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DANGEROUS PHTHALATES IN CHILDREN'S ENVIRONMENT

NIEBEZPIECZNE FTALANY W ŚRODOWISKU ŻYCIA DZIECI

Abstract: There are many chemicals in children's environment that are dangerous to their health. Some of them are eg phthalates used as plasticizers in plastic toys and in production of other articles intended for use by children. Phthalates do not form persistent connections with the polymer and migrate to the surface of the product. Children exposure to these toxic chemicals has been found to occur during licking and sucking of the product taken to the mouth by a child and during long term contact with skin. Results of tests aimed at determination of phthalates content in toys and childcare articles made in the years 2009-2011 in accredited Laboratory of Material Engineering and Environment at KOMAG according to the requirements of REACH regulation, which include limitations as regards use of the following dangerous phthalates: DEHP, DBP, BBP, DINP, DIDP and DNOP, are discussed in the paper.

Keywords: phthalates, children articles, toys, childcare articles, environment

Children are exposed to many dangerous chemical substances. Industrial activity, road transportation [1, 2], food containing preservative additives and residual pesticides [3-5] as well as consumer products made of materials containing toxic chemicals eg esters of *benzene*-1,2-*dihydrocarboxilic* acid known as phthalates [6], are the source of those chemicals. Phthalates are used in personal hygiene articles, paints, medicals and pharmaceuticals as well as in plastics used to produce floor covering, household goods, food packaging, foils and toys [7-9].

Phthalates are the plasticizers, which make polymer material eg *polyvinyl chloride* (PVC) elastic to make production of the final product easier. The following chemicals belong to phthalates: *di-2-ethyl hexyl phthalate* (DEHP), *dibutyl phthalate* (DBP), *benzylbutyl phthalate* (BBP), *diisononyl phthalate* (DINP), *diisodecyl phthalate* (DIDP) and *di-n-octyl phthalate* (DNOP) [10, 11]. DEHP and DINP phthalates are most commonly used in plastics for children due to their properties and low production costs [12-15].

Phthalates do not form covalent bonds with the polymers with which they are mixed, so they can migrate to products surface and then they can be released to the environment [16].

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Broad range of applications as well as easiness of release to the environment cause that phthalates are one of most common pollutants in rooms, where children stay. Presence of phthalates in house air and dust has been confirmed by numerous research projects [17-19].

Phthalates penetrate children organisms through digestive system and through contact with skin. Inhalation of phthalates through children respiratory system is minor source of exposure. Phthalates can be consumed together with food and swallowed dust as well as in the result of licking and sucking of objects made of plastics eg toys [20-24]. Dermal exposure is also an important way of phthalates penetration [25].

Phthalates have especially negative impact on growth and development of human fetus, infants and toddlers. They cause dysfunction of hormonal system by disturbing synthesis, concentration and action of natural hormones responsible for growth and development of sex organs, especially male genitals, and even can cause elimination of hormones [26]. Phthalates have also negative impact on children immune system causing increase of asthma and allergy cases [27].

To be cautious the European Union has introduced limitation in use of some phthalates in toys and childcare articles, including those intended for sleep facilitation, relaxation, hygiene, feeding or sucking [28]. According to the European Commission Regulation (EC) No. 1907/2006 (REACH) such phthalates like DEHP, DBP, BBP, DINP, DIDP and DNOP cannot be used in toys and childcare articles, which can be put into the mouth in a concentration higher than 0.1% in relation to the weight of the material with plasticizer. As regards other toys and childcare articles the above-mentioned limitation concerns only the following phthalates: DEHP, DBP and BBP [29].

Despite introduced restrictions, the products containing dangerous phthalates in concentration higher than accepted, intended for children, are still sold. That fact is documented in published information [30] as well as in the reports issued by Trade Inspection and by RAPEX - *Rapid Alert System for non-food dangerous products* [31, 32]. Identified cases concern exceeded content of DEHP and DINP phthalates in toys.

The published data do not provide, except a few cases, namely: soothers, feeding bottles, and cosmetics for children [33], information on the presence of dangerous phthalates in childcare articles, especially in equipment for relaxation and health, toiletries and accessories for drinking and feeding, with concentrations exceeding the limit value. Considering the fact that the above products are made of thermoplastics, including PVC, like in toys, one would expect that also in this case toxic phthalates may be present.

