

# Melamine Acetate Preparation as a Urea-Formaldehyde Resin Additive for Particleboard Production

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**Abstract.** Urea-formaldehyde (UF) resin binders for wood-based panel production often use melamine as an additive to improve both mechanical properties and environmental compliance. Direct fortification for efficient use of melamine remains a problem due to low solubility. Pizzi & coworkers suggested the use of more soluble melamine salts and demonstrated success in terms of product performance. However, their preparation method showed low productivity and inefficient use of material and energy. In their scheme, a batch reactor fed with 1 kg of water and 75 g of a stoichiometric amount of melamine and acetic acid produced only around 25 g of solid melamine acetate crystal. In contrast, the remaining 50 g remained dissolved in 991 g of water, which requires high of energy to evaporate. This paper reports an attempt to increase batch productivity and work towards the more efficient use of material and energy. The experiment showed that the successive addition of melamine and acetic acid to the batch up to the solubility limit of melamine at the same reaction condition increased melamine and acetic acid fed from 75 g to 165 g. This was followed by a significant increase in dry crystal yield from 25 g to 117 - 132 g. Feeding the mother liquor to the next batch decreased the water use to only 8% of the original requirement. This resulted in a highly efficient process, eliminating the need for energy-intensive melamine acetate recovery from the mother liquor. The addition of 2% - 4% wt. of the product to UF resin resulted in particleboard with significantly lower thickness swelling, an increase in MOR & IB strength, and lower formaldehyde emission.

**Keywords:** Melamine Acetate, Urea-Formaldehyde Resin, Particleboard

## INTRODUCTION

Urea-formaldehyde (UF) resin is the most important binder used to produce wood-based panels because of its reactivity, fast curing, water solubility, and low cost. UF resin can be tailored to produce high bonding strength panels or low formaldehyde emission panels by adjusting its urea-to-

formaldehyde molar ratio, leading to different morphology and crystallinity (Li & Zhang, 2021; Lubis & Park, 2021). The only undesired property of panels bonded with UF resin is their low hydrolytic stability, causing them to deteriorate under wet or moisture conditions easily; thus, their use is limited to interior applications (Mouratidis et al., 2000).

Melamine is the most used additive to

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improve water-resistant properties, thus extending the use of UF resin to semi-exterior or even exterior applications. Due to its higher functionality, melamine improves bonding strength and offers higher capacity as a formaldehyde scavenger (No & Kim, 2004). Low-viscosity melamine urea formaldehyde (MUF) resin is used for the bulking treatment of wood (Ali et al., 2020). MUF resin adhesion performance depends on the synthesis method and melamine content (Jeong et al., 2019). Hence, they can be tailored to possess high boiling water resistance and exhibit low formaldehyde emission (Wang et al., 2019). The problem of formaldehyde emission was first recognized in 1980, and more stringent standards and regulations have been applied ever since (Popovic et al., 2020). Formaldehyde emission is also affected by hardener formulation (Kantieva et al., 2020; Liu et al., 2018)

The addition of melamine to UF resin presents several problems in resin synthesis. Compared to urea, melamine solubility is exceptionally low. Even for the preparation of low melamine content resin, an additional additive, usually dimethylformamide (DMF), is often used to increase solubility, thus enabling melamine to react in the resin solution. The competitive polycondensation mechanism of MUF resin synthesis under alkaline conditions was analyzed (Liang et al., 2020) Usually, a substantial amount of unreacted melamine is present in the final resin product, causing melamine-containing resins to exhibit lower storage stability, i.e., their viscosity rapidly increases during storage at room temperature. To overcome this problem, Mao et al. (2013) suggested feeding melamine early at the beginning of the resin preparation stage. However, the problem can only be solved partially. The storage stability of MUF resin can be

improved by increasing temperature (Lee et al., 2012) but storing resin at higher than ambient temperature is considered impractical.

In a reaction mixture of urea, melamine, and formaldehyde, initially, urea and melamine react with formaldehyde resulting in hydroxymethylureas and hydroxymethylmelamines, respectively. While polymerization of hydroxymethylureas is exclusively acid-catalyzed, hydroxymethylmelamines polymerization can occur in acid and base conditions. Hydroxymethylureas tend to react further with other hydroxymethylureas than to react with hydroxymethylmelamines, and vice versa. Thus, it is a block copolymer, rather than the desired random copolymer (which is required for effective property improvement) dominates in melamine-modified UF resin (Zanetti & Pizzi, 2003).

In addition to its higher price, melamine addition to UF resin should also be limited because melamine promotes unwanted properties concerning resin handling and processability. The buffering capacity of melamine slows down the pH decrease required for resin curing, thus increasing the hot-pressing time required in panel production, causing higher energy consumption, and decreasing panel production capacity, thus, contributing to higher production cost. The gel time of MUF resins is longer than that of UF resins because of the combined effect of melamine moiety's acidic buffering and MUF condensation reactivity (Ding et al., 2021).

