc oxide was added by cutting the rubber on both sides six times. After milling for 2 minutes, stearic acid was added by cutting the rubber on both sides three times. The rubber was milled for 1 minute and then sulfur was

added by cutting the rubber on both sides four times. The rubber was milled for an additional 1.5 minutes and then the sheet was removed. The roll spacing was adjusted to  $0.8 \pm 0.2$  mm and thin sheeting was performed for 1.5 minutes before the sheet was removed. The roll spacing was further adjusted to 1.5-2 mm and the rubber was milled for 1 minute before the sheet was removed. The curing temperature for the mixed rubber was  $150$  °C, with a curing time of 1.3 times the normal curing time ( $t_{c90}$ ), to prepare vulcanized rubber.

**Table 1. Formulation for Preparing Recycled Rubber (phr)**

| Formulation      | 1   | 2   | 3   | 4   |
|------------------|-----|-----|-----|-----|
| Rubber powder    | 105 | 100 | 100 | 100 |
| Rubber oil       | 3   | 3   | 3   | 3   |
| Accelerator TBBS | 0.8 | 0.8 | 0.8 | 0.8 |
| ZnO              | 2.5 | 2.5 | 2.5 | 2.5 |
| Sulfur           | 1.2 | 1.2 | 1.2 | 1.2 |
| CCP              | -   | 5   | -   | -   |
| KH570@CCP        | -   | -   | 5   | -   |
| SA@CCP           | -   | -   | -   | 5   |

**Table 2. CCP Composition**

| Composition                    | Content (wt%) |
|--------------------------------|---------------|
| CaCO <sub>3</sub>              | 45.716        |
| SiO <sub>2</sub>               | 34.302        |
| Fe <sub>2</sub> O <sub>3</sub> | 6.1038        |
| CaO                            | 1.0985        |
| MgO                            | 0.6101        |
| MnO                            | 0.0182        |
| Na <sub>2</sub> O              | 0.1201        |
| K <sub>2</sub> O               | 3.9992        |
| TiO <sub>2</sub>               | 2.435         |
| P <sub>2</sub> O <sub>5</sub>  | 0.1865        |
| SO <sub>3</sub>                | 0.6646        |
| N                              | 0.3315        |
| CO <sub>2</sub>                | 4.0726        |
| Others                         | 0.3419        |

