

SOLUTIONS FOR THE DESTRUCTION OF POPs WASTES



In many parts of the world stocks of legacy persistent organic pollutants (POPs) – e.g. polychlorinated biphenyls (PCBs), organochlorine pesticides, brominated flame retardants, and fluorinated chemicals – are posing threats to human health and the environment. These POPs wastes must be destroyed or irreversibly transformed by sustainable methods that, to the greatest extent achievable, meet the following criteria:

- An effective destruction efficiency of 100 % – taking into account all inputs and outputs (gaseous, liquid and solid);
- Complete containment of all process outputs to enable testing – and reprocessing if necessary – to ensure an effective destruction efficiency of 100 %;
- No uncontrolled releases from the process.

Hazardous waste incinerators

Over the past four decades, incineration has been the principle method for the destruction of organic pollutants. However, there are several problems associated with this technology.

Ideally, incineration of organic compounds at temperatures higher than 850 °C with sufficient turbulence and excess oxygen should lead to non-toxic endproducts, e.g. CO₂ and H₂O. However, side reactions leading to toxic by-products inevitably occur especially when incinerating waste containing halogens and PCDD/Fs precursors (especially PCBs, chlorophenols, chlorobenzenes and other chlorinated aromatic compounds) (Blumenstock et al., 2000; Huang and Buekens, 2001; Jiang et al., 1997; McKay, 2002) and similar brominated compounds lead to the formation of PBDD/Fs and PXDD/Fs (Weber and Kuch, 2003; Schüler and Jager, 2004). Increasingly stringent regulations that require more effective air pollution control and monitoring of gaseous emissions have resulted in considerable reduction of releases to air of PCDD/Fs but, in some circumstances, increased releases to air pollution control residues. However releases from waste handling together with unintentionally-formed products of incomplete combustion released to air and contained in residues

from hazardous waste incinerators can still lead to contamination around the incinerator plant and add to reservoirs of these by-products, depending on the technology used, facility operation conditions and the management of solid residues (Weber et al., 2008). The released toxic by-products, e.g. PCDD/Fs (known human carcinogens), can enter the food chain (Malisch et al., 1999). Several such cases are described in the scientific literature (Goovaerts et al., 2008; Holmes et al., 1994, 1998; Kim et al. 2006; Lovett et al., 1998).

A limiting factor of incineration is the relatively high cost (DOE, 1999; Haglund, 2007), especially when operating incinerators with state-of-the-art pollution control devices, monitoring and residue treatment. Costs also rise with the transport of hazardous wastes to the incineration facility. Cement kilns operate at temperatures of 1450 °C or higher but only a few meet the necessary technical requirements for the incineration of POPs, and high destruction efficiencies have not been demonstrated. Furthermore expensive blending, feeding and monitoring equipment is needed for effective operation (Rahuman et al., 2000).

A comprehensive evaluation of destruction efficiencies is missing for incineration (hazardous waste incinerations as well as cement kilns) up to now (Weber, 2007). However, some tests suggest that incinerators achieve destruction efficiencies that are lower than those achieved by certain non-combustion technologies (Rahuman et al., 2000).

These complications together with public opposition to incineration of hazardous and POPs wastes have led to the investigation of alternative and non-combustion destruction methods.

Non-combustion destruction methods

These technologies use physical and chemical processes of converting POPs wastes to less harmful substances. There are non-combustion, high-temperature technologies operating now at a commercial scale in one or more countries, where these technologies are licensed to destroy POPs stockpiles. Four of them are briefly

described here. Further, two very interesting technologies are also presented:

- A destruction technology that operates under mild reaction conditions (room temperature and pressure); and
- A technology for removing brominated flame retardants from waste plastics.