Pizzi and his coworkers proposed a different approach to increase the effective use of melamine. Rather than reacting formaldehyde with urea and melamine at once, as in the conventionally prepared UMF resin, they first convert melamine to a more

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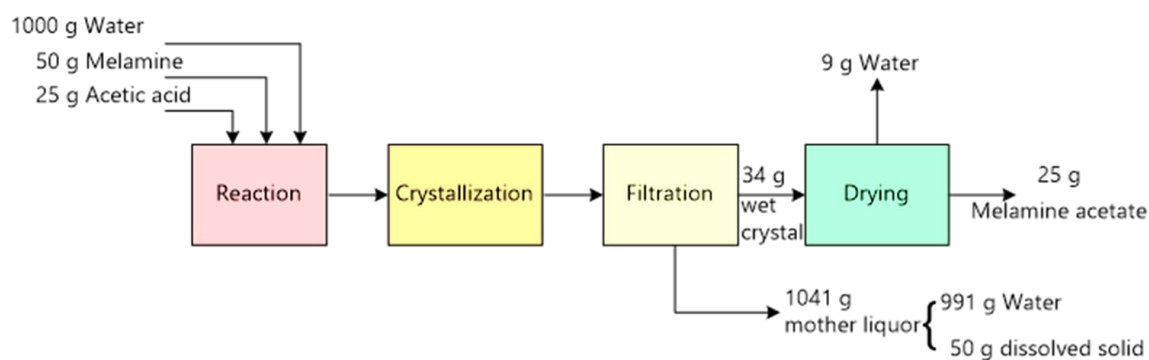
soluble melamine salt and use the resulting salt as an additive for the UF resin. This strategy offers at least three advantages. First, technical difficulties caused by reactivity differences between melamine and urea in UMF resin synthesis can be avoided using the existing UF resin and an appropriate amount of melamine salt, which can be prepared separately. Second, the problem of the low shelf life of UMF resin is no longer the issue. The last and most important advantage is the increase in melamine effectiveness, which allows the use of lower melamine content to improve the same improvement.

Inorganic melamine salts such as melamine phosphate, melamine sulfate, and melamine nitrate do not exhibit higher solubility than melamine (Crews et al., 2002). Therefore, only salts derived from organic acids have the potential to be developed as additives. After producing plywood with UF resin supplemented with additives such as melamine formate, melamine acetate, melamine oxalate, melamine citrate, and melamine adipate. Festifilippo et al. (1996) demonstrated that only melamine format and melamine acetate consistently provided improvements in tensile strength due to their higher solubility compared to other melamine salts. Melamine is a weak base with a  $pK_{a1}$  of 5.10. Organic acids with  $pK_a$  values lower than this are required to produce sufficiently stable salts. However, organic acids with excessively low  $pK_a$  values will form salts that are too stable and difficult to decompose. Decomposition at hot press temperatures is desired to generate organic acids that act as hardeners (Weinstabl et al., 2001). Based on this reasoning, acetic acid ( $pK_a = 4.76$ ) will provide a relatively stable salt but is more easily decomposed compared to formic acid ( $pK_a = 3.77$ ). Therefore, further research focuses only on melamine acetate.

In a two-year weathering test, Cremonini & Pizzi (1999) demonstrated that plywood prepared from UF resin of formaldehyde to urea molar ratio of 1.5 fortified with melamine acetate at the glue mix stage showed better weather resistance than plywood prepared using traditional phenol-melamine-urea-formaldehyde resin containing higher melamine content after 2 years of aging, although initially showed lower strength. Zanetti & Pizzi (2003) demonstrated that particleboard made with resin supplemented with small amounts of melamine acetate exhibited better water resistance than with conventional MUF resin.

Despite the abovementioned success in melamine acetate application as a resin additive for wood-based products, particularly plywood production, these publications pay less attention to the effectiveness of the preparation process of the melamine acetate itself. We have replicated the preparation process in accordance with the procedure of Zanetti & Pizzi (2003) and performed material balance calculations to gain insight into the process performance, which was not described in their papers. The results are summarized in Fig. 1. As depicted in Fig. 1, a 1:1 mol ratio of acetic acid was used for every 50 g of melamine feed, and 1000 g of water was added as a solvent. The quantity of water used was exceedingly high, corresponding to melamine solubility in boiling water of 50 g/L. The maximum amount of melamine acetate produced was only 75 g. Crystallization was performed by cooling the reaction mixture overnight at room temperature. After filtration and subsequent drying, only one-third (25 g) of the melamine acetate crystal was obtained because the remaining 50 g remained dissolved in the mother liquor.

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**Fig.1:** The original mass balance for melamine acetate preparation

We defined batch productivity as the ratio of dried melamine acetate mass obtained to the total mass of batch feed. Since 25 g of dried product were obtained in this scheme, and the total mass of reactor feed was 1075 g, the batch productivity of this scheme was only  $25\text{g}/1075\text{g} = 0.023 \text{ g/g}$  or 2.3%. Thus, each batch produces only 2.3% of melamine acetate crystal out of the total mass fed, which is extremely low. This study aimed at performing process modification: (a) to increase batch productivity of this melamine acetate preparation toward the more efficient use of material and energy by applying simple principles of chemical engineering process design, and (b) to assess the efficacy of melamine acetate as an additive for UF resin in the production of particleboard.

## MATERIALS AND METHODS

### Materials and Equipment

Melamine was obtained from Nissan Chemical (Japan), while glacial acetic acid was obtained from Indo Acidatama (Indonesia). Both were used directly without any treatment.

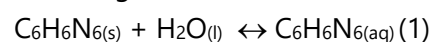
The reactor was a 2 L Erlenmeyer flask with a thermometer and a glass condenser. A heater-magnetic stirrer maintained a

moderate stirring rate of 300 rpm.

Crystallization was carried out by cooling the reaction mixture overnight at room temperature. Laboratory vacuum filtration with a Büchner funnel apparatus separated melamine acetate crystals from the mother liquor. Subsequently, sun-drying was performed to remove water from the moist crystals through evaporation.

