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Mixed Natural Rubber Latex and Co_2SiO_4/SiO_2 Derived from Silica of Oil Palm Leaves and Cobalt Nitrate

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ABSTRACT. Cobalt nitrate solution is combined with oil palm leaf powder to produce a semisolid mixture. The resulting mixture is heated in a furnace at 500 °C for 5 h, producing a grey powder. X-ray diffraction (XRD) analysis of the product showed distinct peaks at 2θ values of 31.2°, 36.8°, 44.8°, 59.2°, and 65.1°, which were consistent with the presence of Co_3O_4/SiO_2. When a similar mixture was heated at 1000 °C for 5 h, a purple powder containing Co_2SiO_4/SiO_2 was obtained. XRD patterns of this material contained peaks at 2θ values of 25.4°, 32.2°, 34.1°, 35.6°, 36.5°, 52.2°, and 62.3°, which corresponded with Co_2SiO_4. The latter product was selected for further analysis, including Fourier-transform infrared (FTIR) spectroscopy which identified characteristic absorption bands at 588 cm^{-1} and 672 cm^{-1}, alongside additional peaks at 817 cm^{-1} and 1117 cm^{-1}, reflecting the formation of cobalt silicate. Energy-dispersive X-ray (EDX) analysis confirmed the presence of cobalt (20.7%), silicon (14.0%), and oxygen (45.5%), alongside minor quantities of other elements, while scanning electron microscopy (SEM) showed irregularly shaped, non-spherical particles with some aggregation and a cloudy network. Following characterization, the synthesized cobalt silicate was applied as a colorant in natural rubber latex. Preliminary observations suggest that it functioned effectively as a colorant and remains stable during extended storage.

Keywords: Cobalt silicate, cobalt oxide, oil palm leaf (OPL), rubber latex, silica.

INTRODUCTION

Various methods have been developed for synthesizing Co_3O_4/SiO_2 composites, each tailored to achieve specific properties depending on the chosen approach (Vaidya et al., 2011; Ivetic et al., 2016). These methods address both material characterization and practical applications, including selective solar coatings (Barrera et al., 2008), carbon monoxide oxidation catalysts (Jia, 2011), Fischer–Tropsch synthesis (Xie, 2014), imidazolone synthesis (Asadi, 2018), photocatalysis (Collard et al., 2015; Baghban et al., 2016; Zha et al., 2019), humidity sensing (Fouad et al., 2012), and peroxidase-mimicking activity (Kandula and Jeevanandam, 2015). Co_3O_4/SiO_2 nanocomposites may exhibit two reduced energy band gaps, influenced by particle size (Gomaa et al., 2013). Tetraethyl orthosilicate (TEOS) is frequently used for in situ silica formation during synthesis. Alternatively, commercially available silica has also served as a starting material (Baghban et al., 2016). A combination of hydrothermal synthesis and calcination, using TEOS and cobalt acetate, has yielded cobalt silicate with strong cyclic stability and favorable rate performance, affirming the potential as a lithium-ion battery anode (Guo and Wang, 2015). Continued interest in cobalt silicate is driven by both its fundamental properties and diverse applications. Recent work employed the Pechini method using TEOS, cobalt acetate, and selected capping agents to enhance catalytic performance (Bayat et al., 2016). In another study, Co_2SiO_4 gel was synthesized from cobalt (II) acetate tetrahydrate, TEOS, ammonia, and carbohydrates, then calcined at 700 °C for 5 h under optimized conditions (Bayat et al., 2018).

This study introduces a modified approach to Co_3O_4/SiO_2 synthesis with greater emphasis on the formation of cobalt silicate (Co_2SiO_4). The synthesis of cobalt with silica has been carried out previously using both synthetic silica and silica extracted from natural materials. The synthesis results show that these two precursors produce two different forms at different temperatures. The materials formed are Co_3O_4-Silica and Co_xSiO_2 (Adam, 2011; Mawaddah & Utomo, 2022). Previous reports have affirmed the potential of oil palm leaves as a biogenic silica source for various applications (Onoja et al., 2018; Yudha S et al., 2020).

Building on this, the present study offers an alternative route by utilizing oil palm leaves as a sustainable silica precursor. This approach not only

addresses environmental considerations but also contributes to the novelty of the study by replacing synthetic sources with naturally derived materials. Furthermore, the simplicity of the synthesis procedure enhances its practicality and accessibility for routine laboratory use. The straightforward application of the resulting cobalt silicate as a colorant for natural rubber latex further reinforces the advantages of this study.

