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Formulation of Alkyd Resin from Palm Oil and Tuna Fish Oil:Effect of Acid Value and Non-Volatile Content as Surface Coating

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Abstract *– Resin is a polymer chemical substance that is made up of monomers from carbon and nitrogen. Alkyd resin, a surface coating resin, produced by polycondensation reaction between polybasic and polyhydric alcohols, modified with monobasic acids or their anhydrides, and drying oil at controlled temperatures. Alkyd resin is widely used in the paint and surface coating industries. The advantages of this resin are its strong chemical and thermal resistance, quick drying power, and inexpensive cost. Alkyd resins can be produced from natural oil (palm and tuna fish oil). Oil derived from palm and tuna fish is known as natural oil. Tuna fish oil includes 40.04 % oleic acid. The oleic content is suitable for use in the alcoholysis process in the production of alkyd resins. The major ingredients in this study were tuna fish oil and palm oil. The aim of this research is to determine the influence of a combination of palm oil:tuna fish oil ratio, amount of phthalic anhydride, and temperature of alcoholysis process on the value of acid number and non-volatile content can be used an alternative material for making alkyd resin was investigated using a Completely Randomized Design (CRD). Alkyd resin produced in various ways using tuna fish oil has too high acid value and brown tint, making it unsuitable for use as a surface coating resin.*

Keywords: *alkyd resin, drying oil, non-drying oil, palm oil, tuna fish oil*

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INTRODUCTION

Polymers have a significant role in human life today. Humans require huge quantities of polymers such as plastics, adhesives, and other materials. Resin is a polymer chemical substance that is made up of carbon and nitrogen-based monomers. Resin can be synthesized by interacting various substances to produce resin polymers, either naturally or synthetically. Epoxy resins, polyester resins, vinylester resins, acrylic resins, phenolic resins, silicone resins, polyurethane resins, polyethylene resins, polystyrene resins, polycarbonate resins, polyamide resins, polyproylene resins, and alkyd resins are the 13 types of resins commonly used today (Danesh, 2021). Polycondensation reactions between polybasic and polyhydric alcohols, modified with monobasic acids or their anhydrides, and drying oil at controlled temperatures produce alkyd resins (Uzoh et al., 2016). This is accomplished through the use of natural oil or fatty acids. Heiskanen, (2010) explained that alkyd resins are extensively used in the surface coating application because of their excellent mechanical properties, reliability, superior drying speed, high gloss, lower cost than the acrylic binder. Alkyd resins are produced in two ways: alkyd resins made from natural oil and alkyd resins made from fatty acids. The second is fatty acids, also known as fatty acids, which are oil derivatives that are divided into two types: natural and synthetic, each of which is further divided into fatty acids and unsaturated fatty acids. Linoleic acid is an example of a natural fatty acid, while versatic acid is an example of a synthesized fatty acid. Oil derived from palm and tuna fish is known as natural oil. The oils utilized in the production of alkyd resins from natural oils are typically triglycerides, which lack functional groups that can react with polyols and acids to create polyester (Kesuma, 2021). The natural excess of Oil than Fatty Acid is a relative price, easily sought and low production costs.

Palm oil is edible plant oil derived from the fruit of oil palm trees. Two types of oil can be produced; from squeezing the freshly fruit and palm kernel oil which comes from crushing the kernel or stone in the middle of the fruit. Palm stearins are more solid severe in comparison to oil seeds crops. The fatty acid composition of palm kernel stearin is approximately 60% saturated and 40% unsaturated fatty acid. therefore, palm kernel stearin is classifies as saturated fatty acid due to its very low degree of unsaturated and alkyd resind, which are based on palm kernel stearin (Uzoh et al., 2016).

Tuna fish oil is one of the non-drying oil types that has three fat acid is, fat acid, non-fat acid, and fat acid is not a dual saturation. Tuna fish oil includes 40.04 % oleic acid. The oleic content is suitable for use in the alcoholysis process in the production of alkyd resins.

