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TECHNOLOGICAL FACTORS AFFECTING THE EMISSION OF FORMALDEHYDE FROM PARTICLEBOARDS

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ABSTRACT

The present work aimed to determine the combined effects of the manufacturing variables on the emission of formaldehyde from particleboards produced from low formaldehyde emission (E1 type) adhesive, while maintaining acceptable physical and mechanical properties. The manufacturing variables studied included pressing temperature (160, 170 and 180 °C), moisture content of the wood – glue mixture before pressing (10, 12 and 14 %) and the density of the boards produced (700, 750 and 800 kg/m^3) while keeping constant the glue and the time variables. The effects of these manufacturing variables on the formaldehyde emission and internal bonding strength were investigated. The results show that the release of formaldehyde from particleboards bonded with urea-formaldehyde resin can be reduced significantly through manipulations and adjustments of the manufacturing variables, i.e. the press temperature, the board density and the moisture content of the wood-glue mixture before entering the press. According to the results, the emission of formaldehyde increases with the moisture content of the wood-glue mixture before entering the press, decreases with the density of the products and, within certain limits, the temperature of the press, has a positive effect (reduces emission). Further, the internal bond strength of the final product increases with the density of the boards and the moisture content of the wood-glue mixture before entering the press, whereas the temperature, similarly to the emission of formaldehyde, has within limits, a positive effect.

KEY WORDS: particleboard, moisture, density, pressing temperature, formaldehyde emission, internal bond strength

INTRODUCTION

The urea-formaldehyde resin has been one of the best available materials, as a bonding agent, for the industrial production of wood composites. However the urea formaldehyde adhesive used to bond these products emits formaldehyde. Wood based panels such as particleboard and medium density fibreboard, were, for many years, a major source of formaldehyde emissions in domestic dwellings. In general, the emission of VOC's to the indoor air from wood-based panels appears to depend on a number technological factors and manufacturing process variables (Larsen et al. 1999, Wang et al. 2003). Over the past years, mainly through improvements in adhesive technology, emissions from these products have been reduced up to 90% (Sudin 1986), although, in many markets (Sakai et al. 2004, Wang et al. 2004), even today the problem of formaldehyde emission is very acute. Hydrolysis of cured resin from urea resin bonded boards appears to contribute the most in the formaldehyde release in product use (Elbert 1995, Kavvouras et al. 1997, Kazakeviés and Spedding 1979, Myers 1990). The sensitivity of urea-formaldehyde polymers to the hydrolytic action is considered as the key factor responsible for the formaldehyde emission (Meyer 1979, Meyer et al. 1980, Myers 1983). The reduction in formaldehyde emission levels from products bonded with urea-formaldehyde adhesives has been achieved through different technological approaches (Dunky 2000, Dziurka et.al. 2003, Meyer et al. 1980, Myers 1985, Myers 1989, Myers 1990, Tomitaa and Hseb 1998, Wang et al. 2003, 2004), such as: (i) changing the formulation of the urea-formaldehyde adhesive resin (e.g., lowering the F/U ratio) (ii) adding special hardeners/crosslinkers during the final setting of the resin (iii) adding formaldehyde-scavenging materials directly to the urea-formaldehyde adhesive resin (iv) separately adding formaldehyde-scavenging materials to the wood finish (v) treating panels after their manufacturing either with a formaldehyde scavenger or by the application of coatings or laminates, and (vi) changing to an entirely different adhesive resin system.

The most widely used approach for reducing formaldehyde emission levels was the mole ratio of F/U. Ratios of about 1,6 that were common 20-25 years ago (Mari 1983) have now been reduced to values as low as 1,0 and, in some cases, lower. Unfortunately, lowering the F/U ratio has as a consequence (Myers 1984, Roffael and Dix 1991) that these urea-formaldehyde adhesives can not be stored very long (shorter shelf-life), require relatively longer pressing times (higher gel time) and produce panels that often have poorer physical and mechanical properties (tendency towards more swelling and low transverse tensile strength). Virtually all related treatments create trade-offs, either in increased expenses or board quality. As a result, the panel manufacturers had to use adhesive resins with a higher F/U ratio and to employ other post manufacturing treatment methods (Myers 1986, Pease 1993) to achieve the necessary reduction in formaldehyde emission levels.