EXPERIMENTAL SECTION

Materials

Hydrochloric acid, nitric acid, and ethanol were obtained from Merck and used without further purification, while cobalt nitrate hexahydrate was sourced from Sigma Aldrich. Oil palm leaves biomass and natural latex were collected from a local plantation in Bengkulu Tengah, Indonesia. All glassware was pre-cleaned using a 3:1 solution of hydrochloric acid and nitric acid, followed by thorough rinsing with distilled water.

Procedure

Three grams of oil palm leaves powder were placed in a crucible and combined with 25 mL of ethanol. The mixture was stirred for 30 minutes using a magnetic stirrer to ensure uniform dispersion. Separately, cobalt nitrate was dissolved in 25 mL of ethanol and added to the leaf suspension while stirring continued. The mixture was stirred for an additional 30 minutes, then left undisturbed at room temperature overnight to allow gradual evaporation of the ethanol. The resulting semisolid was subjected to thermal treatment in a furnace. The temperature increases from room temperature to 500 °C was set 2 h, then maintained for 5 h. After heating, the sample was allowed to cool gradually over 24 h. A similar preparation was conducted using the same procedure, but subjected to 1000 °C.

The resulting solid products were characterized using various analytical instruments. Phase composition was analyzed using X-ray diffraction (XRD MiniFlex, Rigaku). Furthermore, morphological features were examined using scanning electron microscopy (SEM), while elemental composition was determined via energy-dispersive X-ray spectroscopy (EDX, JEOL JED-2300). Functional group analysis was performed using Fourier-transform infrared spectroscopy (FT-IR, Compact FT-IR Alpha 2, Bruker).

For the latex coloration test, one gram of natural rubber latex in dilute ammonia solution was placed in a vial containing a magnetic stir bar. A total of 10 mg Co_2SiO_4/SiO_2 powder were added gradually while stirring continued until the mixture became homogeneous. The mixture was poured into a 2.5 cm petri dish and left to dry under sunlight until a rubber film formed. As a control, a similar film was prepared using latex without the addition of Co_2SiO_4/SiO_2.

RESULTS AND DISCUSSION

Synthesis of Co_3O_4/SiO_2 and Co_2SiO_4/SiO_2

The synthesis of Co_3O_4 and Co_2SiO_4 embedded in oil palm leaves ash, which contains silica, entailed three main stages. These include thorough cleaning of oil palm leaves with distilled water followed by drying and grinding using a juice mixer, mixing cobalt nitrate in ethanol with an ethanol suspension of the processed leaves, aging the mixture to allow slow ethanol evaporation, followed by calcination at two different temperatures, as presented in **Figure 1**.

The reaction conducted at 500 °C and 1000 °C produced a black solid and purple product, respectively. The color difference serves as a preliminary indication of distinct compounds or material being formed.

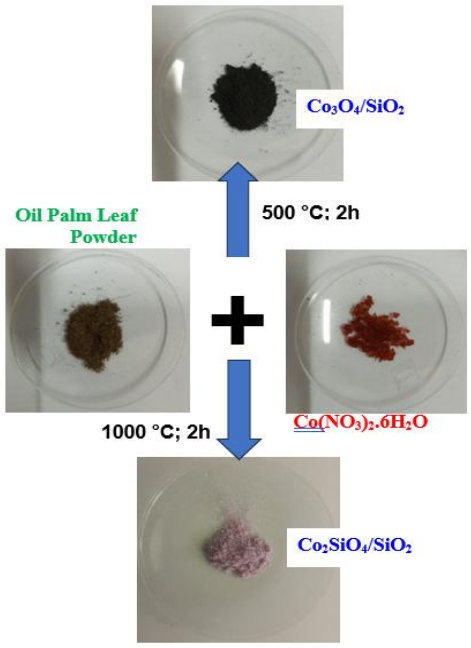

Figure 1. Synthetic Pathway for the synthesis of Co_3O_4/SiO_2 and Co_2SiO_4/SiO_2

Characterization

The XRD patterns of the prepared samples are shown in **Figure 2-3**. All observed peaks at 2θ values of 31.2°, 36.8°, 44.8°, 59.2°, and 65.1° corresponded to the characteristic reflections of Co_3O_4 with a face-centered cubic (FCC) crystal structure, consistent with JCPDS card 80-1541. A broad peak around 20.8° signified the presence of amorphous silica. The diffraction pattern confirmed the formation of Co_3O_4, with no observable peaks for Co_2O_3. The transformation of Co_2O_3 to Co_3O_4 was presumed to occur during the heating process and was in line with previously reported results (Ma et al., 2010).

