D-limonene from orange (Citrus Maxima) peel extraction as destructive agent of styrofoam waste

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Abstract. Styrofoam is widely used for food packaging because it has good weight, stability, and stiffness but it cannot be decomposed naturally by microorganisms. D-Limonene, one of the monoterpen derivatives, is an environmentally friendly styrofoam solvent. This monoterpen is the main component of essential oil that can be extracted from orange peel. The aim of this study is to destruct styrofoam wastes (foods packaging, electronics packaging, foam boards and cup noodles) using d-limonene from orange (Citrus Maxima) peel extraction by water distillation process. The orange peel after size reduction is extracted by water distillation process at 94°C for 7 hours to produce essential oils. It was used to destruct styrofoam waste at the different volume ratio (1:0:0 ; 1:0:3 ; 1:1:2 ; 1:2:1) of the essential oil mixture (essential oil : ethanol : water). The result of this study shows that styrofoam wastes can be destructed using d-limonene from orange (Citrus Maxima) peel extraction. D-limonene of essential oil from orange (Citrus Maxima) peel can be used as destructive agent of styrofoam waste. The fastest destruction time of styrofoam waste occurs at electronic packaging with essential oil mixture by volume ratio of 1:1:2.

1. Introduction

Styrofoam or EPS (Expanded Polystyrene) is a synthetic polymer consisting of 5% polystyrene (PS) and 95% air. Because of this composition, styrofoam has good weight, stability, and stiffness. However, styrofoam cannot be decomposed naturally by microorganisms then it is polluting the environment. Several methods were developed to overcome the environmental problems caused by styrofoam waste. Liu et al studied EPS recycle by breaking down PS into its monomers by pyrolysis methods at a temperature of 450-700 °C [1]. Recycle EPS using the pyrolysis method requires high energy, so it is less economical to process styrofoam waste. Conventional methods of recycling EPS have the disadvantages of molecular degradation caused by oxidation and contamination by other materials, resulting in a reduction of the quality of the recycled polymer. A method of destruction using a natural solvent called d-limonene is promising for improving the quality of recycled PS [2].

Limonene is one of the most common essential oil constituents of aromatic plants. D-limonene is the main compound in the essential oils of the peels of Citrus sp. Limonene is widely used as a flavor and fragrance additive in consumer products such as perfume, beverages, detergents, and soaps [3]. D-limonene has been clinically used to dissolve cholesterol-containing gallstones. It has also been used for the relief of heartburn, because of its gastric acid neutralizing effect and improvement of peristalsis. D-limonene has well-established chemo-preventive activity against many types of cancers [4]. D-limonene is the main component of essential oils from orange peels with 82-99 % content [5–8]. The amount of
limonene in orange peels has not been utilized properly. Orange fruits are only used for consumption as a source of vitamin C while the peels and up in the trash. The peels of orange (Citrus Maxima) which are mostly produced from Nambangan, Madiun is one of them that has not been widely used. This type of orange has a unique characteristic, has a large size, fresh taste, and long shelf life which can reach 4 months [9]. The long shelf life of orange (Citrus Maxima) peels is a supporting factor in the development of research to increase its economic value.

The high limonene content of orange (Citrus Maxima) peels essential oil (77.7 – 95.4%) has the potential to be converted into more useful products [10,11]. One of the uses of limonene as a destructive agent of styrofoam waste.

Several methods have been developed to produce essential oils from orange peels ranging from simple technology to the latest technology. Kurniawan et al compared distillation, pressing, and leaching methods to produce essential oil of orange peel with the best quality and highest yield. From the result of this research, the best quality of citrus peels essential oil obtained by the distillation method [12]. Giwa et al using steam distillation, water distillation, and solvent extraction to produced essential oil from orange peels. From the results obtained, it was discovered that the orange peels could give the maximum yields of essential oil 3.47% when water distillation was employed [13].