### Increasing Melamine & Acetic Acid Fed

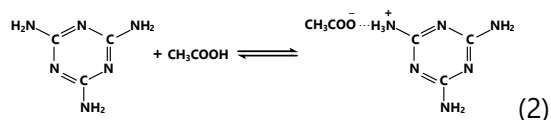
The quantity of water used in the original recipe was 1 kg for each 50 g of melamine, corresponding to melamine solubility in boiling water of 50 g/L:



A large quantity of water was used to ensure the complete dissolution of all the melamine. However, the role of water in this process was merely a solvent that need not be recovered in the product. Thus, the higher the quantity of water, the lower the batch productivity. In other words, water quantity should be decreased to increase batch productivity. However, melamine should stay dissolved completely.

An equivalent statement applied in the context of this work was: at the same amount of water, one needs to increase melamine and acetic acid fed, but the solution should stay clear, indicating no existence of undissolved

melamine. This idea should be workable because as soon as dissolved in water containing enough acetic acid, the melamine would immediately be converted to melamine salt according to the equilibrium aqueous phase reaction (Suzuki et al., 1997):



Because this reaction consumed melamine, the solution was no longer saturated with melamine. Thus, the solution recovers its ability to dissolve more melamine. Additional acetic acid is then fed to the reactor and readily consumes the melamine, and again, the solution gains its ability to dissolve extra melamine. This process of providing extra melamine and acetic acid can be repeated until the solution was saturated so it cannot dissolve more melamine, indicated by solution turbidity, which was easily monitored visually (Fig. 2).

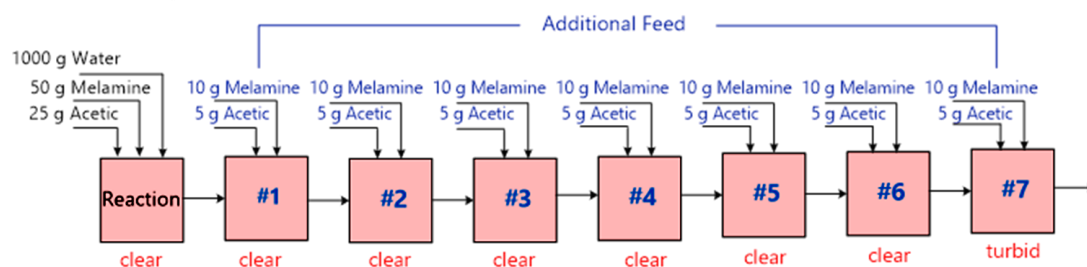
### Reuse of Mother Liquor

Increasing melamine and acetic acid fed as described above would increase solid crystal quantity after cooling crystallization. Still, the remaining melamine acetate concentration in the mother liquor was expected to be constant because solubility

remains constant at a constant temperature. Referring to Fig. 1, if the crystallization were carried out down to 5°C in a chiller, our study showed that the obtained crystal from the original recipe could be increased by 15 g due to lower solubility of melamine acetate at a lower temperature so that the resulting dry product increase to 40 g (corresponding to batch productivity of only 3.7%). Thus, cooling improves batch productivity. However, this cannot be considered effective.

Recovery of the rest of the melamine acetate from the mother liquor needs extensive water evaporation. This approach is incorporated with the expense of additional costly equipment and extensive energy use for water evaporation. Even if all the dissolved solids were recovered by evaporating all water in the mother liquor using appropriate equipment, such as a vacuum evaporator or spray dryer, the batch productivity can only reach the limit value of 75 g/1075 g, i.e., less than 7%.

A simple yet effective method of solid recovery from the mother liquor is to recycle it by using the mother liquor as additional feed for the next batch. This way, using expensive equipment for energy-extensive water evaporation can be avoided. Not only melamine acetate would be recovered, but the solvent as well. Thus, the need for freshwater as a solvent would decrease



**Fig. 2:** Successive feeding of additional melamine & acetic acid at the reaction stage

drastically. The concept of reusing mother liquor in this research aligns with reducing water consumption and wastewater generation by maximizing the utilization of the process water, as described by Tan & Cruz (2005).

### Particleboard Production

To demonstrate the efficacy of melamine acetate as a urea-formaldehyde resin additive, three samples of 350 mm x 350 mm x 100 mm particleboards with a target density of 0.700 g/cm<sup>3</sup> were made using (a) only UF resin, (b) UF resin fortified with 2% wt., and (c) UF resin fortified with 4% wt. of melamine acetate.

The particleboards were three-layered, i.e., surface-core-surface, with a weight proportion of 25%, 50%, and 25%, respectively.

## RESULTS AND DISCUSSION

### Maximum Feed Quantity

As in the original recipe, 1000 g of water, 50 g of melamine, and 25 g of acetic acid were initially charged to a stirred glass reactor. The reaction occurs at the boiling point of the mixture, which is 100°C, under atmospheric pressure. Heating to boiling point changes the appearance of the reaction mixture from a white suspension to a clear and transparent solution, indicating that all melamine dissolved and converted to melamine acetate. In the original recipe, this mixture was then cooled to room temperature for cooling crystallization (Fig.1). In this experiment, additional feed #1 containing 10 g of melamine and 5 g of acetic acid, was added to the boiling mixture. Soon after the addition, the mixture turns clear and transparent. This phenomenon demonstrated that, as expected, the mixture gained its

ability to dissolve more melamine which was then immediately converted to melamine acetate.

The process of feeding additional melamine and acetic acid was repeated several times until additional feed #7 (Fig. 2), which showed that the mixture remained turbid even after 15 minutes, indicating that the solution had been saturated with dissolved melamine. Thus, it was concluded that the maximum quantity of melamine and acetic acid has been reached at additional feed #6, corresponding to a total charge of 165 g. This result was equivalent to 220% of the original recipe.

