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CONTENT OF FORMALDEHYDE IN LIGNOCELLULOSIC RAW MATERIALS FOR PARTICLEBOARD PRODUCTION

The article contains the results of tests for formaldehyde content in selected wood species, alternative raw materials from experimental and agricultural plantations, particles dried under industrial conditions, recycled chips, and in boards made of elements of furniture, at the end of its life cycle. An unexpectedly high content of formaldehyde was determined in particleboards produced a few dozen years ago. It was observed that the amount of formaldehyde in particles dried under industrial conditions may have a bearing on the content of this compound in finished products and/or its emission into the environment. Control tests of raw materials may be performed using the flask method.

Keywords: formaldehyde content, wood raw material, conventional, alternative, recycled

Introduction

In 2006 the International Agency for Research on Cancer (IARC), which belongs to the World Health Organization (WHO), tightened up the assessment of the incidence of cancer caused by formaldehyde through recognizing this compound as a 1. group substance, i.e. a substance that is considered carcinogenic to humans [Christianson 2004; Kupczewska-Dobecka 2007]. Before that date, the IARC considered formaldehyde a compound that was "probably carcinogenic to humans" [International Agency for Research on Cancer 2004]. The recommendation of the IARC has no legal efficacy, nevertheless it brought about an intensification of research aimed at the reduction of formaldehyde emissions, as well

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as a discussion on the sources of formaldehyde emissions and the permissible level as regards production processes and products. In 2011, the US government added "formaldehyde used, among others, for the production of synthetic resins" to a list of carcinogenic factors, which resulted in the tightening of regulations concerning formaldehyde content in and emission from wood-based materials [In the USA formaldehyde …]. In Europe the IARC assessment resulted in the classification of formaldehyde as a category 3. substance, i.e. a substance that may be carcinogenic to humans [Kupczewska-Dobecka 2007, Reduction of formaldehyde 2010]. Discussions concerning the harmful effect of formaldehyde often raise the argument that formaldehyde is ubiquitous in nature, i.e. in most living organisms such as people, animals, trees, plants etc. [Reduction of formaldehyde 2010, Timber Trades Journal]. The research on formaldehyde content in and emission from natural wood has been conducted for years. Within this field, the formaldehyde emission from wood and the changes of this emission due to thermal technological processes were tested [Marutzky, Roffael 1977; Sachsse, Roffael 1993; Roffael 2000], the emission of formaldehyde from birch, aspen and pine was determined [Ellbert 1995], as well as the effect of the storage period of pine and spruce wood on formaldehyde release [Schäfer 1996]. Schäfer, Roffael [2000], Roffael [2006] conducted broad research on the role of major components (lignin, cellulose, hemicelluloses, and wood extracts) as potential sources of formaldehyde.

One of the most recent publications [Weigl et al. 2009] presents the results of tests of formaldehyde content in various European wood species: Scots pine, Norway spruce, birch, poplar, and oak. Different classes of wood and samples of early and late wood were tested. The content of formaldehyde was determined by the extraction method based on tests of particles obtained, dried and stored in constant conditions. The principle of the perforator method, according to EN 120:1992 standard, was adopted with only some of its parameters modified. The formaldehyde content was within the range of 0.15-0.71 mg/100 g oven dry wood. Coniferous wood was characterised by a higher content although wood species seemed irrelevant. Clear differences between juvenile and mature wood were observed. The lowest formaldehyde content was observed in juvenile red beech wood (0.15 mg/100 g), and the highest in mature pine wood, where it was approximately 0.70 (mg/100 g). The tests of formaldehyde content in natural wood, conducted by Meyer and Boehme [1997], resulted in similar values, from within the above range. Within the framework of prefatory research conducted in the Wood Technology Institute in 2005, it was observed that the formaldehyde content in so-called "dry particles", which had been sampled randomly in one particleboard plant, was 1.8-2.1 mg/100g of oven dry particles, approximately 25% of the permissible formaldehyde content in E1 class particleboards. The approximate value of the perforator, i.e. 1.6 mg/100 g of oven dry particles after conversion to a moisture content of 6.5%, was determined based on the tests of particle samples after drying in industrial conditions in 2009 [Danecki 2009]. More and more diverse raw wood material has been used in the production of wood-based panels, especially particleboards. The situation on the wood market and an attempt to minimise waste, essential for sustainable development, are the reasons for the drive to use production waste and post-consumer wood, including recycled furniture, as a source of material. This article presents the results of tests for formaldehyde content in and emission from: natural wood of selected domestic species, alternative lignocellulosic raw materials for wood-based panel production, so-called post-consumer wood", and particles used in the production of boards in various plants.