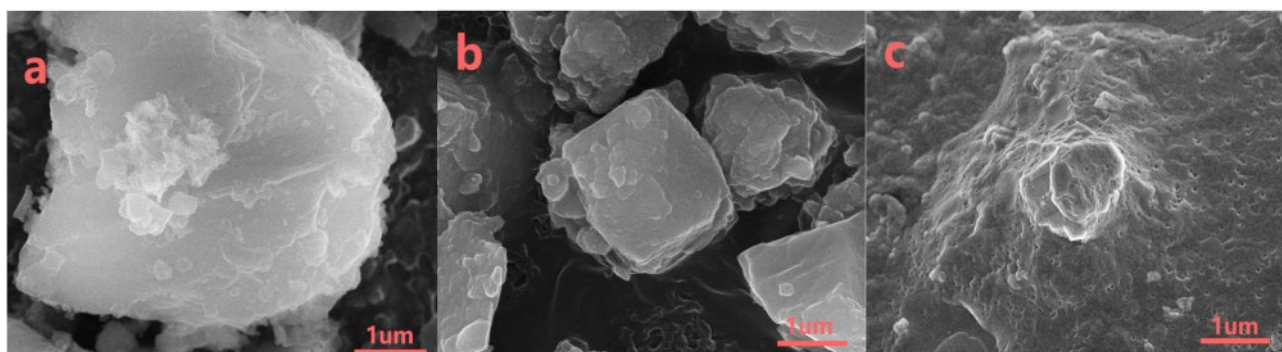
## Results and Discussion

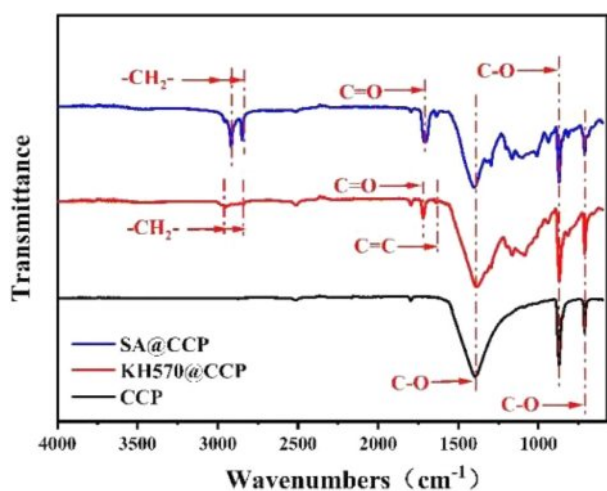
**Analysis of CCP Composition.** The X-ray fluorescence spectroscopy (XRF) measurement results of CCP are shown in Table 2, indicating the presence of a significant amount of calcium carbonate (45.716% wt%) and silicon dioxide (34.302% wt%) in the powder. In recent years, silicon dioxide has been widely used as a filler in rubber products due to its excellent properties. Additionally, heavy calcium carbonate has also gradually been applied in rubber products, becoming one of the potential rubber fillers due to its low cost. Therefore, theoretically, CCP can be used as a filler material in rubber compounds.

**Material Characterization.** Figure 3 shows the scanning electron microscope (SEM) images of CCP, KH570@CCP, and SA@CCP. Figure 3(a) displays the surface morphology of CCP, which appears as irregular-shaped polyhedrons with small irregularities or tiny angular structures on the surface. Figure 3(b) illustrates the surface morphology of KH570@CCP, where the material surface appears relatively smooth. This is due to the treatment with the silane coupling agent KH570, which forms a layer of organosilicon compounds on the surface of cal-

cium carbonate. This layer can form chemical bonds or adsorb onto the surface of CCP, resulting in a stable bond. This leads to a closer arrangement of CCP particles and a smoother, flatter surface at the microscopic scale. Figure 3(c) shows the surface morphology of SA@CCP, where CCP is coated with stearic acid, presenting a smooth and protruding surface. This is because stearic acid forms a monolayer or multilayer coverage on the surface of CCP and adheres to the particles through electrostatic attraction or intermolecular forces. This stearic acid coating layer provides higher lipophilicity, making the modified CCP easier to disperse in organic matrices.

In order to study the possible reaction mechanisms, FTIR spectra of CCP, KH570@CCP, and SA@CCP are presented in Figure 4. The main absorption peaks of CCP are observed at 710 cm<sup>-1</sup>, 875 cm<sup>-1</sup>, and 1394 cm<sup>-1</sup>. The absorption peak at

**Figure 3.** SEM micrographs: (a) CCP; (b) KH570@CCP; (c) SA@CCP.



**Figure 4.** FTIR spectra of CCP, KH570@CCP, and SA@CCP.

710  $\text{cm}^{-1}$  is attributed to C-O deformation vibration, while the absorption peak at 875  $\text{cm}^{-1}$  can be assigned to C-O anti-symmetric vibration. The absorption peak at 1394  $\text{cm}^{-1}$  is due to C-O symmetric vibration.<sup>16</sup>

After grafting with KH570, KH570@CCP exhibits a doublet structure around 2952  $\text{cm}^{-1}$  and 2836  $\text{cm}^{-1}$ , corresponding to the asymmetric and symmetric stretching vibrations of  $-\text{CH}_2-$ , respectively.<sup>17</sup> The peaks at 1718  $\text{cm}^{-1}$  and 1631  $\text{cm}^{-1}$  can be attributed to the stretching vibrations of C=O and C=C, respectively, indicating the successful grafting of KH570 onto the surface of CCP.<sup>18</sup>

After grafting with SA, SA@CCP shows peaks at 2915  $\text{cm}^{-1}$  and 2849  $\text{cm}^{-1}$ , which correspond to the asymmetric and symmetric stretching vibrations of  $-\text{CH}_2-$ , respectively. The peak at 1703  $\text{cm}^{-1}$  can be attributed to the stretching vibration of C=O, demonstrating the successful grafting of SA onto the surface of CCP.<sup>19-20</sup>