Previous research from Athawale and Nimbalkar (2011), using oil of soy oil and tuna fish oil is sardine fish oil acquired from Jantha Fish Meal (Mangalore). The method used is the alcoholicism process and the esterification process. The resin is synthetic in the reactor using the batch process at 220 ºC temperature for 8 to 10 hours. At the adhesion testing on all samples indicate a good result is 80% of it. While, on the test of chemical endurance test the entire sample has not been experienced. The alkid resins from sardine fish oil has a lower dried trait than soy oil. The excess of this research is the resin Alkid who uses sardine oil that is more saving sardine oil and can function as a surface-upper resin, whose flaws are less than soy oil.

METHODOLOGY

Material and Tools

Tuna fish oil and palm oil will be used as the key ingredients in this study. Palm oil is obtained from PT Wilmar Nabati Indonesia, Tuna fish oil (99% purity) is obtained from tuna fish waste. The supporting material is glycerine (99.5% purity) obtained from PT Wings Surya, phthalic anhydride from PT Petrowidada, and NaOH from PT Asahimas Subentra Chemical. The catalyst used is LiOH derived from PT ChengTok Lithium Indonesia.

This research aims to determine the influence of a combination of palm oil:tuna fish oil ratio, amount of phthalic anhydride, and temperature of alcoholysis process on the value of acid number and non-volatile content of alkyd resin was investigated using a Completely Randomized Design (CRD).

Completely Randomized Design (CRD) is a type of design the simplest experiment. This design is common used for experiments that have media or environment uniform or homogeneous experiments (Mattjik, 2006).

Run	PO:TFO	G	PA
A1	100 % : 0 %	180	180
A2		170	200
A3		160	220
B1	30 % : 70 %	180	180
B2		170	200
B3		160	220
C1	50 % : 50 %	180	180
C2		170	200
C3		160	220
D1	70 % : 30 %	180	180
D2		170	200
D3		160	220
E1	0 % : 100 %	180	180
E2		170	200
E3		160	220

Table 1. Completely Randomized Design (CRD)

PO = Palm Oil
TFO = Tuna Fish Oil
G = Glycerine (gr Glycerine/ml Oil)
PA = Phthalic Anhydride (gr PA/ml Oil)

Method

Preparation of Resin Alkyd

A two-stage method was used in this investigation, namely alcoholysis and esterification, which were both done in batches in a three-neck flask.

The alcoholysis process begins with a solution of oil and glycerine, which is then heated to 240°C. After the alcoholysis process had been finished for 45 minutes at 200ºC, the esterification process was carried out (Pasaribu et al., 2019).

LiOH catalyst is added to the esterification process to speed up the reaction. During the procedure, nitrogen is pumped into a three-neck flask to keep the resin from becoming oxidized or black. An alkyd resin with c/c (clear and clean) criteria is created when the alcoholysis and esterification procedures are completed. Tuna fish oil, glycerine, and phthalic anhydride were the altering factors in this investigation. The goal of this research was to find the best alkyd resin from a mixture of palm oil and tuna fish oil, as well as the effects of glycerine and phthalic anhydride on acid value, non-volatile content, viscosity, and resin color.

Acid Value Tester

Analysis of Acid Number refers to (Kesuma, 2021). Weight 20 g of sample solution in a 250 ml Erlenmeyer flask. Then add 50 ml of 95% alcohol. Heat in a water bath, stirring, until boiling. Titrate the solution with 0.1 N KOH using phenolphtalein indicator until it turns pink.

$$\text{Acid Value} = \frac{ml\ KOH\ x\ N\ KOH\ x\ 56{,}1}{gram\ sample} \quad (1)$$

Non-Volatile Content (Solid Content)

Analysis of non-volatile content (solid content) based on the article (Okparanta et al, 2016). About 100 g of the short oil resin were weighed and placed in a pre-weighed crucible and heated until the water content and any other volatile components were completely vaporized leaving behind the solid. The crucible with its contents was allowed to cool and re-weighed. Percentage solid content (non-volatile component).

$$\text{Non volatile} = \frac{W1-W2}{W} \times 100\% \quad (2)$$

Viscosity

Analysis of viscosity refers to (Paparingan et al., 2019). Sample of 500 mL of solution is poured into a beaker glass and then mounted on a viscometer. The viscometer is turned on so that the rotor spindle rotates up to a speed of 50 rpm. The rotor spindle is inserted into the sample solution until the rotor spindle head rotates halfway through the solution. The Erlenmeyer is allowed to stand for a while, let the rotor spindle rotate for a few seconds until the displayed number on the spindle stabilizes, then the value is recorded.

