Towards composites with formaldehyde emission at natural wood levels

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ABSTRACT

The acceptable levels of formaldehyde emission from composite panel products have been continuously reduced over the last decades. The driving forces have been the increased public awareness and consumer demand for non-hazardous products as well as the corresponding governmental regulations. The recent re-classification of formaldehyde by the International Agency for Research on Cancer as "carcinogenic to humans", has triggered further concern and reactions by worker and consumer associations, "green" organisations, regulatory authorities and the industry itself. New studies on formaldehyde health effects were initiated since 2005 in both the USA and Europe and corresponding decisions on reclassification have been postponed until the results are available.

CHIMAR HELLAS, wood chemicals technology provider, has pioneered in developing products reducing the panel formaldehyde release. With the aim to protect people's health and to help the industry satisfy the acceptable formaldehyde levels, CHIMAR has developed novel wood adhesive system technologies, which provide composite panels conforming to the most stringent standards. Even panel grades with emission at the level of natural wood (Super E0, the Japanese F****) are obtained, while simultaneously maintaining acceptable bonding performance.

The proposed adhesive systems are the results of many years' research and experimentation in laboratory, pilot and industrial environments. In this work, the advantages of such systems are presented. In parallel, the leading standards and regulations concerning formaldehyde emission from wood composites are reviewed and mention is made of the Occupational Exposure Limits worldwide.

INTRODUCTION

The issue of formaldehyde release from composite wood panels is mainly related to the use of urea-formaldehyde (UF) resins as bonding adhesives for their production. These high reactivity and low cost polymers are contributing to the panel formaldehyde emission by their low resistance to hydrolysis and the presence of free non-reacted formaldehyde. Resin copolymers produced with the use of melamine either at low (urea-melamine-formaldehyde resins, UMF) or at high levels (melamine-urea-formaldehyde resins, MUF) have improved hydrolytic stability but yet questionable performance in regard to very low formaldehyde emission levels.

Urea-formaldehyde resins have traditionally been used in the production of wood-based panels (mainly particleboard, fibreboard, plywood) and related products for decades. Indoor air quality and formaldehyde emissions from composite wood products first became subject to broad public and governmental concern in the late 1970s, when the energy crisis encouraged heat conservation through tight sealing of homes. This reduced the rate of outdoor air infiltration and overall ventilation rates leading to the entrapment of gaseous pollutants inside home air atmosphere. With Europe, North America and Japan as pioneers, test methods to accurately measure formaldehyde emissions from panels were developed and product emission guidelines were established. These were combined with work place exposure limits for formaldehyde. A total change in the formulation of UF resins was made by the resin industries to meet the low panel emission guidelines. Moreover, competitive bonding systems such as phenol-formaldehyde resins or polymeric diphenyl methane diisocyanate (PMDI) binders were proposed.

Until recently, formaldehyde was classified by the World Health Organization (WHO) as probably carcinogenic to humans (Group 2A). In 2004, the International Agency for Research on Cancer (IARC) of WHO, decided to recommend the reclassification of formaldehyde as "carcinogenic to humans (Group 1)", on the basis of available scientific data (IARC 2004). This recommendation, although not legally binding, was received with concern and immediate reactions from worker and consumer associations, "green" organisations, regulatory authorities and the industry (producers and users of formaldehyde) associations (FormaCare and European Panel Federation in Europe, Formaldehyde Council and Composite Panel Association in N. America). The latter indicated that the decision of IARC had been based on studies regarding the exposure in 30-60 years ago, while the work place emission levels have declined dramatically the past three decades due to the technological progress made (Composite Panel Association 2004, Formaldehyde Council 2004). Moreover, the IARC classification is a "hazard identification" and not a full risk assessment.

In 2005, new toxicological and cancer studies were initiated by FormaCare and the Formaldehyde Council, involving various independent research institutes in Europe and the USA. In the meantime, there were proposals to reclassify formaldehyde in Europe by the French institute for occupational risk prevention (INRS) and the German Federal Institute for Risk Assessment (BfR). The European Chemicals Bureau (ECB) has postponed its decision on the reclassification of formaldehyde until the results of the new studies are available (FormaCare 2006). Within the EU, formaldehyde is currently classified as a Category 3-R40 substance ("limited evidence of carcinogenic effect"), which is the lowest available EU category for suspected carcinogens. Also the U.S. Environmental Protection Agency (EPA) has delayed formaldehyde reclassification until the completion of the follow-up study of the National Cancer Institute. The current classification by EPA is that of a probable human carcinogen.