### The Expected Mass Balance

Fig. 3 depicts the expected material balance if the experiment was conducted using the same equipment and at the same conditions as those of Fig. 1, except that the quantity of melamine and acetic acid was increased according to the findings. For simplicity, in Fig. 3, the stagewise feeding of melamine and acetic acid is not depicted as those of Fig. 2. Compared to Fig. 1, the total reactor feed was expected to increase only by 8.4% from 1075 g to 1165 g, while the expected melamine acetate crystal produced increased by almost five folds from 25 g to 117 g. This corresponds to a significant batch productivity increase from 2.3% to 117g/1165g = 10%. This value is even higher than the original recipe's maximum limit of 7%.

Assuming the same moisture content of the wet crystal of 26.5% and the same solid content in the mother liquor of 4.8% as those of Fig. 1, the quantity of mother liquor in Fig. 3 was expected to decrease slightly from 1041 g to 1006 g. It should be noted that the value of 1041 g in Fig. 1 was not measured. Rather, it was deduced from the difference

between the total amount of feed and the obtained wet crystal, assuming no water loss during the process. Consequently, the expected value of 1006 g in Fig. 3 was an overestimate, since water loss due to evaporation cannot be avoided in the actual process because the reactor would be opened frequently for feeding additional melamine and acetic acid.

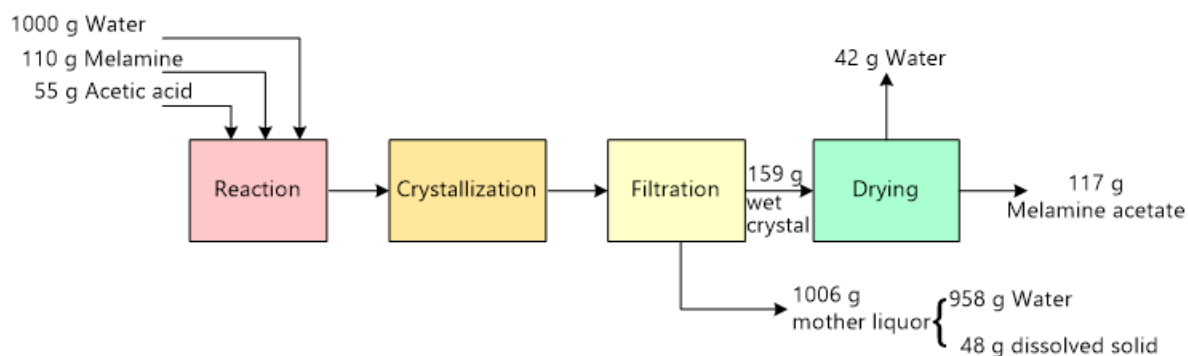
### The Experimental Mass Balance

The results of the experimental reaction using feed composition, as shown in Fig. 3, were allowed to cool overnight until reaching room temperature. The reduction in melamine acetate solubility caused by the temperature decrease led to solid melamine acetate crystals forming. Subsequently, laboratory vacuum filtration separated the generated crystals from the solution at room temperature.

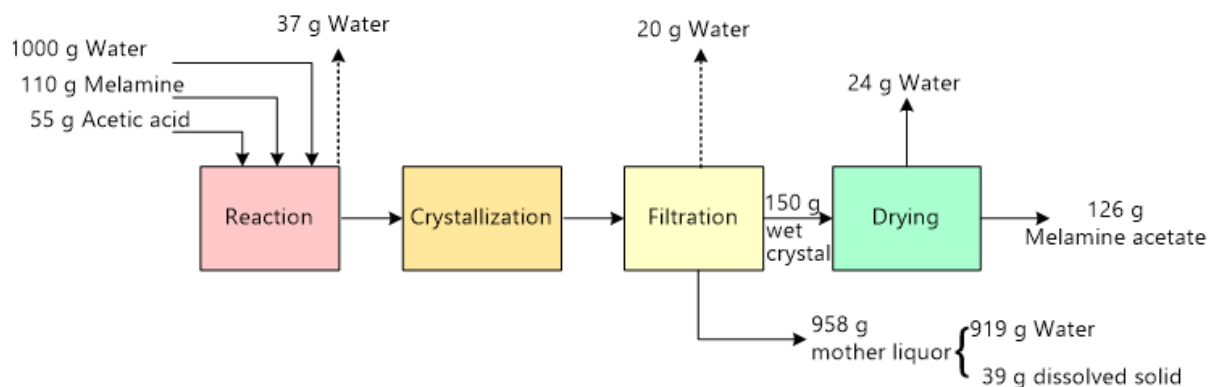
Fig. 4 shows the actual experimental data, which indicated a similar result as the expectation, except for the occurrence of water loss in the reaction and filtration step. In the experiment, 37 g of water escaped to the atmosphere during reactor openings for additional melamine and acetic acid feeding at the reaction step. On the one hand, water loss from the reactor is unwanted because

decreased water in the reactor means the decreased ability to dissolve melamine. Thus, if there were no water loss, more melamine and acetic acid could be fed. Thus increased batch productivity would have been expected. On the other hand, water evaporation decreases the amount of available solvent, thus increasing the amount of crystal formed. Therefore, the experiment obtained 126 g of melamine acetate, higher than the expected value of 117 g as depicted in Fig. 3.

The experiment used the same laboratory vacuum filtration equipment as the original process, where the mixture was poured into the filter funnel. The accumulation of moist melamine acetate in the filtration funnel slowed the filtration process by impeding filtrate flow from the solution. The amount of wet crystal formed in this experiment was 150 g (Fig. 4), much higher than that of the original process (34 g in Fig. 1). Consequently, the filtration process took longer to complete. It took more than 1 hour to filter one batch of melamine acetate solution in this experiment, compared to less than 10 minutes in the original process. During this filtration period, the moist melamine acetate was exposed to open air, leading to evaporation of water content.



