Mutagenicity of Teakwood 2-Methylanthraquinone and Its Catalytic Effects on Pulping for Food Packaging Paper

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ABSTRACT

The application of anthraquinone (AQ) (9,10-anthracenedione) as cooking additive has been well documented in scientific studies. It is effective for accelerating delignification and stopping carbohydrate degradation, enabling a reduction in cooking time, temperature, and chemical charge. Thus, AQ has beneficial effects in the pulp and paper industries for incremental pulp production, energy reduction, and protecting wood resources. AQ derivatives are widely found in higher plants. In plants, Mycelium of *Pyrenophora graminea* consists of two oxygenated AQ. The oxygenated AQ structures do not have the catalytic effects on cooking. AQ also can be synthesized at least three different methods, oxidation of anthracene (AQ-OX), Friedel-Craft reaction (AQ-FC), and Diels-Alder chemistry (AQ-DA). In assessing the potential biological activity of productions of AQ, it is critical to be aware of how their synthesize and the potential contaminants inherent with different synthesis processes. Based on a report by the International Agency for Research on Cancer (IARC), some AQ impurities are potentially mutagenic or carcinogenic and may impact negatively on human beings. In 2011, IARC classified AQ as “Group 2B, possibly carcinogenic to humans”, based on a report in 2005 by the National Toxicology Program (NTP). On the other hand, in 2013 German Federal Institute for Risk Assessment (BfR) removed AQ from its approved list of chemicals for

use in the manufacture of paper and paperboard that contact food. Based on the above concerns, AQ has become less frequently used in the paper industry. The U.S. Food and Drug Administration (FDA) allowed an AQ charge at addition level as 0.1% on oven dried wood for a cooking additive in the alkaline cooking of wood material, which is at the maximum amount for food contact paper.

Some AQ derivatives, such as 2-methylanthraquinone (2-MAQ), are also effective cooking additives. Tectoquinone (natural 2-MAQ) has been found in teak (*Tectona grandis L.f.)*. Teak grows naturally in Southeast Asia and other tropical and subtropical regions such as Australia, Africa, and Latin America. Teak wood is one of the most important valuable timber in Indochina region and the most highly valued hardwoods. Quinones are major secondary metabolites which are present in teak wood in the form of naphthoquinones and anthraquinones. Teak wood extract mainly contains chemical compounds, such as lapachol, deoxylapachol, 5-hydroxylapachol, 2-methylanthraquinone (2-MAQ), 1-hydroxy-2-methylanthraquinone.

Generally, 2-MAQ in industry is synthesized from 1,4-naphtaquinone and isoprene by Diels-Alder reaction. It also can be synthesized from phthalic anhydride and toluene by Friedel-Crafts reaction.

In Chapter 2, to overcome the AQ issues raised by the BfR and others, we explored the use of Myanmar teak wood extract in kraft cooking. Determining the effect of teak extractive on pulp quality using eucalyptus wood chips and the amounts of residual 2-MAQ in pulp is an aim of the study, which is important for ensuring food safety. Kraft cooking with the acetone extract of Myanmar teak wood increased pulp yield by 1.6% but decreased kappa number by two points compared to that obtained using commercial 2-MAQ. The addition of 0.03% 2-MAQ to kraft cooking had benefits of saving forest resources and fossil energy in industry with increased pulp yield by 0.7–1.2% at the same kappa number (17–20) compared to no addition. Ames testing clarified that 2-MAQ was not mutagenic. Unbleached and oxygen-bleached pulp contained 0.40–2.90 and 0.21–0.39 ppm residual 2-MAQ, respectively, while 2-MAQ was not detected in fully bleached pulp; therefore, this pulp should be safe for food-packaging use.

The goals of the research described in the Chapter 3, here were to isolate and quantify 2-MAQ and deoxylapachol from Indonesian teak wood, and to compare the cooking effectiveness of the teak extract to that of natural 2-MAQ and natural deoxylapachol. Secondly, to determine the amounts of residual 2-MAQ and deoxylapachol in unbleached for ensuring food safety as food-packaging materials. Indonesian teak wood has three major compounds while Myanmar teak wood has one major compound (2-MAQ). Based on the analysis data from GC-MS, it was shown Indonesian teak wood contains deoxylapachol, isodeoxylapachol (its isomer), and 2-MAQ. The deoxylapachol and its isomer are the highest-amount components in Indonesian teak wood compared to 2-MAQ. The deoxylapachol and 2-MAQ were isolated and purified by column chromatography. These compounds were subjected as additive in kraft cooking in comparison with the Indonesian teak wood extractive. The resulted pulps showed the deoxylapachol from Indonesian teak wood extract also gave slightly effect in pulp yield increment.

The overall results showed that the Myanmar teak wood extract gave the best effect in kraft cooking (high pulp yield and low kappa number) compare to Indonesian teak extractive. Presumably, because 2-MAQ in Myanmar teak extract is major compound , meanwhile in Indonesian teak wood extract has three major compounds, such as deoxylapachol , its isomers and 2-MAQ. It might be three major compunds in Indonesian teak extract have inhibition effect when they used in kraft cooking. Interestingly, when Indonesian teak extract was purified further to get deoxylapachol compunds, the effect of deoxylapachol was similar with 2-MAQ as catalyst in kraft cooking. The order of delignification catalytic effectiveness was Myanmar teak extract> Natural or synthesized 2-MAQ>deoxylapachol>Indonesian teak extract.