End-of-life treatment of HBCD-containing polystyrene insulation foams

Large-scale demonstration of the treatment of Expanded Polystyrene Foam (EPS) and Extruded Polystyrene Foam (XPS) containing Hexabromocyclododecane (HBCD) as a flame-retardant by co-incineration in the Würzburg Municipal Solid Waste Incinerator

Technical Summary Report

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The Waste Association Würzburg Area (ZVAWS) was founded in 1979 as a public corporation of the city of Würzburg, the district of Würzburg, and the district of Kitzingen. It manages and operates the MHKW waste-to-energy facility in Würzburg.

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Summary report

Introduction

Polystyrene insulation foams are durable materials, designed to offer superior, constant insulation performance over their entire service life of more than 50 years. The use of polystyrene foam reduces significantly heat losses and related CO_2 emissions from heating and/or air conditioning, thus contributing strongly to mitigation of the effects of climate change by improving energy efficiency.

HBCD, the flame retardant most used so far for polystyrene insulation boards, has recently been classified by the EU authorities as a substance subject to Authorisation under REACH; it is also being listed as a POP (Persistent Organic Pollutant) under the UNEP Stockholm Convention.

Determining and implementing safe end-of-life treatment options for polystyrene foam boards containing HBCD is essential to meet these possible regulatory requirements; a demonstration trial was carried out in 2013 to evaluate the co-incineration of these foams with municipal solid waste to assess if it could be a suitable option of choice in this respect.

This document provides a technical overview of this demonstration trial and of its results.

Rationale and Objectives of the trial

The high lifetime value of insulation foam for energy savings has been widely recognised and accepted by legislators, consumers and society at large. Organic polystyrene foams (PSF) have a large market share based on performance and cost efficiency. Due to national fire regulations in Europe a large proportion of PSF has been and is currently manufactured with flame-retardant properties. The chemical of choice for foam suppliers for decades is Hexabromocyclododecane (HBCD) because of its long-term proven performance. In 2008 HBCD was classified as PBT (persistent, bioaccumulative and toxic) under the REACH Regulation, and it is currently subject to a REACH Authorisation process. It was also recently classified as a POP (persistent organic pollutant) under the UNEP Stockholm Convention.

Industry is of the opinion that there should be a non-discriminatory approach to the End of Life (EoL) scenarios for PS foams, in which the preferred sequence of options for optimal eco-efficiency and risk reduction would be incineration with energy recovery, recycling and landfill.

Indeed, the co-incineration of PSF from building and construction (B&C) markets together with municipal solid waste is an eco-efficient and practical option for the safe treatment of such waste, along with the recovery of energy. A controlled one-week co-incineration trial was conducted by a broad consortium of stakeholders to evaluate the effects of PSF containing HBCD on the performance of the large-scale energy recovery incinerator in Würzburg, Germany (MHKW).

The demonstration trial was completed within the time-scale and objectives set by the consortium partners: MHKW/Würzburger Versorgungs- und Verkehrs GmbH (WVV), Zweckverband Abfallwirtschaft Raum Würzburg (ZVAWS), MARTIN GmbH, Walhalla Kalk, Regensburg, and PlasticsEurope & EXIBA. Representatives of LfU, the Bavarian Environmental Protection Agency and local governmental authorities visited the MHKW during the test.

Key objectives of this trial demonstration were:

1. to prove high combustion efficiency and energy recovery of PS foams with municipal solid waste (MSW);
2. to meet emission limits at high PSF feed rates with existing flue gas treatment;
3. to show any influence of high PS foam rates on operations;
4. to prove a high destruction efficiency of HBCD.