Based on the above description, it is necessary to develop research on the use of d-limonene from orange (Citrus Maxima) peels as a styrofoam destruction agent. This study aims to destruct styrofoam waste using d-limonene from orange (Citrus Maxima) peel extraction by water distillation process. Styrofoam wastes from foods packaging, electronics packaging, foam boards, and cup noodles were destructed at the different volume ratios (1:0:3; 1:1:2; 1:2:1) of the essential oil mixture (essential oil : ethanol: water). The destruction time is measured to obtain the fastest destruction time and the best composition of the destructive agent.

2. Materials and methods

D-limonene, the highest essential oil component from orange peel, was extracted by water distillation process. The essential oils were used to destruct styrofoam wastes from foods packaging, electronics packaging, foam boards, and cup noodles in certain compositions of the essential oil mixture with ethanol and water. The essential oil was analyzed by GCMS (Gas Chromatography-Mass Spectrometry) analysis to determine the essential oil content. Density and viscosity of the essential oil were also analyzed. The essential oil mixture (Essential Oil : Ethanol : Water) was used as destructive agent for styrofoam wastes. The destruction time for each type of styrofoam waste was determined at various volume ratios of the essential oil mixture.

The experiment can be divided into four steps, i.e.:
- Pretreatment of orange (Citrus Maxima) peel
- Extraction of essential oil by water distillation process
- Destruction of styrofoam wastes
- Analysis

2.1. Materials

Materials used in this research included orange (Citrus Maxima) peel that collected from local market, aquadest, ethanol with the composition of 96%, styrofoam wastes (foods packaging, electronics packaging, foam boards, and cup noodles) that collected from household waste.

2.2. Methods

2.2.1. Pretreatment of orange (Citrus Maxima) peel. The collected orange peel was separated from the impurities manually then it was processed by size reduction using a blender.

2.2.2. Extraction of essential oil by water distillation process. After size reduction, 220 grams of the orange peel were put into the erlenmeyer and added 220 ml of aquadest. Then, it was processed by
distillation to extract the essential oil at temperature of 94°C for 7 hours. After the distillation process, the distillate containing the water-essential oil mixture (hydrosol) was separated using a separating funnel. The water distillation apparatus can be seen in Figure 1.

![Water distillation apparatus](image)

**Figure 1.** The water distillation apparatus.

2.2.3. *Destruction of styrofoam wastes.* The types of styrofoam waste used in this study were foods packaging, electronics packaging, foam boards, and cup noodles. The wastes cut into uniform sizes (1 x 1 x 0.25 cm) then put it into a bottle sample containing the essential mixture as destructive agent. Styrofoam wastes were destructed using essential oil mixture with ethanol and water at various volume ratios, i.e. 1:0:0; 1:0:3; 1:1:2; 1:2:1. The destruction time was measured from the beginning of mixing until the styrofoam was completely destructed then it was determined the best composition of the destructive agent.

2.2.4. *Analysis.* The essential oil extracted from orange (*Citrus Maxima*) peel was analyzed using GCMS analysis to determine the content of the essential oil. The density of essential oil from orange peels was measured by pycnometer and the viscosity of essential oils was measured by analyzing the intrinsic viscosity using the Ostwald viscometer instrument.

3. Results and discussion

3.1. *The components of essential oil from orange (*Citrus Maxima*) peel extraction*  
The essential oil product from orange (*Citrus Maxima*) peel extraction using water distillation process was analyzed by GCMS to determine the constituent components. The results of GCMS analysis as presented in Table 1 show that d-limonene is the highest essential oil component with the composition of 90.43% w/w. It is consistent with the result of other studies that analyzed the essential oil content of various varieties [5-7]. The highest essential oil component from orange peel extraction is d-limonene. Therefore, the essential oil is also known as limonene oil [8].

| Peak | Component     | Content (% w/w) | Peak | Component     | Content (% w/w) |
|------|---------------|-----------------|------|---------------|-----------------|
| 1    | α-Pinene      | 1.12%           | 6    | Sabinene      | 0.48%           |
| 2    | Sabinene      | 0.31%           | 7    | α-Terpineol   | 0.23%           |
| 3    | Myrcene       | 4.96%           | 8    | Geranyl acetate | 0.42%        |
| 4    | α-Phellandrene| 0.58%           | 9    | β-Caryophyllene| 0.19%           |
| 5    | Limonene      | 90.43%          | 10   | Germacrene-D  | 1.28%           |

Table 1. The results of GCMS analysis of essential oil from orange (*Citrus Maxima*) peel extraction.

