Press-forming Molded Pulp from Repulped Liquid Packaging Board: Role of Heat Input, Pressing Force, and Defect Formation

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Laboratory sheets were prepared from repulped liquid packaging board (LPB) for press-forming experiments and tensile tests to study convertibility and mechanical properties of molded pulp material developed from the repulped LPB. Maximum forming depth was utilized as a convertibility indicator, and defect formation in press-formed molded pulp samples was observed with a visual analysis. Female mold temperature and pressing force were altered among press-forming parameters. The results showed that the laboratory sheets had a limited convertibility. The fragile structure of the laboratory sheets was connected to negative effects from the presence of plastic particles in the material. Increased heat input and decreased pressing force improved the convertibility, and the defect formation during the press-forming was aggravated by flattening of the material in flange and bottom regions of the samples. The use of repulped LPB as the raw material in the manufacturing of molded pulp by press-forming was found viable, and the presented solution offers an ecological alternative to conventional recycling of LPB.

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INTRODUCTION

Food and beverage packages require a combination of functions, which can be achieved with multilayer packaging materials. The multilayer structure of liquid packaging board (LPB) is made up of paperboard, plastic, and aluminum material layers to provide the combined functions. The paperboard layer offers stability, while the plastic and the aluminum layers provide barrier properties. Liquid repellence of the paperboard layer can be improved with additives to reinforce its barrier properties (Zhang *et al.* 2020).

The multilayer structure of LPB complicates its recycling process. LPB waste is conventionally repulped, after which the bulk of its fibrous fraction can be reused. Repulping reject consisting of plastics, aluminum, and some fibers is incinerated. Mechanical or chemical recycling of the repulping reject is an alternative to incineration, and the alternative treatment of LPB waste can be used to lower environmental impacts from handling of the reject (Khan *et al.* 2021). In addition to the recycling method, the environmental impact of recycling LPB is influenced by composition of the recycled LPB (Sun *et al.* 2021).

The presence of fibers in the repulping reject makes it difficult to reuse the plastic and the aluminum fractions of LPB as a fully recyclable secondary raw material, and fiber recovery from the repulped LPB is affected by separation of the material layers. Cleaner separation of material layers with polyethylene-coated paperboard can be achieved with a nanofibrillated cellulose (NFC) layer (Al-Gharrawi *et al.* 2021), yet a simple screening of repulp of coated paperboards is enough to recover most of its fibrous fraction (Sridach *et al.* 2006). Hwang *et al.* (2006) recycled LPB without the separation of the material layers; they found that the reuse of LPB in composite board applications was a viable alternative to its recycling *via* the conventional repulping route.

The reuse of unseparated material fractions of LPB provides an alternative upcycling route for its fibrous and plastic fractions. Molded pulp materials for electronic equipment packages were developed from a pulp-plastic composite (PPC) by Noguchi *et al.* (1998). The unseparated material fractions of LPB can be similarly considered as a suitable raw material source for molded pulp applications. Usability of molded pulp packages is affected by their static and dynamic strength. Rounded molded pulp shapes yield a larger compression strength than square or rectangular shapes (Hoffmann 2000), and the optimal shape design is also necessary to reduce material usage and maximum stresses (Bahlau and Lee 2022). The increased thickness in molded pulp products is connected to improved modulus under bending and tensile modulus (Curling *et al.* 2017). The thickness of the molded pulp products is important in manufacturing thin-walled items (Semple *et al.* 2022), and geometrical accuracy of thermoformed molded pulp products is influenced by their pulp type (Didone and Tosello 2018).

Research about the manufacturing of molded pulp is limited, and press-forming of molded pulp has not been investigated before. Studies about mechanical recycling of multilayer materials have focused on multilayer plastic films. In a previous work by Jönkkäri *et al.* (2020), the recycled multilayer plastic films yielded sufficient mechanical properties and processability in comparison to virgin low-density polyethylene (LDPE). Sufficient mechanical properties and processability of molded pulp developed from the unseparated material fractions of LPB are necessary for a viable upcycling and reusage of repulped LPB in molded pulp applications. Advancing the upcycling and reusage of repulped LPB in molded pulp applications is needed to enhance the recycling of multilayer packaging materials.