**Fig. 3:** The expected (calculated) first batch mass balance after modification



**Fig. 4:** Experimental batch #1 mass balance after modification

In this experiment, 20 g of water evaporated from the wet crystal before it was weighed. While cooling the feed before filtration can minimize water evaporation, it was not performed in this experiment due to its benefits in reducing energy consumption during the subsequent drying process.

Due to water evaporation during the filtration process, the calculated moisture content of the wet crystal in Fig. 4 was only 16.0%. If the 20 g of water loss in the filtration step were considered as the additional water content in the wet crystal, the weight of the wet crystal would be 170 g, and the moisture content of the obtained wet crystal would be 25.9%, which is close to the value of 26.5% in Fig. 3. The actual mother liquor obtained was 958 g, almost 5% lower than the expected value of 1006 g due to water loss by evaporation.

The solid content in the mother liquor was found to be 4.1%, which is slightly lower than the expected value of 4.8%. The experimental value was lower, possibly because the room temperature at which this experiment was conducted was slightly lower than that of the original process from which the expected value in Fig. 3 was obtained. Lower temperature decreases the solubility of melamine acetate. This leads to a greater amount of solid melamine acetate being

obtained during filtration, ultimately resulting in a higher yield (Dermawan, 2018).

### **Drying and Thermogravimetric Analysis of Melamine Acetate**

The original recipe condition for oven drying is at 75°C for 72 h, but our experiments showed that these recommended conditions are causing melamine acetate decomposition. The decomposition was evident from a) a strong acetic acid odor emanating from the oven b) visual observation of the physical appearance: part of the solid, which was originally a hard salt-like crystal, turned into powder-like melamine, and c) compared to the hard crystal, the powder part was hardly dissolved in water. Thus, in this study, drying was conducted by sun-drying until no noticeable weight loss. Fig. 5 showed that this can be achieved within 4-5 days of sun drying under the ambient temperature of 31 - 34°C.

A thermogravimetric analysis was conducted to demonstrate the efficacy of the drying process in this analysis, 10.82 mg of the sample was initially kept at 30°C for 1 minute as atmospheric air was passed over it, then heated at 10°C/min. Initially, heating will evaporate volatile matter such as moisture and free acetic acid in the sample. Fig. 6 showed that the first onset temperature at



which the weight loss started to accelerate began at 70.43°C when the remaining sample weight was 10.78 mg, corresponding to the initial decomposition of melamine acetate, in agreement with the inference that 0.37% of the melamine acetates decomposed when oven-dried at 75°C. If the weight loss until this first onset temperature was due to water evaporation, it can be inferred that the sample's water content was 0.35%. This finding justifies the assumption taken in the mass balance calculation performed previously that within the experimental accuracy, the product's water content was low enough to be ignored. Further heating resulted in melamine stagewise decomposition with the release of ammonia, as described by Liu et al. (2016).

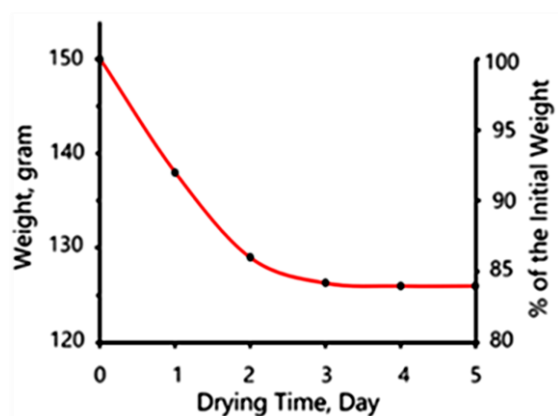


Fig. 5: Weight of product during sun-drying

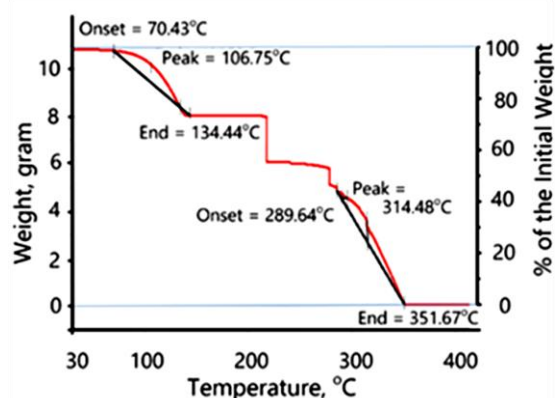


Fig. 6: TGA of dried melamine acetate

### The Reuse of Mother Liquor

As depicted in Fig. 4, the mother liquor contained 39 g of valuable dissolved melamine acetate and 919 g of water which can be reused as a solvent for the next batch. The reuse of this mother liquor, as depicted in Fig. 7, would very significantly reduce the quantity of fresh water required for the next batch from 1000 g to only 81 g, which equals the total quantity of water evaporation loss and water evaporated during the drying process. The mother liquor's 39 g of melamine acetate originated from 26 g of melamine and 13 g of acetic acid. Thus, this quantity subtracts the quantity of melamine and acetic acid initially fed from 50 g and 25 g to 24 g and 12 g, respectively. After this first feeding, followed by stirring and heating until all solids dissolved, reactor condition and composition would be the same as those of the original recipe, and the additional feeding can be conducted from additional feeding #1 to #6 as performed in Batch #1.

