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FORMALDEHYDE, PHENOL AND AMMONIA EMISSIONS FROM WOOD/RECYCLED POLYETHYLENE COMPOSITES

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ABSTRACT

Wood-plastic composites (WPC) from recycled polyethylene and wood particles provide an improved sustainability compared to conventional wood-based composites such as plywood, particleboard and medium density fibreboard. In fact, the implementation of these materials into industrial applications is hindered by the missing knowledge on the environmental properties of such WPC. Therefore, the main aim of this study was to determine the release of formaldehyde, phenol, and ammonia from flat pressed WPC. The emissions were evaluated by chamber method. Various recycled polyethylene/wood particles ratios (20/80; 40/60; 60/40; 80/20; 100/0) were used for manufacturing WPC in a laboratory scale. It was found that formaldehyde, phenol and ammonia emission of flat pressed WPC are much lower than steady-state emission. As a consequence, such WPC create significant ecological impact as green composites.

Key words: wood plastic composites, volatile organic compounds, emission, chamber method.

INTRODUCTION

Wood plastic composites (WPC) are new generation of wood composites without using thermosetting formaldehyde resins such as urea formaldehyde (UF) or phenol formaldehyde (PF). Different types of polymers are used for the production of WPC, mainly they are polyethylene, polypropylene and polyvinylchloride (EDER 2010, NISKA *et al.* 2008, ROWELL 2005). Basically, virgin polymers are used for manufacture of WPC but recycled polymers can also be used. The use of these thermoplastic polymers can be explained by their low melting temperature (110–160 °C). Such low temperatures are also important for the manufacture of wood-based composites, as the thermal stability of wood becomes worse when higher temperatures (more than 200 °C) are used (KLYOSOV 2007, NISKA *et al.* 2008). Moreover, elevated temperatures cause the decomposition of the wood macromolecules and negative changes in wood properties.

Mostly WPC are produced by extrusion which gives an opportunity to obtain composites of endless profiles with high durability, water resistance, chemical resistance and ability to re-use (EDER 2010, NISKA *et al.* 2008, ROWELL 2005). Alternative technology is making WPC in flat presses (LYUTYY *et al.* 2014). This technology is less studied and less explored but it has several advantages over extrusion: a relatively lower pressure is required, the saving of wood raw material due to lower density of composite panels, higher productivity of flat pressing technology. Flat pressing technology is widely used for the production of conventional wood-based composites such as particleboard, medium density fibreboard (MDF) and oriented strand particleboard (OSB). The thermosetting formaldehyde resins usually are used for producing these composites. That leads to the emission of free formaldehyde, phenol, ammonia and other volatile compounds during the pressing and over the service life of the composites (SALEM *et al.* 2011, ROFFAEL *et al.* 2010).

In 2004, the International Agency for Research on Cancer (IARC) decided to reclassify formaldehyde ($H_2C=O$) as "carcinogenic to humans (Group 1)", on the basis of available scientific data (IARC 2006). Predominant signs of short-term exposure to formaldehyde in humans are irritation of the eyes, nose and throat, together with concentration-dependent discomfort, lachrymation, sneezing, coughing, nausea, dyspnoea. Formaldehyde has been shown to be toxic in vitro in a variety of experimental systems, including human cells. It reduced growth rate, cloning efficiency and the ability of cells to take up neutral red and to exclude trypan blue while inducing squamous differentiation of cultured human bronchial epithelial cells (WHO 2001).

Phenol (C_6H_6O) is not classifiable as to its carcinogenicity to humans, but according to toxic reaction on humans is considered to Group 3. Phenol poisoning can occur in humans after skin absorption, inhalation of vapours or ingestion. Acute local effects are severe tissue irritation and necrosis. At high losses, the most prominent systemic effect is central nervous system depression (IARC 1999).

Inhalation of ammonia (NH_3) will rapidly cause irritation to the nose, throat and respiratory tract. Increased lacrimation, coughing and increased respiratory rate as well as respiratory distress may occur (IPCS 1986). Ammonia is water soluble and is therefore absorbed by the mucosa of the upper respiratory tract; this protects the lungs from exposure to low concentrations of ammonia (IPCS 1986).

That's why the WPC, which is produced without using thermosetting resins, can be the alternative to particleboard, OSB, MDF and in future can displace them (ŠATANOVÁ *et al*. 2015, MIRSKI *et al*. 2015). According to EN 13986: 2015, the test requirement does not apply to wood-based panels to which no formaldehyde containing materials were added during production or in post-production processing; these may be classified E1 without testing. But wood as a natural material contains formaldehyde. The steady-state emissions vary between 2 to 9 ppb (0.001 ppm or 0.00125 mg/m^3) after a test period of 240 to 384 hours (MEYER *et al.* 1996). Although, information about the VOCs composition and releases of harmful substances from the WPC would be of great interest and importance not only to industry but also to the consumers, there are only a few studies devoted to this subject (FÉLIX *et al.* 2013).