The aim of this work was to provide a proof of concept for use of repulped LPB as a raw material in manufacturing of molded pulp. Convertibility of laboratory sheets prepared from repulp of LPB was examined with a press-forming toolset, and the suitability of press-forming for manufacturing of molded pulp was evaluated. Pressing force and heat input were altered in the press-forming experiments to study their effects on the convertibility, and maximum forming depth was used as a convertibility indicator. Tensile tests were conducted for mechanical characterization of the laboratory sheets, and defect formation in press-formed molded pulp samples was observed.

EXPERIMENTAL

Materials

Separately collected commercial milk cartons were used as a raw material for repulping. The composition of the milk cartons was 75% fibers and 25% polyethylene (PE). The milk cartons were shredded with a 5-mm screen mesh using a Shini SG-1635V-CE

Granulator (New Taipei City, Taiwan). Repulping was done with an L & W Pulp Disintegrator (ABB, Stockholm, Sweden) by soaking 20 g of the shredded milk carton particles in 2 L of water before repulping for 1500 revolutions. Composition of the repulp remained 75% fibers and 25% PE as material losses during the repulping were minimal. Laboratory sheets were prepared from the repulp according to a modified ISO 5269-1 standard (2005). The modifications included a 275-g/m^2 target grammage, the use of an auxiliary 90 g/m^2 polyester satin wire on top of a sheet mold wire, wet pressing of the sheets for 5 min at 0.4 MPa, and drum drying of the wet pressed sheets for 2 h at 60 °C.

The prepared laboratory sheets were stored inside a humidity chamber at 80% relative humidity (RH). The moisture content of the stored laboratory sheets was measured with an Adams Equipment PMB 53 Moisture Analyzer (USA). The measured moisture content was 10.7%, and the moisture content value was deemed suitable for press-forming the laboratory sheets based on the recommended moisture content values, 8% to 11%, for the press-forming of fibrous materials (Tanninen *et al.* 2014). Tensile tests of the laboratory sheets were conducted with an L & W Tensile Tester (ABB, Stockholm, Sweden) according to the ISO 1294-3 (2005).

Methods

Convertibility of the laboratory sheets was tested with a MiniMould press-forming tool set of LUT Packaging Line (Lappeenranta, Finland). A detailed description of the MiniMould tool and the press-forming process were presented in a previous work by Tanninen *et al.* (2017) in which the MiniMould (Fig. 1) was validated as a suitable tool for experimenting the convertibility of fibrous materials by using maximum forming depth as a convertibility indicator.

Fig. 1. Female mold (1), blank holder (2), and male mold (3) of MiniMold press-forming tool set

Pressing force and female mold temperature were altered in the press-forming experiments. Blank holding force, pressing speed, and dwell time were kept constant (Table 1). The maximum female mold temperature was kept at 100 °C to avoid melting of the PE from the material-tool contact. The maximum forming depth was verified with an accuracy of 0.5 mm by press-forming five defect-free molded pulp samples. The obtained forming depth values are averages of the five samples, which were press-formed with different pressing force and female mold temperature. Defect formation in the samples was observed with a visual analysis which was carried out by taking images above the samples.

Table 1. Variables and Constants in Press-forming Experiments

Pressing Force	7.5 kN	15 kN	30 kN
Female Mold Temperature	23 °C	100 °C	
Blank Holding Force	0.72 kN		
Pressing Speed	40 mm/s		
Dwell Time	1 s		

RESULTS AND DISCUSSION

Defect-free molded pulp samples (Fig. 2) were successfully press-formed from the mixed pulp-plastic material of the laboratory sheets. The press-forming tool set had a limited dewatering capability, and consequently the dewatering of the laboratory sheets by wet pressing and drum drying were found necessary to enable a pulp molding process with the used press-forming tool set.