The sample synthesized at 1000 °C featured a distinct set of peaks at 2θ values of 25.4°, 32.2°, 34.1°, 35.6°, 36.5°, 52.2°, and 62.3°, which match the diffraction pattern of Co_2SiO_4 based on JCPDS card 76-1501 (Phuong et al., 2021; Hideki, et al., 2002). The emergence of different crystalline phases at the two calcination temperatures confirmed that reaction mechanism played a critical role in determining the final product. The Co_2SiO_4 mechanism reaction could be explained using solid-state reaction approach as summarized in **Figure 4**.

The reaction mechanism for the formation of Co_2SiO_4 was explained using a temperature-dependent solid-state transformation model, as presented in **Figure 4**. At 500 °C, thermal decomposition of the organic matter in the oil palm leaves initiated the formation of silica through the oxidation of organosilicon compounds, as detailed in **Figure 4a**. Simultaneously, cobalt nitrate undergoes decomposition and oxidation to form Co_2O_3, which subsequently transformed into Co_3O_4, as shown in **Figure 4b**. At 1000 °C, the elevated temperature facilitated a solid-state reaction between cobalt oxide and silica through accelerated ionic diffusion, leading to the formation of Co_2SiO_4 embedded in silica, as presented in **Figures 4c** and **4d.**

Figure 5 presents the FTIR spectrum of the Co_2SiO_4/SiO_2 composite. Two distinct bands at 588 cm^{-1} (υ_1) and 672 cm^{-1} (υ_2) were attributed to Co–O stretching vibrations in Co_3O_4. The υ_1 band was associated with Co^{3+}–O vibrations, while υ_2 corresponded to Co^{2+}–O bonds, in line with earlier reports (Ma et al., 2010). Additionally, a weak band at 817 cm^{-1} was assigned to Si–OH stretching, and a strong absorption at 1117 cm^{-1} signified the presence of Si–O–Si linkages (Ghasemzadeh et al., 2017).

