Ambiente & Água - An Interdisciplinary Journal of Applied Science
ISSN: 1980-993X
ambi-agua@agro.unitau.br
Universidade de Taubaté
Brasil

Raboni, Massimo; Viotti, Paolo

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Ambiente & Água - An Interdisciplinary Journal of Applied Science, vol. 11, núm. 1, enero-marzo, 2016, pp. 4-12
Universidade de Taubaté
Taubaté, Brasil

Ambiente & Água - An Interdisciplinary Journal of Applied Science
ISSN 1980-993X – doi:10.4136/1980-993X
www.ambi-agua.net
E-mail: ambi.agua@gmail.com

Formation and destruction of Polycyclic Aromatic Hydrocarbons (PAHs) in the flaring of the biogas collected from an automotive shredded residues landfill

doi:10.4136/ambi-agua.1758

Received: 17 Sep. 2015; Accepted: 15 Dec. 2015

Massimo Raboni[1]; Paolo Viotti[2*]

[1]University LIUC - Cattaneo, 21053 Castellanza, VA, Italy
School of Industrial Engineering
[2]University of Roma “La Sapienza”, Via Eudossiana 18, I-00184 Rome, Italy
Department of Civil and Environmental Engineering
[*] Corresponding author: e-mail: paolo.viotti@uniroma1.it,
mraboni@liuc.it

ABSTRACT

The paper shows the results of the combustion in an enclosed flare of the biogas collected from an automotive shredded residues landfill. The results demonstrate that at 1,000°C and long combustion, several synthesis reactions lead to the formation of 4 to 6 rings of PAHs. This formation also involves the formation of compounds such as benzo(g,h,i)perylene, indeno(1,2,3-cd)pyrene and dibenzo(a,h)anthracene not present in raw biogas. However, the compounds most likely to form in combustion are benzo (a) anthracene and benzo (b) fluoranthene. The only exception is chrysene which is significantly destroyed. The experience has proved the total lack of formation of PAHs with only 2 and 3 aromatic rings.

Keywords: automotive shredded residues, biogas, combustion, PAHs.

Formação e destruição de hidrocarbonetos aromáticos policíclicos (HAP) na combustão do biogás coletado de um aterro de resíduos de veículos desmantelados

RESUMO

O artigo mostra os resultados da combustão em um flare enclausurado de biogás coletado de um aterro de resíduos de veículos desmantelados. Os resultados demonstram que a 1,000°C e combustão longa, várias reacções de síntese conduzem à formação de 4 a 6 anéis aromáticos policíclicos. Esta formação também envolve a formação de compostos, tais como o benzo(g,h,i)perileno, indeno(1,2,3-cd)pireno e dibenzo(a,h)antraceno não presentes no biogás bruto. No entanto, os compostos que mostraram a maior formação em combustão são o benzo(a)antraceno e benzo(b)fluoranteno. A única excepção é o criseno que sofre um efeito significativo de destruição. A experiência provou a total falta de formação de HAP com apenas 2 e 3 anéis aromáticos.

Palavras-chave: biogás, combustão, HAP, resíduos de veículos desmantelados.

1. INTRODUCTION

Automotive Shredded Residues (ASRs) are generally disposed of in landfills, resulting in significant environmental and health issues (US-DHHS, 1995; Raboni et al., 2014). However, in some cases thermal treatment plants are used (Ciacci et al., 2010; Mancini et al., 2014). The ASR is characterized by the presence of hazardous inorganic compounds (particularly lead, cadmium, and copper), along with hazardous organic compounds derived from fuels, lubricating oils, paints, combustion residues, and solvents used in vehicle maintenance and cleaning (Torres et al., 2013). The composition of the ASR and the presence of hazardous inorganic compounds are well documented in the literature (Raboni et al., 2014; Santini et al., 2012; Mancini et al., 2014; Cossu et al., 2014). The leachate produced by these landfills has a significant amount of organic compounds and heavy metals and its treatment is carried out in a manner similar to the leachate from municipal solid waste landfills; in particular physical-chemical processes (Raboni et al., 2013) and biological activated sludge processes, and also MBR are applied (Torretta et al., 2013).

Biogas production from ASR landfills represents a very small share of the total production of biogas from different sources, which in Europe amounts approximately to 10 Mtoe y^{-1} (Eurobserv'Er, 2012; Raboni and Urbini, 2014; Raboni et al., 2015). However, it is characterized by the presence of a broad spectrum of hazardous organic compounds, in the categories of VOCs and PAHs, which are typical ubiquitous compounds as they are normally found in urban areas and along automotive routes (Mohamed et al., 2002). PAHs are typically generated by combustion processes, whereas the VOCs detected in the air are also connected to many to industrial activities; in some cases the VOCs are found in the exhaust air of water and wastewater stripping processes (Raboni et al., 2013).

