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Biodegradability Properties of Polyurethane Film Made from *Eucalyptus pellita* Wood Polyol

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ABSTRACT

The properties of polyurethane (PU) film are greatly influenced not only by the raw materials but also by the compatibility of polyol and isocyanate. This paper aimed to evaluate the effect of the isocyanate index (NCO/OH ratio) on the biodegradability properties of *Eucalyptus pellita* PU film. *E. pellita* wood polyol and polymeric methylene diphenyl diisocyanate (pMDI) were mixed at different NCO/OH ratios (1.8 – 3.0). The PU film was produced through the one-shot method. The effect of the NCO/OH ratio on the biodegradability properties of PU film was evaluated. The rate of biodegradation of PU film by soil burial test decreases proportionally to the NCO/OH ratio. The biodegradation rate is the highest (14.02%) when the NCO/OH ratio is the lowest (1.8). The results of water solubility showed that PU films with low NCO/OH ratios are easily soluble in water. The band associated with the ester compound was detected at nearly 1,060 cm^{-1}. Based on the findings of this study, increasing the NCO/OH ratio made the PU film from *E. pellita* less degradable. Therefore, lowering the NCO/OH ratio is an ideal option to produce films with better biodegradability.

Keywords: polyurethane film, biodegradability, water solubility, FT-IR

INTRODUCTION

Polyurethane (PU) is a versatile polymer that can be used in a variety of applications such as foam, elastomer, film, and adhesive (Acik et al., 2018; Mizera & Ryszkowska, 2016; Tuohedi & Wang, 2021; Zhang et al., 2020; Zheng et al., 2011)bio-based epoxy resins were prepared from polyhydric-alcohol-liquefied cotton stalk by glycidyl etherification. The cotton stalk was liquefied in a polyethylene glycol/glycerol cosolvent under H2SO4 catalysis. Epon 828 and cotton-stalk-based epoxy resins could be cured using methylhexahydrophthalic anhydride as the curing agent, and the curing process was exothermic. The thermal properties and tensile strength of cured resins were investigated to examine the effect of adding cotton-stalk-based resin on the performance of the copolymerized epoxy resin. Further, the liquefied-cotton-stalk-based epoxy resin was blended with Epon 828 at different ratios (10, 20, and 30 mass%. PU is typically made from non-renewable materials such as petroleum or fossil fuels. However, as we move into the twenty-first century, other alternative materials should be thoroughly researched to replace our reliance on petroleum-based materials for PU production. The most important goal is to balance or reduce the environmental effects, as well as to be concerned about the depletion of fossil fuels in future generations. Furthermore, many abundant renewable materials were discovered to have the potential for PU synthesis, which is great for reducing our reliance on petroleum sources.

To date, PU obtained from various biomass sources has been successfully recorded. Wheat straw (Wang et al., 2008), oak and alder wood sawdust (Gosz et al., 2021; Kurimoto et al., 2001), *Mesua Ferrea* L. seed oil (Dutta & Karak, 2006), palm oil components (Amran et al., 2021; Yeoh et al., 2020), *Acacia mangium* (Palle et al., 2023) and castor oil (Panda et al., 2017) are among the biomass that has been used for PU production. Along with the green transition movement, the biodegradability of future PU materials and products is important because a lot of waste ends up in the natural environment and waste management facilities. (Hammer et al., 2012).

It has been reported that PU derived from bio-based materials is degradable. Dutta et al. (2010) discovered that after 180 days of biodegradation of film from *Mesua ferrea* L. seed oil, the intensity of bands for ester linkages decreased. Lopes et al. (2022) concluded from FT-IR and SEM analysis that polyurethane made of linseed oil with NCO/OH ratios of 0.8 and 1.2 was susceptible to microbiological deterioration. Zhang et al. (2013) discovered that after 12 months buried in soil, PU foam degraded to the wood components and ether soft segment via an oxidation reaction. According to Sahoo et al. (2018), soil biodegradation of bio-based polyester PU with the aid of soil microorganisms resulted in the greatest weight loss.