Provided that all conditions were identical, it can be expected that the material balance in the rest of the process would be identical to those depicted in Fig. 4. Thus, following the scheme of Fig. 7, 37 g of water loss was expected during the reaction; 20 g of water evaporates from wet crystal during filtration, and drying would evaporate 24 g of water to obtain 126 g of melamine acetate crystal, and the resulting mother liquor would consist of 919 g of water and 39 of dissolved melamine acetate were ready to be reused in the next batch.

Fig. 8 depicts the experimental data of Batch #2 with a simplified reaction stage as those conducted in Fig. 4. In this batch, 58 g of total water loss was observed compared to 57 g in Batch #1. If the water evaporated during filtration was considered part of the original wet crystal, then the water content in

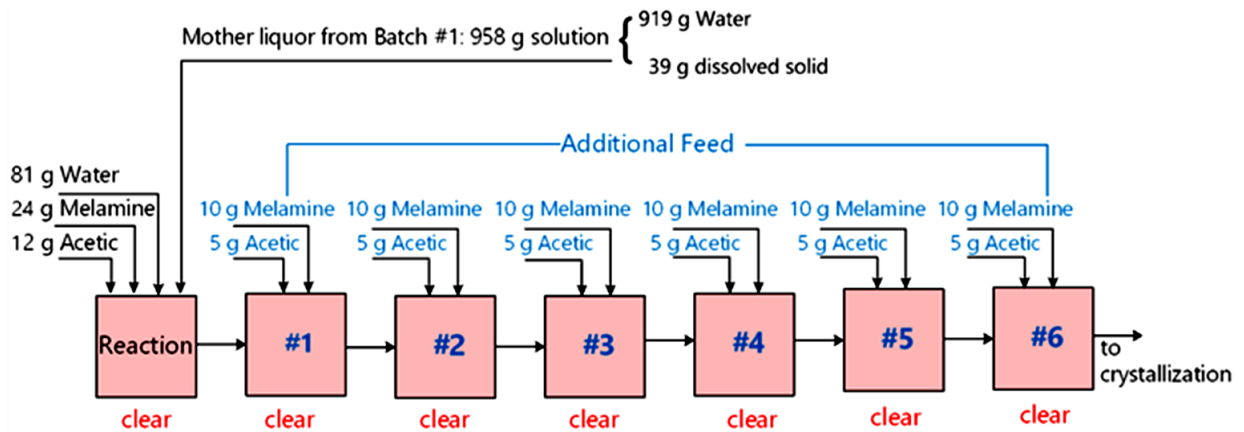


Fig. 7: Raw material feeding scheme in batch #2 and the next batches

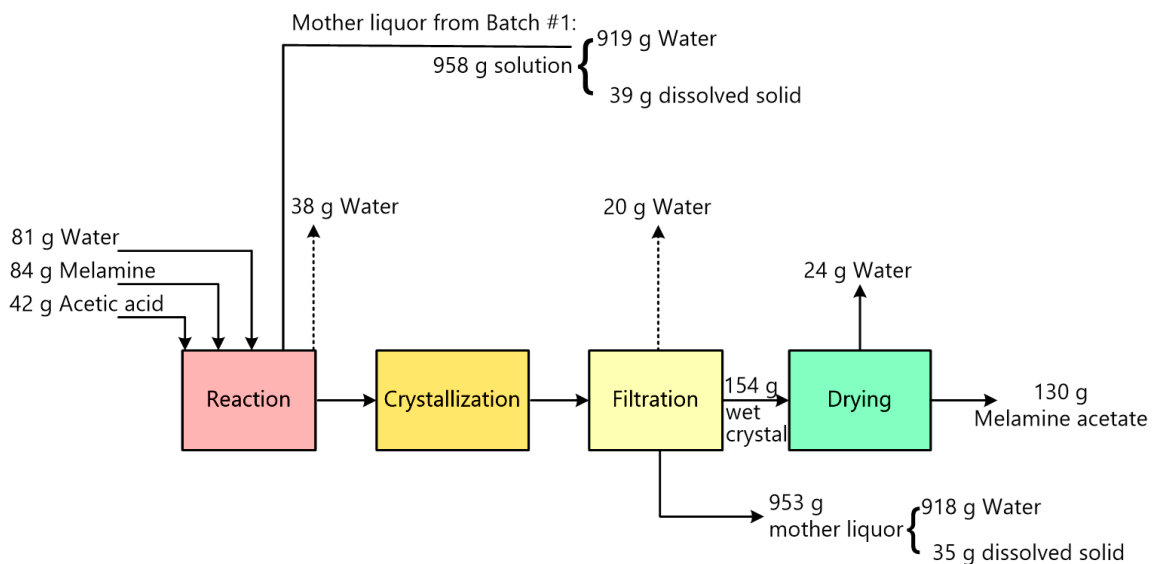


Fig. 8: Experimental batch #2 mass balance after mother liquor reuse from the previous batch

the wet crystal in this batch was 25.3%, like the value of 25.9% in the previous batch. A total of 953 g of mother liquor was obtained in this batch, which was comparable to the value of 958 g in the previous batch. The solid content of the mother liquor was found to be 3.7%, comparable to the value of 4.1% in the previous batch. In conclusion, within the limit of experimental accuracy, all the experimental measurements in Batch #2 can be concluded to be indistinguishable from those of Batch #1.

### Process Consistency

As discussed earlier, the material balance for Batch #2 and the next batches, can be expected to be similar. Hence, mother liquor from Batch #2 was fed to Batch #3 along with the calculated amount of make-up water, melamine, and acetic acid feed. Then this mixture can be processed at the same conditions as the preceding batch. This experiment was conducted and conducted repeatedly until Batch #5.