Gas Phase Chemical Reduction (GPCR)

This technology involves the gas-phase chemical reduction of organic compounds by hydrogen at temperatures of 850°C or greater and low pressure. Organic compounds are ultimately reduced to methane, hydrogen chloride (which is later neutralized), and minor amounts of low molecular weight hydrocarbons. It is able to treat both liquid and solid wastes of high contents of DDT, hexachlorobenzene (HCB), PCDD/Fs and PCB transformers, capacitors, and oils. Pre-treatment is needed for both solid and liquid wastes. The configuration of the methodology is modular: it can be fixed as well as transportable.

All emissions and residues may be captured and reprocessed if needed. PCDD/Fs have not been detected in the product gas from the process, but have been detected at low levels from natural gas burner used to heat the reaction vessel (UNEP, 2004). Solid residues are generated from solid waste inputs, but these should be suitable for disposal in a landfill, although UNEP (2003) indicates that traces of POPs may remain.

The destruction efficiency data provided by the Eco Logic company from facilities run in Australia and Canada report DEs of > 99.9999 % for PCBs, DDT and HCB and > 99.9995 % for PCDD/Fs (Eco Logic, 2002). The throughput of 150 tons per month or 1800 tons per year can be doubled due to modular design (IHPA, 2002). EPA (2005) stated that this technology was not considered to be cost-effective but more recently reported that it is currently "being modifying to improve its cost effectiveness" (EPA, 2010).

Base Catalysed Decomposition (BCD)

This US EPA developed process consists of two separate processing steps: first indirectly heated thermal desorption at temperatures of 200–400°C is

used for the decontamination of treated media, then the POPs in form of pure chemicals or concentrates obtained by the desorption are destroyed by a chemical reaction. The dehalogenation occurs when selected chemicals including sodium hydroxide (a base) are mixed with the condensed contaminants and heated to 236°C in a reactor. If the carrier oil does not meet the disposal criteria, it is returned to the reactor and reheated. This technology is able to remediate liquids, soils, sludge and sediments contaminated especially with PCBs and PCDD/Fs. The treated soil can be used as backfill on site. The technology is fixed or designed as mobile units (EPA, 2005; Rahuman et al., 2002; UNEP, 2004).

Compared to older facilities, today's plants are equipped with off-gas scrubbing and filtration. Emission concentrations are reported to be low and the total mass of emitted off-gas is orders of magnitude smaller than incinerators (UNEP, 2004).

A BCD reactor can process 2–12 tonnes of POPs per day and higher throughputs can be achieved by increasing the number of modules. High destruction efficiencies (4–6 nines) have been demonstrated for PCBs, OCPs and PCDD/Fs in treatability trials and routine operations (UNEP, 2004). The BCD treatment time is short, energy requirements are moderate and operation and maintenance costs are relatively low. The operation costs of a full-scale BCD system are considered to less than 25 % of the incineration operating costs (Rahuman et al., 2000).

Super Critical Water Oxidation (SCWO)

Many chemical destruction technologies are based on the reaction of the organic compounds with hydroxyl radicals. In supercritical water, organic pollutants become highly water-soluble and react rapidly with added oxidants. Final decomposition products are carbon dioxide, water and mineral acids and salts. The supercritical phase of water occurs under high temperature and pressure conditions, e.g. 374°C and 24–28 MPa (EPA, 2005; Rahuman et al., 2000; Veriansyah et al., 2007). Effluent gases do not contain nitrogen oxides, acid gases, or particles and carry less than 10 ppm carbon monoxide

(Environment Australia, 1997). SCWO is suitable for the treatment of soil, sludge and liquid wastes contaminated with e.g. PCBs and pesticides as well as hazardous military wastes in low or high concentrations. The technology is highly transportable (IHPA, 2008; Marulanda, 2010).

Current SCWO plants use corrosion resistant materials. All emissions and residues may be captured for reprocessing, if needed (UNEP, 2004).