Monoterpene derivatives, such as d-limonene, isoamyl acetate, benzyl propionate, and ethyl butyrate, are good solvents for styrofoam. D-limonene, which is a natural vegetable oil extracted from the orange.
peel, is the best destructive agent for styrofoam because of its high solubility, stable supply, and fragrance [2]. The results of the previous studies showed that the essential oil could destruct styrofoam [13,14].

The physical properties of the essential oil from orange (Citrus Maxima) peel extraction were also determined the density and viscosity. Based on the measurement, the density of the essential oil is 0.8494 g/mL and the viscosity is 8.636 x10^{-3} g/cm.s.

3.2. Destruction of styrofoam wastes
Essential oil product by water distillation process was used as destructive agent for styrofoam waste. It was mixed with ethanol and water with various volume ratios, i.e. 1:0:0; 1:0:3; 1:1:2; 1:2:1. Ethanol was added to the mixture to facilitate the penetration of essential oil particles into the styrofoam waste. The mixture of essential oil, ethanol, and water was used to destruct styrofoam wastes from foods packaging, electronics packaging, foam boards, and cup noodles. The destruction time of styrofoam wastes at various volume ratios of the mixture are presented in Figure 2.

![Figure 2](image_url)

**Figure 2.** The destruction time at various volume ratios with the various styrofoam waste types.

Figure 2 shows that the destruction time tends to decrease at the composition of 25% v/v ethanol addition (at volume ratio of 1:1:2) but increase at the composition of 50% v/v ethanol addition (at volume ratio of 1:2:1). It is consistent with the research of Noguchi et al who reported that ethanol is a poor destructive agent for styrofoam. The addition of 3% by weight of pure limonene decreased the viscosity of limonene however maximized the penetration of the essential oil mixture to styrofoam and accelerated the destruction time [2]. The fastest destruction time occurs at the essential oil mixture by the volume ratio of 1:1:2 (Essential Oil: Ethanol: Water). The best composition mixture is determined on the composition with the minimum amount of essential oil because the price of essential oil is high. The composition of 1:0:0 is only for comparison however it is not recommended because the pure essential oil is expensive.

3.3. Effect of styrofoam waste types on the destruction percentage
Destruction percentage of styrofoam waste can be used to determine the amount of the destructed styrofoam waste at the various styrofoam waste types with the same volume ratio of the essential oil
mixture. The destruction percentage was calculated by the mass ratio of the destructed styrofoam to the destructive agent. The results of this calculation be seen in Table 2.

| Volume Ratio | Styrofoam Waste Types | Destruction Percentage (%) |
|--------------|------------------------|-----------------------------|
| 1:1:2        | Foods Packaging        | 8.48                        |
|              | Electronics Packaging  | 9.37                        |
|              | Foam Boards            | 6.95                        |
|              | Cup Noodles            | 7.20                        |

Table 2. Destruction percentage of styrofoam waste.

At the same volume ratio of essential oil mixture, Table 2 shows that the lowest percentage of destruction occurs at cup noodles with the percentage of 7.2%. The electronics packaging result the highest percentage of destruction with the percentage of 9.37%. It is caused by the density of the polymer in the styrofoam varies depending on the types of styrofoam.

4. Conclusion
Styrofoam wastes can be destructed using d-limonene from orange (Citrus Maxima) peel extraction by water distillation process. D-limonene of the essential oil (90.43% w/w) can be used as destructive agent of styrofoam wastes from foods packaging, electronics packaging, foam boards, and cup noodles. The fastest destruction time of styrofoam waste occurs in essential oil mixture by volume ratio of 1:1:2. The highest destruction percentage (9.37%) of styrofoam waste occurs at electronics packaging.

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