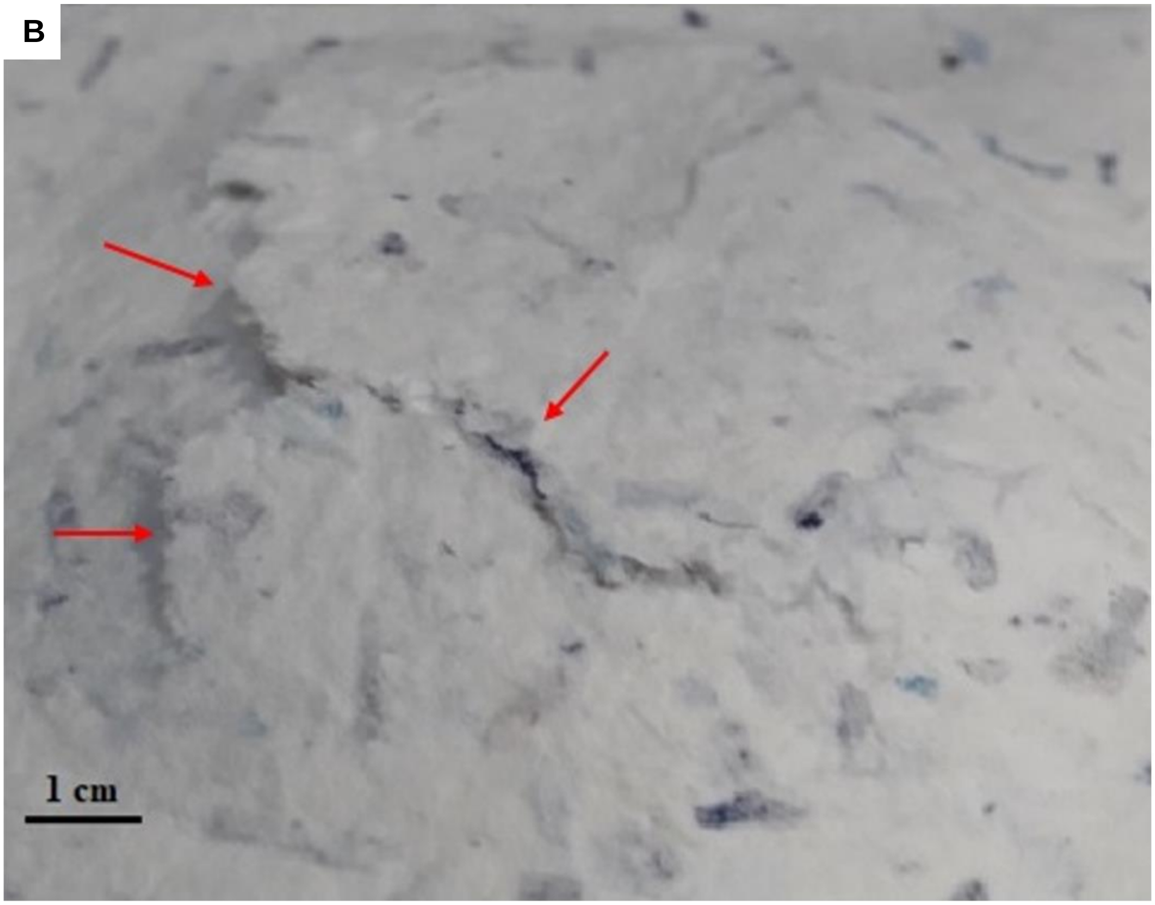

Fig. 2. A defect-free (A) and a damaged (B) molded pulp sample, defects in the damaged sample are highlighted with arrows

The obtained tensile strength of the laboratory sheets was 2.43 kN/m, and the measured strain at break was 1.19%. The convertibility evaluation results connected increased heat input and decreased pressing force with increased forming depth (Fig. 3). The molded pulp material developed from the repulped LPB displayed limited mechanical properties and convertibility. By comparison, previous testing of a fibrous material with the Minimould tool yielded a maximum strain at break of 4.5% and a maximum forming depth of 35 mm (Tanninen *et al.* 2017).