Materials and methods

The test were performed on selected domestic wood species most often used in wood-based panel production, as well as on alternative raw materials, which were proved useful for the production of composite panels for the furniture industry and construction, based on the results of statutory research carried out in the Wood Technology Institute in the period 2007-2009 [Frąckowiak 2007, 2010]. Post-consumer wood, mainly from particleboards originating from elements of furniture, which after a long period of use was at the end of its life cycle, was tested as well. The test samples were cut out in the Institute. The samples of recycled chips, intended for tests for formaldehyde content and emission, were randomly taken from a chip bunker at one of the domestic panel producers. The tests also took into account particles industrially-dried in diaphragm and non--diaphragm dryers, and sampled at various particleboard producers in Poland and Germany.

The characteristics of the tested materials are given in tables 1-4.

The particles were tested according to the methods described in the following standards:

- PN EN 120:1994 Wood-based panels – Determination of formaldehyde content – Extraction method called the perforator method,
- PN EN 322:1999 Wood-based panels – Determination of moisture content,
- PN EN 717-3:1999 Wood-based panels – Determination of formaldehyde release – Part 3: Formaldehyde release by the flask method (modified by the authors with particle testing in mind).

  The particles for emission testing were put into a receptacle made of thick, thin and transparent polyamide fabric. The mass of a particle sample, due to the relatively large volume, was reduced to 15±1 g. The amount of water was in accordance with the assumptions of the standard method. Formaldehyde release was calculated taking into consideration the mass and moisture content of the particles.

Results and discussion

Table 1 presents the results of the tests for formaldehyde content in and emission from pine, alder, birch and alternative lignocellulosic raw materials for the production of particleboards. Pine sapwood was characterised by the highest content and emission of formaldehyde amongst the tested wood species. A relatively high formaldehyde content in birch bark and yellow rape straw, i.e. fresh and, tested before storage, also draws attention. The lowest formaldehyde content was found in the wood from two-year-old root offshoots of the locust tree (*Robinia pseudoacacia*).

Table 1. Average values of formaldehyde content in and emission from various raw material assortments – wood from forest plantations and plantations, agricultural waste
Tabela 1. Średnie wartości zawartości i emisji formaldehydu z różnych sortymentów surowca – drewno z upraw leśnych i plantacji, odpady rolnicze

Characteristics of the tested material [%] *Charakterystyka badanego materiału [%]*		Moisture content when formaldehyde content was tested *Wilgotność w chwili badania zawartości formaldehydu* [%]	Formaldehyde content converted into dry mass of wood *Zawartość formaldehydu po przeliczeniu na suchą masę drewna* [mg/100 g]	Moisture content when formaldehyde emission was tested *Wilgotność w chwili badania emisji formaldehydu* [%]	Formaldehyde emission converted into dry mass of wood *Emisja formaldehydu po przeliczeniu na suchą masę drewna* [mg/kg]
1		2	3	4	5
Pine/*Sosna Pinus sylvestris L)* 52 years old/ *lata*	bark/*kora*	16.4	0.41	-	-
	sapwood/*biel*	77.9	1.42	15.4	0.52
	heartwood/ *twardziel*	19.0	0.83	10.8	0.62
Alder/*Olcha Alnus glutinosa (L.) Gaertn* 66 years old/ *lat*	bark/*kora*	26.7	0.51	-	-
	peripheral wood/ *drewno obwodowe*	44.0	0.35	44.0	0.12
	wood from the middle part of trunk/ *drewno ze środkowej części pnia*	57.0	0.38	57.0	0.00
Birch/Brzoza *Betula verrucosa Ehrh* 37 years old/ *lat*	bark/*kora*	38.6	1.11	-	-
	peripheral wood/ *drewno obwodowe*	12.3	0.26	12.3	0.21
	wood from the middle part of trunk/ *drewno ze środkowej części pnia*	13.0	0.15	13.0	0.19

