Original Research Article

The Acalypha siamensis leaf as natural dye in eco-print

NATALIA DIYAH HAPSARI - https://orcid.org/0000-0002-5661-1631

Chemistry Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia RISNITA VICKY LISTYARINI - https://orcid.org/0000-0002-7959-9786

Chemistry Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia LUISA DIANA HANDOYO - https://orcid.org/0000-0002-3625-581X

Biology Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia YOANNI MARIA LAUDA FERONIASANTI - https://orcid.org/0009-0001-2470-3371

Biology Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia INTAN PUTRI HAPSARI - https://orcid.org/0009-0008-0750-3766

Biology Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia ANDREAS FELIX TAN - https://orcid.org/0009-0002-1681-8926

Chemistry Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia

ELSIE GRACIA PUTRI DARMANTO

Chemistry Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia

AGNES SELFIANA IRMA ABUK

Chemistry Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia

HELENA FITRIANI

Chemistry Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia

BRIGITTA DELLA OCHA KRISTA

Biology Education Study Program, Faculty of Teacher Training and Education, Sanata Dharma University, Yogyakarta, 55281, Indonesia

Corresponding authors: Natalia Diyah Hapsari (e-Mail: nataliadiyah@usd.ac.id)

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ARTICLEINFO	A B S T R A C T The use of synthetic dyes in the textile industry has a negative impact on the environment. Therefore, it is necessary to explore natural dyes that are more environmentally friendly. The aim of this research is to find natural dyes from <i>Acalypha siamensis</i> leaves. In this research, compounds contained in <i>A. siamensis</i> extract were identified using FT-IR and GC-MS instruments, then	
Keywords:		
A. siamensis;		
Cinnamaldehyde;		
Eco-print;		
Natural dye	simulations were carried out of the reactions that occurred between cinnamaldehyde (E), cellulose in cotton, and alum mordant using quantum chemistry software. ORCA 5.0.2 and Visual Molecular	
	Dynamics (VMD). The cinnamaldehyde compound is the main constituent compound in <i>A.</i> siamensis leaf extract. Based on the simulation results: the reaction between cinnamaldehyde (E),	
History:	cellulose in cotton fabric, and alum mordant is energetically favorable. The reaction simulation	
• Received - 10 Dec 2024	results are in line with experimental research, the reaction of cellulose in cotton fabric, alum	
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Introduction

The use of synthetic dyes in the textile industry has been widespread since 1960. Synthetic dyes are widely used by the public because they offer several advantages, including a wide range of colors, rapid dyeing process, efficient use of water, and rapid dyeing process (Bide, 2014). However, these synthetic dyes have negative effects on the environment. Synthetic dye waste contains dangerous chemicals that can pollute water and soil. In addition, wastes from synthetic dyes containing amine compounds are carcinogenic and may pose a threat to human health (Gürses et al., 2016).

To reduce the negative impact of synthetic dyes, several natural dyes have been developed. Natural dye has the benefit of not harming aquatic ecosystems or the environment (Yusuf et al., 2017) Furthermore, the use of natural dyes promotes sustainable economic growth for future generations, as demonstrated by their widespread availability in nature, which is more cost-effective and environmentally friendly (Rather et al., 2024).

Plant parts such as roots, stems, leaves, fruits and flowers are used for the natural dyeing process. Because rosella flower crowns (*Hibiscus sabdariffa*) contain anthocyanins with the predominant compound cyanidin-3-sambubioside, they can



impart a deep red hue (Maciel et al., 2018). Plants containing anthocyanins can produce shades of red, blue and purple. The bonds that form between the sugar and the hydroxyl groups of the structure determine these colors (Nurtiana, 2019). Mulberry leaves can be used as a natural dye as they can produce a yellow to brownish color. The yellow to brownish color is due to the presence of flavonoids and phenolic compounds in mulberry leaves (Chetia et al., 2023). Flavonol and flavone compounds can impart a cream color, anthocyanins an orange color, carotenoids a yellow color, and chlorophyll a green color (Meijica et al., 2022). Each plant species is able to produce different color pigments depending on the secondary substance content contained in the plant.