**Table 1.** Key process variables in the sequential batches of the modified process

Batch #	Fresh Feed, gram			Loss of Water, gram			Mother Liquor, gram	Wet Crystal, gram	Evaporation Duty, gram	Batch Productivity
	Water	Melamine	Acetic Acid	Reaction	Filtration	Total				
1	1000	110	55	37	20	57	958	126	24	10.8%
2	81	84	42	38	20	58	953	130	24	11.2%
3	82	87	43	33	23	56	960	134	23	11.5%
4	79	89	45	35	21	56	958	131	20	11.2%
5	76	87	44	32	20	52	953	133	21	11.4%

Table 1 shows the comparison of key process variables in all the batches. It can be concluded that, within experimental accuracy, the modification process consistently increases melamine acetate production from only 25 g to around 130 g per batch, corresponding to batch productivity increase from only 2.3% to about 11%, while at the same time, practically recover all the raw materials at the next batch, and reduce water consumption to about 80 g per batch.

Figure 9 shows ATR-FTIR spectra of dry melamine acetate products from all the batches, which are barely indistinguishable, showing qualitatively the consistency of the product quality. General observation showed that FTIR spectra of melamine acetate resemble those of melamine in a wide range of wave numbers, particularly between 1700 – 4000  $\text{cm}^{-1}$  because both melamine and melamine acetate structures are dominated by N-H, C-N, and C=N bonds in the triazine ring. However, a closer observation showed the existence of a small peak around 1720 – 1730  $\text{cm}^{-1}$  originating from the C=O bond in melamine acetate, which does not exist in melamine. Between 800 – 1700  $\text{cm}^{-1}$  melamine acetate showed more complex originating from the existence of C-H (1250 – 1450  $\text{cm}^{-1}$ ) and C-O (1050 – 1200  $\text{cm}^{-1}$ ) bonds in melamine acetate which do not exist in the molecular structure of melamine structure. Unfortunately, these differences are not easily

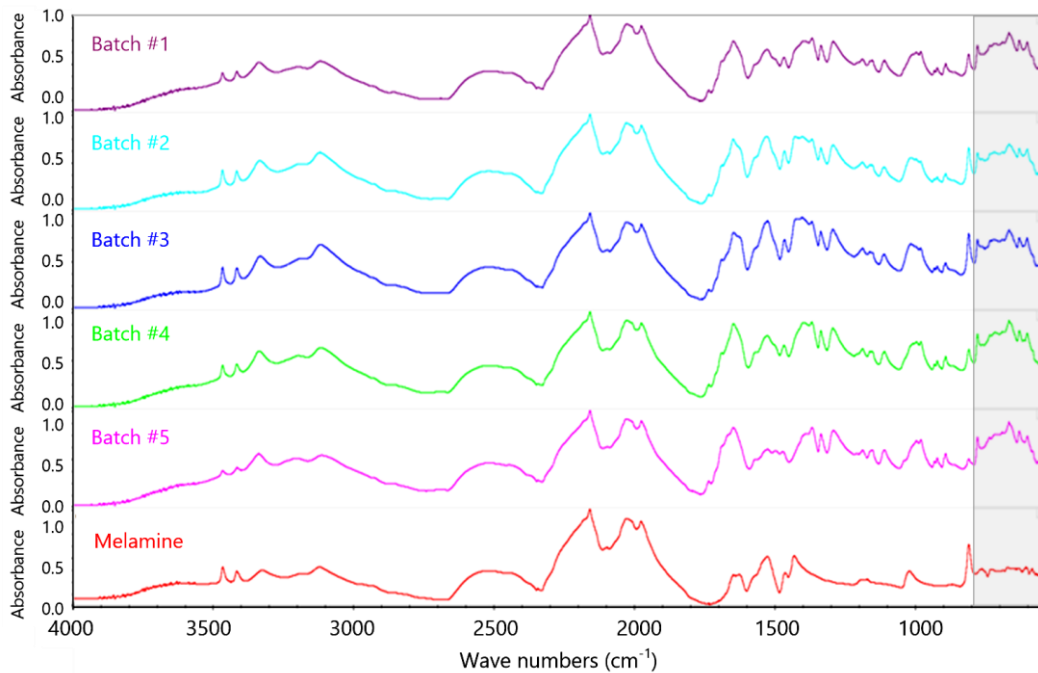
observed due to overlap. However, in the wavenumber range of 800-500  $\text{cm}^{-1}$ , (shaded area in Fig. 9) which falls within the fingerprint region, the vibrational interactions in melamine acetate produce a unique shape distinctly different from that of melamine. As a result, FTIR spectra within this range can be used to differentiate between melamine acetate and melamine easily.

### Application Test

The characteristics of UF resin used were listed in Table 2, specifically formulated for Base particleboard Type 13 according to Japanese Industrial Standard JIS A 5908 (2005). "Type 13" refers to the minimum modulus of rupture of 13.0 MPa, i.e., one of the most important items for particleboard specification.

**Table 2.** Characteristics of the UF Resin

Specification	Value
F/U mol ratio	1.20
Viscosity @25°C, cP	200
Specific Gravity @25°C	1.30
pH	7.70
Solid content, %	67.17
Gel time, sec	52.95
Free formaldehyde content, %	1.37



**Fig. 9:** FTIR spectra of melamine acetate & melamine: the main distinctions are easily observed between 800 – 500 cm<sup>-1</sup> (shaded)

**Table 3.** Particle size distribution in the core and surface layer of particleboard

Particle Size	Core Layer, wt. %	Surface Layer, wt. %
> 4 mm	79.03	-
> 3.15 mm	0.90	-
> 2.00 mm	1.97	-
> 1.25 mm	7.31	15.36
> 800 μm	6.30	17.17
> 315 μm	3.31	47.83
< 315 μm	1.18	19.64

The wood used in this study was a blend referred to as MLH (Mixed Light Hard Wood), which includes Meranti (*Shorea*), a tropical hardwood, plantation hardwood, and Sengon (*Falcata*) provided by PT Dover Chemical, Cilegon. Table 3 shows the particle size distributions of wood particles used. These wood particles conform to the specifications typically employed by a particleboard manufacturer in Indonesia for producing commercial particleboards. Finer particles

were placed in the surface layer to obtain a board product with a smooth surface.