In the Laboratory of Material Engineering and Environment at the KOMAG Institute of Mining Technology the research project aiming at determination of phthalates contents in toys and childcare articles as well as at assessment if they meet the requirements of REACH (**R**-Registration, **E**-Evalution, **A**-Authorisation of **C**h-Chemicals) regulation was undertaken to identify the problem and to complete information in that range.

The project was realized within research task entitled: “Methodology for shaping safe environment and safe children’s life” [34]. The results of tests on toys safety, carried out in the KOMAG Laboratory within years 2009-2011, were the project merits. The results of the research project are presented in the paper.

Testing material and methodology

Determination of content of the following phthalates: DEHP, DBP, BBP, DINP, DIDP and DNOP in toys and childcare articles as well as their assessment as regards conformity with the requirements of REACH regulation were the project objective.

In total 228 of products for children were tested, including 172 toys intended for children under 36 months as well as 56 childcare articles and materials for their manufacture.

The following types of toys were tested:

- for bathing (rubber ducks and inflated toys),
- activating toys for domestic use (swings),
- to play in sand (plastic car, sand kits: bucket, shovels, rakes, molds, sandbox),
- thematic (kitchen set with plates and cups),
- on which a child can sit (rockers, vehicles),
- a child can enter (houses),
- presenting different scenes and ready models (farmyard, set vehicles),
- equipment for sports (skittles, balls),
- dolls,
- trumpets and balloons,
- simple plays and puzzles.

Childcare articles and materials for their manufacture were divided into the following groups depending on their use:

- articles for relaxation and hygiene (poufs, chairs, foils used for manufacture of mattresses in beds, strollers and for change of baby),
- articles of toilet (brushes, cups, soap dishes, containers for toothbrushes, vanity bags),
- feeding and drinking accessories (cups, cutlery, bottles, plates).

Share of each type of tested products is given in Figure 1.

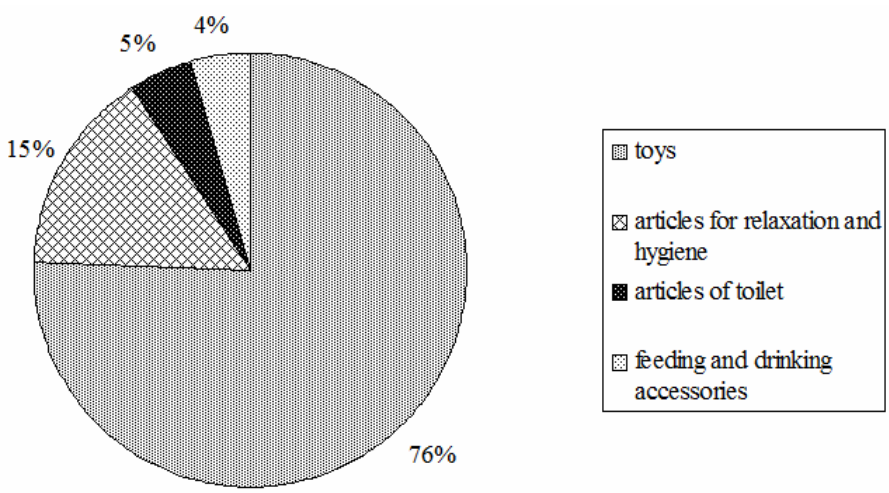

Fig. 1. Share of each type of tested products [own source]

Tested samples of products or materials used for manufacture of those products were made of different types of plastics like: *polyvinyl chloride* (PVC), *polypropylene* (PP), *polyethylene* (PE), *polyethylene of high and low density* (HDPE and LDPE), *polystyrene* (PS), *poly(acrylonitrile-co-butadiene-co-styrene)* (ABS), *poliethersulfone* (PES), *phenol-formaldehyde resin* (PF), *thermoplastic elastomer* (TPE) and rubber. Among the tested samples there were also materials, which were not identified due to lack of enough information and improper marking on the product. List of materials of tested samples of products for children are shown in Table 1.