Physical Properties Analysis

Color Test: the color of the short oil resin was examined visually using the sense of sight.

RESULTS AND DISCUSSION

The effect of combining palm oil with tuna fish oil, adding glycerinee, and using phthalic anhydride on the quality of the alkyd resin produced was investigated in this study. Table 2 provides the following information regarding the findings of the analysis of alkyd resin products.

Table 2. Research Data

PO : TFO	G	PA	AV	NV	μ
0 % : 100 %	180	180	55,73	88,57	946
	170	200	67,95	84,76	987
	160	220	72,41	81,88	1212
30 % : 70 %	180	180	40,33	73,89	230
	170	200	47,61	74,91	276
	160	220	54,72	75,23	297
50 % : 50 %	180	180	50,9	86,40	84
	170	200	56,3	87,22	160
	160	220	61,7	88,04	236
70 % : 30 %	180	180	51,92	86,25	120
	170	200	67,41	86,93	183
	160	220	82,9	87,60	246
100 % : 0 %	180	180	7,94	77,29	98
	170	200	8,46	76,91	112
	160	220	9,58	75,21	120

PO = Palm Oil
TFO = Tuna Fish Oil
G = Glycerine (gr Glycerine/ml Oil)
PA = Phthalic Anhydride (gr PA/ml Oil)
AV = Acid Value (mgKOH/g)
NV = Non Volatile (%)
μ = Viscosity (mPa.s)

Impact on Acid Number of Combining Palm Oil and Tuna Oil, Adding Glycerine, and Adding Phthalic Anhydride

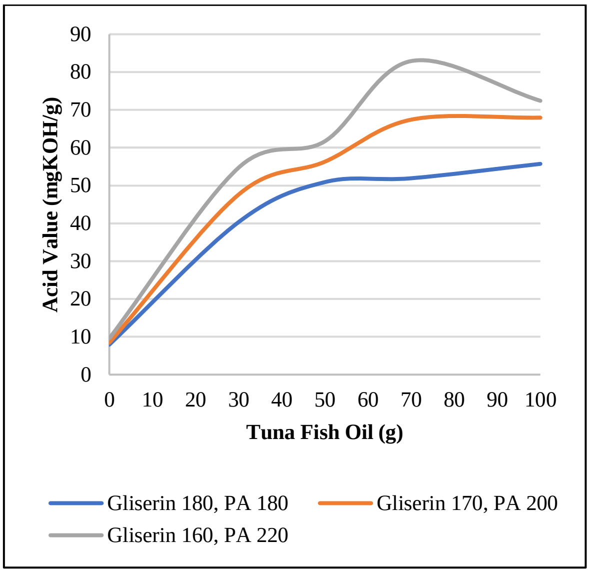

Figure 1. Impact on Acid Number of Combining Palm Oil and Tuna Oil, Adding Glycerine, and Adding Phthalic Anhydride

According to Figure 1, the combination of 180 glycerine (gr glycerine/ml oil) and 180 PA (gr PA/ml oil), glycerine 170 ; PA 200, and glycerine 180 ; PA 180 at all ratios of vegetable and tuna fish oils, respectively, indicated the lowest to greatest acid number values.

The acid number will rise as the fish oil ratio increases. This can be explained by pointing out that the high concentrations of free fatty acids (FFA) in fish oil are what cause the increase in acid number at a high ratio of the oil (Atimuttigul dkk., 2006). In addition, the amount of glycerine solvent used decreased the amount of acid created. This is because there are more reactants present, which raises the frequency of intermolecular collisions and enhances the conversion of carboxyl groups. As a result, there are fewer leftover -COOH groups in the alkyd resin product, which results in a lower acid number value (Nurandini dkk., 2018). In the experiment where phthalic anhydride was added, it was discovered that the value of the acid number created increased with increasing phthalic anhydride.

According to the Indonesian National Standard (SNI 06-0504-1989), the resulting alkyd has a maximum acid number of 20. The alkyd produced in the experiment of the ratio of palm oil:tuna fish oil 100%:0% in all ratios of glycerine and phthalic anhydride has an acid number < 20 so that it can be used as a surface coating. For the purposes of making special paints, the value of the acid number can be higher. However, alkyds with low acid values produce non-corrosive products and are suitable for surface coatings.