**Table 4.** Parameters in particleboard production

Parameter	Core Layer	Surface Layer
Glue dosing,		
Dry resin to dry chip wt.	5.0%	8.5%
Hardener dosing,		
Dry resin to dry chip wt.	3.0%	0.2%
Initial mat moisture content	4.0%	12.0%

The glue dosing, hardener dosing, and the initial mattress moisture content in each layer are listed in Table 4. The finer particle size for the surface layer requires higher glue dosing. During hot-pressing (Fig. 10), the average temperature at the surface layers would be higher than those at the core layer, causing a higher resin curing rate at the surface layer if the same hardener dosing was used. Hardener dosing for the core layer was

significantly higher to force a higher curing rate at a lower temperature. Higher moisture content in the surface layer assures no overheating in this layer, particularly those in direct contact with hot platen during hot pressing at 140°C due to the cooling effect of water evaporation.

The boards were then conditioned for 24 h at room temperature before trimming and sanding. After cutting to form a wood specimen for testing, the samples were again conditioned at 20°C until a constant weight was achieved and ready for testing.

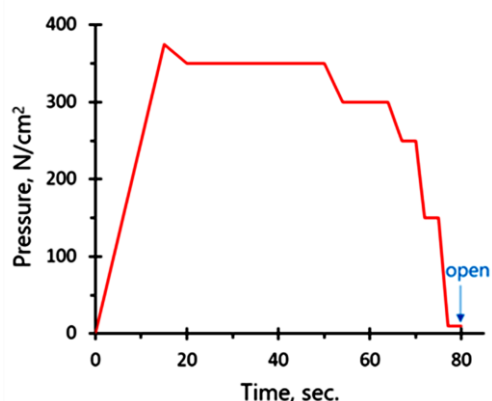
Table 5 summarizes the test results. Board densities are remarkably close to the target value of 0.700 g/cm<sup>3</sup> with a barely identical moisture content of around 7.5%.

**Table 5.** Effect on particleboard properties

Property	Melamine Acetate Added		
	0%	2%	4%
Density, g/cm <sup>3</sup>	0.703	0.687	0.684
Moisture content	7.59%	7.37%	7.58%
Thickness swelling, 2 h @20°C (max. 12%)	14.89%	11.90%	10.02%
Thickness swelling, 24 h @20°C	19.10%	17.81%	16.40%
Modulus of rupture, (min. 13.0 MPa)	13.417	13.383	15.257
Internal bonding strength, (min. 0.3 Mpa)	0.453	0.553	0.593
Formaldehyde emission, mg/L	4.16	4.22	3.25

The board specimen was immersed in water at specified conditions in the thickness swelling test. The addition of melamine acetate was effective in increasing water resistance, demonstrated by the significant decrease in thickness swelling due to water absorbance, both in the 2h and the 24h tests. The maximum value of thickness swelling after 2 h immersion in water at 20°C for

commercial particle board was 12%, which cannot be achieved using UF resin only, while adding 2% wt. of melamine acetate, based on the resin wt. was proven to overcome the problem.



**Fig. 10:** Pressure profile during hot-pressing

The modulus of rupture (MOR) is a parameter that measures the strength of a material when subjected to external forces. It is the maximum stress a material can withstand before breaking or rupturing. The resin acts as an adhesive that binds wood particles into a board. Thus, the mechanical and physical properties of the particleboard depend greatly on the properties of the resin. Melamine-containing resin is more highly cross-linked than purely urea formaldehyde resin, resulting in stronger bond strength and more rigid resin networks. Therefore, incorporating melamine acetate into UF resin is expected to increase the MOR value. As expected, adding melamine acetate results in better mechanical properties, demonstrated by the increase in both the modulus of rupture (MOR) and the internal bonding strength. Nevertheless, in this case, the minimum expected value of MOR and IB strength, was 13.0 MPa and 0.3 MPa, respectively, which was achieved using UF resin only.

Likewise, adding of melamine acetate

also tends to lower formaldehyde emission. However, the improvement was considered unimportant because all boards produced are still in the same classification of emission quantity of formaldehyde.

## CONCLUSIONS

Melamine acetate is a special form of melamine that was shown to have potential as a urea-formaldehyde resin additive for wood-based panel binders. To improve melamine acetate process preparation, specifically to increase batch productivity, this study has successfully modified the original process of Zanetty & Pizzi (2003), resulting in a higher productivity and more energy-efficient production process. The modification was conducted by feeding raw materials at a higher quantity than the solubility limit, increasing batch productivity from only 2.3% in the original process to around 11%. The reuse of mother liquor for the next process batch recovers all the raw materials as a product while significantly reducing freshwater make-up to only about 8% of the required quantity in the original process. The modification of oven-drying to lower temperature sun-drying process diminishes heat requirement while the process still achieved practically required degree of dryness with less product decomposition. The application test by adding 2% - 4% wt. of melamine acetate to UF resin prepared for particleboard production showed that the most important improvement effect of melamine acetate addition is the increased water resistance of particleboards produced, showed by the decrease in thickness swelling. Improvements in MOR, IB strength, and formaldehyde emission were also observed.

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