Therefore, the main aim of this study was to investigate the emission of formaldehyde, ammonia and phenol from WPC experimental panels produced by conventional flat pressing technology, which is widely used in manufacturing wood-based panels.

MATERIALS AND METHODS

Recycled polyethylene (rPE) and wood particles (WP) were used in this study to manu facture WPC. Commercial WP were produced and received from the particleboards mill. T he wood species composition was according to the mixture during the manufacture of parti cleboard and the ratio between coniferous and deciduous species was about 60/40. The rPE film was cleaned from food particles and shredded. The fraction analysis of WP and rPE is presented at Table. 1.

Tab. 1 Fraction analysis (by % weight).

Components	Screen hole size (mm)						
	-/5	5/4	4/2	2/1	1/0.63	0.63/0.315	0.315/0
WP	4.75	12.2	15.79	40.28	15.67	9.13	2.18
rPE	9.53	3.04	53.14	32.45	1.83	-	-

Various WP/rPE (%/%) ratios (20/80; 40/60; 60/40; 80/20; 100/0) were used to prepare WPC mat and to press panels.

WP and rPE (in the natural dry state) were manually mixed based on appropriate ratios and after that the WPC mat without having any resin or additives was initially cold pressed and then pressed in the hot press by pressure of 3.5 MPa, temperature of 200 °C and time of 1.2 min/mm. No adhesive was used since there is polyethylene which melts and acts as an adhesive to bond the mat in panel. At the end of the press cycle, the panel was removed from the press for cooling to the temperature of 30–40 °C in cold press. The WPC thickness was 16 mm and the density of WPC was 800 kg/m^3. Finally, the experimental WPC panels were conditioned in a climate room with a relative humidity of 65 ± 5% and a temperature of 20 ± 2 °C before being cut into test specimens. WPC panels without rPE (100% wood particles) were made at the same pressing parameters for comparison of formaldehyde, ammonia and phenol emissions.

The formaldehyde emission was determined in chamber according EN 717-1. The standard parameters of testing were modified according to MHU (2012) to be able to determine not only the formaldehyde emission, but also ammonia and phenol emissions and they are described in Table 2. The amount of volatile organic compounds in the WPC specimens were determined by photocolorimeter in the cuvette with layer thickness 20 mm at the wave length of 412 nm. The emission (C) of volatile organic compounds was defined by the formula (mg/m^3):

$$C = \frac{m}{V} \tag{1}$$

where, m – the amount of volatile organic compounds in the specimen, mg;

V – the volume of air taken for analysis and brought to normal conditions, m^3.

Tab. 2 The parameters of testing for formaldehyde, ammonia and phenol emissions.

Conditions	Specification
Temperature, °C	20 and 40
Relative humidity, %	65
Loading factor, m^2/m^3	0.4
Edges sealing	all edges
Air exchange rate, L/h	from 0.5 to 1.0
Test time, hours	24

RESULTS AND DISCUSSION

The investigation results of formaldehyde, ammonia and phenol emissions from WPC and 100% wood particles are presented in Table 3.

According to the Order of the Ministry of Health of Ukraine the emission of formaldehyde from non-coated wood based panels should be not more than 0.01 mg/m^3 (MHU 2012). The formaldehyde emission form investigated WPC is two times less than steady-state emission. The same results were observed also for ammonia and phenol emissions. Respectively such materials could be used for furniture manufacture, and as building material in all types of constructions (external and internal).

Tab. 3 Emission of formaldehyde, ammonia and phenol from WPC.

Substance	Emission at temperatures (20 ± 2) and (40 ± 2) °C mg/m^3 of WPC panels at different WP/rPE ratios (%/%)		Steady-state emission, mg/m^3, according to MHU (2012)
	20/80; 40/60; 60/40; 80/20	100/0	
Formaldehyde	< 0.005*	< 0.002*	0.010
Ammonia	< 0.020*	< 0.020*	0.040
Phenol	< 0.002*	< 0.002*	0.003

* - values are lower than the sensitivity of the measuring device

WPC have a little bit higher emission of formaldehyde at different WP/rPE ratios in comparison with composite without rPE (100% wood particles). It can be explained by some degradation of alkyl radicals/groups which are in rPE (BOLDIZAR *et al.* 2000). More than 140 VOC compounds were observed in the raw materials and industrially and experimentally manufactured WPC samples (FÉLIX *et al.* 2013). The most abundant compounds quantified were furfural, α-pinene, 2-ethyl-1-hexanol, 2-methoxyphenol, N-methylphthalimide, butylated hydroxytoluene, 2,4-di-tert-butylphenol and diethyl-phthalate. But it should be noted that FÉLIX *et al.* (2013) used higher temperature (80 °C) for VOC compounds quantification in comparison with this study (temperature 20 and 40 °C). The temperature of 80 °C is an extremely high temperature for any application of WPC where the consumer could be directly exposed to the vapours. That's why it could be explained that at temperature 20 and 40 °C the release of VOC was lower in this study.