**Performance of Recycled Rubber.** Melt flow index (MFI) is a method used to measure the fluidity of liquids or semi-solid materials, typically used to describe the resistance and viscosity of a material during flow. In the context of rubber compounds, MFI can be utilized to assess the flowability of the rubber.<sup>21</sup> Lower MFI values indicate better flowability of the rubber, with lower viscosity, allowing for easier processing and filling during manufacturing. Higher MFI values indicate poorer flowability with higher viscosity, resulting in a more dense material that requires greater external force, such as high shear rates or elevated temperatures, to facilitate flow or shape change.<sup>22</sup>

Table 3 demonstrates that the recycled rubber prepared using

**Table 3. Performance of Recycled Rubber**

| Performance  | No CCP | CCP   | KH570@CCP | SA@CCP |
|--------------|--------|-------|-----------|--------|
| MFI (150 °C) | 76.39  | 83.59 | 80.7      | 71.2   |

stearic acid-modified CCP exhibits the lowest MFI compared to the recycled rubber prepared without the addition of CCP, and the recycled rubber prepared using unmodified CCP or KH570-modified CCP. This indicates that the stearic acid-modified CCP imparts superior processing performance to the recycled rubber. The improved flowability can be attributed to the stearic acid coating on the surface of CCP, promoting more uniform dispersion within the rubber compound and acting as a plasticizer, thereby enhancing the flowability of the rubber.

On the contrary, the recycled rubber prepared using unmodified CCP as a partial replacement for waste rubber powder exhibits the highest MFI, indicating poor processability and formation. This may be due to the difficulty of achieving uniform dispersion of unmodified CCP in the recycled rubber matrix, possibly leading to agglomeration.

The recycled rubber prepared using KH570-modified CCP as a partial replacement for waste rubber powder presents higher MFI compared to the recycled rubber prepared without the addition of CCP. Although the KH570-modified CCP improves the dispersion of CCP in the recycled rubber, it lacks the plasticizing effect seen with stearic acid modification. Therefore, its processing performance is compromised.

**Crosslinking Density.** In order to test the crosslinking density of recycled rubber prepared by different methods of modifying CCP as a replacement for waste rubber powder, the toluene dissolution method was used to measure the crosslinking density of the cured recycled rubber by measuring the volume and mass of the treated rubber solution.<sup>23-24</sup> From Table 4, it can be seen that the recycled rubber prepared by using stearic acid-modified CCP as a replacement for waste rubber powder has the highest crosslinking density. This is because waste rubber typically contains partially reacted vulcanizing agents and sulfur crosslinks. Stearic acid reacts with the sulfur crosslinks and vulcanizing agents in the waste rubber through substitution reactions, reconnecting some of the sulfur crosslinks and forming additional crosslink structures through chemistry with the

**Table 4. Crosslinking Density of Recycled Rubbers**

| Performance                                     | No CCP | CCP | KH570@CCP | SA@CCP |
|---|--------|-----|-----------|--------|
| Crosslinking density ( $\text{g}/\text{cm}^3$ ) | 0.6    | 0.4 | 0.7       | 0.9    |

unreacted vulcanizing agents. The long carbon chain structure and polar functional groups of stearic acid allow it to adsorb onto the surface of rubber particles and create a block effect, promoting the progress of crosslinking reactions. Additionally, stearic acid dissolution during the preparation of the recycled rubber has a certain swelling effect on the rubber powder, reducing the energy barrier of the s-s and c-s bonds and improving the reclamation rate of the waste rubber powder. The recycled rubber prepared by using KH570-modified CCP as a replacement for waste rubber powder has a slightly lower crosslinking density. This may be because KH570 possesses hydrophilic and hydrophobic functional groups, which can form a stable adsorption layer on the surface of calcite. This improves the compatibility between the CCP and the matrix material and solvent, enhances its dispersibility, and enables more uniform dispersion of CCP in the matrix. The recycled rubber prepared by using unmodified CCP as a replacement for waste rubber powder has the lowest crosslinking density. This may be due to the angular structure on the surface of unmodified CCP, which affects its dispersion in the recycled rubber. Additionally, the hydrophilicity of CCP prevents effective crosslinking from occurring in the waste rubber matrix. The formula for calculating crosslink density is as follows:  $V_c = (mw_2 - mw_3) / (mw_2 - mw_1) \times V_s$  ( $V_c$  represents the crosslink density of the rubber composite,  $V_s$  denotes the volume of the rubber composite,  $mw_1$  is the weight of the filter membrane after dissolving the rubber composite,  $mw_2$  refers to the dry weight of the rubber composite after drying treatment, and  $mw_3$  is the weight of the rubber composite after toluene treatment and drying).