Table 1. Continued
Tabela 1. Ciąg dalszy

1	2	3	4	5
Miscanthus straw *Słoma z miskanta olbrzymiego* *Miscanthus sinesis giganteus*	13.3	0.34	13.3	0.1
Two-year-old shoots of fast-growing willow/*Pręty dwuletnie szybko rosnącej wierzby* *Salix Viminalis* L.	7.3	0.21	7.4	0.3
Two-year-old root offshoots of locust tree/*Odrośle korzeniowe dwuletnie robiniii akacjowej* *Robinia pseudacacia* L.	13.5	0.11	13.0	0.6
Yellow rape straw/ *Słoma rzepakowa zółta* *Brasica napus* L.	14.9	1.9	-	-
Grey rape straw/ *Słoma rzepakowa szara* *Brasica napus* L.	8.9	0.9	-	-

Unexpected test results were obtained for particleboards from the elements of furniture, at the end of its lifecycle. The highest (27.0 mg/100 g of oven dry board), but also the lowest formaldehyde content was determined in the case of panels produced in the 1970s. The samples of the panels came from furniture used in conditions familiar to the authors and from materials sent to the Institute for testing. Therefore, the influence of the conditions in which the tested furniture panels and materials were used or stored on the content of formaldehyde in the panels may be excluded. The level of formaldehyde content seems connected rather with the technology than with the time that had passed since the panels were produced. Laminating or veneering of the panels' surface, which hindered formaldehyde release from the material, could have some influence on the high formaldehyde content. The panel samples were tested after removing the coatings.

The content of formaldehyde in the tested recycled chips ranged from 1.6 to 4.1 mg/100 g of chips.

Fig. 1 illustrates the interrelation between the formaldehyde content in the panels and the emission from them tested by the flask method. Data presented in Fig. 1 indicates a strong correlation between the test results obtained using these methods, thus it confirms the conclusions from previous research [Frąckowiak 2003], i.e. that the flask method is useful (and at the same time inexpensive and environmentally-friendly) for the testing of particleboards stored in different periods, produced using various technologies and raw materials.

Table 2. The results of the tests for formaldehyde content in and emission from post--consumer wood
Tabela 2. Wyniki badań zawartości i emisji formaldehydu z drewna poużytkowego

Characteristics of the tested material *Charakterystyka badanego materiału*	Moisture content *Wilgotność*	Formaldehyde content *Zawartość formaldehydu*		Formaldehyde emission *Emisja formaldehydu*	
		at moisture content of the samples when tested *przy wilgotności próbek w chwili badania*	at moisture content of the samples after conversion into moisture content of 6.5% *przy wilgotności próbek po przeliczeniu na wilgotność 6,5%*	test results *wyniki badania*	average value *wartość średnia*
	[%]	[mg/100 g z.s.p.]		[mg/kg]	
1	2	3	4	5	6
Double-side laminated particleboard (liquor cabinet from 1970s) *Płyta wiórowa dwustronnie laminowana (barek z lat 70.)*	8.1	34.5	27.0	37.7	40.1
				32.0	
				42.5	
Double-side laminated particleboard – thickness 18 mm (1990s) *Płyta wiórowa laminowana dwustronnie – grubość 18 mm (lata 90.)*	7.0	7.4	6.9	5.5	5.9
				6.2	
				4.5	
Double-side laminated board – thickness 18 mm (particles with an addition of ground laminate); 1970s *Płyta dwustronnie laminowana – grubość 18 mm (wióry z dodatkiem rozdrobnionego laminatu); (lata 70.)*	8.6	22.3	16.0	21.9	23.9
				25.3	
				24.5	
Double-side laminated particleboard – thickness 17 mm (cabinet from 1970s) *Płyta wiórowa dwustronnie laminowana – grubość 17 mm (szafka z lat 70.)*	6.9	12.2	11.5	10.9	11.1
				10.0	
				12.3	
Double-side laminated particleboard – thickness 16 mm (cabinet from 1970s); made from debarked particles *Płyta wiórowa dwustronnie laminowana – grubość 16 mm (szafka z lat 70.) z wiórów bez kory*	7.4	16.4	14.4	17.0	16.0
				14.0	
				12.0	