EPA (2005) as well as Rahuman et al. (2000) claim no data on the destruction efficiency have been found, however, UNEP (2004) report a demonstrated high efficacy of the technology. IHPA (2008) mention a DE of six to eight 9s for very low pesticide concentrations. For waste containing maximum 20 % of organic carbon, SCWO is considered to be far less costly than incineration (Boock, 1996). Due to some operation problems, the industrial status of SCWO is currently limited however, Veriansyah and Kim (2007) claim that enhanced investment into novel technologies potentially superior to incineration could remove the current technical limitations of the technology. More recently Marulanda (2010) suggests that the economics and performance of a mobile unit in South America might make SCWO an economically viable alternative to incineration in South American countries.

Sodium reduction

This transportable or fixed technology has been used widely for the in-situ removal of low to high contents of PCBs from transformer oils. The basic principle is the reduction of the PCBs with dispersed metallic sodium in mineral oil which leads to the final products of non-halogenated biphenyls, sodium chloride, petroleum based oil and water. The capacity for treating transformer oils has been reported to be 15 000 litres per day. No destruction efficiency has been reported for this technology and there is insufficient information available on the characterisation of residues. Also, as with any process which does not involve destruction of the transformer itself, there is concern about residues of PCBs in the porous materials of transformers

when transformer oils are treated in situ. However, this approach has been widely used for treating PCBs for more than two decades (UNEP, 2004).

Ball milling/Mechanochemical dehalogenation (MCD)

Ball mills are mechanochemical dehalogenation reactors for the destruction of PCBs and other organic chlorinated pollutants to their parent hydrocarbons in high yields. The reductive dehalogenation occurs in the presence of an alkali metal and a low acidic hydrogen source. It can be applied to contaminated materials as well as highly concentrated or pure chemicals regardless of their state. The pollutants are eliminated directly inside a contaminated material (Birke et al., 2004; UNEP, 2004). Although the mechanochemical degradation is performed at low temperatures, inside the milled material temperatures up to several thousand degrees Celsius occur when a grain collides at high velocity with a solid surface (Heinicke, 1984). The final product powder may require further treatment (IHPA, 2008).

Ball mills are available in different sizes and constructions such that treatment of materials up to several tons is possible. Mechanochemical reduction is cost-effective and offers also environmental benefits due to its low energy consumption. Due to mild reaction conditions and the design of a closed system, no harmful emissions to the environment are expected (Birke et al., 2004; UNEP, 2004).

IHPA (2008) mentions the DE of ball milling to be 4–5 nines. Birke et al. (2004) state that PCBs in solid or liquid matrices can be destroyed (based on laboratory studies) to non-detectable levels in a period of minutes to hours. However, as there is currently very limited commercial experience with this technology, limited information exists on its emissions, efficacy, intermediate breakdown products and other important parameters under industrial operation. Main questions also remain regarding the destruction efficiency of the technology and the amount and toxicity of agents needed in the process (IHPA, 2008). When applied to pesticide-contaminated soils at a site in New Zealand, ball mill-

ing reduced pesticide levels in the soils by less than 90 percent. There were also concerns about possible PCDD/Fs releases to the air during processing as well as potentially problematic levels of process agents that remained in the treated soil (PCE, 2010; PCE, 2008).

CreaSolv® Process

Brominated flame retardants ('BFRs') are removed from waste plastics by the CreaSolv® process. Specific polymers in the waste plastics are selectively dissolved by proprietary solvents and then precipitated by another proprietary formulation. This follows pre-separation of other wastes so that the feed material entering the solvent extraction contains a high proportion (generally $\geq 75\%$) of the type of plastic to be recovered (Malcolm Richard, 2011). The developers – CreaCycle GmbH, in co-operation with the Fraunhofer Institute IVV – report that the volume of solvent used is very small in relation to the treated plastic ($< 1\%$), because the solvents are recycled. The only solvent removed from the process is the small fraction in which BFRs and other contaminants are separated and concentrated. The final products of the process are usable polymer recyclate, BFR-rich concentrate, and, if present, a metals-rich insoluble fraction (Creacycle; Mäurer and Schlummer, 2004).