Tuna fish oil and palm oil combined, with glycerine and phthalic anhydride added to make a non-volatile percentage

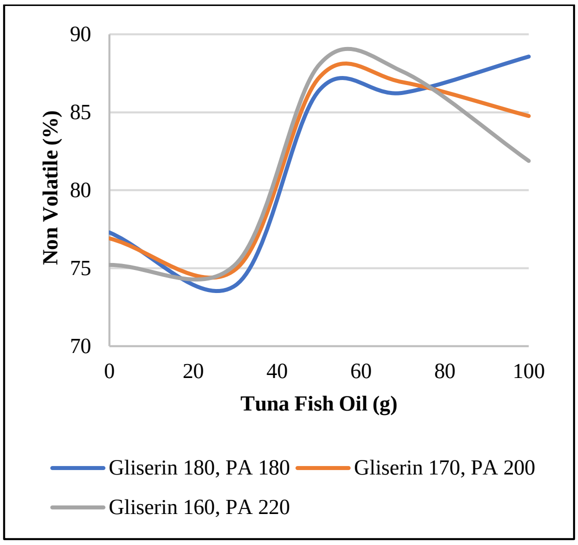

Figure 2. Tuna fish oil and Palm Oil Combined, with Glycerine and Phthalic Anhydride added to Make a Non-Volatile Percentage

The combined effects of palm oil, tuna fish oil, glycerine, and phthalic anhydride on non-volatile percentage are depicted in Figure 2. The figure explains that the percentage of non volatile oil decreased in the sample ratio of palm oil to tuna fish oil from 100%:0% to 70%:30% but tended to grow at a higher ratio (palm oil:tuna fish oil). Because palm oil has a larger molecular weight than tuna fish oil (848 g/mol), the percentage of non-volatile materials has increased. The larger the solid content, the longer the alkyd oil contained.

With the addition of glycerine:phthalic anhydride 180:180, there was a rise in the percentage of non-volatile in the ratio of palm oil:tuna fish oil 0%:100%. With the addition of glycerine:phthalic anhydride 170:200 and 160:220, the ratio of palm oil to tuna fish fat, which is 0%:100%, exhibited a drop in non-volatile content. Adiningsih & Fauziati, (2016) study, which claimed that phthalic anhydride is a form of polybasic acid added to alkyd resin, is at odds with these findings. The amount of non-volatile content increased as polybasic acid's reactions with oil and glycerol increased. This is due to the increased production of polyester. 30-80% is the typical non-volatile percentage for alkyd resins (Aidha & Jati, 2017). Figure 2 displays the percentage of non-volatile in samples with a palm oil:tuna fish oil ratio of 100%:0% and 30%:70% that satisfies the criteria. The sample is a short oil alkyd with a fatty acid content that is less than 35% of what is found in palm oil (Aidha & Jati, 2017). Additionally, samples with more glycerine will result in a more liquid alkyd resin, lowering the alkyd resin's non-volatile content. The drying process will take a long period if resin is applied.

Viscosity Effect of Combining Tuna Oil and Palm Oil with Glycerine and Phthalic Anhydride

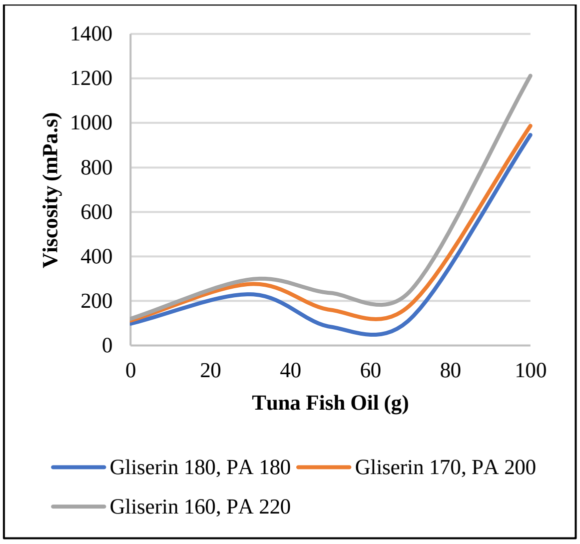

Figure 3. Illustrates the Impact on Viscosity of Combining Palm Oil and Tuna Oil with The Addition of Glycerine and Phthalic anhydride