Table 2. Continued
Tabela 2. Ciąg dalszy

1	2	3	4	5	6
Particle-flax board – thickness 21 mm (80% particles, 20% harl of flax); from 1970s *Płyta wiórowo-paździerzowa – grubość 21 mm (80% wióry; 20% paździerze); (lata 70.)*	7.7	7.3	6.1	5.0 5.6 4.2	5.3
Double-side veneered particleboard – thickness 22 mm; 1980s *Płyta wiórowa dwustronnie oklejana – grubość 22 mm; (lata 80.)*	8.3	17.4	13.2	14.6 10.9 16.7	15.7
Particleboard (wardrobe from the end of 1950s) – thickness 19 mm *Płyta wiórowa (szafa – koniec lat 50.) – grubość 19 mm*	10.4	11.2	-	13.1 8.5 10.4	11.8
Chips from post-consumer wood, from a bunker in a particleboard plant – batch 1 *Zrębki z drewna poużytkowego, z zasobnika w zakładzie płyt wiórowych – partia 1*	9.9	2.3	-	-	-
Chips from post-consumer wood, from production line in a particleboard plant – batch 2 *Zrębki z drewna poużytkowego, z linii produkcyjnej w zakładzie płyt wiórowych – partia 2*	11.7	4.1	-		-
Chips from post-consumer wood, from production line in a particleboard plant – batch 2 *Zrębki z drewna poużytkowego, z linii produkcyjnej w zakładzie płyt wiórowych – partia 2*	7.5	1.6	-		-

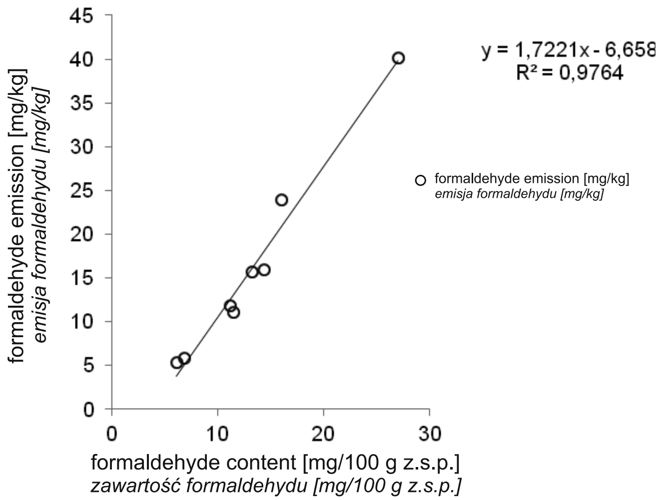

Fig. 1. The interrelation between the formaldehyde content determined by the perforator method and its emission tested by the flask method – based on post-consumer wood in the form of particleboards from furniture elements
Rys. 1. Zależność między zawartością formaldehydu oznaczoną metodą perforatora i jego emisją badaną metodą butelkową – na podstawie drewna poużytkowego w postaci płyt wiórowych z elementów mebli

Table 3 contains the results of the tests for formaldehyde content in the particles randomly sampled in various production plants and prepared in the Institute's laboratory.