Because of its rapid growth and potential for profit, *Eucalyptus pellita* is now widely planted in Malaysia, particularly in Sabah and Sarawak (Zaiton et al., 2020). The main products of these planted Eucalyptus are timber, firewood, and pulp and paper (Hassan et al., 2021; Hii et al., 2017). Unused industrial waste in the form of chips and sawdust has potential in the bio-chemical industry. As a woody plant, the chemical content of *E. pellita* such as cellulose, hemicellulose, and lignin, contain two

or more hydroxyl groups (Ertaş et al., 2014), which demonstrate their capacity as a raw material (polyol) for PU production. It has potential applications in industries such as food packaging, biomedical appliances, plastics, and agricultural technology. To the best of our knowledge, no studies on the biodegradation of PU film made from *E. pellita* wood polyols have been published. Therefore, this paper may provide important insight into the biodegradation behaviour of PU film made from *E. pellita* as a local species. In this study, the previously obtained *E. pellita* wood polyol (Nurul Hazwani et al., 2023) was used as raw material for producing PU films via reactions with pMDI. The stoichiometric ratio of OH (*E. pellita* wood polyol) to NCO was varied, and the biodegradability potential was evaluated to assess the environmental effects of using non-renewable sources. The findings aimed to highlight the biodegradability properties of *E. pellita* PU film as influenced by the NCO and OH ratio. The polymer structure was assessed using Fourier transform infrared (FT-IR) spectra, and the rate of biodegradation of these films was assessed using a water absorption test and indoor soil burial via weight loss measurement.

Materials

Eucalyptus pellita wood chips obtained from Sabah Softwood Berhad, Sabah, Malaysia were used in this study. The wood chips were ground with a Wiley mill and sieved, and particle sizes greater than 120 mesh were used. The samples were dried at 105 ± 3°C before liquefaction. Polymeric methylene diphenyl diisocyanate (pMDI) was supplied by Sigma-Aldrich and used as the crosslinker. Polyethylene glycol (#400), glycerol, distilled acetone, methanol, and other chemicals were reagent grade and used as received.

Film Preparation Methods

PU films were prepared by blending about 0.5 ± 0.3 g of *E. pellita* wood polyol with pMDI at different NCO/OH ratios through the casting method. Initially, the polyol was mixed with distilled acetone twice its weight before being agitated for 2 min. Following the procedure specified by Kurimoto et al., (2001), the predetermined weight of pMDI (equation 1) was added to the blend and continued stirring for 30 s. It was then cast in a petri dish (diameter = 9 cm) and dried overnight in a dark place. The films were peeled off before oven drying for another 5 h at 105 ± 3°C to rid of excess acetone and moisture. Prior to characterization, the films were conditioned at room temperature for 48 hours at 65% relative humidity. The ingredients of *E. pellita* PU films are listed in Table 1.

$$NCO{:}OH\ ratio = \frac{MpMDI \times WpMDI}{Mpolyol \times Wpolyol + Wwater \times 2 / 18 \times 1000} \quad (1)$$

where M_{pMDI} is the amount of isocyanate group in pMDI, W_{pMDI} is the weight (g) of pMDI, M_{polyol} is the value of hydroxyl group in polyol (227.67 mg KOH/g), W_{polyol} is the weight (g) of EP polyol, W_{water} is the weight (g) of water in EP polyol.

Table 1 *E. pellita* PU films ingredient at different NCO/OH ratio

Eucalyptus pellita PU Films	Weight (g)		NCO/OH ratio
	Eucalyptus pellita Wood Polyol	pMDI	
PU 1.8	0.5739	0.4261	1.8
PU 2.2	0.5243	0.4757	2.2
PU 2.6	0.4825	0.5175	2.6
PU 3.0	0.4469	0.5531	3.0

Indoor Soil Burial Degradation

The test was conducted in accordance with the technique reported by Deshmukh et al. (2021), with slight modifications. Films from each ratio were cut into small pieces with the dimension as follows: length of 10 ± 0.5 mm; width of 10 ± 0.5 mm and thickness of 0.25 ± 10 mm. The sample was oven dried for 24 h at 105 ± 3°C, prior to initial weighing. It was then buried in a perforated tube (50 mm) at a depth of 25 mm from the topsoil (Figure 1) before being watered (1 mL) every alternated day to preserve the moisture of the soil. The films were buried for 1 month. After the predetermined time, the samples were taken out and washed with distilled water then oven-dried for 24 h at 105 ± 3°C, before the final weight was taken. The rate of biodegradation was measured according to the weight loss after the soil burial, following equation 2.

$$Rate\ of\ biodegradation = \frac{Wi - Wf}{Wi} \times 100 \quad (2)$$

where *Wi* is the initial weight of films after oven drying and *Wf* is the final weight of films after 1 month of soil burial.