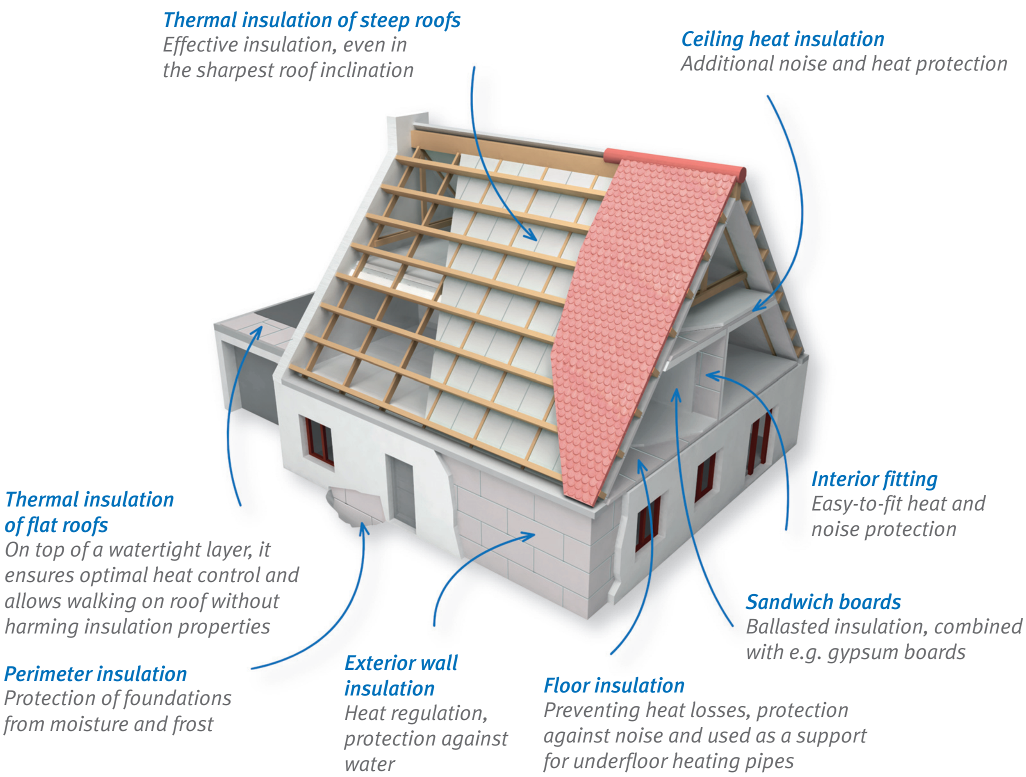
PS Foam Insulation

Polystyrene foam is used for a wide range of insulation applications, in the residential, commercial, institutional and industrial building sectors as well as for civil engineering. From roof to floors to walls, from cavity fill to perimeter insulation and anti-frost layers, polystyrene foam provides versatile insulation solutions, adapted to every situation. The largest application is thermal insulation to prevent heat transfer. Buildings last longer and have less maintenance requirements because of the durability and moisture resistance of PS foam.

Due to its insulation performance, light weight, rigidity and flexible shape design, PS foam reduces space requirements for walls and roofs, and hence maximises internal volume. This is especially important when existing buildings are being renovated to meet improved insulation standards.

The two types investigated in this trial demonstration are Extruded and Expanded Polystyrene, abbreviated XPS and EPS respectively. Almost all XPS and EPS insulation boards for the building and construction markets, where fire-retardant properties are required, contain the flame-retardant HBCD, which has a very high Br content of 74.7 wt%. The actual level of the HBCD content depends very much on the application and the country. Different flame-retardant testing methods and requirements exist in Europe and hence various HBCD levels exist in the market. It is estimated that 77 % of EPS and 94 % of XPS in construction markets is flame-retarded.

Figures 1a and 1b: Examples of uses of PS foam in insulation: XPS and EPS.



PS Foam Characteristics

The PSF supplied by manufacturers had typical commercial quality and was delivered in bulk with the dimensional properties: EPS 1000/500/100 mm and XPS 1265/615/50-80 mm with board weights 0.75, 1.5 and 2.2 kg for EPS, XPS1 and XPS2. The board dimensional characteristics represent what comes back from end of life as total boards. Smaller size boards or broken up pieces are typical for job site waste or demolition waste. The densities of EPS, XPS1 and XPS2 were 15, 38.5 and 35.5 kg/m^3 respectively.