Research work (Marutzky et al. 1992, Nemli 2002, Sudin 1979, Vasilèv et al. 1988) has shown that the processing conditions employed during the production of those products (process variables) have a decisive effect on both, their mechanical properties and the stability of the urea-formaldehyde resin under environmental conditions favoring the hydrolysis. The basic process variables include the pressing temperature, the pressure, the time of pressing and the moisture content of the wood-glue mixture. Extensive investigations carried out in many countries have shown that the liberation of formaldehyde from wood composites can be reduced significantly, through the proper control of production process variables, during the manufacturing stage of the wood composites (Marutzky et al. 1979, Marutzky et al. 1992, Nemli 2002, Sudin et al. 1992, Szesztay et al. 1993, Vasilèv et al. 1988). It is obvious from above that the optimization of resin formulation should be accompanied by the optimization of board manufacturing parameters. The application of optimized parameters during manufacture will ensure that formaldehyde emissions will meet E1 grade requirements and optimize board mechanical properties without further production cost increase.

The temperature and pressure during the hot pressing and the moisture content of the wood-glue mixture contribute significantly to the thermal degradation and the subsequent hydrolytic cleavage of glue polymers (Wang et al. 2003). The presence of catalyst is considered to be responsible for the acidic environment inside the board, which favors and further facilitates this hydrolytic activity (Szesztay et al. 1993).

In most of the research work carried out so far, the processing conditions and their effect on the formaldehyde liberation were studied individually and mostly during the stage of hot pressing. The target of the study is to provide information about the relationship between particleboard manufacturing process variables and formaldehyde release during its use. This information will help determine whether changes in manufacturing conditions can be used to reduce emission levels during subsequent product use. In the first part of the study an initial qualitative screening has been attempted through the evaluation of the effects of isolated process variables on final product formaldehyde emission. The second part, deals with the study of the selected variables interactions. The aim of the work was not to generate absolute numbers as much as to demonstrate trends. The combined effect of the process variables on the emission of formaldehyde from particleboards bonded with a low molar ratio urea-formaldehyde resin were studied taking into account the corresponding mechanical (strength) properties of the final product as expressed by its internal bond strength (tearing resistance perpendicular to the surface of the board). The internal bond strength is considered to be a good indicator of the overall strength properties of the board. The final objective was the creation of a data basis that will enable to identify the weight of each process variable on the formaldehyde emission, and the elaboration, at a later stage, of a real time control system for the minimization of formaldehyde emission during the use of the product.

MATERIAL AND METHODS

Logs from trees of *Abies cephallonica* from the forest area of Karpenissi were collected and transferred to the laboratory of the Forest Research Institute in Athens. The logs were chipped using a two-knife laboratory scale flake chipper. The flakes were hammer milled, screened and their dimensions were further adjusted so that they had a more or less uniform size of *ca.* 15x5x0,3 mm. The wood flakes were kiln dried to an average moisture content of *ca.* 15-20%, on an oven-dry (o.d.) weight basis, placed in plastic bags and stored outdoors until their use. Experimental particleboards were made using a laboratory scale single opening press. The plates of the press were heated with electricity and the final thickness of the boards was adjusted with stoppers. The wood flakes were thoroughly mixed with U: F glue (7% w/w on the oven-dry weight of wood), placed by hand in a box type 500x500x200 mm mold and pre-pressed. The glue characteristics were: liquid form U:F glue with molar ratio of F:U 1,1, 65 % solids, gel time 2,68 min (20 ml of resin at 90 °C), pH 7,69, viscosity 197 cps (at 20 °C). The catalyst was NH_4NO_3 and it was added in the form of a solution (30% w/v in water), at a proportion of 2% (on the basis of the o.d. weight of wood). Zinc powder was added at a proportion of 5% (w/w on the basis of the solids of the glue) for the adjustment and monitoring of the pH of the glue. The final thickness of the boards was adjusted with stoppers. As mentioned earlier, the present work was carried out in two stages. During the first stage, the basic production variables (moisture, temperature, time and density) were each studied separately for its individual effects on both the formaldehyde emission and the internal bond strength of the produced boards. Each of the above variables was studied in three different levels, i.e. density of the boards 600, 700 and 800 kg/m^3, moisture content of the wood-glue mixture (before pressing) 9, 11 and 13%, time of pressing 4, 5 and 6 min, pressing temperature 140, 160 and 180 °C. The basic, standard, conditions for the preliminary tests, against which each variable was studied, were density 700 kg/m^3, wood-glue moisture content (before pressing) 11 %, time of pressing 5 min and pressing temperature 160 °C. The above design gave 12 “types” of boards (4 variables at 3 levels each) and the production, in total, of 36 boards (each type was produced in triplicate). The boards, after a three-week ageing period, were tested for formaldehyde emission (EN 717-3:1994) and the internal bond strength (B.S. 5669). The variables studied in the second stage of the work were "temperature", "moisture content" of the wood-glue mixture and "density" of the boards. In order to study the full interaction of those variables, it was necessary to have 36 "types" of boards. Each type of board was produced in triplicate so that, in total, 108 boards were produced. Each of the boards produced as above was evaluated, after a three-week ageing period, for its formaldehyde emission using the Flask method (EN 717-3:1994) and the internal bond strength properties (B.S. 5669). The boards, before testing were conditioned at 20 °C and 65% relative humidity for two weeks.