The IARC recommendation was finally published in December 2006 through its monograph series Volume Number 88. In this report, it is stated that "there is sufficient evidence in humans and in experimental animals for the carcinogenicity of formaldehyde" and that "formaldehyde is carcinogenic to humans (Group 1)" (IARC 2006). It is further mentioned that the highest occupational exposure to formaldehyde were measured in varnishing, production of textiles, garments, furs, in certain jobs in board mills and foundries. Lower exposure levels have been encountered in formaldehyde production (mean concentration < 1ppm). A wide range of exposure levels has been observed in the production of resins production (all data were derived from the 1980s). In wood products manufacture, exposure occurs during glue mix preparation, laying of mat, hot pressing and sanding – all data were derived from the 1960s-70s-80s. The reported mean concentrations in the air were greater than 1ppm in particleboard mills and approximately 2ppm in plywood, however, recent studies reported concentrations lower than 0.4ppm in plywood and less than 0.16ppm in OSB and fibreboard plants.

In September 2007, an International Formaldehyde Science Conference took place in Barcelona, Spain, organized by FormaCare (the European Formaldehyde Industry Association, sector group of CEFIC, the European Chemical Industry Council). The results of newly available scientific studies on the epidemiology and toxicology of formaldehyde were presented and discussed by representatives of institutes from Europe, the USA and Brazil, European Commission representatives and industry scientists (FormaCare 2007, Press Release). Main conclusions include the following points: a) the evidence for NasoPharyngealCancer formation is highly ambiguous, b) the leukemia formation related to formaldehyde exposure is highly improbable, c) no mutagenic effects were observed in experiments and d) in the normal living environment or at the workplace, formaldehyde exposure is not expected to lead to sensory irritation. The threshold for sensory irritation is clearly lower than that leading to cell death. Concentrations of 0.5 ppm or 0.3 ppm with peaks of 0.6 ppm will not lead to objective signs of sensory irritation. The bottom line of the conference was that “the common use of formaldehyde in consumer products and other applications does not pose a risk to human health”. The IARC recommendation was challenged by presenting the weak points of the studies on which it was based (old data, lack of statistical robustness of data analysis, no consideration of confounding effects) (also in FormaCare 2007, Scientific fact sheet). Such weak points and the newly available data suggest that there is no clear connection between formaldehyde exposure at current levels of exposure and cancer in humans.

Occupational Exposure Limits

The Occupational Exposure Limits (OELs) for formaldehyde are presented in Table 1. In countries with higher limits there are discussions to lower them following the recommendation of IARC (e.g. in Australia, the OEL of 1.0ppm will be set to 0.3ppm in 2008).

Table 1: Occupational Exposure Limits (OELs) for formaldehyde (IARC 2006, FormaCare 2007, Q&A on formaldehyde)

Country	Concentration, ppm	Type[a]
Australia	1.0	TWA
Austria	0.3	TWA
Belgium	0.3	Ceiling
Brazil	1.6	Ceiling
Canada-Alberta	2.0	Ceiling
Canada-Ontario	0.3	Ceiling
Canada-Quebec	2.0	Ceiling
Denmark	0.3	TWA & STEL
Finland	0.3	TWA
France	0.5	TWA
Germany	0.3	TWA
Greece	2.0	TWA
Hong Kong	0.3	Ceiling
Ireland	2.0	TWA
Italy	0.3	Ceiling
Japan	0.5	TWA
Malaysia	0.3	Ceiling
Mexico	2.0	Ceiling
Netherlands	1.0	TWA
New Zealand	1.0	Ceiling
Norway	0.5	TWA
South Africa	2.0	TWA
Spain	0.3	STEL
Sweden	0.5	TWA
Switzerland	0.3	TWA
United Kingdom	2.0	TWA
USA-ACGIH[b]	0.3	Ceiling
USA-NIOSH[b]	0.016	TWA
USA-OSHA[b]	0.75	TWA

[a]*TWA: time-weighted average, STEL: short-term exposure limit.*

Formaldehyde emission standards for wood-based panels

In Table 2, the classification of wood-based panels in respect to formaldehyde emission according to the European, Australian, U.S.A. (also valid in Canada) and Japanese standards is given.