The total concentrations of regulated heavy metals found according to the EU Waste Incineration Directive (WID) are rather low and range between 100 and 200 mg/kg, which can be compared to average concentrations in other plastics waste streams found in municipal solid waste (MSW). The amount of other hetero atoms such as the halogens Cl, Br and F and sulfur were measured to understand the mass flux of acid gases released into the raw gas, that require neutralisation and are subject to emission limitations under EU legislation.

Up until the mid-1990s, XPS foam was made using blowing agents that have since been classified as ozone depleting substances (ODS). In 1993/94 the safe destruction of ODS gases in XPS was studied and demonstrated in the Tamara pilot plant. At that time the environmental behaviour of HBCD was not directly part of the co-incineration tests. The high degree of destruction efficiency for ODS compounds such as CFCs, HCFCs or HFCs was shown. None of the PSF types used nowadays contain such ODS gases. The blowing agents used for the foams tested during this trial were respectively pentane for EPS, and CO_2 without halogen co-blowing agent for XPS.

Sample	Summary of HBCD and hetero atom concentrations in PSF				
	Br, wt%	Cl, wt%	F, wt%	S, wt%	HBCD data from manufacturer, wt%
EPS	0.41	0.046	<0.005	0.005	0.7
XPS1	0.74	0.025	<0.005	<0.005	1.3
XPS2	1.53	<0.005	<0.005	0.032	2.4

Table 1: PSF Chemical Parameters

PS Foam Handling and MSW Mixing

The type of in-line mixing of loose and non-compacted MSW with PSF as conducted for this trial does not represent a real-life operation. This specific procedure was selected to ensure a precise input of PSF with a known HBCD content. Such special dosing ensures that the exact amount of HBCD during the period of sampling and measurement is known. A high degree of confidence in the destruction efficiency of HBCD can therefore be expected. PSF/MSW blending provides a measure of the appropriate mixing of MSW and PSF. No special care was taken to distribute the PSF boards on the MSW. The typical loading time for the process conditions chosen allowed the feeding and dosing procedure to be maintained through all test conditions.