RESULTS AND DISCUSSION

The results from the first stage of work showed that the pressing temperature, the board density and the wood-glue moisture content have had a decisive effect on both properties of the boards studied, i.e. the internal bond strength and the formaldehyde emission. When the pressing temperature was raised from 140 °C to 180 °C, the other variables being kept constant (density 700 kg/m^3, wood-glue moisture content 11 %, and time of pressing 5 min) the boards showed an increase in the emission of formaldehyde and a reduction of the internal bond strength values. This may be the result of the thermal degradation of resin (Szesztay et al. 1993). The board density also appeared to have a significant effect on the internal bond strength of the boards. An increase in board density from 600 to 800 kg/m^3, the other variables being kept constant (temperature 160 °C, wood-glue moisture content 11 %, and time of pressing 5 min) had as a result an increase in the internal bond strength by approx. 63%. The formaldehyde emission appears to increase when the board density increases from 700 kg/m^3 to 800 kg/m^3. At the higher level of board density, the results from the preliminary tests showed that the formaldehyde emission was increased. This is contrary to previous reports (Marutzky et al 1979, Sudin et al. 1992), where it was found that boards with lower density liberate higher proportion of formaldehyde. The moisture content of the wood-glue mixture (before pressing) appeared to exercise a strong influence on the emission of formaldehyde. An increase in moisture content from 9% to 13%, the other variables being kept constant (temperature 160 °C, board density 700 kg/m^3, and time of pressing 5 min) resulted to an increase of 113% in the formaldehyde emission. The moisture content had an almost negligible effect on the internal bond strength. The variable "time of pressing " had, in all cases, a minor effect on both the emission of formaldehyde and the internal bond strength, when compared to the effect of the other three variables. As a result this variable was not included in the second stage of tests. These preliminary tests supplied the basis for defining the final range of the variables employed for study of the multiple, combined, effects of those variables on the properties of the boards. The design of the experimental work and the results obtained during the second stage of the experimental work are shown in Table 1. As it can be seen from these results, the moisture content of the wood-glue mixture appears to have a rather weak influence on the properties of the boards. Within the range of 8 to 14 % moisture content studied, a range of pressing temperatures between 160-190 °C and board density 700 to 800 kg/m^3, the values of formaldehyde emission appear fluctuating erratically around the mean value in each case (see Figures 1 and 2). In contrast, as it will be noted from Table 1, both the pressing temperature and the final density of the board appear to influence strongly the emission of formaldehyde and the internal bond strength of the boards (see Figures

3 and 4). The moisture content of the wood-glue mixture appears to be critical on the emission of formaldehyde above the level of 12%. Below that level the other two factors (temperature and board density) have the decisive effect upon the properties of the boards.