The amount of double bonds has an inverse relationship with viscosity, which is directly proportional to the length of the carbon chain. The viscosity of fatty acids and alcohols increases with increasing carbon chain length. In contrast, the oil becomes more saturated the higher the viscosity (UMY, 2014). Figure 3 demonstrates that viscosity increases with increasing tuna fish oil ratio. This is because fish oil contains more double bonds than palm oil does. A more prevalent form of fatty acid found in fish oil is one with a carbon atom number of 20 or 22, which is particularly unsaturated due to the presence of 5 or 6 double bonds per molecule (Galer & Hasselt, 2011). Saturated and unsaturated fatty acids are both in balance in palm oil.

The amount of glycerine added is inversely correlated with the viscosity measurement. The viscosity value decreases as glycerine is added more. Due to glycerine's ability to absorb moisture from the air and increase the water content of the resin, it is hygroscopic, which results in a drop in viscosity (Wicaksana, 2016).

The higher the phthalic anhydride ratio, the higher the viscosity value was in the experiment with the addition of phthalic anhydride. This is caused by the addition of high levels of phthalic anhydride, which has more benzene rings and a stiffer structure, making it harder for the resulting alkyd resin to flow than samples with low levels of phthalic anhydride (Isaac & Nsi, 2013).

Effects of Glycerine and Phthalic Anhydride, as well as Tuna Fish Oil and Palm Oil Additions, on Color

Table 3. Effects of Glycerine and Phthalic Anhydride, as well as Tuna Fish Oil and Palm Oil Additions on Color

PO : TFO	Color
0% : 100%	Dark Brown
0% : 100%	Dark Brown
0% : 100%	Dark Brown
30% : 70%	Dark Brown
30% : 70%	Dark Brown
30% : 70%	Dark Brown
50% : 50%	Light Brown
50% : 50%	Light Brown
50% : 50%	Light Brown
70% : 30%	Dark Brown
70% : 30%	Dark Brown
70% : 30%	Dark Brown
100% : 0%	Light Yellow
100% : 0%	Light Yellow
100% : 0%	Light Yellow

Table 3 displays the color analysis results for alkyd resins for tuna fish oil variables and demonstrates that the darker the hue, the higher the ratio of tuna fish oils used. Due to the original hue of tuna fish oil, which is darker than palm oil, the product has a dark tint (palm oil). In addition, the growing number of oxidation reactions that take place during the process may also contribute to the dark color of the alkyd resin. During the alcoholysis process, the three-neck flask's oxygen content also causes the alkyd resin to turn brown, necessitating a constant flow of N_2 as an inert gas that serves to flush the flask of oxygen.

CONCLUSION

In this study, a variety of tests were conducted to investigate the effects of a combination of glycerine, phthalic anhydride, palm oil:tuna fish oil ratio, glycerine amount, acid number analysis, and non-volatile analysis on alkyd resin formulations. According to the results of this investigation, formulations of 100% palm oil: 0% tuna fish oil and the amounts of glycerine and phthalic anhydride as follows are the optimum formulations for manufacturing alkyd resins with a maximum acid number value of 20 mgKOH/g. Tuna's tuna fish oil is ineffective when employed as a component in the production of alkyd resins. A 100% palm oil sample yielded the best acid number in alkyd resin according to SNI 06-0504-1989, which is <20 mgKOH/g. Samples with a palm oil: tuna fish oil ratio of 100%:0% and 30%:70% were found to have non-volatile percentages that met the requirement. The sample is a short oil alkyd, which means it has less than 35% of the fatty acids found in palm oil. The viscosity value decreases as glycerine is added more. The viscosity of alkyd resin can be raised by adding phthalic anhydride. Alkyd resin produced in various ways using palm oil has a brown tint, making it unsuitable for use as a surface coating resin.

NOTATION

PO = Palm Oil
TFO = Tuna Fish Oil
G = Glycerine (gr Glycerine/ml Oil)
PA = Phthalic Anhydride (gr PA/ml Oil)
AV = Acid Value
NV = Non Volatile (%)
μ = Viscosity (mPa.s)
W1 = Weight of crucible + resin, before heating
W2 = Weight of crucible + resin, after heating and cooling
W = Weight of resin before heating

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