Table 2: Formaldehyde emission standards for wood-based panels in Europe, Australia, the U.S.A. and Japan

Country	Standard	Test method	Board class[a]	Limit value
Europe	EN 13986	Chamber EN 717-1 Perforator EN 120	E1-PB, MDF, OSB	≤0.1ppm ≤8mg/100g
		Chamber EN 717-1 Gas analysis EN 717-2	E1-PW	≤ 0.1ppm ≤ 3.5mg/h*m^2
		Chamber EN 717-1 Perforator EN 120	E2-PB, MDF, OSB	>0.1ppm >8 - ≤30mg/100g
		Chamber EN 717-1 Gas analysis EN 717-2	E2-PW	> 0.1ppm > 3.5 ≤÷ 8.0mg/h*m^2
Australia	AS/NZS 1859-1 & 2	Desiccator AS/NZS 4266.16	E0-PB, MDF	≤0.5mg/L
			E1-PB	≤1.5mg/L
			E1-MDF	≤1.0mg/L
			E2-PB, MDF	≤4.5mg/L
U.S.A.	ANSI A208.1 & 2 (PB & MDF)	Large chamber ASTM E1333	PB, MDF	≤0.3ppm
		Large chamber ASTM E1333	Industrial PW	≤0.3ppm
		Large chamber ASTM E1333	PW wall panels	≤0.2ppm
Japan	JIS A 5908 & 5905 (PB & MDF)	Desiccator JIS A 1460	F** F***/E0 F****/SE0	≤1.5mg/L ≤0.5mg/L ≤0.3mg/L

[a]PB: particleboard, MDF: medium density fibreboard, OSB: oriented strand board, PW: plywood

In Europe, Germany pioneered in issuing regulations as well as in reducing panel formaldehyde emissions in actual industrial practice. Currently the German regulation requires compliance with E1 emission limits of 6.5mg/100g for particleboard and 7.0mg/100g for fibreboard (tested by the perforator method) (Roffael 2006). Also other European countries like Austria, Denmark and Sweden followed Germany in producing boards of only these E1 levels. However, most European countries still have legislation allowing the production and distribution of E2 boards (according to EN 13986). Recently, the members of EPF agreed to only produce E1 boards and that compliance should be monitored through a system of internal and external checks (EUWID 2007). At the same time, the members firmed up the E1 limit values for ongoing production monitoring.

The emission limit of the Japanese F** class is more or less equivalent to European E1-class according to the German regulations, while the F*** and F**** emission limits are much lower than the E1-class. The emission of F**** boards is close to the emission of solid untreated wood (e.g. between 0.008-0.01ppm or 0.5-2.0mg/100g for spruce wood flakes, Marutzky et al. 2004).

The Air Resources Board of California (CARB) adopted a new regulation in April 2007 to reduce the formaldehyde emissions from composite wood products, including particleboard, MDF and hardwood plywood (Airborne Toxic Control measure, ATCM) (CARB 2007). This regulation proposes the reduction of formaldehyde emission standards in two phases (Figure 1). Phase 1 limits (effective from January 2009) are roughly equivalent to the European E1 emission class that is the Japanese F** class (Figure 2). Phase 2 limits (effective from January 2011) are comparable to the Japanese F*** limits, the so-called E0 levels in Europe. The new regulation will establish the most stringent formaldehyde emission limits on wood products in the United States. The measure requires that wood panels and products manufactured from wood panels be certified by a "third party" laboratory approved by the CARB.