The particles sampled at the panel producers (table 3) were tested after various storage periods at the Institute. Hence the formaldehyde content might have changed over a period, i.e. decreased, in relation to the value characteristic of particles directly after drying. This especially concerns the particles from sample 3 and 5, the moisture content of which was much higher than that characteristic of so-called "dry" industrial particles. The content of formaldehyde in the tested particles was within a range of 0.7-2.7 mg/100 g of oven dry particles. Based on the obtained results, one cannot draw conclusions as to the influence of the drying method on formaldehyde content in particles. The suggestions that particles dried in a diaphragm dryer will not contain formaldehyde at all or will contain only trace amounts of it were not confirmed. After a thorough organoleptic assessment of the particles dried in a diaphragm dryer (samples 5 and 6), parts of ground panels were detected in the particles, which explains a relatively high content of formaldehyde. Table 3 also contains the results of the tests for formaldehyde content in particles dried to a constant mass under laboratory conditions. As a result of additional drying, the content of formaldehyde decreased many times. The results of the tests for formaldehyde emission from the particles are given in table 4. The decrease in the formaldehyde emission resulting from additional drying is illustrated in fig. 2.

Table 3. Formaldehyde content in particles
Tabela 3. Zawartość formaldehydu w wiórach

Sample of particles *Próba wiórów*	Particle origin/ /dryer type *Pochodzenie wiórów/ /typ suszarki*	Moisture content of particles *Wilgotność wiórów*	Formaldehyde content *Zawartość formaldehydu*		
			at moisture content when tested *przy wilgotności w chwili badania*	at moisture content after conversion into oven dry particles *przy wilgotności po przeliczeniu na z.s.w.*	at moisture content after drying to constant mass *przy wilgotności po wysuszeniu do stałej masy*
		%	mg/100 g z.s.w./of oven dry particles		
1	Polska/Poland A/1	2.1	1.8	1.8	-
2	Polska/Poland A/1	2.6	2.6	2.7	-
3	Polska/Poland B/1	7.4	0.7	0.8	-
4	Polska/Poland B/1	3.3	0.9	0.9	0.3
5	Niemcy/Germany A/2	5.1	2.2	2.3	0.4
6	Niemcy/Germany A/2	2.7	2.0	2.0	-
7	Polska/Poland A/1	2.7	1.5	1.5	0.6
8	Polska/Poland A/1	2.9	1.2	1.2	0.6
9	Polska/Poland A/1	3.9	1.6	1.7	0.2
10	Pine wood particles cut and dried in ITD/3 *Skrawane i suszone z drewna sosny w ITD/3*	5.3	0.8	0.8	0.2

1 – non-diaghragm/*bezprzeponowa*
2 – diaghragm/*przeponowa*
3 – chamber, eletrically heated/*komorowa ogrzewana elektrycznie*

Table 4. Formaldehyde emission from particles
Tabela 4. Emisja formaldehydu z wiórów

Sample marking acc. to Table 3 *Oznakowanie próby wg tabeli 3*	Moisture content of particles *Wilgotność wiórów*	Number of analysis *Nr analizy*	Formaldehyde emission *Emisja formaldehydu*			
			at moisture content when tested *przy wilgotności w chwili badania*		after drying to constant mass *po wysuszeniu do stałej masy*	
	x		x	x_s	x	x_s
	%		mg/kg			
1	2	3	4	5	6	7
6	7.4	6a	3.4	3.4	0.6	0.4
		6b	3.0		0.3	
		6c	3.7		0.3	

Table 4. Continued
Tabela 4. Ciąg dalszy

1	2	3	4	5	6	7
7	3.4	7a	2.0	2.4	0.5	0.4
		7b	2.6		0.4	
		7c	2.6		0.3	
8	5.1	8a	1.5	1.5	0.1	0.1
		8b	1.4		0.1	
		8c	1.5		0.2	
9	5.3	9a	2.9	2.6	0.8	0.7
		9b	2.4		0.6	
		9c	2.5		0.4	
10	12.6	10a	0.1	0.0	0.0	0.0