**Sulfurization Characteristics.** The difference in MH (maximum torque) and ML (minimum torque) signifies the crosslinking density of the rubber material, with a higher difference indicating a higher density.<sup>25-26</sup> Table 4 shows that regenerated rubber prepared with stearic acid-modified CCP exhibits a 9.1%, 15.2%, and 5.2% higher MH-ML value compared to regenerated rubber prepared without CCP, unmodified CCP, and KH570 silane coupling agent-modified CCP, respectively. This indicates that the stearic acid-modified CCP leads to a higher crosslinking density and better desulfurization effect in the preparation of regenerated rubber, in line with the observed trend in crosslinking density testing.

Furthermore, Table 5 demonstrates that regenerated rubber prepared with stearic acid-modified CCP exhibits a lower cure time ( $t_{c90}$ ) compared to regenerated rubber prepared without CCP, unmodified CCP, and KH570 silane coupling agent-modified CCP. This suggests that stearic acid modification

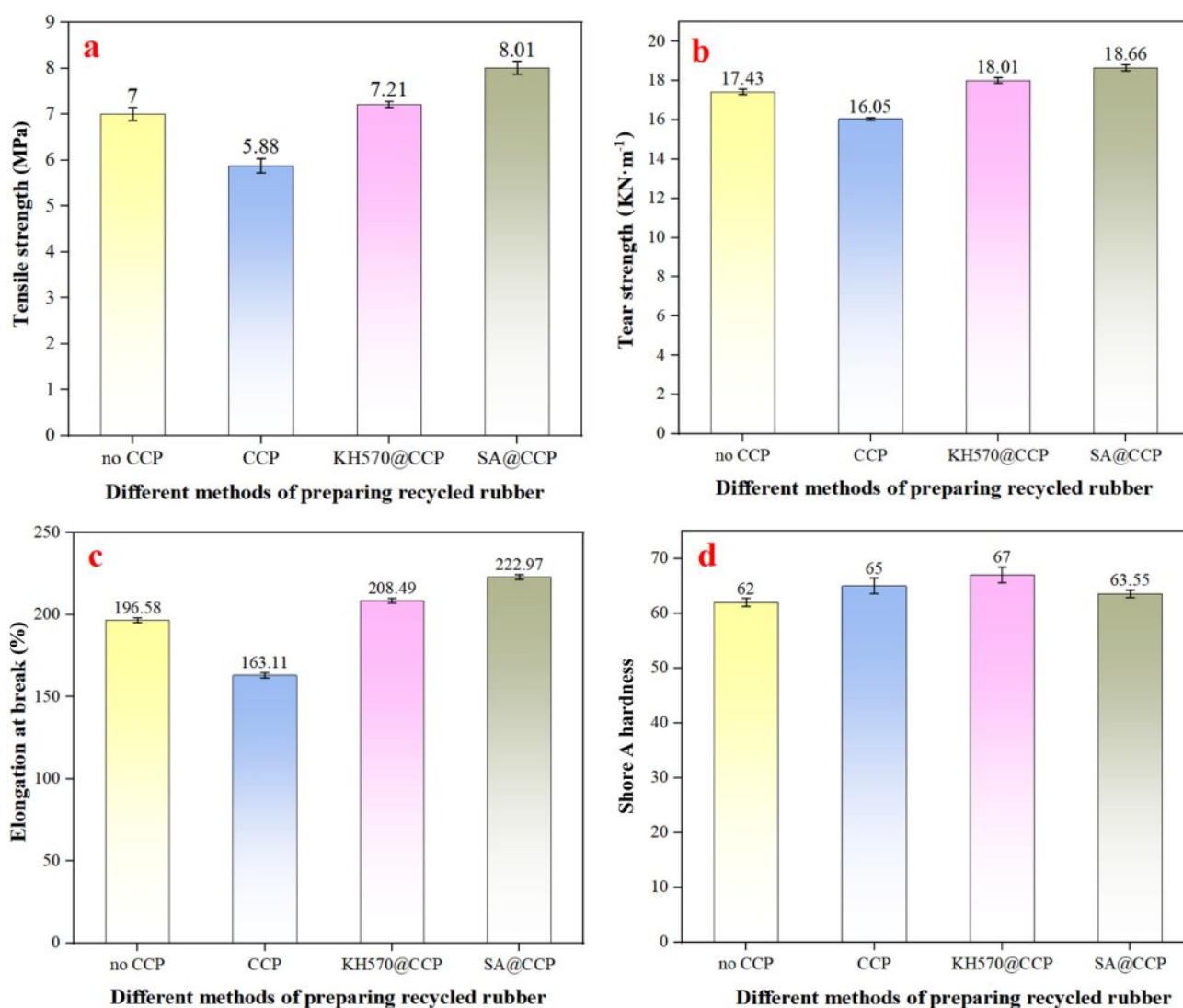
**Table 5. Sulfurization Characteristics of Composite Materials**