Natural dyes can be applied directly to cotton fabrics through an eco-printing process or through an extraction process. Eco-printing technology involves dyeing fabric by directly transferring color and shape (Bintrim, 2008). The dyes used come from nature and can reduce the negative impact on the environment. The color that results from dyeing in eco-printing is influenced not only by the compound of secondary material compounds, but also by the interaction between the cellulose contained in the fabric and the mordant used. The aim of the mordant process using alum (aluminum sulfate), and iron (FeSO4) to emphasize the original color (Sofyan & SY, 2015). The use of alum and iron as a mordant is safe for the environment (İşmal, 2017). The substance combines with compounds contained in natural dyes by forming hydrogen bonds. Hydrogen bonds are formed between the hydroxyl groups in phenols found in natural dyes from turmeric and the hydroxyl groups in cellulose (Ragheb et al., 2017). In addition, covalent bonds are formed between quinones in natural dyes and the corresponding groups in cellulose (Ragheb et al., 2017).

A. siamensis leaves are widely used in medicine. *A. siamensis* leaves contain flavonoids, alkaloids, tannins and saponins (Frizqia et al., 2020; Fadilah et al., 2024). One of them is used as an anti-cancer drug (Frizqia et al., 2020). However, the use of Acalypha siamensis leaves as a natural dye in eco-prints has not been widely studied. The *A. siamensis* leaves is shown in Fig-1.



Fig-1 Acalypha siamensis plant

Therefore, in this study, we explored the compounds in in *A. siamensis* leaves as a natural dye. We analyzed the compounds in *A. siamensis* leaves, then predicted the reactions that occurred between cotton fabric, alum mordant and the dominant compounds in *A. siamensis* leaves. *A. siamensis* extract was analysis using FT-IR and GC-MS instruments. Then, the most abundant compounds in *A. siamensis* leaves were reacted with alum (alumunium sulfate) mordant using calculations with quantum chemistry software ORCA 5.0.2 (Zhao and Truhlar, 2008) and Visual Molecular Dynamics (VMD) was used to represent the structure (Humphrey et al., 1996). Furthermore, we applied *A. siamensis* leaves with eco-print coloring in cotton fabric.

Materials and Methods

Materials

The materials used in this research were *A. siamensis*, alum (aluminum sulfate), 70% ethanol and TRO (Turkey Red Oil) are the materials.

Sample Preparation

A. siamensis leaves were harvested from the garden of Sanata Dharma University in Yogyakarta. *A. siamensis* leaves were washed and dried at 35°C using oven for 3 days (Alsaud and Farid, 2020). Then the leaves of *A. siamensis* are ground to obtain *A. siamensis* leaf powder.

Extraction of A. siamensis

The samples were macerated in 70% ethanol solvent for 72 h at room temperature. The sample was then filtered using Whatman filter paper. *A. siamensis* extract was evaporated using a rotary evaporator at 50-55°C and a speed of 200 rpm (Fadilla and Kumalaningsih, 2021). The result obtained is a thick extract of *A. siamensis*.

Characterization

The chemical compounds in the leaves of *A. siamensis* were characterized using a Shimadzu Spirit 8000 infrared spectrometer and GC-MS.

Determination of the reaction

In order to evaluate the interaction of metal ion in alum, Al^{3+} with cellulose in cotton fabric and abundant compound in *A. siamensis* leaf, the interaction energy have been determined. The structures have been optimized for energy minimization at M06-2X (Zhao and Truhlar, 2008), level of theory using def2-TZVP (Weigend and Ahlrichs, 2005) basis set with the inclusion of dispersion correction (D3) (Grimme, 2006; Grimme et al., 2010) with the damping function (D3ZERO) (Grimme et al., 2010).

The interaction energy Eint (Reis et al., 2020) has been determined as the energy difference between the complex and the isolated cellulose, metal ion (Al^{3+}) and cinnamaldehyde energy via:

$$E_{\text{interaction}} = E_{\text{complex}} - (E_{\text{cellulose}} + E_{\text{metal ion}} + E_{\text{cinnamaldehyde}})$$

All calculations were performed using ORCA 5.0.2 (Zhao & Truhlar, 2008), with structure visualizations generated using VMD (Humphrey et al., 1996).