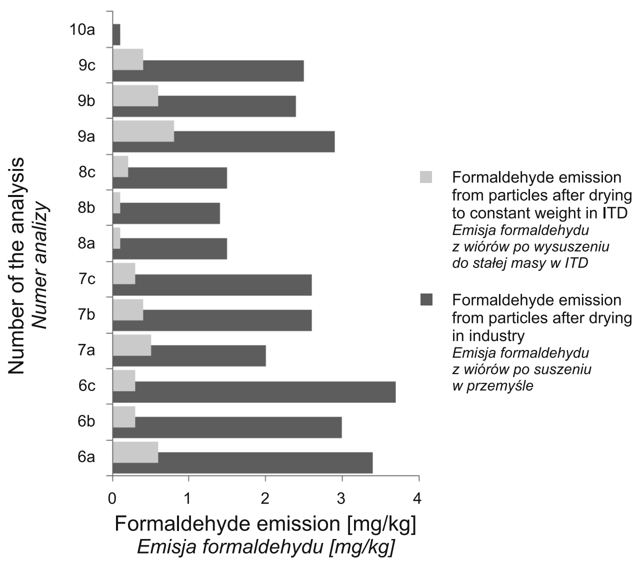

Fig. 2. The influence of the additional drying of particles on the formaldehyde emission
Rys. 2. Wpływ dodatkowego suszenia wiórów przemysłowych na emisję formaldehydu

The interrelation between formaldehyde content in and emission from the particles (fig. 3) was calculated based only on the results of the tests of the particles, which were not additionally dried in the Institute. It should be stressed, that although the method of emission testing, modified for the needs of this research, was used, the obtained correlation of the tested interrelation was very good. The coefficient of determination indicates that the formaldehyde emission was 99% connected with the content of formaldehyde in the particles.

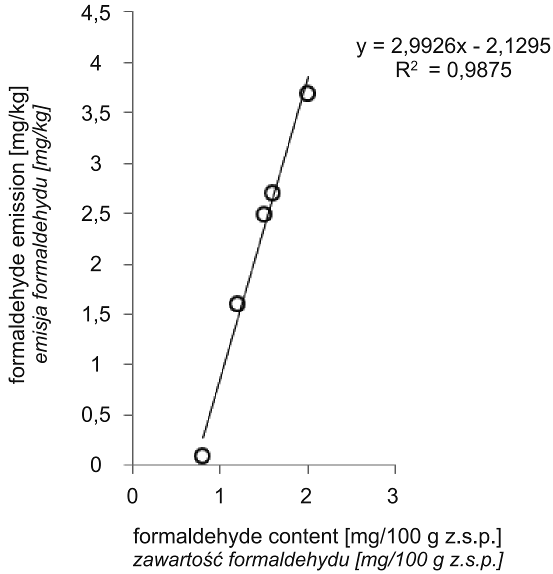

Fig. 3. The interrelation between the formaldehyde content determined by the perforator method and its emission tested by the flask method – based on the particles from industrial plants and obtained in laboratory conditions
Rys. 3. Zależność między zawartością formaldehydu oznaczoną metodą perforatora i jego emisją badaną metodą butelkową – na podstawie wiórów z zakładów przemysłowych i pozyskanych w warunkach laboratoryjnych

Conclusions

The research carried out proved that the amount of formaldehyde contained in natural wood of domestic species, which are commonly used in the production of wood-based panels, reaches a maximum of approximately 1 mg/100g of panel. The range defined, based on, tests of natural wood, also encompasses alternative raw materials (fast-growing willow, locust tree, miscanthus giganteus straw, and grey rape straw, i.e. straw tested after storage). The content of formaldehyde in yellow rape straw was relatively high – 1.9 mg/100g of oven dry particles. The panels from elements of furniture manufactured a few dozen years ago contained a large amount of formaldehyde – even 27 mg/100g of oven dry panels (after conversion to moisture content 6.5%). The particles randomly sampled at various producers were characterised by different formaldehyde content, but higher than in the case of natural wood. Therefore, it seems purposeful to broaden the research on the phenomena connected with the recycling of materials containing formaldehyde in the panel industry. The results of the tests confirmed the usefulness of the flask method for the control of formaldehyde content in particleboards and the possibility of its adaptation for control tests of raw material.