Figure 1 *E. pellita* PU films in perforated tubes for soil burial test

Water Solubility

Water absorption of the films was tested in accordance with Halimatul et al., (2019). Films were cut into 10 mm 10 mm dimensions and oven-dried at 105 ± 3°C for 24 h before the initial weight was taken. Each sample was immersed in 30 mL of distilled water for 24 h without any stirring. The samples were re-weighted after 24 h of oven drying, and the percentage of water solubility (WS) was calculated following equation 3.

$$Water\ solubility = \frac{(Mf - Mi)}{Mi} \times 100 \quad (3)$$

where *Mi* is the initial weight (g) of film samples and *Mf* is the final weight of film samples after immersion and oven drying.

Fourier Transform Infrared (FT-IR) Spectra

The prepared films were analyzed using Bruker Alpha II FTIR Spectrometer. About 24 scans were accumulated at 4 cm^{-1} within the frequency range of 4000 – 1000 cm^{-1}.

RESULTS AND DISCUSSION

The effect of the isocyanate index on the biodegradability rate of *E. pellita* PU films after 1 month of soil burial is shown in Figure 2. The biodegradability properties of PU films were decreased proportionally with increasing NCO/OH ratios, with the highest rate being 14.02%. The reason for this observation might be due to the chemical structure of the films. According to Lopes et al., (2022), as the ratio of NCO to OH increased, the polymer matrix tightened, resulting in the formation of crystalline regions, and eventually limiting the accessibility to microorganism attack. Furthermore, higher NCO/OH may increase intermolecular interactions between hard-to-hard segments, such as hydrogen bonds, resulting in PU films with high rigidity. As a result, PU films with high ratios (2.6 – 3.0) are more resistant to microorganism action and deterioration. Meanwhile, low-ratio PU films (1.8 – 2.2) contained more EP polyol (OH) than NCO. Because of the lower intermolecular reactions and high soft segments, this has resulted in the loosening of polymer arrangements and the formation of amorphous regions, resulting in soft and flexible films. Serrano et al., (2020) also stated that soft segments or woody components (EP polyol) contributed to the biodegradability of polyurethane films. Furthermore, the amorphous region was known to be a selective site for microbiological degradation, allowing enzymes easier access to the polymer chain (Rutkowska et al., 2002). PU films (NCO/OH 1.8) may have achieved the highest biodegradability rate due to their loose polymer arrangements, which are attributed to higher soft segments and lower intermolecular interactions. As a result, lower NCO/OH ratios are more vulnerable to microorganism attacks in the soil. In terms of physical state, *E. pellita* Biofilms with low NCO/OH proportion were found to be flexible and soft, whereas higher ratios were found to be brittle. After one month of soil burial testing, no visible cracks, pores, or holes were found in any of the film samples (Figure 3).