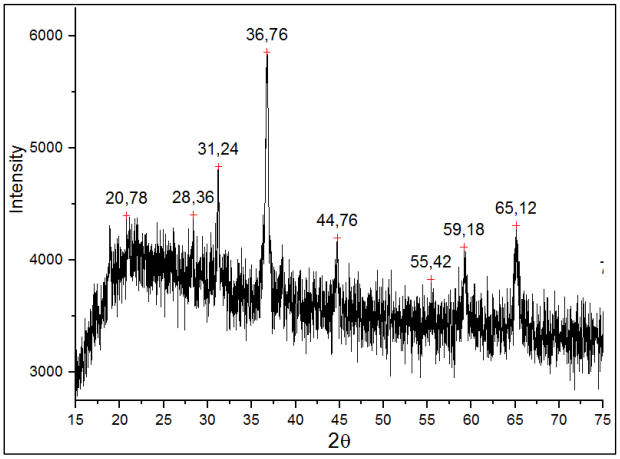

Figure 2. XRD pattern of Co_3O_4/SiO_2

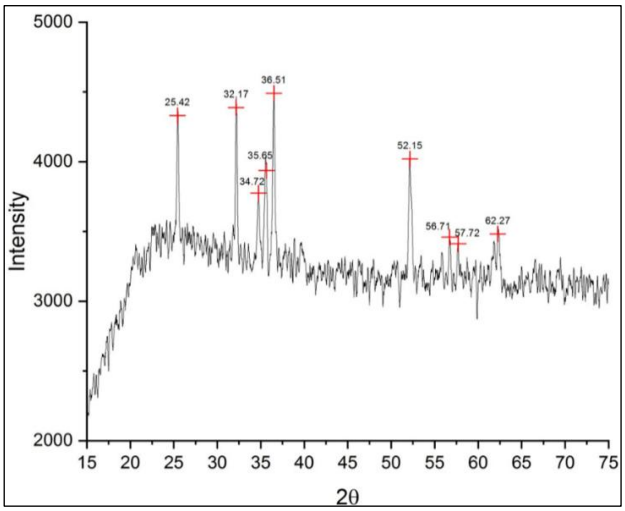

Figure 3. XRD pattern of Co_2SiO_4/SiO_2

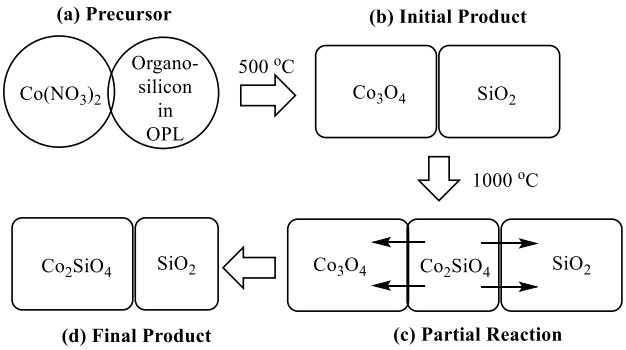

Figure 4. Plausible solid state reaction mechanism of cobalt nitrate and organosilicon in oil palm leaves dependent on temperature to form Co_3O_4/SiO_2 and Co_2SiO_4/SiO_2

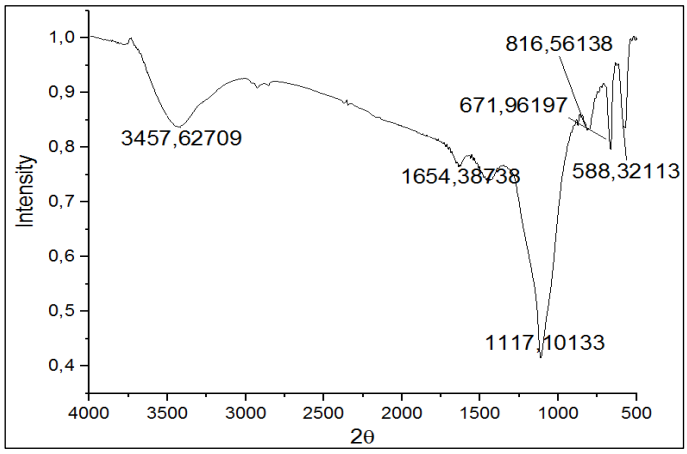

Figure 5. FTIR spectrum of Co_2SiO_4/SiO_2

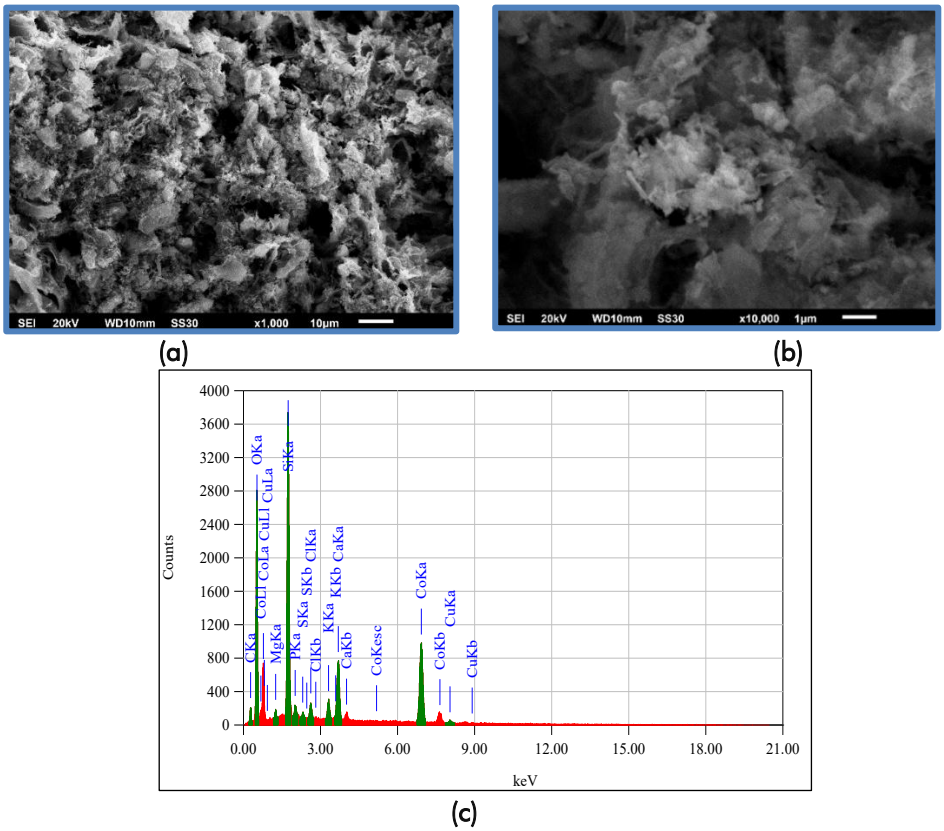

Figure 6. SEM Pattern of Co_2SiO_4/SiO_2 (**a**) 1000× (**b**) 10000× and (**c**) EDX pattern of the as prepared material