Tab. 1: Experimental design and results obtained for the study of the combined effects of the process variables on formaldehyde emissions and internal bond strength of boards

Board No	Production Process variables			Board properties	
	Wood-glue Moisture content %	**Density of boards** kg/m³	**Pressing temperature** °C	**Formaldehyde emission** (Flask method) mg/kg	**Internal Bond Strength** MPa
1	8	700	160	15,52	0,39
2			175	11,03	0,43
3			190	12,23	0,51
4		750	160	14,12	0,55
5			175	12,53	0,51
6			190	10,18	0,63
7		800	160	12,20	0,56
8			175	11,24	0,51
9			190	9,39	0,35
10	10	700	160	15,96	0,27
11			175	14,57	0,32
12			190	10,80	0,48
13		750	160	13,87	0,39
14			175	12,44	0,36
15			190	11,25	0,44
16		800	160	12,19	0,56
17			175	10,97	0,60
18			190	11,09	0,57
19	12	700	160	17,41	0,34
20			175	15,22	0,33
21			190	11,76	0,36
22		750	160	12,92	0,40
23			175	13,48	0,44
24			190	13,36	0,45
25		800	160	13,16	0,51
26			175	12,13	0,54
27			190	12,89	0,47
28	14	700	160	18,27	0,19
29			175	15,67	0,35
30			190	12,35	0,21
31		750	160	15,97	0,23
32			175	13,18	0,43
33			190	13,25	0,59
34		800	160	12,86	0,34
35			175	12,22	0,34
36			190	12,30	0,35

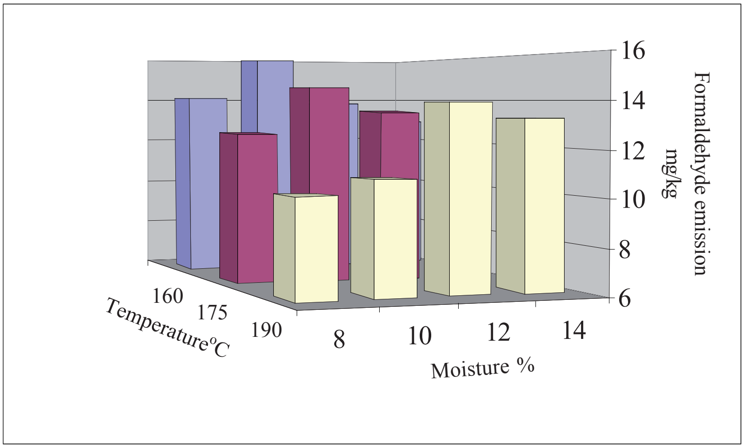

Fig. 1: Combined effect of wood-glue and pressing temperature on the formaldehyde emission, at a calculated average board density of 750 kg/m³

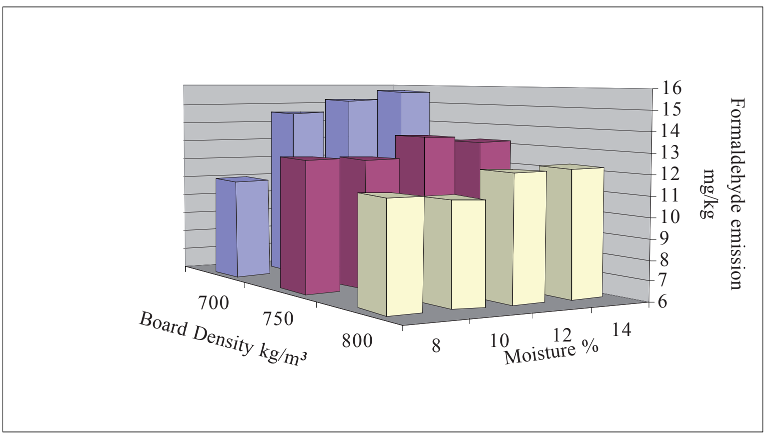

Fig. 2: Combined effect of wood-glue and board density on the formaldehyde emission, at a calculated average pressing temperature of 175 °C

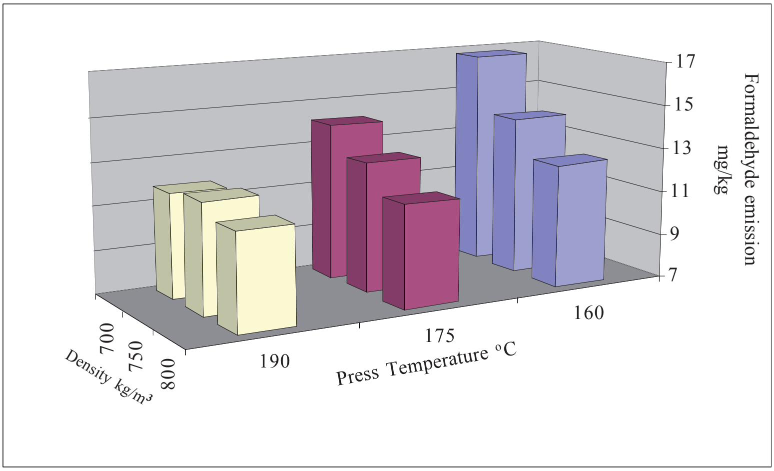

Fig. 3: Effect of pressing temperature and board density on the formaldehyde emission at a calculated average moisture content of wood-glue mixture of 11%