Application in eco-print

The aim of this scouring process is to remove various types of contaminants that can affect the dyeing process in eco-printing (Naini and Hasmah, 2021). The fabric used is washed and soaked with a TRO (Turkey Red Oil) solution of 0.3 grams/liter. The cloth and TRO solution are then boiled for 8 minutes. The cloth is then rinsed with water and air dried (Naini and Hasmah, 2021). After scouring process, the cotton fabric to be used is treated with a mordanting process. This mordanting process uses alum. The mordanting process is made by dissolving 12 grams of alum in 1.5 liters of distilled water. Next, put the cotton cloth in the solution prepared above for 5 minutes. Next, the cloth is squeezed and aired (Sofyan and SY, 2015).

Then, the cotton fabric is ready to be applied using eco-print plants. *A. siamensis* leaves are arranged on a plastic-covered cotton cloth. The fabric is then rolled up with a paralon and tied tightly. The rolled cloth is steamed for one hour and 30 minutes. After the steaming process is completed, the remaining leaves of *A. siamensis* are cleaned and the cotton cloth is dried in the sun (Sofyan and SY, 2015). Then leave it for one week and fix it using alum.

Results and Discussion

The focus of this research is to identify compounds contained in *A. siamensis* leaves that have the potential to be used as natural dyes in eco-prints. In this study, a simulation was carried out of the reaction that occurred between the most dominant compounds in *A. siamensis* leaves, cotton fabric and the alum mordant used. The *A. siamensis* are then applied to cotton fabric for eco-print dyeing. The extraction process produced the green extract of *A. siamensis* leaves. The *A. siamensis* extract is shown in Fig-2. The resulting extract was then analyzed using FT-IR.



Fig-2. Extract of A. siamensis leaves

Spectra analysis of A. siamensis extract

The FT-IR analysis finding several peaks as shown in Fig-3. A peak at 3500–2500 cm⁻¹, shows the stretching vibration of OH (hydroxyl) groups. This outcome is similar to the identification result of the extract of *Ficus amplissima* leaves, which displays the band at 3414 as a result of the OH's stretching vibration (Kumbhar et al., 2019). A peak at 3050–3000 cm⁻¹ due to stretching vibrations is corresponds to the C(sp2)-H (alkene group). A peak at 3000–2900 cm⁻¹, this peak indicates the presence of a C(sp3)-H (alkyne). A peak at 1750–1700 cm⁻¹, it shows the presence of the C=O (carbonyl) (Gultom et al., 2017). The peak observed at 1650–1600 cm⁻¹ indicates the presence of a C=C stretching group. Furthermore, a peak at 1150–1000 cm⁻¹ corresponds to the C-O stretching group. These results are not clear enough to specifically explain the content contained in the leaves of *A. siamensis*. Therefore, to obtain more detailed information, analysis using GC-MS was carried out.

The GC-MS analysis revealed that the *A. siamensis* extract contained the main components as shown in Fig-4 and Table 1. Compounds that are found in *A. siamensis* leaf extract are cinnamaldehyde (81.26%), cinnamyl ester (2.11%), Caryophyllene (1.80%), Benzofuran, 2-methyl-(1.13%), and phenol, 2-methoxy-4- (2-propenyl)-, acetate (0.95%). The most dominant compound in Acalypha siamensis extract is the compound cinnamaldehyde, (E). The cinnamaldehyde (E) compound is included in the group of phenylpropanoids or phenolic compounds (Helmalia et al., 2019) or flavonoids (Banerjee and Banerjee, 2023). The results of this research are in line with research conducted by Adriani and Atmajayanti (2023), plants that contain flavonoid and tannin compounds can be used as eco-print materials.