The finding about positive effect of heat input on the convertibility was consistent with previous studies by Vishtal *et al.* (2012, 2013). The increased heat input promoted a softening effect on fibers, which enhanced elongation of the material during the press-forming. The decreased pressing force facilitated sliding of the laboratory sheets during the press-forming. Due to the sliding of the laboratory sheets, the press-forming occurred by folding of the material in addition to its stretching. The simultaneous folding and stretching of the material enable the press-forming of deeper shapes (Tanninen *et al.* 2017). The enhanced sliding of the material from the use of low pressing force thereby positively influenced the maximum forming depth of the material.

Presence of heterogenous particles and incompatibility of material fractions can degrade mechanical properties of recycled plastic fractions (Dintcheva *et al.* 2001). The fragile structure of the molded pulp material was associated with the presence of plastic particles. Bonding of fibers greatly affects strength properties of paper (Przybysz *et al.*

2016), and bonding of fibers has additionally been linked to improved convertibility of paperboard (Hauptmann *et al.* 2015). Plastic particles disrupted the structure of the fiber network and the bonding of the fibers in the molded pulp material, and the disruptive effect degraded the convertibility and the mechanical properties of the laboratory sheets.

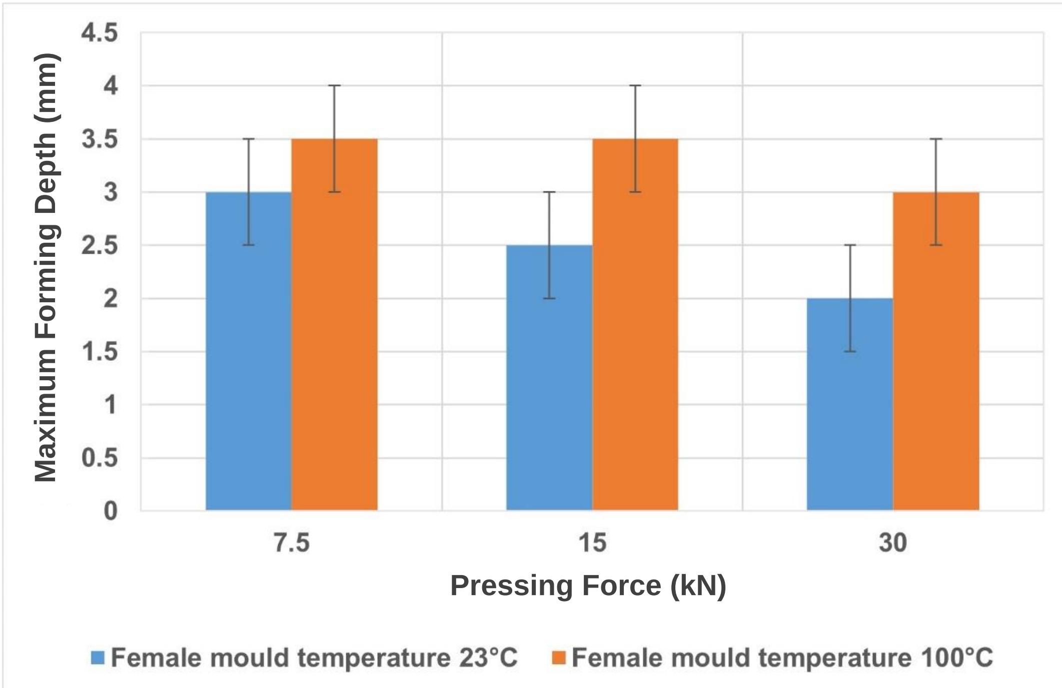

Fig. 3. Effect of female mold temperature and pressing force on maximum forming depth

Mechanical properties of composite materials with fibrous and plastic fractions can be enhanced with use of fillers and compatibilizers (Hosseini *et al.* 2016; Effah *et al.* 2018). The use of fillers and compatibilizers in the molded pulp material developed from the repulped LPB thereby provides an interesting reference for future research. Increasing the fibrous content of the laboratory sheets by mixing the repulp with virgin pulp and the press-forming of moister laboratory sheets are in addition possible research interests. Uneven visual appearance and color defects of the molded pulp samples were sourced to the plastic particles. Visual quality of fiber surfaces and cleanliness of pulp can be improved by deinking (Viesturs *et al.* 1999). Deinking consequently offers an important approach for obtaining a more even visual quality with mixed pulp-plastic materials.