	-------------- Standard Concentrations (ppm) --------------				
Phase 1 Formaldehyde Emission Standards					
Eff. Date	HWPW-VC	HWPW-CC	PB	MDF	tMDF
Jan 2009	0.08	-----	0.18	0.21	0.21
July 2009	-----	0.08	-----	-----	-----
Phase 2 Formaldehyde Emission Standards					
Eff. Date	HWPW-VC	HWPW-CC	PB	MDF	tMDF
Jan 2011	0.05	-----	0.09	0.11	-----
Jan 2012	-----	-----	-----	-----	0.13
July 2012	-----	0.05	-----	-----	-----

(1) "ppm" = parts per million; "Eff. Date" = effective date; "HWPW-VC" = hardwood plywood – veneer core; "HWPW-CC" = hardwood plywood – composite core;"tMDF" = thin medium density fiberboard (8-mm or thinner). "Standard Concentrations" are allowable limits based on measurements made using the American Standards & Testing Method E1333-96.

Figure 1: The California Air Resources Board new standards (CARB 2007)

P1 (ppm)	E1	F☆☆☆	F☆☆☆☆
HWPW (0.08)	More	Comparable	Less
PB (0.18)	Less	Less	Less
MDF (0.21)	Less	Less	Less
P2 (ppm)	E1	F☆☆☆	F☆☆☆☆
HWPW (0.05)	More	More	Comparable
PB (0.09)	More	Comparable	Less
MDF (0.11)	Comparable	Less	Less

(1) Values in parentheses are the proposed Phase 1 or Phase 2 standards in ppm. "More" means the proposed standard is "more stringent" that applicable E1, F☆☆☆, or F☆☆☆☆ standards.

Figure 2: CARB versus European and Japanese standards (CARB 2007)

Discussions on a new emission class are currently under way in Europe. Industry insiders believe that the limit will be similar to that of the Blue Angel certification for environmentally-friendly products and services (EUWID 2007). Wood-based panels meeting this certification have to remain at least 50% below a formaldehyde emission value of 0.1ppm set out in class E1 and thus produce emissions of less than 0.05ppm. This new emission value should be fixed as a voluntary guideline outside of standardisation. It may, however, serve as the basis for a future standard.

THE CHIMAR HELLAS APPROACH TO THE FORMALDEHYDE EMISSION REDUCTION

CHIMAR HELLAS S.A. is an innovating **technology provider** for the resin and wood-based panel industries and a pioneer in industrial research and development in the field of formaldehyde emission reduction. Following a process of continuously experimenting at laboratory, pilot and industrial scale, CHIMAR has concluded that the formaldehyde emission problem can be solved by the use of:

- **Innovative gluing systems** of resins and chemical additives, consisting of high performance urea-formaldehyde resins (with/without melamine) in combination with a special additive-formaldehyde scavenger, the synergistic action thereby provides panels with the desirable properties and low formaldehyde emission.
- **Advanced technologies for resin synthesis,** by better exploiting the active ingredients during resin synthesis, to provide resins for boards with very low formaldehyde emission (even at the level of the Super E0 class, i.e. emission at the level of natural wood) and acceptable mechanical strength and water resistance properties.
- **Resins based on biomass products or by-products** (e.g. soya, tannin, starch, lignin from paper making, pyrolysis oil) that combine the utilisation of renewable raw materials with high bonding performance and reduced formaldehyde emission at the same time.

All of the above-mentioned techniques have been optimised to offer cost savings to the resin and wood-based panels industries as well as to their end users-the consumers of such products.

The present work aims to report findings from recent tests at the pilot and mostly at the industrial scale, to produce low formaldehyde emission panels by applying the above first two approaches. Yet, CHIMAR already in 1993 reported results from industrial particleboard production using a system of UF resin and formaldehyde scavenger (formaldehyde catcher), that provided panels with formaldehyde emission at the level of natural wood (1.8mg/100g of board, Markessini 1993).

MATERIALS, METHODS AND RESULTS

Pilot MDF trial at the Chinese Academy of Forestry (CAF)

China is a country with large timber industry. As low quality products are generally produced in this country, China is trying to escape from this situation and produce more reliable and environmentally friendly products. Hence there is an imminent need for the timber industry, to be in the position to produce and offer E1 quality boards. Today, the Chinese MDF plants can produce low E2 level boards but the E1 emission level cannot be met easily. There are two obstacles for this as far as MDF production is concerned:

- It is thought that the Chinese wood species provide fibres which are problematic when E1 resins are applied.
- The Chinese industry believes that E1 MDF production is more costly than the low E2 MDF.