Table 1

List of materials of tested samples of products for children [34]

Item	Type of material	Number of tested products from a given type of material			
		Toys	Articles for relaxation and hygiene	Articles of toilet	Feeding and drinking accessories
1.	PVC	22	20	-	-
2.	PP	23	1	5	10
3.	PE	9	-	-	-
4.	HDPE	27	-	-	-
5.	LDPE	17	-	-	-
6.	PS	2	1	5	
7.	ABS	4	-	-	-
8.	PES	1	-	-	-
9.	PF	1	-	-	-
10.	TPE	1	-	-	-
11.	PA	10	-	-	-
12.	rubber	4	-	1	-
13.	not identified	43	21	-	-

Samples of materials for determination of phthalate content were taken from toys and childcare articles or raw materials used for their manufacture by cutting out, ensuring full quantitative and qualitative compatibility of their composition to the composition of analyzed product. The taken samples were then fragmented or ground using a cryogenic mill. Two parallel samples were taken from that fragmented material and they were subjected to extraction process by the Soxhlet method. Extraction was carried out for 16 hours using dichloromethane. Two analytic samples for chromatographic analysis were taken from the obtained extract. Analytic measurements of phthalates content in the above-mentioned samples were taken on *gas chromatograph-mass spectrometer* (GC-MS). Before measurements gas chromatograph was calibrated using reference solutions of DBP, DEHP, DNOP, BBP, DIDP and DIBP phthalates. Identification of chemical compounds was made on the basis of mass spectra and retention time, and quantitative analysis was made using proper analytic algorithm. The obtained results in a form of phthalate concentration in the extract given in [$\mu g/dm^3$] were converted into phthalate mass content [mg] in the analytic sample, and then into percentage content of phthalate in relation to the material of tested product [% by weight].

General algorithm for determination of phthalate content in toys and childcare articles is shown in Figure 2.

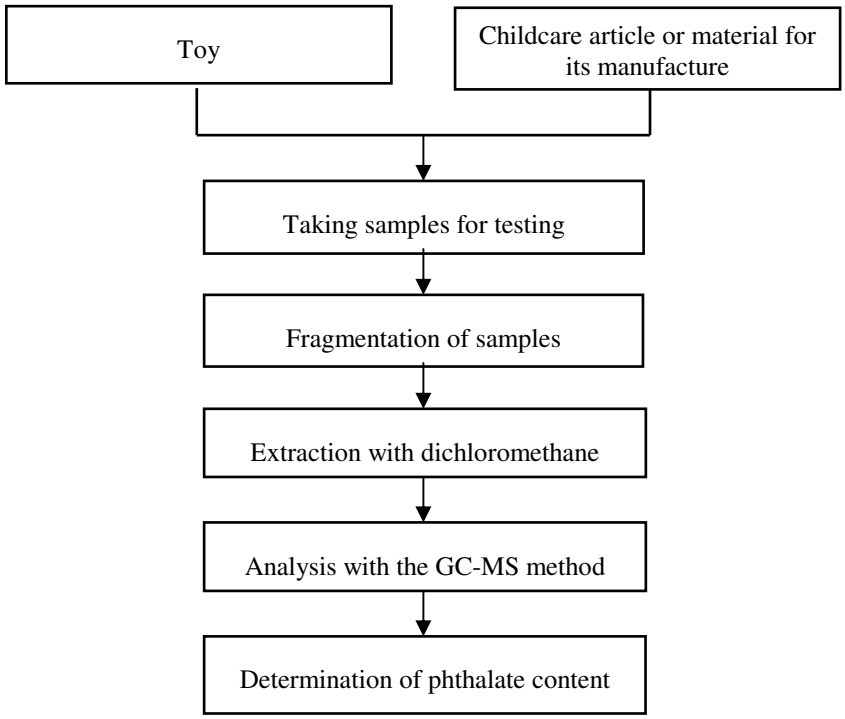

Fig. 2. Algorithm for determination of phthalate content in toys and childcare articles [own source]

Results and discussion

Analysis of determination of phthalates content in toys and childcare articles has proved presence of toxic chemical compounds in about 10% of tested samples, including: di-2-ethylhexylphthalate (DEHP), diisononylphthalate (DINP) (Fig. 3) and di-*n*-octylphthalate (DNOP).

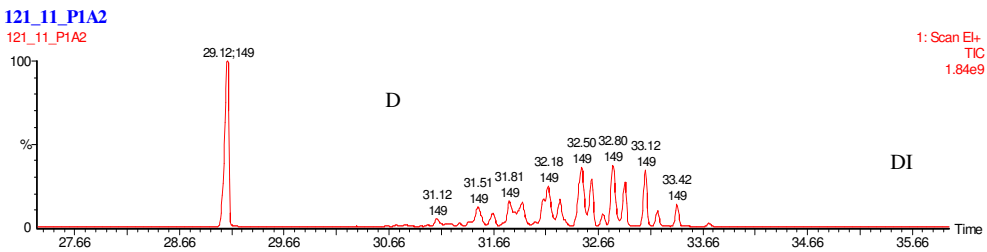

Fig. 3. GC-MS chromatogram showing the presence of DEHP and DINP phthalates in samples of PVC foil used for manufacture of mattresses for change of baby [35]

Concentration of the above-mentioned phthalates exceeded accepted value, which is 0.1%, as specified in REACH regulation. Results of analytic measurements in the rest of samples did not show phthalates presence or their concentration was below 0.05%.