Wood/Recycled polyethylene composites are suitable, sustainable and biodegradable green materials to achieve durability without using toxic chemicals. The extensive use of wood fibres is because of their properties such as low density, relative high strength, modulus and stiffness, non-abrasive nature, high level of filler loading, biodegradability and safe working environment (ASHORI 2008). On the other hand, the growing volume of plastic solid waste is necessary to find the process for high added value products.

CONCLUSIONS

The experimental results obtained show that formaldehyde, phenol and ammonia emissions from flat pressed WPC were much lower than steady-state emission. This information can greatly expand the different application (indoor and outdoor) of such composites. As a consequence, WPC is a very promising, sustainable material without emission of toxic chemicals.

REFERENCES

ASHORI, A., 2008. Wood-plastic composites as promising green-composites for automotive industries. Bioresour. Technol. 99: 4661–4667.

BOLDIZAR, A., JANSSON, A., GEVERT, T., MOLLER, K. 2000. Simulated recycling of post-consumer high density polyethylene material. Polymer Degradation and Stability, 2000, 68: 317–319.

MIRSKI, R., DZIURKA, D., WIERUSZEWSKI, M. 2015. Properties of OSB boards after a few cycles of aging tests. Wood Research, 60(4): 633–644.
EDER, A. 2010. Wood-plastic composite markets in Europe. In The Fourth China International Summit of WPC, 2010, 22 p.
EN 717-1: 2005. Wood-based panels. Determination of formaldehyde release. Part 1: Formaldehyde emission by the chamber method.
EN 13986: 2015. Wood-based panels for use in construction. Characteristics, evaluation of conformity and marking.
FÉLIX, J.S, DOMEÑO, C., NERÍN, C. 2013. Characterization of wood plastic composites made from landfill-derived plastic and sawdust: volatile compounds and olfactometric analysis. Waste Management, 2013, 33(3): 645–655.
IARC. 1999. Monographs Vol 71: Re-evaluation of some organic chemicals, hydrazine and hydrogen peroxide. France, Lyon, 1999.
IARC. 2006. Monographs Vol 88: Formaldehyde, 2-Butoxyethanol and 1-*tert*-Butoxypropan-2-ol. France, Lyon, 2006.
IPCS. 1986. International Programme on Chemical Safety. Environmental Health Criteria. Vol 54: Ammonia. Geneva, Switzerland, 1986.
KLYOSOV, A.A. 2007. Wood-Plastic Composites. New Jersey: John Wiley & Sons, Inc., 2007, 698 pp.
LYUTYY, P., BEKHTA, P., SEDLIAČIK, J., ORTYNSKA, G. 2014. Properties of flat-pressed wood-polymer composites made using secondary polyethylene. Acta Facultatis Xylologiae Zvolen, 2014, 56(1): 39−50.
MEYER, B., BOEHME, C. 1997. Formaldehyde emission from solid wood. Forest Products Journal, 1997, 47(5): 45–48.
MHU. 2012. Ministry of Health of Ukraine. Order №1139. On approval of the State sanitary norms and rules; Polymer and Polymer materials, products and structures used in construction and furniture manufacturing. Hygiene requirements. Kyiv, Ukraine, 2012.
NISKA, K.O., SAIN, M. 2008. Wood-polymer composites. Woodhead Publishing Limited, Cambridge, 2008.
ROFFAEL, E., JOHNSSON, B., ENGSTROM, B. 2010. On the measurement of formaldehyde release from low-emission wood-based panels using the perforator method. Wood Science and Technology, 2010, 44: 369–377.
ROWELL, R.M. 2005. Handbook of wood chemistry and wood composites. Boca Raton : CRC Press, 2005, 487 pp.
SALEM, M.Z.M., BÖHM, M, BARCÍK, Š., BERÁNKOVÁ, J. 2011. Formaldehyde Emission from Wood-Based Panels Bonded with Different Formaldehyde-Based Resins. Drvna Industrija, 2011, 62(3): 177–183.
ŠATANOVÁ, A., ZÁVADSKÝ, J., SEDLIAČIKOVÁ, M., POTKÁNY, M., ZÁVADSKÁ, Z., HOLÍKOVÁ, M. 2015. How Slovak small and medium manufacturing enterprises maintain quality costs: an empirical study and proposal for a suitable model. Total Quality Management and Business Excellence, 26(11-12): 1146–1160.
WHO. 2001. Air Quality Guidelines for Europe: second edition. Formaldehyde. World Health Organization, 2001, 91: 87–91.

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