The process has been applied at laboratory and pilot plant scales. For example, after battery removal, post-consumer mobile phones were treated, yielding polymer particles suitable for extrusion and injection molding processes (Mäurer and Schlummer, 2004). In another example, expanded polystyrene (PS) waste was successfully treated at pilot scale to produce re-expandable PS that is comparable to virgin polystyrene in usability (Mäurer and Knauf, 2005). In a small-scale feasibility study, both BFRs and PBDD/Fs, which were present as co-contaminants, were successfully removed from plastic wastes from Canadian WEEE dismantling plants (Schlummer et al, 2008).

The Waste and Resources Action Programme (WRAP) funded a study by Freer (2005) to assess the potential environmental impacts of four new processes for

recovering electrical and electronic plastic waste containing BFRs in comparison to landfill and incineration with and without energy recovery. Among these, the CreaSolv® Process was ranked best with respect to energy consumption and photochemical oxidation potential, and was second only to incineration with energy recovery in having the lowest global warming potential. The study concluded that the two solvent-based processes, CreaSolv® and Centrevap® were the two best environmentally performing processes and also noted that the CreaSolv® Process was particularly distinguished by "low solvent losses and high solvent recovery efficiencies."

A recent review by Nnorom and Osibanjo (2008) of the management of BFR-containing plastics noted that other WRAP-funded studies found the CreaSolv® Process to perform better in removing BFRs from WEEE polymers than the Centrevap® process. However both processes were said to provide financially viable alternatives to landfill and incineration as options in the management of WEEE plastics. The reviewers concluded that these solvent-based methods of removing BFRs "presently offer the best commercial and environmental option in the sound management of waste BFR-containing plastics," and went on to suggest that commercialization of these processes will help to reduce export of WEEE wastes to developing countries.

The concentrated BFRs recovered from the process can be destroyed by other non-combustion technologies or irreversibly transformed as reagents in industrial processes.

Questions to be answered

One important criterion for the assessment of POPs destruction technologies is the potential formation of new POPs and other toxic by-products during the process. PCDD/Fs can be formed during combustion of hazardous wastes (see above). However, the operation conditions for a number of non-combustion technologies have also the potential to generate PCDD/Fs if relevant precursors are present (Weber, 2007). One example is GPCR, where if either the product gas or the ambient air used for the combustion contains chlorin-

ated species, PCDD/Fs may be generated. In order to meet the fundamental technical criteria for POPs destruction, both the product gas and combustion air must be suitably treated (Rahuman et al., 2000). Also the BCD treatment process may result in an increased concentration of lower chlorinated species, where this is of potential concern for PCDD/Fs, where the lower congeners are significantly more toxic than the higher congeners. It is therefore essential that the process is appropriately monitored to ensure that the reaction continues to completion (Rahuman et al., 2000). A laboratory study of PCBs destruction by SCWO has shown that in lower temperature operations considerable amounts of PCDD/Fs can be formed (Weber, 2004). Operators must ensure that processes are operated at conditions (particularly temperature and residence times) which avoid such formation. In many cases, however, detailed assessments of non-combustion technologies with respect to PCDD/Fs formation are lacking (Weber, 2004, 2007).

How to choose the most suitable technology?

There is likely no "perfect" technology for the destruction of POPs. Many criteria play a role when deciding on a suitable technology to be applied in each specific case. A basic performance standard for POPs destruction technologies is the efficiency of destruction. Destruction efficiency (DE) is a comparison of the amount of a specific POP that fed into a process with the total amount of that POP that is released in all the process outputs, e.g. gaseous and liquid emissions as well as solid residues. However, another measure that is sometimes used is destruction and removal efficiency (DRE), which takes into account only the amount of a specific POP that is released in air emissions. Unfortunately, many of the reports on a technology's efficiency report only DREs and sometimes DREs are erroneously reported as DEs (Costner, 2004). Both measures must be understood with respect to non detectable POPs concentrations. These are reported as "less than" detection limit values, not as zero. As a consequence, calculated

DE and DRE values approach, but never reach 100 %. Therefore, a destruction efficiency of >99.9999 % can be considered as effectively 100 % (Rahuman et al., 2000). Although chemical and toxicological analyses of all the outflow streams are expensive, they must be carried out with a frequency sufficient to ensure compliance of the technology with the basic criterion of DE >99.9999 % during all operation conditions, i.e. start-ups, shutdowns, routine operation and eventual critical events (Rahuman et al., 2000). Weber (2007) states, that DEs have to be determined by long-term monitoring lasting up to months and should be performed for the whole duration of POPs destruction projects.