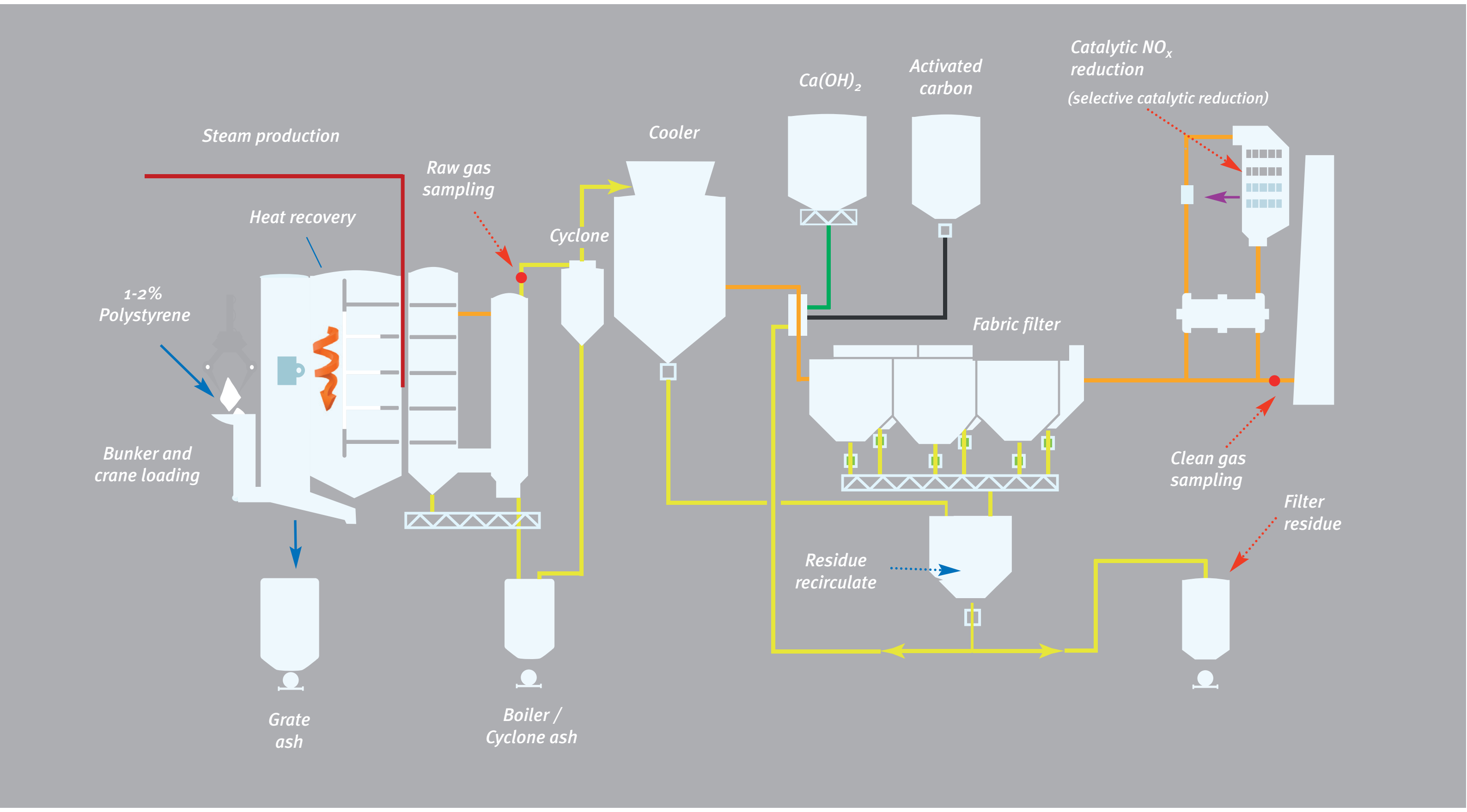
Co-Combustion Tests of PS Foam and MSW

The specific incinerator plant site was selected for the same reasons as it was for the plastics packaging co-combustion test in 1993/94, and the ASR and ESR tests in 1997 and 2004 respectively. The MHKW (Müllheizkraftwerk Würzburg) has a cost-efficient operation, a reliable dry scrubbing system, a long residence furnace time leading to good burnout in the gas phase, a proven grate boiler design leading to excellent residue characteristics, and well documented emissions.

MHKW Incinerator Facility Description and Operations

The MSW incinerator design and operation has been extensively described in public literature (www.zvaws.de).

The following chart provides an overview of the key processes involved.



Test Programme and Conditions

The programme consisted of a one-week testing period in which the test conditions were coded as follows:

A	Base case tests – without PSF	A1, A2
B	Medium level of 1 wt% PSF addition	B1, B2
C	Higher level of 2 wt% PSF addition	C1, C2
A	Base case tests – without PSF	A3, A4

The operation of the plant was maintained as constantly as possible and conditions were kept close to normal operation. The plant process control computer was operated fully automatically during the PSF tests without modifications or special process setting.

Solid residue sampling was carried out on the following streams: grate ash, combined fly ash and fabric filter ash. Clean and raw gas sampling was carried out by an external contractor.

Operation: Online Analysis

The MHKW incinerator is operated by a fully automated process control system. A data logger was used to document online data at different interval sizes: raw data, averaged for 10, 300 and 900 seconds. The plant online operating data, averaged for respectively six and four hours of sampling, are shown in Table 2.