| Performance        | No CCP | CCP   | KH570@CCP | SA@CCP |
|--------------------|--------|-------|-----------|--------|
| $M_L$ (dN·m)       | 1.403  | 1.362 | 1.751     | 1.441  |
| $M_H$ (dN·m)       | 8.587  | 8.168 | 9.068     | 9.283  |
| $M_H - M_L$ (dN·m) | 7.184  | 6.806 | 7.317     | 7.842  |
| $t_{c90}$ (min)    | 7.363  | 7.363 | 7.083     | 6.723  |

results in a shorter cure time and higher processing efficiency due to its catalytic or promoting effect in the sulfurization reaction, enhancing the reaction rate and efficiency.

**Mechanical Properties.** From Figure 5(a), it can be seen that the tensile performance of the regenerated rubber prepared using stearic acid modified CCP reached 8.01 KPa, which is 14.2%, 36.2%, and 11% higher than that of the regenerated rubber prepared without adding CCP, unmodified CCP, and KH570 modified CCP, respectively. The analysis suggests that stearic acid coating on the surface of CCP improves the dispersion and filling performance of calcite in the regenerated rubber, thereby enhancing the structural strength and mechanical properties of the regenerated rubber. Stearic acid, as a surfactant, has good hydrophilicity and wetting ability, enabling it to interact with the surface of CCP. The dispersion of stearic acid modified CCP in the colloid is significantly improved, and agglomeration is reduced. Additionally, the long-chain structure of stearic acid can increase the adhesion between the filler and the matrix material, effectively improving the compatibility and adhesion between the filler and matrix.<sup>27-28</sup> The regenerated rubber prepared with unmodified CCP exhibited the poorest tensile performance. The analysis suggests that unmodified CCP, due to its poor adhesion and large gaps between solid particles, is prone to aggregate and form clusters, resulting in poor dispersion in the colloid. This agglomeration phenomenon adversely affects the mechanical properties of the regenerated rubber and reduces its tensile performance. Furthermore, the hydrophobic nature of unmodified CCP's surface prevents it from forming a tight cross-linking network with the regenerated rubber, leading to decreased tensile performance. This trend aligns with the MH-ML data trend. The tensile strength of the regenerated rubber prepared with KH570 modified CCP, as an alternative to waste rubber powder, is higher than that of the regenerated rubber prepared without adding CCP and unmodified CCP. This is because the surface of the CCP is covered with a layer of oil-friendly functional groups, increasing the cross-linking density of the regenerated rubber and ensuring better dispersion.<sup>29</sup>

From Figure 5(b), it can be seen that the tear performance of



**Figure 5.** Mechanical properties of the regenerated rubbers: (a) tensile performance; (b) tear performance; (c) elongation at break; (d) shore A hardness.