Definitely greater number of exceedances of the limit content of phthalates was found in samples of childcare articles.

Shares of phthalate concentration exceedances cases in each group of tested products for children are presented in Figure 4.

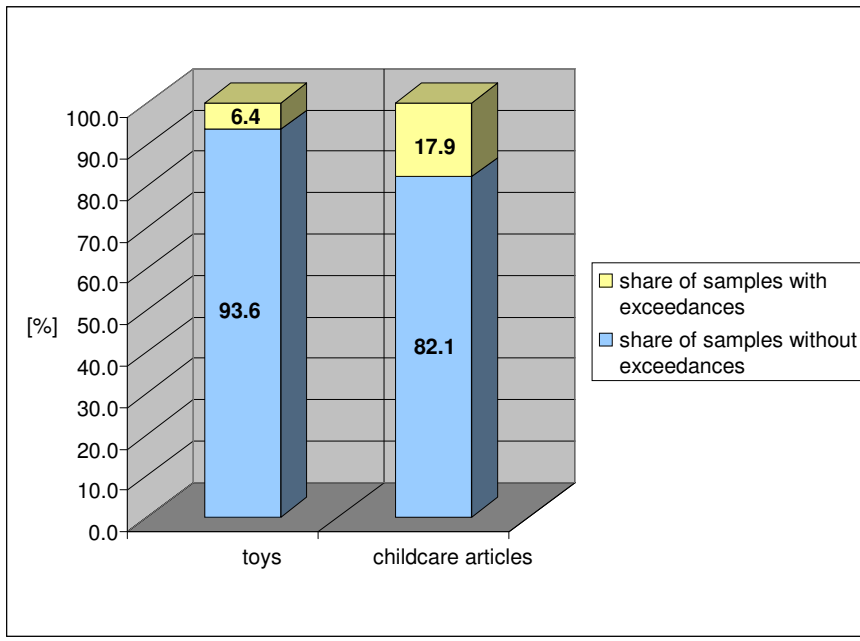

Fig. 4. Shares of exceedances of acceptable phthalate content in samples of toys and childcare articles [own source]

Polyvinyl chloride (PVC) and plastics, which were not able to be identified due to lack of any information on the product, were the materials, in which exceedances of acceptable phthalate content were reported in all tested samples.

The highest phthalate content was reported in samples of the following toys:

- inflating toys, including valves,
- toys packaging, which is a part of the toy (bag for toy carrying),
- balls,
- vehicles.

Detailed list of results of phthalates determination in toy samples, in which exceedances of acceptable values were reported are given in Table 2.

In the group of childcare articles, exceedances of permissible phthalate content were reported first of all in the samples of articles for relaxation and hygiene as well as in the samples of material used for their manufacture.

The following tested samples had the highest phthalate content:

- poufs and chairs for children,
- foils for manufacture of mattresses for children beds, strollers and for change of babies.

Detailed list of results of phthalates determination in toy samples, in which exceeding of acceptable values are given in Table 2.

Table 2

List of results from tests, which indicate exceeding of acceptable values of phthalates content in toy samples [34]

Item	Type of material	Phthalates content in toys material											
		DEHP [% by weight]		DBP [% by weight]		BBP [% by weight]		DINP [% by weight]		DIDP [% by weight]		DNOP [% by weight]	
		$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */
1	PVC	<0.05	-	<0.05	-	<0.05	-	**0.70**	**0.05**	<0.05	-	<0.05	-
2	PVC	<0.05	-	<0.05	-	<0.05	-	**1.08**	**0.03**	<0.05	-	<0.05	-
3	n.i. **/	<0.05	-	<0.05	-	<0.05	-	**4.13**	**0.07**	<0.05	-	<0.05	-
4	n.i. **/	**0.18**	**0.02**	<0.05	-	<0.05	-	**1.62**	**0.10**	<0.05	-	<0.05	-
5	n.i. **/	**14.63**	**0.14**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
6	n.i. **/	**0.37**	**0.12**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
7	n.i. **/	**14.8**	**0.8**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
8	n.i. **/	**12.6**	**0.8**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
9	PVC	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05		**2.02**	**0.14**
10	n.i. **/	**>20**	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
11	PVC	**0.17**	**0.02**	<0.05	-	<0.05	-	**7.67**	**0.17**	<0.05	-	<0.05	-