The costs of a technology are, of course, important limiting factors. They vary significantly due to the variation in the POPs content of the material to be treated, the volume of this material and transport distances (when applying ex situ technologies). Initial investment costs can be high, however, the treatment costs of the recently developed and less optimized technologies can decrease in future which is not likely for the mature approach of incineration (Haglund, 2007).

Other economical, social, environmental and technical criteria should be considered when deciding on a destruction technology, too. These are, among others (Environment Australia, 1997; IHPA, 2008; Lodolo, 2002; Rahuman et al., 2000; Veriansyah and Kim, 2007; Weber, 2007):

- Capability of the technology to treat the waste in the physical form(s) present (liquid, solid, size of grains), with the determined organic carbon content and necessity for the pretreatment of the waste
- Capacity of the installation
- Local applicability
- Ability of the waste to be transported and mobility of the facilities
- Reliability and maintenance of the facility
- Volumes of secondary waste streams and the content of toxic reaction by-products under all operation conditions including unstable periods, closed system design
- Risks connected to the technology's operation (load flexibility, transient

control, emergency management, dismantling of the facility)

- Public acceptability

When considering green chemistry principles (which makes the use of environmentally conscience manufacturing and production technologies) processes at room temperature and atmospheric pressures should be preferred. This ensures the lowering of energy consumption and increasing the prospects for the facility to be transported. Further, any reagents or starting materials should be inexpensive, non-toxic and easily stored, toxic by-products should be avoided and last, but not least, the method should exhibit a high destruction efficiency (Laine and Cheng, 2007). This underlines the need for the design of new reactors and further research.

Conclusions

Although the incineration of old POPs stocks is still the most used destruction technology, it cannot be considered "clean", there is concern about its high costs, comprehensive information on its destruction efficiency is missing and it is not transportable. A technology that is transportable, locally applicable, sustainable and destroys obsolete POPs at a reasonable cost would mean a breakthrough in the destruction of these chemicals (IHPA, 2008). Innovative non-combustion technologies have the potential to become such technologies, however, specific conditions have to be guaranteed:

- long-term resource availability,
- strong environmental awareness and
- sound scientific research sustained by political support and appropriate funding to ensure continuous technological progress.

Also, as some necessary information (e.g. potential PCDD/Fs formation during the destruction process) is currently provided almost exclusively by the developing and selling companies (Weber, 2007), independent evaluations of the technologies should be conducted and data made publicly available. The feasibility and durability of the technologies should be proven by real field demonstration tests in countries/regions with limited infrastructure and lack of necessary conventional supply services (IHPA, 2008).

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IPEN Dioxin, PCBs and Waste Working Group

The IPEN Dioxin, PCBs and Waste Working Group was established in May 2001 in Sweden, after the text of the Stockholm Convention was agreed. The Working Group, within its capacity and resources, works to assure that measures addressing dioxins, PCBs and wastes are appropriately interpreted and fully incorporated into each country's Stockholm Convention Enabling Activities and National Implementation Plans. Furthermore, it works to promote policies and practices in every region and country aimed at the elimination of dioxins and PCBs; and aimed at reduction and elimination of wastes, and appropriate waste management for the residues.

IPEN Dioxin, PCB and Waste Working Group c/o Arnika Association

Toxics and Waste Programme

Chlumova 17

Prague 3

Czech Republic

130 00

phone/fax: +420 222 781 471

e-mail: ipen-dioxin@arnika.org

website: IPEN: <http://www.ipen.org>

<http://english.arnika.org>