Defects were observed in bottom (Fig. 4) and flange (Fig. 5) regions of the molded pulp samples.

Defect formation during the press-forming of the molded pulp material was ascribed to a breakage of the fibers and the fiber bonds from stress and strain localization in the fiber network. Defects in tensile deformation of paperboard can form in weak regions of the material, and the weak regions are associated with thinner or less dense locations in the deformed samples (Johansson *et al.* 2021). The bottom and the flange regions of the molded pulp samples appeared weaker due to the location of the defects, and the defect formation was connected to the stress and strain localization in the weaker regions. Thickness of the bottom and the flange regions of the molded pulp samples was influenced by material-tool contact during the press-forming.

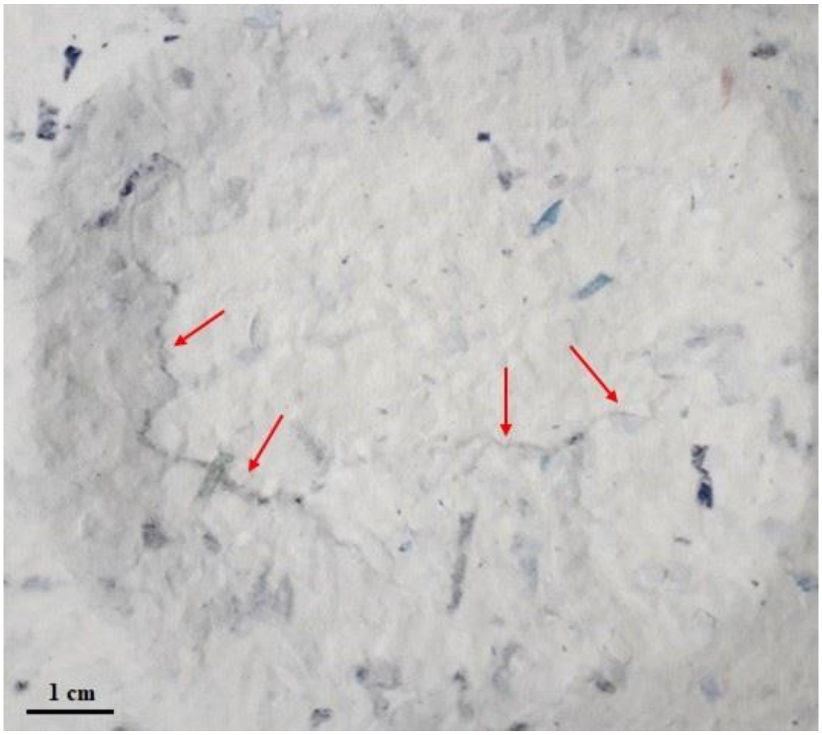

Fig. 4. Defects in bottom region of a molded pulp sample, the defects are highlighted with arrows