Fig-4. GC-MS chromatogram of A. siamensis extract

Table 1. List of the 20 dominant components in A. siamensis extract			
Retention Time	Area (%)	Name	
9.763	81.26	Cinnamaldehyde, (E)-	
12.200	2.11	Acetic acid, cinnamyl ester	
10.720	1.80	Caryophyllene	
3.665	1.13	Benzofuran, 2-methyl-	
10.914	0.95	Phenol, 2-methoxy-4-(2-propenyl)-, acetate	
3.510	0.58	Cobalt, tetracarbonylsilyl-	
4.120	0.54	Propanoic acid, pentafluoro-, ethyl ester	
3.460	0.53	Iron, tricarbonyl[(1,2,3,4eta.)-7-methylen	
3.871	0.47	1,1,1,2,2,3,3-Heptafluoro-3-methoxypropan	
4.374	0.47	2-Butene ozonide	
3.785	0.46	Semioxamazide	
4.000	0.42	4-Methylpentan-2-yl propyl carbonate	
3.831	0.41	1,4,7,10-Tetraoxacyclododecan-2-one	
3.715	0.40	Manganese, acetylpentacarbonyl-, (OC-6-2	
3.567	0.36	1H,1H,9H-Hexadecafluoro-1-nonanol	
9.491	0.36	Benzaldehyde, 4-methoxy-	
8.205	0.34	2,4,7-Octanetrione	
4.080	0.33	Succinic anhydride	

Determination of the reaction

3.950

The reaction that occurred between cinnamaldehyde (a compound that is most abundant in A. siamensis extract), alum mordant, and cotton fabric was simulated to see the reaction that occurred. The reaction between cinnamaldehyde, cellulose and aluminum sulfate (Al₂(SO₄)₃) is carried out using ORCA 5.0.2 (Zhao and Truhlar, 2008), with structure visualizations generated using VMD (Humphrey et al., 1996). The cellulose is modeled using two monomeric units of glucose monomers with β -1,4-glycosidic bonds (Kaviani et al., 2019; Reis et al., 2020; Zhao and Truhlar, 2008) (Fig-5).

0.31

3-(2-Hydroxyethyl)-2-oxazolidinone



Fig-5. Structural formula of cellulose (C12H22O11)

The formation of the complex referring to cellulose, Al3+ and cinnamaldehyde complex involves the interaction of Al3+ with hydroxyls of cellulose as well as carbonyl of cinnamaldehyde as shown in Fig-6. This finding is in line with previous study of the interaction between cellulose with Cu²⁺, Cd²⁺ and Cr³⁺ metal ions that occurred with more than an interaction of hydroxyls of cellulose and the ion (Reis et al., 2020). The Al³⁺ metal ion binds to two oxygen atoms of cellulose hydroxyls at

1.94 and 2.01 Å, respectively. In addition, the Al³⁺ metal ion binds the oxygen atom of the carbonyl of cinnamaldehyde at 1.73 Å.



Fig-6. Structure of cellulose, Al3+ and cinnamaldehyde complex



Fig-7. Cotton fabric after dyeing using A. siamensis

The interaction of metal ion, Al^{3+} with cellulose and cinnamaldehyde is evaluated from its corresponding interaction energy Eint that describes the stability of the complex. The interaction energy E_{int} is determined as the total energy difference between the complex and the isolated molecules. The calculated interaction energy E_{int} obtained at M06-2X level of theory is -3210.1 kJ mol⁻¹. The formation of complex is energetically favorable, as shown by the respective negative interaction energy.

Application of A. leaves as a natural dye in eco-print

To find out the real results of the reaction that occurs between *A. siamensis* leaves, alum mordant, and cotton fabric. *A. siamensis* leaves are used as natural dyes in eco-prints. The results of eco-print coloring with *A. siamensis* leaves as shown in Fig-7. The result of this staining is brownish green *A. siamensis* leaf marks. The results of this research are in line with the results of research conducted Deo and Prasad (2007), fabric that is treated using alum in the mordanting process will produce a darker color. Coloring using *A. siamensis* leaves produces very clear coloring, leaf traces and shapes. The fixation process using alum mordants produces very clear shapes and motifs, with a 100% assessment from the panelists (Rasmi and Nelmira, 2024).

Conclusion

Eco-print coloring using *A. siamensis* leaves can produce clear colors, marks and shapes. The results of this coloring are also in accordance with the results of simulation calculations between cellulose in the fabric, alum mordant compounds and cinnamaldehyde, the formation of a complex that is energetically favorable. These results are in line with the results of experimental research in this study which produced clear shapes, trace, and color. The resulting eco-print dye are brownish

green. However, further research is still needed regarding the use of other mordants to determine the color results and traces produced.

Conflict of Interests

The author declares that there is no conflict of interest in this research and manuscript.

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