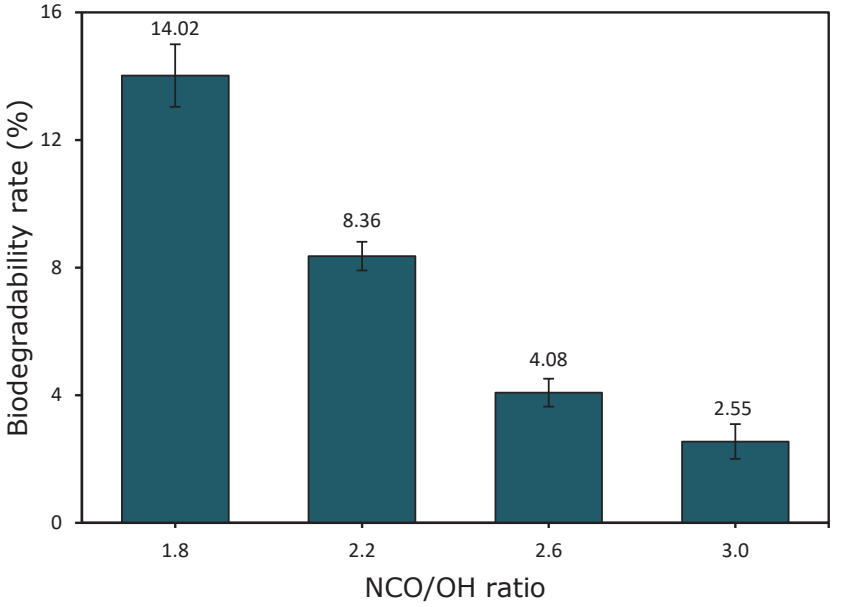

Figure 2 Biodegradability rate of E. pellita PU films at different NCO/OH ratios

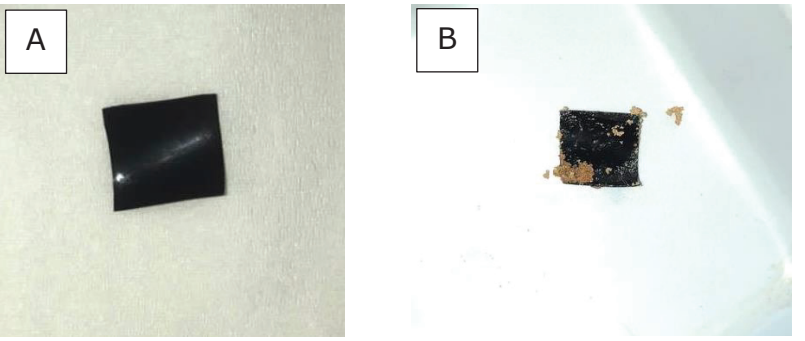

Figure 3 *E. pellita* PU film (A) before and (B) after soil burial test

Figure 4 depicts the rate of water solubility (WS) of *E. pellita* PU films at various NCO/OH ratios. The rate of WS decreases with increasing isocyanate index, with the highest obtained at 1.8 ratio. As stated by Velayutham et al., (2009) and Cakić et al., (2010), films with lower NCO/OH ratio contain higher soft segments due to lower hydrogen bonds and crystallinity that led to loose polymer packing, which made the water penetrations into the film easier (Elnaggar et al., 2019). The crystallinity of the films increased with increasing isocyanate index, as did the density of the matrix arrangement, making the films less soluble in water. This is demonstrated in this study, where *E. pellita* PU films with low NCO/OH ratios (1.8 to 2.2) exhibit higher water solubility while increasing the isocyanate index reduces weight loss. Palle et al., (2023) show that a high NCO/OH ratio had enhanced the interconnectivity among the hard segments and consequently reduced the solubility of film. The WS is known to

be related to the biodegradation characteristics (Serrano et al., 2020). Increasing the NCO/OH ratio reduced the WS and biodegradability rate of *E. pellita* PU films. After water immersion, films with a low NCO/OH ratio may have less crystallinity and loose polymer arrangements, resulting in lower resistance to microorganism attack.

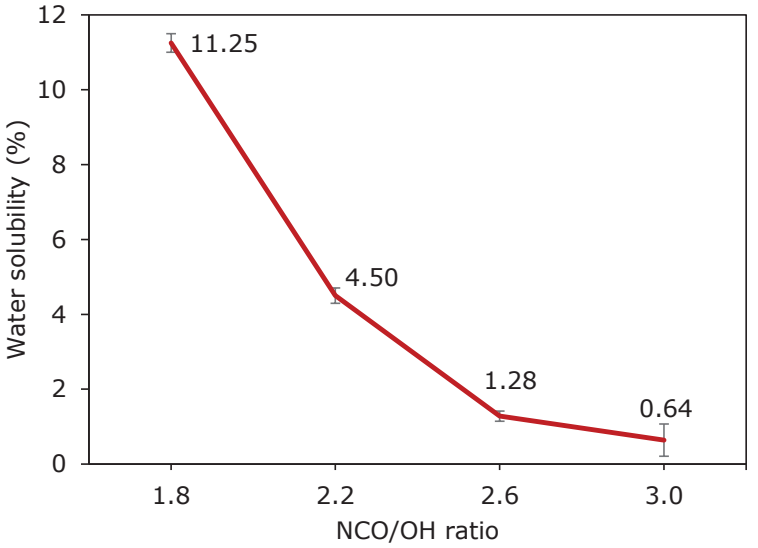

Figure 4 Water solubility rates of E. pellita PU films at different NCO/OH ratios