Raw gas HBr concentrations for base conditions stayed at a very low level of 5 mg/m³. Raw gas concentrations during PSF feed for the key acid gases stayed in the expected ranges up to 12 mg/m³ for HBr, 220 to 1145 mg/m³ for HCl, and 60 to 220 mg/m³ for SO_2. NO_x values from 300 to 350 mg/m³ in the raw gas and CO levels of 7 to 13 mg/m³ were proof of efficient and rather stable combustion.

Furnace/Boiler	A1/2	B1/2	C1/2	A3/4	Comments
Steam actual, t/h	27.3	26.9	26.5	27.8	95.2 % max
O_2, mean furnace, vol%	7.8	7.45	7.95	7,65	Set point 8.09 vol%
Primary air, Nm³/h	26552	25829	25931	25483	66.5 %
Secondary air, Nm³/h	13632	12892	12875	12819	33.5 %
Mean furnace temperature, °C	912/924	913/912	900/903	933/932	900-932 °C at furnace roof

Table 2: MHKW online operating parameters

Raw Gas Conditions: Dioxins/Furans

The raw gas concentration of PCDD/Fs ranged in a very narrow span between 1.8 and 3.3 ITEQ ng/m³. This very small difference between high and low concentrations confirms the stable combustion conditions, which were achieved through good MSW mixing, the PSF feeding and controlled automated process operation. It demonstrates that good combustion control with high combustion efficiency was achieved. The formation of mixed halogen PBCDD/Fs occurs before similar low halogenated congeners of PBDD/Fs types are formed. The PBDD/F concentration was, as expected from earlier studies, very low and no single congener was detected.

Raw gas PCDD/Fs	A1	B1	C1
ITEQ ng/m³ excl. LOQ	1.84	3.05	3.31
ITEQ ng/m³ incl. LOQ	1.65	2.67	3.01
Raw gas: PBCDD/Fs			
ng/m³ excl. LOQ	11.6	59.4	110.8
ng/m³ incl. LOQ	< 15.8	< 63.1	114.2
Raw gas: PBDD/Fs			
ng/m³ excl. LOQ	n.d.	n.d.	n.d.
ng/m³ incl. LOQ	0.02-0.5*	0.02-0.5*	0.02-0.5*

Note: n.d. = not detected
* No single congener was detected, LOQ of single congeners is in the given range as reported by GfA
Table 3: Dioxins/Furans concentrations in the raw gas in ITEQ or ng/m³ at 11 vol% O_2

Efficient Raw Gas Cleaning by Emission Control

The emission control performed well, confirmed by the clean gas data comparison with the emission limit values (ELV) in Table 4. Heavy metal concentrations also remained far below the ELV. The addition of lime and active carbon (AC) was controlled by a number of lead online measured parameters. Acid gas levels of HCl, SO_2 and NO_x were drastically reduced to a maximum of 9 mg/m³ for HCl and SO_2 and 75 mg/m³ for NO_x. There was no increase in the use of the lime-based neutralisation agent. No effect of PSF addition on the combustion can be seen on the basis of the clean gas analytical results.

Component	A1* and A3 * (mean value for 6 hrs)	B1* and C1 * (mean value for 6 hrs)	Min/max daily (month March)	Emission Limit Value (Daily)
CO, mg/m³	14.6	18.3	5.7/15.5	50
C, total mg/m³	< 0.1	< 0.1	0.01/0.03	10
HCl, mg/m³	7.3	5.1	4.8/7.3	10
Hg, µg/m³	< 0.01	< 0.01	0.0001/0.0004	0.03
NO_x, mg/m³	62.5	76	58/82	200
SO_2, mg/m³	6.7	1.7	0.16/13.8	50
Dust, mg/m³	0.13	1.6	0.01/0.9	10

Note: * max ½ hr mean value
Table 4: Emission data and the respective ELV

HBCD destruction

When calculating the destruction efficiency of HBCD, all mass flows throughout the incineration plant as well as the HBCD concentrations in all of these streams have to be known as precisely as possible. The input streams are MSW, PSF, and combustion air. The MSW feed has been recorded by the crane load scale, the respective PSF amounts have been taken according to a prepared input list, with accuracies expected to be ± 5 %. The combustion air has been in the order of 40.000 $m^3 h^{-1}$.