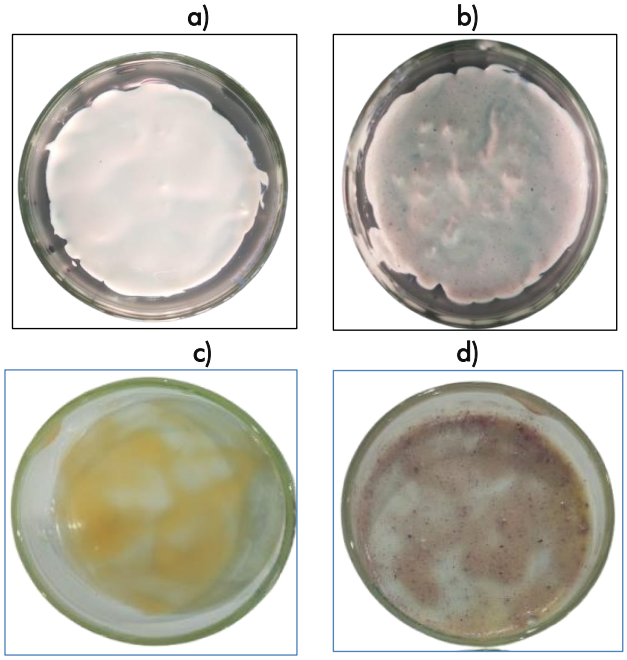

Figure 7. Appearance, **a**) natural rubber, **b**) natural rubber-Co_2SiO_4/SiO_2, **c**) natural rubber after drying, **d**) natural rubber-Co_2SiO_4/SiO_2 after drying

The SEM analysis of the synthesized Co_2SiO_4/SiO_2 composite showed the morphology and particle structure of the material. The observed particles featured irregular, non-spherical forms resembling sheet-like structures. As shown in **Figure 6**(**a**) and **6**(**b**), partial aggregation and the formation of a cloudy network were evident. This morphology suggests that Co_2SiO_4 was successfully embedded within the silica network derived from the decomposition of organosilicon complexes present in oil palm leaves. **Figure 6**(**c**) shows the result of EDX analysis of Co_2SiO_4/SiO_2 and gives in % weight (cobalt) 20.7; (silicon) 14.0 and (oxygen) 45.5. This result clearly shows evidence of the presence of cobalt and silicon elements in the sample.

Colorant Applications

The synthesized cobalt silicate (Co_2SiO_4) was evaluated for its potential use as a colorant in natural rubber. This application is motivated by the demand for stable pigments that can be integrated into rubber-based materials, as presented in **Figure 6**.

This effort was driven by the need for stable colorants suitable for natural rubber derivatives. Natural rubber is commonly utilized either in its raw form or as modified derivatives. As shown in **Figure 7**, the application of Co_2SiO_4/SiO_2 led to a visible color change, transitioning from one appearance when the material is wet to another when dry. This shift may be attributed to differences in light reflection between the wet and dry states of the rubber. Finally, the coloration shows considerable durability, affirming the potential as a viable coloring agent for natural rubber and related products. Previous research has shown that adding silica to latex improves the colouring of the latex and also its durability (Bokobza, 2019). The use of copper biphthalocyanine and iron oxide compounds has a significant effect on the latex modification process, making colouring one of the important factors for latex (Abd El-Ghaffar et al., 1990).

CONCLUSIONS

In conclusion, palm leaves were successfully used as a natural source of silica for high temperature reactions with cobalt nitrate precursors. The final product depended on the reaction temperature applied. At 500 °C, the reaction produced Co_3O_4/SiO_2, while increasing the temperature to 1000 °C led to the formation of Co_2SiO_4/SiO_2. The solid-state reaction mechanism played a significant role in the formation of these products. The Co_2SiO_4/SiO_2 product were utilized simply as a natural rubber coloring agent.

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