The chemical structure of the *E. pellita* PU films was analyzed via FT-IR spectra from 4,000 cm^{-1} to 1,000 cm^{-1} (Figure 5). A huge and shallow band from 3,480 cm^{-1} and 3,310 cm^{-1} is attributed to the -NH group of urethane linkages which is the main backbone of polyurethane polymer (Amran et al., 2021)chemical and compressive properties of the liquefied EFB-based PUF (EFBPUF. Approximately at 3,310 cm^{-1}, the band intensity seems to deepen as the NCO/OH ratio increases to 3.0, whilst vice versa for the band at approximately 3,470 cm^{-1}. This occurrence could be due to the increment of hydrogen bonding as more reactions between NCO and OH groups occurred (Dutta & Karak, 2006). There are two distinct peaks located at 2,900 cm^{-1} and 2,800 cm^{-1} that are asymmetrical and symmetrical, respectively. These peaks correspond to –CH groups that are connected to the EP wood characteristics (soft segment) (Palle et al., 2023). Commonly bands in the range of 1,700 cm^{-1} to the below are called fingerprint regions, which are usually intensified in polyurethane polymer due to huge formations of intermolecular reactions from the increased NCO/OH ratio (Dutta & Karak, 2006). In this region, bands and peaks that are commonly associated with urethane linkages are around 1,750 – 1,700 cm^{-1} (C=O), 1530 – 1,500 cm^{-1} (N-H bending vibrations), 1,220 cm^{-1} (C-N stretching vibration), and 1,065 cm^{-1} (C-O-C). Meanwhile, a shoulder peak observed at 1,665 cm^{-1} indicates the formation of urea groups in PU films and the intensity enhanced with increasing isocyanate index revealed that a high NCO/OH ratio induced the increment of hard segment formation (Garca-Pacios et al., 2010).