The mass flows of grate ash, boiler ash, as well as of APC residue represent average values with an estimated standard deviation of ± 20 %.

For the analysis of the HBCD concentrations in the residue streams, investigations concerning the analytical LOQ in the various matrices were conducted. The respective figures were 1 ng m^{-3} for the flue gas and 1,4 µg kg^{-1} for the solid residues. The analytical error is estimated to be around ± 10 %.
With these data at hand, the HBCD substance flows have been calculated. The results are compiled in Table 5.

	Unit	A1	B1	C1
EPS	gh^{-1}	0	366 ± 46	671 ± 84
XPS1	gh^{-1}	0	333 ± 48	610.5 ± 88
XPS2	gh^{-1}	0	648 ± 63	1,188 ± 115
Accumulated inputs	gh^{-1}	0	1,347 ± 96	2,470 ± 145
Grate ash	mgh^{-1}	3.08 ± 0.70	3.18 ± 0.75	5.94 ± 1.32
Boiler + cooler ash	mgh^{-1}	0.44 ± 0.10	0.82 ± 0.18	0.55 ± 0.11
APC residue	mgh^{-1}	0.18 ± 0.04	0.18 ± 0.04	0.20 ± 0.04
Flue gas	mgh^{-1}	0.031 ± 0.006	0.18 ± 0.04	0.36 ± 0.07
Accumulated outputs	mgh^{-1}	3.80 ± 0.70	4.36 ± 0.82	7.04 ± 1.33
Destruction effiency	%	–	99.999	99.999

APC = air pollution; EPS: expanded polystyrene; XPS: extruded polystyrene

Table 5: Substance flows of HBCD and its destruction efficiency

From the above data, practically identical destruction efficiencies (DE) of ›99.999 % for the B1 as well as for the C1 tests have been calculated. The findings provide evidence for a virtually complete destruction of HBCD.

Energy Balance/Boiler Efficiency

The energy balance for the MHKW resulted in an efficiency of 74 to 75 %. The differences in ranges of the MSW lower heating value (Hu) depend on the waste collection and separation system. In previous tests Hu ranged from 10 to 13 MJ/kg. These changes of total Hu feed value are due to the normal variations of heat values experienced with MSW of different composition delivered during the test.

The small amount of PSF (1 to 2 wt%) added to the MSW does not significantly raise the MSW Hu value (max 6 %). The EPS and XPS range between 39 and 38 MJ/kg for the Hu. This overall boiler efficiency of the MHKW rated the MSWI as an energy recovery plant according to the WFD 2008/98/EC. The amount of steam and electricity produced over the year has been published. 25 % of this energy is used for internal purposes and 75 % is supplied to the electricity and heating grid of the city of Würzburg. This energy amount represents 14 and 16 % of the total electricity and heat produced by WVV GmbH.

Summary of Analytical Emission Results

Clean Gas Emissions: Dioxins/Furans

It is important to note that all clean gas results are far below the emission limit value of 0.1 ng/m³ expressed in International Toxic Equivalents (ITEQ). The average clean gas data from Line No. 2, which was used in all earlier full-scale trial demonstrations, fall very much in the range of clean gas values from Line No. 1 as shown in Table 6. The range between high and low values is not significantly different between PSF and ESR.

The range of variation is due to the extremely low concentration levels measured. The clean gas concentrations of PCDD/Fs during the PSF test were in the order of 1 % of the emission limit value (ELV) of the EU WID Directive. This confirms again the high reliability and high performance of the MHKW operation to reduce dioxin/furan concentrations from the raw gas level to the clean gas level.
The addition of active carbon absorbs more than 99% of the PCDD/Fs.