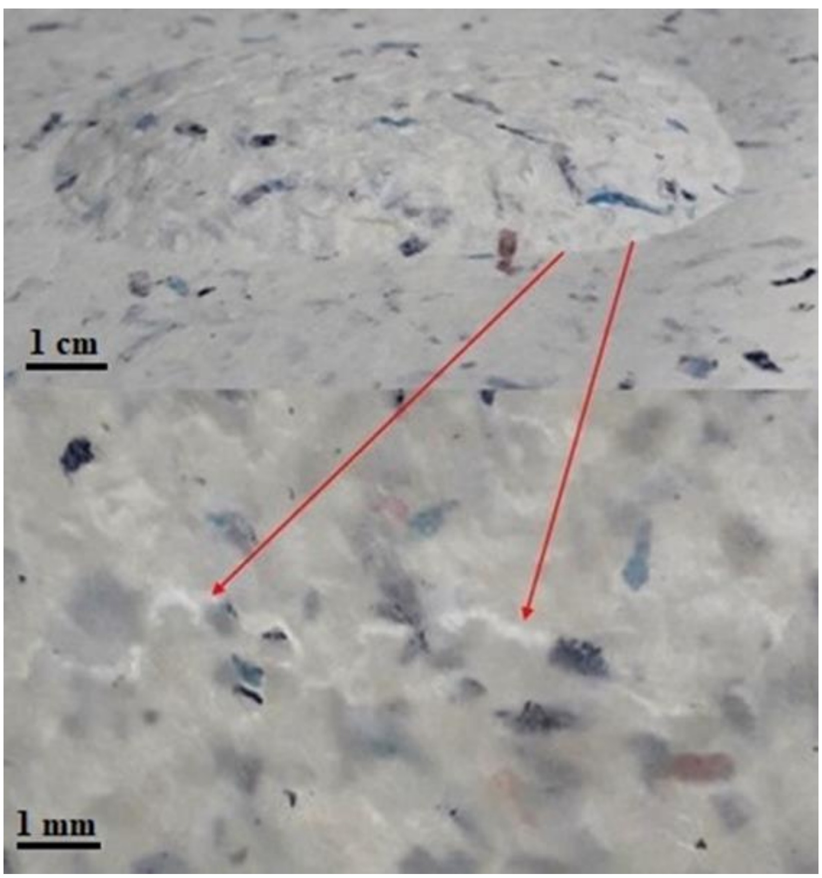

Fig. 5. Defects in flange region of a molded pulp sample (top), the defects are highlighted with arrows in an enlarged section (bottom) of the same sample

The flange region of the samples was flattened between the female mold and the blank holder, whereas the bottom region of the samples was flattened between the female and the male mold. The stress and strain localization in the bottom and the flange regions was connected to the flattening of the material. The flattening of the material from the material-tool contact induces thickness reduction in the bottom and the flange regions of the molded pulp samples. The thickness reduction in the bottom and the flange regions aggravated the defect formation in the samples. The flattening of the material during the press-forming could be mitigated with the use of a lower blank holding force. However, decreasing the blank holding force increases the folding and decreases the stretching of the material. The utilized blank holding force should thus be adjusted with consideration for desired material elongation in the press-forming of molded pulp products.

The press-forming experiments validated the viability of using repulped LPB as the raw material in the manufacturing of molded pulp. The convertibility and the mechanical properties of the laboratory sheets prepared from the repulped LPB were limited by the discussed disruptive effect from the plastic particles of the material. The optimal forming parameters improved the material performance with a minor effect. The use of fillers and compatibilizers in the molded pulp material was recommended to advance the alternative upcycling and reusage of repulped LPB in molded pulp products. As the mechanical recycling of LPB provides a lower environmental impact in comparison to the incineration of its plastic fraction (Khan *et al.* 2021), the manufacturing of molded pulp products from the repulped LPB can be exploited to reduce the environmental impact by reusing the plastic fraction of LPB instead of incinerating it.

CONCLUSIONS

1. Press-forming was deemed suitable for the manufacturing of molded pulp on the condition that the dewatering of the material during the pulp molding process is considered. In the case of laboratory sheets, the dewatering could be done by wet pressing and drum drying.
2. The presence of plastic particles in the molded pulp material degraded its convertibility and mechanical properties. The limited mechanical properties and convertibility were also attributed to a weak structure of fiber network and poor bonding of fibers in the material.
3. Defects were observed in the flange and the bottom regions of the damaged molded pulp samples. The defect formation during the press-forming was associated with reduced thickness in the flange and the bottom regions of the molded pulp samples.
4. A proof of concept was outlined for manufacturing molded pulp by using repulped LPB as the raw material. The outlined concept can be utilized to upcycle repulp of LPB in molded pulp applications. Reusing the plastic fraction of LPB in molded pulp applications gives an alternative to its incineration, and the alternative recycling route can be considered for industrial or consumer waste streams of LPB.

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