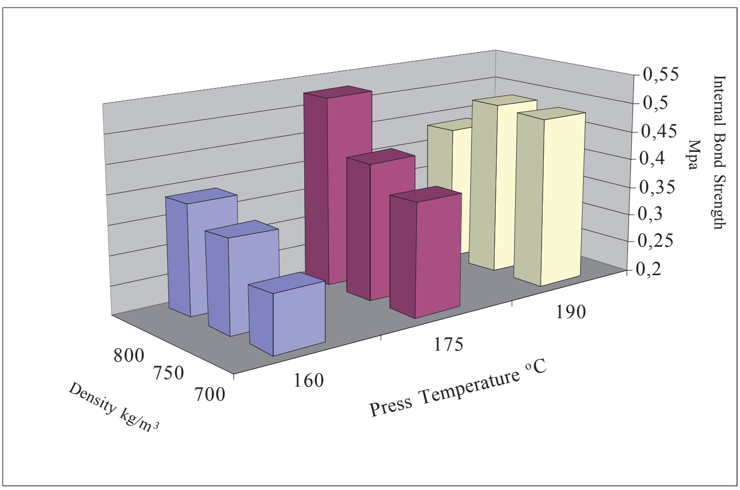

Fig. 4: Effect of pressing temperature and board density on the internal bond strength at a calculated average moisture content of wood-glue mixture of 11%

The density of the boards appears to be the most significant of the variables studied in the present work. The internal bond strength of the boards increases with the density. The emission of formaldehyde decreases with increasing the density of the boards. However, when the pressing temperature increases to 190 °C, the positive effect of the board density on the reduction in formaldehyde emission disappeared. The same is also valid for the internal bond strength. The reason may be that the temperature, above certain limits, has a negative effect on the properties of the boards. The results of the present work indicate that the release of formaldehyde from wood composites bonded with urea-formaldehyde resin systems can be reduced significantly through manipulations and adjustments of the process variables during the stage of the production of the boards, i.e. the press temperature, the board density and the moisture content of the wood-glue mixture before entering the press. More specifically, the emission of formaldehyde:

- increases with the moisture content of the wood-glue mixture before entering the press
- decreases with the density of the products
- temperature of the press, within certain limits, has a positive effect (reduces emission)

Further, the internal bond strength of the final product increases with the density of the boards and the moisture content of the wood-glue mixture before entering the press, whereas the temperature, similarly to the emission of formaldehyde, has within limits, a positive effect.

After statistical processing of the results obtained in the present work, the following linear interactions were established:

	Moisture Content %	**Pressing Temperature** °C	**Board Density** kg/m³
Formaldehyde emission	F=9,26+0,345MC	F=29,3933-0,0933PT	F=30,664-0,0235D
Internal Bond strength	IBS=0,6894-0,0212MC	IBS=0,0277+0,0023PT	IBS=-0,523+0,0013D

where F is the emission of formaldehyde (Flask value in mg HCHO/kg of oven-dry weight of board), IBS is the internal bond strength, and MC, PT and D are the production variables moisture content of the wood-glue mixture before entering the press (%), temperature of the press (°C) and density of the boards (kg/m^3) correspondingly. The above simple linear regressions were used in the multiple regression analysis for the calculation of the liberation of formaldehyde and the internal bond strength properties.

The emission of formaldehyde after one week of ageing can be calculated from the equation:
F = 0,52584 (34,6135 – 0,069783 MC + 0,019783 PT – 0,03233 D)

Whereas the internal bond strength can be calculated from the equation:

IBS = -0,50728929 + 0,000866 MC + 0,002255 PT + 0,000109 D

In conclusion, the present work shows that the liberation of formaldehyde from wood composite materials of low formaldehyde emission type, glued with urea-formaldehyde glues, can be further significantly reduced though the optimization and monitoring of the production process variables, i.e. the pressing temperature, the board density and the moisture content of the wood-glue mixture before the pressing stage.

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