MHKW Lines	Clean gas A1/A2	Clean gas B1/B2	Clean gas C1/C2	Comments
March 2013 (line n° 1)				
ITEQ ng/m³ excl. LOQ	0.0019/0.0007	0.0004/0.00005	0.00002/0.00005	PSF added to MSW
ITEQ ng/m³ incl. LOQ	0.0021/0.0018	0.0004/0.00016	0.00003/0.00016	
August 2004 (line n° 2)				
ITEQ ng/m³ incl. LOQ	0.0023	0.004	0.013	Electrical & Electronic Shredder Residue

Table 6: Clean Gas PCDD/Fs concentrations

Heavy Metals/Halogens

The level and range of heavy metals within the clean gas was extremely low and for many elements close to the detection limits. The levels of all elements were safely below the specified limits according to the EU WID. There was no influence of PSF addition on the clean gas emissions.

HBCD

Clean gas HBCD data ranged in a magnitude from 1 to 8 ng/m³. There was no detectable influence of HBCD concentration in the clean gas from PSF addition. The limits of quantification were 0.09 ng/m³ and significantly lower than the measured HBCD concentrations in the clean gas. Hence the confirmation that there is no contradiction between what was measured within the residues on the one hand and in the gas phase on the other. HBCD concentrations in the two emission paths, gas and solids are also independent of the actual amount of PSF fed to the MSWI. It can therefore be concluded that the usual HBCD input from other products in the MSW was more significant than the HBCD input from the PSF.

The grate ash residue is rather heterogeneous in its nature due to the many different MSW fractions contributing to the grate ash. The limit of quantification ranged between 1.2 and 1.6 µg/kg. HBCD concentrations are given as the sum of the three isomers and any differentiation about different isomer distributions in PSF or emission residues and gas has not been considered.

Grate Ash Characteristics

Grate ash characteristics were investigated in depth to support the technical objectives: (1) Good combustion performance is directly linked to high burn out and low residual carbon, and (2) high destruction efficiency of HBCD. Residues of unburnt matter, metals and stones/inert were hand-separated and weighed. No influence of PSF co-incineration could be detected. Residue weight values ranged as normal for the hand-sorted fractions and confirmed the stable combustion.

Grate Ash Leaching Tests: EU/EN 12457 1-4 and CH/TVA

Different leaching test requirements exist in Europe for landfill, and the International Ash Working Group deals with the collection and development of know-how. The leaching tests applied differ in their goal and therefore in the procedure employed. The EU standardised leaching tests EN 12457-1 to 4 basically describe the national requirements. Selected results of the screening leaching tests are shown in Table 7. Due to the heterogeneous character of the grate ash, significant variations within the same test series have to be accepted. The regulatory limits for the EU leaching test are also shown in Table 7.

The differences between the reference tests A and the co-feeding test C were not significant and it is clear that the quality of the grate ash is not influenced negatively by the co-combustion of the PSF material. In the case of Pb leaching it is known that grate ash aging for several months reduces the leachability strongly.

Sample	EN 12457-4		EN 12457-3				CH-TVA		EU-limits for granular non-hazardous waste	
	A1	C1	A1, 1st eluate	A1, 2nd eluate	C1, 1st eluate	C1, 2nd eluate	A1	C1	L/S=2	L/S=10
pH	12.2	11.8	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Arsenic	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	0.4	2
Lead	0.27	< 0.005	2.1	0.77	0.021	< 0.005	< 0.005	< 0.005	5	10
Cadmium	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	0.004	0.01	0.6	1.0
Copper	0.024	0.14	0.077	0.031	0.55	0.063	0.13	0.22	25	50
Nickel	< 0.005	< 0.005	0.014	< 0.005	0.018	< 0.005	0.17	0.24	5	10
Mercury	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	0.05	0.2
Zinc	0.09	0.02	0.23	0.06	0.02	< 0.01	3.3	7.8	25	50
Chloride	520	660	2500	130	3400	170	n.a.	n.a.	100000	150000
Sulfate	920	630	1400	680	720	140	n.a.	n.a.	10000	20000
TDM	3100	2400	10000	2200	8400	820	n.a.	n.a.	40000	60000