the regenerated rubber prepared using stearic acid modified CCP reached 18.66  $\text{KN}\cdot\text{m}^{-1}$ , which is 7%, 16.2%, and 3.6% higher than that of the regenerated rubber prepared without adding CCP, unmodified CCP, and KH570 modified CCP, respectively. The tear strength of the regenerated rubber is related to the cross-linking density. The analysis suggests that stearic acid modified CCP can effectively improve the tear performance of the regenerated rubber by enhancing filling effect, providing support, and improving interface interactions. Firstly, the enhanced filling effect is achieved as stearic acid modified CCP has good surface activity and dispersibility, which allows it to effectively combine with the particles of the regenerated rubber.<sup>30</sup> By filling the gaps and increasing the vol-

ume of the rubber material, it enhances the filling effect, thereby increasing the tensile strength and tear resistance of the regenerated rubber. Secondly, it provides enhanced support by forming a structurally stable network in the regenerated rubber, which improves the tear performance of the rubber material by providing enhanced support. This network can prevent the propagation of tears and disperse stress, thereby improving the tear resistance of the regenerated rubber. Thirdly, it improves interface interactions as the surface of stearic acid modified CCP contains oil-friendly functional groups, which can undergo physical adsorption and chemical bonding with the rubber chains in the regenerated rubber. By improving interface interactions, it strengthens the adhesion between the filler and the

rubber material, allowing for effective transmission of force. This helps prevent the propagation of tearing stress in the rubber material and improves its tear resistance. The regenerated rubber prepared with unmodified CCP, as an alternative to waste rubber powder, exhibited the poorest tear performance. This is because the CCP forms agglomeration in the rubber matrix, greatly reducing the tensile strength of the regenerated rubber. The regenerated rubber prepared with KH570 modified CCP showed improved tear performance compared to the regenerated rubber prepared without adding CCP. This is due to its better dispersibility and cross-linking density, which partially enhances the tear performance of the regenerated rubber.

From Figure 5(c), it can be seen that the elongation at break of the regenerated rubber prepared using stearic acid modified CCP is 222.93%, which is 13.4%, 26.8%, and 6.9% higher than those of the regenerated rubber prepared without adding CCP, unmodified CCP, and KH570 modified CCP, respectively. The regenerated rubber prepared with unmodified CCP exhibited the poorest elongation at break, which may be due to the low surface activity of CCP when not subjected to modification, leading to weak interface adhesion between the powder and the regenerated rubber. This makes it susceptible to interface separation during stretching, resulting in a decrease in elongation at break. The regenerated rubber prepared using stearic acid modified CCP showed the largest increase in elongation at break, which may be attributed to the following two reasons: Firstly, improved dispersion was achieved as stearic acid modification acts as a surfactant in the regenerated rubber, improving the dispersion of the additive in the matrix material. This allows for more uniform distribution of the particles in the matrix material, reducing stress concentration caused by particle aggregation and localization, thereby improving the strength and elongation of the rubber material. Secondly, the rubber structure was adjusted as stearic acid exhibits good plasticizing effects, allowing for changes in the colloidal structure of the regenerated rubber, resulting in better flexibility and ductility. The introduction of stearic acid can alter the interaction forces in multiphase systems, regulate the arrangement and spatial orientation of molecular chains, thus affecting the flowability and deformability of the rubber material. The regenerated rubber prepared with unmodified CCP, as an alternative to waste rubber powder, exhibited the poorest elongation at break due to agglomeration of the CCP in the regenerated rubber matrix. The regenerated rubber prepared with KH570 modified CCP showed better tensile properties compared to the regenerated

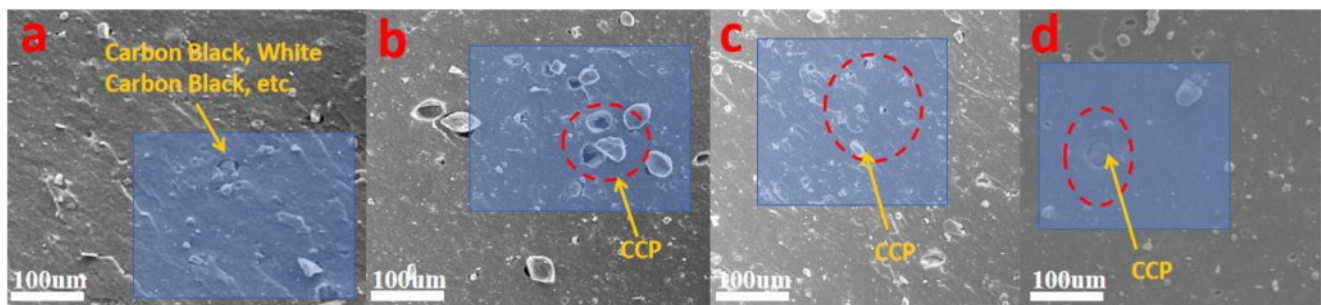
rubber prepared without adding CCP, but inferior to the regenerated rubber prepared with stearic acid modified CCP. This is because although the addition of KH570 enhances the dispersion effect and cross-linking density of CCP in the regenerated rubber, it does not have a plasticizing effect.