Pilot scale tests were run by CHIMAR at the Chinese Academy of Forestry to overcome both of these obstacles. A straight UF resin providing low E2 boards was used for the trial in combination with a formaldehyde scavenger and the target was to produce E1 quality boards. Both products (resin and scavenger) were of CHIMAR technology. The scavenger was used in substitution of the resin solids at levels of 10 and 12% w/w. Industrial fibres from a local plant were provided. The wood species was 66% pine plus various local species. The resin loading used was 15% w/w and no paraffin was added. The target board density was 650kg/m^3 and the target thickness was 8.8mm. The analysis of the boards' properties was performed by CAF and the results are shown in Table 3.

Table 3: Properties of pilot MDF produced at CAF

Sample	Scavenger, %	Density, kg/m^3	Swelling, %	IB, N/mm^2	Emission, mg/100g
Control	0	700	17.2	0.74	9.9
Glue mix 1	10	770	19.2	0.97	5.6
Glue mix 2	12	760	22.1	0.83	4.6

Industrial E0 MDF trial in Europe

Industrial scale tests were performed by applying the CHIMAR resin technology in a European MDF mill with the aim to produce panels of E0 emission quality. A UMF resin was applied in MDF with target density and thickness of 720kg/m^3 and 14mm respectively. Urea was used as formaldehyde scavenger. The wood fibre species consisted of 50% beech and 50% softwood. The results from the boards' analysis that was carried on-site are shown in Table 4.

Table 4: Properties of industrial E0 MDF produced in Europe

Sample	Resin content, %	Urea	Thickness, mm	Density, kg/m^3	IB, N/mm^2	Emission, mg/100g
Plant control	10.5	yes	13.9	710	0.81	6.56
CHIMAR UMF	12.5	no	14.1	704	0.67	3.36
CHIMAR UMF	12.5	yes	14.3	696	0.62	2.50

Industrial MDF trials in Australia

In Australia, industrial trials were performed to produce moisture resistant (MR) thin MDF with formaldehyde release lower than 0.5mg/L (E0 grade). An MUF resin of CHIMAR technology was applied in combination with a CHIMAR formaldehyde scavenger. The scavenger was used in substitution of the resin solids at levels of 10, 15 and 20% w/w. The total resin loading used was 18% w/w. The target board thickness was 4mm. The analysis of the boards' properties was performed on-site and the results are shown in Table 5, representing average values of series of samples.

Table 5: Properties of industrial E0-MR MDF produced in Australia

Requirements		10% Scavenger	15% Scavenger	20% Scavenger
Density,	<850kg/m^3	832	795	818
IB,	>1.35N/mm^2	1.89	1.36	1.30
Swelling,	<11%	4.2	6.3	9.1
Emission,	<0.5mg/L	0.45	0.38	0.31

Furthermore, trials were performed to produce industrial MDF with emission lower than 0.3mg/L (Super E0 grade). A CHIMAR system of UMF resin and formaldehyde scavenger was applied at two levels of total loading: 16% and 18% w/w. The scavenger was used in substitution of the resin solids at levels of 15 and 20% w/w in both levels of loading. The target board thickness was 16mm. The analysis of the boards' properties was performed on-site and the results are shown in Table 6, representing average values of series of samples.

Table 6: Properties of industrial SE0 MDF produced in Australia

Requirements		15% Scavenger 16% Loading	15% Scavenger 18% Loading	20% Scavenger 16% Loading	20% Scavenger 18% Loading
IB,	>0.8N/mm^2	1.1	1.0	0.9	0.9
Swelling,	<12%	7.2	7.1	7.0	7.4
Emission,	<0.3mg/L	0.30	0.28	0.29	0.27

Industrial scale trials aiming at producing SE0 MDF (emission lower than 0.3mg/L) of a target thickness of 3.2mm were performed too. A CHIMAR system of UMF resin and formaldehyde scavenger was applied at total loading level of 18% w/w. The scavenger was used in substitution of the resin solids at levels of 15 and 20% w/w. The analysis of the boards' properties was performed on-site and the results are shown in Table 7, representing average values of series of samples.