Note: n.a. not applicable, TDM is Total Dissolved Matter, all numbers in mg/l except pH

Table 7: Selected important Leaching Limit Parameters and Analytical Results

Conclusions and Recommendations

The PSF co-incineration tests validated important findings on emissions and operation. The test has been done with current commercial-quality PSF in order to ensure the correct and consistent PSF input during the measurement periods. The following recommendations for practical handling of EoL PSF waste in MSWI are not only relevant to current manufacturing scrap and job site waste, but also, more importantly, to demolition waste. In addition, appropriate mixing of MSW and PSF boards is important and should be performed by experienced operators.

PSF can be handled in large-scale MSWI if certain provisions are met:

- Preferably a delivery of mixed/single boards with other construction waste.
- Delivery of complete board packages in plastic wrapping should be minimised as the crane has to break down these parcels.
- Typical commercial-size boards as delivered for this trial can be handled.
- No larger baled PSF parcels should be delivered.

State-of-the-art MSWI operation guarantees the simultaneous high-efficiency destruction of HBCD and ODS. UNEP, EU and national authorities should therefore support the co-incineration of old PSF in MSWI without requiring the use of hazardous waste incinerators.

At the state and local level, EoL practices within the European Union are rather different. This is due to geography, cost, demographics, population density and existing infrastructure of waste management operations like landfill, incinerators, heat users, etc.

The trial clearly demonstrated the suitability of state-of-the-art MSWI installations for the safe end-of-life treatment of PS foam boards containing HBCD. The co-incineration of PS Foam waste from building and construction with MSW is indeed an option of choice that is able to provide a practical, long-term technical solution to the management of end-of-life PS foam waste, including non-HBCD containing boards.

In addition, the results of the trial present a solid, documented reference point for the definition and enforcement of the specific requirements for the classification and handling of EoL PS foams that may in future be adopted under the UNEP Stockholm and Basel Conventions as a consequence of the POP status of HBCD.

Reference

The full Technical Report can be obtained from Plastics*Europe* and EXIBA:

www.plasticseurope.org ***www.exiba.org***

Abbreviations

A	Condition of Base Case with no PSF added
A1	Measurement time period at Condition A, 6 hours
AC	Activated Carbon
ASR	Automotive Shredder Residue
B, C	Conditions with PSF added
C	Carbon
CO	Carbon monoxide
CO_2	Carbon dioxide
DE	Destruction Efficiency
ELV	Emission Limit Value
EoL	End of Life
EPS	Expanded polystyrene foam
ESR	Shredder residue from electrical/electronic end of life goods
HBCD	Hexabromocyclododecane
HBr, HCl	Acid gases hydrogen bromide, hydrogen chloride
HM	Heavy metals, as defined by the WID
Hu	Heating value, lower
ITEQ	International Toxic Equivalent
LOQ	Limit of Quantification, one standard deviation
MSW	Municipal Solid Waste
MSWI	Municipal Solid Waste Incinerator
NO_x	Nitrogen oxides
O_2	Oxygen
ODS	Ozone Depleting Substance
PBDD/Fs	Polybrominated Dioxins & Furans
PBT	Persistent Bioaccumulative Toxic
PCDD/Fs	Polychlorinated Dioxins & Furans
POP	Persistent Organic Pollutant
PS	Polystyrene
PSF	PS Foam (XPS and EPS)
PXDD/Fs	Halogenated (brominated and/or chlorinated) dioxins & furans
SO_2	Sulfur dioxide
TEAP	Technology and Economic Assessment Panel
TEQ	Toxic Equivalent
TOC	Total Organic Carbon
TVA	Technical Ordinance on Waste, Switzerland
UNEP	United Nations Environment Programme
WFD	European Waste Framework Directive
WID	European Waste Incineration Directive
XPS	Extruded polystyrene foam

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