From Figure 5(d), it can be seen that the hardness of the regenerated rubber prepared using stearic acid modified CCP is 64, which is 2 degrees, 1 degree, and 3 degrees lower than that of the regenerated rubber prepared without adding CCP, unmodified CCP, and KH570 modified CCP, respectively. This is because stearic acid increases the flowability of the regenerated rubber material, enhances its flexibility, and decreases its hardness, which is consistent with the trend of elongation at break and Mooney viscosity. The regenerated rubber prepared with KH570 modified CCP exhibited the highest hardness, which is due to the fact that under the action of KH570, CCP is uniformly dispersed in the regenerated rubber matrix and forms a denser cross-linking network, thereby increasing the hardness of the regenerated rubber. The hardness of the regenerated rubber prepared with unmodified CCP as an alternative to waste rubber powder is slightly lower, which is because CCP provides support and increases the hardness of the waste rubber.

#### SEM Cross-Sectional Images of the Regenerated Rubber.

According to Figure 6, we can observe the cross-sectional images of the regenerated rubber composite materials prepared without adding CCP, with unmodified CCP, with KH570 modified CCP, and with stearic acid modified CCP. From Figure 6(a), it can be seen that there are particles of various sizes distributed on the surface, which are fillers such as carbon black and white carbon black contained in the regenerated rubber itself. From Figure 6(b), it can be observed that unmodified CCP forms agglomerates inside the regenerated rubber composite material, exhibiting uneven distribution and detachment phenomena, indicating poor interface bonding. This is because the hydrophobic surface of unmodified CCP makes it difficult to bond with the rubber material, resulting in lower mechanical properties when unmodified CCP is used as a substitute for waste rubber powder. From Figure 6(c), it can be seen that the cross-section of the regenerated rubber prepared with KH570 modified CCP as a substitute for waste rubber powder shows a relatively uniform distribution of CCP, and the interface bonding is better. This is because compared to unmodified CCP, KH570 modified CCP enhances its dispersion and improves the interface bonding between CCP and the waste rubber matrix. From Figure 6(d), it can be observed that the surface of the regenerated rubber prepared with stearic acid modified CCP as a





**Figure 6.** SEM cross-sectional images of the regenerated rubbers: (a) No CCP; (b) CCP; (c) KH570@CCP; (d) SA@CCP

substitute for waste rubber powder is smooth and evenly distributed. Additionally, there is good interface bonding between CCP and the rubber material.

## Conclusions

Regenerated rubber prepared using stearic acid modified CCP exhibits better performance and rheological characteristics compared to regenerated rubber prepared without adding CCP, unmodified CCP, and KH570 modified CCP.

Regenerated rubber prepared by stearic acid modified CCP demonstrates excellent mechanical properties, with higher tensile strength, tear strength, and elongation at break compared to those prepared by unmodified CCP and KH570 modified CCP. Compared to regenerated rubber prepared without adding CCP, unmodified CCP, and KH570 modified CCP, the tensile strength is increased by 14.2%, 36.2%, and 11% respectively, tear strength is increased by 7%, 16.2%, and 3.6% respectively, and the elongation at break is increased by 13.4%, 26.8%, and 6.9% respectively.

Compared to KH570-modified CCP, stearic acid-modified CCP was found to better enhance the mechanical properties, interfacial bonding performance, and processing efficiency of reclaimed rubber. CCP is abundant in resources, low in cost, chemically stable, and environmentally friendly. This research was conducive to promoting the industrial application of CCP in reclaimed rubber, which held significant importance for the reclaimed rubber industry.

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**Conflict of Interest:** The authors declare that they have no conflict of interest.

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