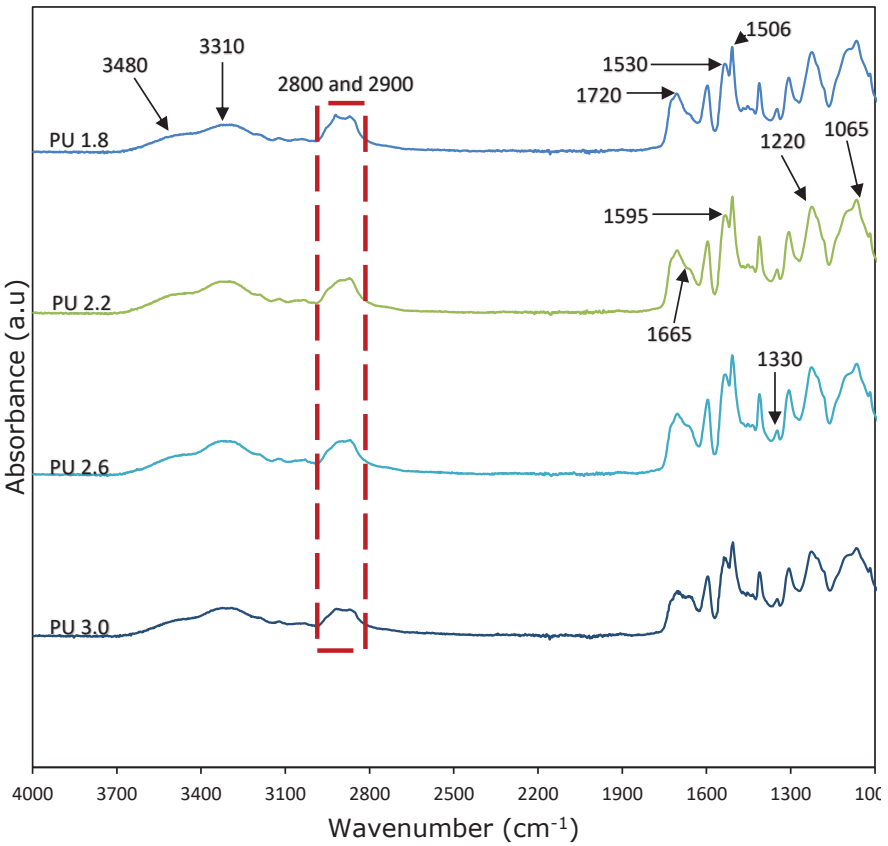

Figure 5 FT-IR spectra of E. pellita PU films at different NCO/OH ratios

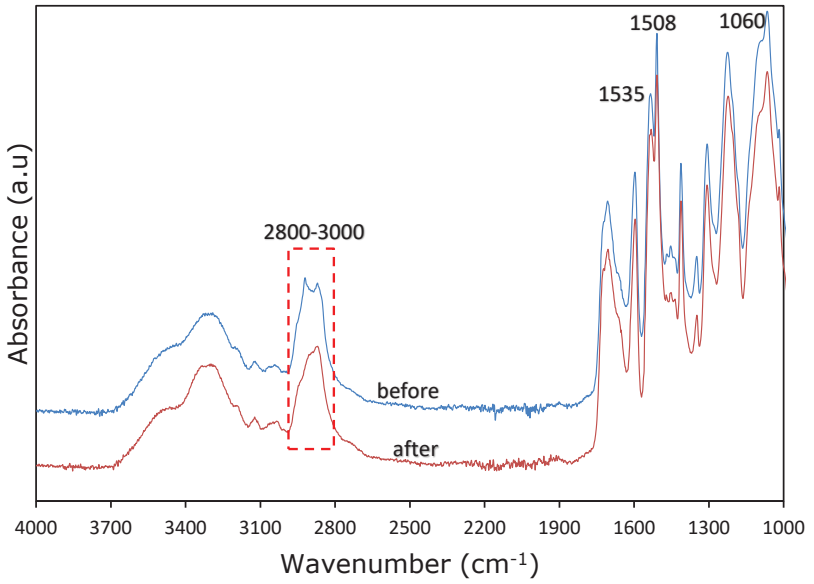

Figure 6 FT-IR spectra of E.pellita PU film at 1.8 NCO/OH ratio before and after 1-month soil burial test

The changes in the chemical structure of *E. pellita* PU film at 1.8 NCO/OH ratio, before and after 1 month of soil burial test could also be observed from the FT-IR spectra (Figure 6). The absorbance bands located at 2,900 to 2,800 cm^{-1} (CH, CH_2, and CH_3 groups from EP wood polyol) show a slight reduction in intensity. The same results obtained by Sahoo et al., (2018the modification of CO was confirmed using proton nuclear magnetic resonance (1HNMR) and Su et al., (2023), suggest that the occurrence might be due to the break in the carbon backbone. A few other important bands that show a minor decrement of intensity are 1,535 cm^{-1} and 1,510 cm^{-1} (-NH groups of urethane linkages) and 1,060 cm^{-1} (-C-C-O bonds of ester linkages) (Gómez et al., 2014; Panda et al., 2017; Yeoh et al., 2020). According to Saha et al., (2021), the ester bonds of *E. pellita* wood polyol appear to be the preferred attack site for microorganisms and fungi, as it is well known to have low resistance to microorganisms, making it easier to degrade in soil. Also, urethane, ester, and other hydrocarbon domains of polyurethane could be degraded in the soil by microbial enzymatic activity (Gómez et al., 2014; Sahoo et al., 2018).

CONCLUSION

From the findings of this study, it is concluded that PU film made from *E. pellita* polyol could be a biodegradable material for industrial applications such as packaging, medical purposes, plantation, coating, and engineered materials. It was discovered that the biodegradability rate of *E. pellita* PU film decreases from 14.02% to 2.55% as the ratio of NCO/OH increases concurrently with the rate of solubility. The absorbance band at 1,665 cm^{-1} attributed to urea is enhanced depending on the NCO/OH ratio implying that a high NCO/OH ratio significantly enhances the interconnectivity among the hard segment and consequently reduces the water solubility of the film. FT-IR analysis of the chemical structure revealed that the bands corresponding to wood characteristics and the hydrocarbon domain had slightly decreased in intensity after 1 month of soil burial, indicating a preferred site of attack for soil microorganisms. Therefore, using a lower NCO/OH ratio in PU film production from *E. pellita* polyol is the best recipe to obtain the highest biodegradability rate compared to that of a higher NCO/OH ratio.

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