Table 7: Properties of industrial SE0 thin MDF produced in Australia

Requirements		15% Scavenger	20% Scavenger
Swelling,	<37%	26.3	26.9
Emission,	<0.3mg/L	0.23	0.26

Industrial particleboard trials in Australia

Industrial scale tests were carried out to produce 3-layer particleboards with formaldehyde release lower than 0.5mg/L (E0 grade). A UMF resin of CHIMAR technology was applied without adding formaldehyde scavenger. The resin loading was 9.5% w/w in the surface and 8.5% w/w in the core. The target board density and thickness were 630kg/m^3 and 16mm respectively. The analysis of the boards' properties was performed on-site and the results are shown in Table 8, representing average values of series of samples.

Table 8: Properties of industrial E0 PB produced in Australia

	Thickness, mm	Swelling, %	IB, N/mm^2	MOR, N/mm^2	Emission, mg/L
Requirement	>12 to 22	15.0	0.30	15.0	0.50
Test samples	16.02	12.3	0.39	15.3	0.30

Furthermore, industrial scale moisture resistant particleboards of E0 emission grade were produced as per the above production parameters but using an MUF resin of CHIMAR technology. The on-site analysis of the boards' properties provided the results shown in Table 9, which actually represent the average values from series of samples.

Table 9: Properties of industrial E0 MR PB produced in Australia

	Thickness, mm	Swelling, %	IB, N/mm^2	MOR-A, N/mm^2	Emission, mg/L
Requirement	>12 to 22	15.0	0.30	4.5	0.50
Test samples	16.02	4.3	0.61	6.4	0.30

DISCUSSION AND CONCLUSIONS

The following points can be made in relation to the pilot production of MDF at the Chinese Academy of Forestry:

- The trial was successful in producing E1 quality MDF by using a low E2 resin and formaldehyde scavenger without deterioration of the board properties or change of the production parameters.
- There was no direct or indirect increase of the production cost and no loss of productivity due to use of the CHIMAR resin system (low E2 resin and formaldehyde scavenger).
- Actually the formaldehyde emission was reduced in the range of 50% while the internal bond strength (IB) was significantly increased.
- The increase of the percent swelling cannot be taken into consideration since the boards were produced without wax due to technical difficulties of the manufacturing procedure at CAF. Nevertheless the swelling results are not disappointing either.

- The above-mentioned results were obtained using fibres from a local plant which are considered to exhibit problems when E1 boards are produced.

By using the CHIMAR resin system, it was thus made possible to produce E1 quality MDF with problematic Chinese fibres and without increasing the production cost as compared to low E2 quality boards.

In regard to the industrial trials in MDF production in both Europe and Australia as well as the industrial particleboard trials in Australia the following can be noted:

- The trials were successful in producing the desired quality of MDF and PB panels in regard to low formaldehyde emission (E0 or SE0 level) by using either an advanced aminoplastic resin or an innovative system of aminoplastic resin plus scavenger.
- There was no need to change the production parameters or plant settings.
- There was no loss in productivity or significant increase of production cost.
- The board properties were not adversely affected by the introduction of low emission resin system. In many cases there was even an improvement of the board properties.
- The low emission quality can be achieved even in very specific cases such as the thin or moisture resistant panel products.

These developments have shown that it is possible to meet the new demands for very low formaldehyde emission from composite panel products with the use of properly formulated aminoplastic resins systems, without any deterioration in panel performance or significant modification of plant operating conditions or need to employ other types of binders. The formaldehyde emission values that can be obtained are **at the level of natural wood.**

CHIMAR HELLAS has succeeded in reducing the panel formaldehyde emissions by developing innovative resin systems, using advanced resin synthesis technologies and components that are well studied and controlled. Through its worldwide experience, network of customers and collaborating research institutes, CHIMAR HELLAS develops and implements integrated solutions to the formaldehyde emission problem.

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