*/ u is extended uncertainty at confidence level 95% and extension coefficient k = 2
**/ n.i.- not identified

Table 3

List of results from tests, which indicate exceeding of acceptable values of phthalates content in materials of articles for relaxation and hygiene [34]

Item	Type of material	Phthalates content in the materials of articles for relaxation and hygiene with plasticizer											
		DEHP [% by weight]		DBP [% by weight]		BBP [% by weight]		DINP [% by weight]		DIDP [% by weight]		DNOP [% by weight]	
		$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */	$x_{śr}$	u */
1	PVC	**2.98**	**0.39**	<0.05	-	<0.05	-	<0.05	-	<0.05		<0.05	-
2	PVC	**5.14**	**0.68**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
3	PVC	**0.19**	**0.03**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
4	n.i. **/	**9.10**	**1.18**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
5	n.i. **/	**8.96**	**1.23**	<0.05	-	<0.05	-	**1.46**	**0.11**	<0.05	-	<0.05	-
6	PCV	**0.49**	**0.14**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
7	PCV	**0.15**	**0.02**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
8	PCV	**4.46**	**0.61**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
9	PVC	**3.52**	**0.04**	<0.05	-	<0.05	-	<0.05	-	<0.05	-	<0.05	-
10	PVC	<0.05	-	<0.05	-	<0.05	-	<0.05	**-**	<0.05	-	**1.48**	**0.2**

*/ u is extended uncertainty at confidence level 95% and extension coefficient k = 2
**/ n.i.- not identified

Conclusions

Plastics used for manufacture of toys and childcare articles contain toxic phthalates, which act as plasticizers that make structure of the material, eg PVC, elastic. Phthalates, due to their chemical properties - lack of covalent bond with the plastic, migrate to the surface of the product's material and are the source of hazard for children. Phthalates that are on the

surface can be consumed by children in a result of sucking and licking as well as they can penetrate children bodies in a result of direct contact with skin.

The tests proved that some of the dangerous phthalates are still in use in products for small children not only in toys, but also in childcare articles.

The tests identified exceedances of permissible content of the following phthalates: di-2-ethylhexylphthalate (DEHP), diisononylphthalate (DINP), di-n-octylphthalate (DNOP). They were present especially in the samples of toys intended for children under 36 months, which can be taken to mouth as well as in the samples of articles for relaxation and hygiene made of PVC and the plastic, information of which was not placed on the product.

The tests carried out so far indicated for a need of further tests for the specified group of materials to verify their conformity with the requirements of REACH declaration.

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NIEBEZPIECZNE FTALANY W ŚRODOWISKU ŻYCIA DZIECI

Abstrakt: W środowisku dziecka występuje wiele niebezpiecznych dla jego zdrowia substancji chemicznych. Jednymi z nich są ftalany, stosowane jako plastyfikatory tworzyw sztucznych w produkcji zabawek i innych wyrobów przeznaczonych do stosowania przez dzieci. Ftalany nie tworzą trwałych połączeń z polimerami i migrują na powierzchnię produktu. Narażenie dzieci na ekspozycję tych toksycznych związków chemicznych występuje przede wszystkim w trakcie lizania i ssania produktu wkładanego do ust przez dziecko oraz podczas długotrwałego kontakt z jego skórą. W publikacji przedstawiono wyniki badań zawartości ftalanów w zabawkach i artykułach pielęgnacyjnych przeznaczonych dla dzieci przeprowadzone w latach 2009-2011 w akredytowanym Laboratorium Inżynierii Materiałowej i Środowiska ITG KOMAG, zgodnie z wymaganiami rozporządzenia REACH, wprowadzającego ograniczenia w stosowaniu niebezpiecznych ftalanów: DEHP, DBP, BBP, DINP, DIDP i DNOP.

Słowa kluczowe: ftalany, produkty dla dzieci, zabawki, artykuły pielęgnacyjne dla dzieci, środowisko