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ZAWARTOŚĆ FORMALDEHYDU W SUROWCACH LIGNOCELULOZOWYCH DO PRODUKCJI PŁYT WIÓROWYCH

Streszczenie

Do produkcji płyt drewnopochodnych, szczególnie wiórowych, stosuje się coraz bardziej zróżnicowany surowiec drzewny. Sytuacja na rynku drewna oraz niezbędna dla zrównoważonego rozwoju minimalizacja odpadów wymuszają materiałowe wykorzystywanie odpadów produkcyjnych i drewna poużytkowego, w tym mebli z recyklingu. Decyzja IARC o zakwalifikowaniu formaldehydu do substancji o rakotwórczym działaniu na ludzi spowodowała intensyfikację badań zmierzających m.in. do poszerzenia wiedzy o źródłach emisji formaldehydu w procesach technologicznych oraz z gotowych produktów. W niniejszym artykule przedstawiono wyniki badań zawartości i emisji formaldehydu z:

– drewna naturalnego – sosny zwyczajnej (*Pinus sylvestris* L.), olszy czarnej (*Alnus glutinosa* Gaertn.), brzozy *(Betula verrucosa Ehrh.)* - powszechnie stosowanego do produkcji płyt wiórowych,
– lignocelulozowych surowców alternatywnych, których przydatność do wytwarzania płyt kompozytowych dla meblarstwa i budownictwa wykazały wyniki wcześniejszych badań wykonanych w Instytucie Technologii Drewna, tj. dwuletnich odrośli robinii akacjowej – *Robinia pseudacacia* L., dwuletnich prętów wierzby szybko rosnącej (*Salix viminals* L.*),* słomy miskanta olbrzymiego *(Miscanthus sinesis),* słomy rzepakowej (*Brasica napus* L.),
– drewna poużytkowego w postaci elementów mebli z płyt wiórowych i zrębków recyklingowych,
– wiórów stosowanych do produkcji płyt w różnych zakładach - w Polsce i w Niemczech.

Zawartość formaldehydu badano metodą perforatora, według PN-EN 120:1994. Emisję formaldehydu oznaczano metodą butelkową według PN-EN 717-3:1999. Metodę butelkową zmodyfikowano w celu jej adaptacji do badania wiórów. Spośród badanych gatunków drewna największą zawartością i emisją formaldehydu charakteryzowało się drewno sosny z części bielastej. Uwagę zwraca również stosunkowo duża zawartość formaldehydu w korze brzozy i w słomie rzepakowej żółtej, tj. świeżej, badanej przed składowaniem. Najwyższą (27 mg/100 g z.s.p.), ale i najniższą (6,1 mg/100 g z.s.p.) zawartość formaldehydu oznaczono w płytach wyprodukowanych w latach siedemdziesiątych ubiegłego wieku. Próbki płyt pochodziły z mebli, których warunki użytkowania były znane, oraz z materiałów przesłanych do badań w Instytucie Można więc wykluczyć wpływ warunków użytkowania czy przechowywania na zawartość formaldehydu w płytach. Poziom zawartości formaldehydu wydaje się związany raczej z technologią niż okresem jaki upłynął od wyprodukowania płyt. Różną, od 0,7 – 2,7 mg/100 g. z. s., zawartością formaldehydu charakteryzowały się wióry pobrane losowo u różnych producentów. Wartość perforatora nie była związana z typem suszarki (przeponowa i bezprzeponowa). Wpływ na nią miały przypuszczalnie cząstki rozdrobnionych płyt zawarte w wiórach. Wióry pobrane u producentów płyt badane były po różnym czasie przechowywania. Zawartość formaldehydu mogła więc w tym okresie ulec zmianie – tzn. zmniejszeniu, w stosunku do wartości charakteryzującej wióry bezpośrednio po suszeniu.

Celowe wydaje się, w związku z powyższym, poszerzenie badań zjawisk związanych z recyklingiem materiałów zawierających formaldehyd w przemyśle płytowym poprzez okresową kontrolę zawartości formaldehydu w stosowanych surowcach. Badania wykazały możliwość adaptacji do tych celów butelkowej metody oznaczania emisji.

Słowa kluczowe: zawartość formaldehydu, surowiec drzewny: konwencjonalny, alternatywny, z recyklingu