With reference to hazardous organic compounds found in the biogas of ASR landfills, two recent studies measured the level of PAHs and VOCs in this biogas (Raboni et al., 2015; Urbini et al., 2014). Due to their high toxicity and carcinogenicity properties, it is essential to minimize the release of these compounds into the environment and, above all, into the atmosphere, in order to protect public health. While physical-chemical and thermal processes (Mancini et al., 2014; Innocenti et al., 2014) are regularly applied for the removal of these compounds from waste air streams, actually many studies and also real-scale applications are exploring biological treatment processes (Copelli et al., 2012; Mathur et al., 2007).

The main objective of the present research is to verify that PAHs may be destroyed by the combustion of biogas at high temperature in an enclosed ground flare. Attention was essentially focused on PAHs, as they represent the major risk to human health due to their toxicological and carcinogenic properties. In fact, dibenz(a,h)anthracene, benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, chrysene and indeno(1,2,3-c,d)pyrene are considered probable human carcinogens (while acenaphthylene, anthracene, benzo(g,h,i)perylene, fluoranthene, fluorene, phenanthrene, and pyrene are not classified as carcinogenic) (US-DHHS, 1995). Dibenz(a,h)anthracene and benzo(a)pyrene especially are assigned the highest values of Toxicity Equivalent Factor (TEF), assessed in relation to carcinogenicity (TEF=5 for dibenz(a,h)anthracene; TEF=1 for benzo(a)pyrene), while significantly smaller values are assigned to chrysene (TEF=0.01) and pyrene (TEF=0.001) (US-DHHS, 1995).

Indeed, several recent studies have shown that the formation and destruction of PAHs are concomitant phenomena, capable of leading to a net increase of these hazardous compounds in the output of combustion systems. This finding has also been verified in the combustion of methane and other light hydrocarbons.

The research was developed in an enclosed flare of a real-scale ASRs landfill.

2. EXPERIMENTAL SECTION

2.1. Combustion flare characteristics

The landfill is equipped with 24 wells of biogas equally distributed over the entire landfill area, each discharging into two parallel, enclosed flares. The research was focused on one of the two flares fed by 700 $Nm^3 h^{-1}$ of biogas.

The main features of the enclosed ground flare were as follows:

- diameter: 1.6 m
- height of combustion area: 4 m
- average temperature 1,000°C
- average retention time: 3.6 s
- combustion chamber lined with refractory (internally) and mineral wool (externally)
- multi-nozzle stainless steel burner
- combustion temperature measured with thermocouple (accuracy ± 1°C)
- Measurement of Oxygen in the combustion chamber with a zirconinum oxide probe suitable for temperatures up to 1,400°C (accuracy: ± 1%); Throughout the experiment, O_2 was kept in the range 11-13% (average 12%)

2.2. Analyzed compounds

The research was conducted in a three-month period during which 60 samples were collected on different days (30 of the raw biogas; 30 of the flare outlet).

The following compounds were detected:

a) sixteen PAHs in raw biogas and at the end of combustion flare: benzo(g,h,i)perylene, indenopyrene, dibenz(a,h)anthracene, benzo(a)pyrene, benzo(k)fluoranthene, benzo(b)fluoranthene, benz(a)anthracene, chrysene, pyrene, fluoranthene, anthracene, phenanthrene, fluorene, acenaphthene, acenaphthylene, naphthalene;

b) In raw biogas: CH_4, O_2, N_2, CO_2 (with in addition, for a better characterization: NH_3, H_2S, Mercaptans, alkyl-sulfides: VOCs).

2.3. Methods of sampling and analysis

Measurement of CH_4, CO_2, N_2 and O_2 in the raw biogas was done with an infrared analyzer LFG 20 (resolution 0.01%). Pressure measurement was done with a micro-electronic manometer (accuracy ± 1% of full scale). Temperature and humidity measurement was done with a thermo-hygrometer (accuracy: T ± 0.45°C: RU ± 1% of full scale).

The sampling and analysis of PAHs, in raw biogas and flare outlets, were performed with specific procedures for each class of compounds after the separation of particulates on filter membranes, and separation of condensate and adsorption of the organic compounds present in the vapor phase on a vial ORBO 43. The determination of PAHs was obtained by HPLC with fluorescence detection so as to combine high sensitivity with sufficient selectivity. The limit of analytical detection of single PAHs was 0.1 ng Nm^{-3}.

3. RESULTS AND DISCUSSION

3.1. Biogas composition

Table 1 shows the macro-constituents (methane, carbon dioxide, oxygen and nitrogen) of the biogas composition, with additional minor components that may cause foul odors (ammonia, hydrogen sulphide, mercaptans and alkyl-sulfides). Biogas temperature is also indicated.

Table 1. Biogas composition and temperature.

Constituents and temperature	Unit of measure	Mean	Standard Deviation	Range min-max
CH_4	%	48.0	± 1.8	45.0-51.0
CO_2	%	38.0	± 2.2	35.0-42.0
O_2	%	1.8	± 0.4	1.30-2.30
N_2	%	12.0	± 1.1	10.5-13.6
NH_3	mg Nm^{-3}	6.5	± 2.2	3.60-11.20
H_2S	mg Nm^{-3}	3.6	± 0.7	2.50-5.40
Mercaptans	mg Nm^{-3}	0.5	± 0.2	0.25-0.80
Alkyl-sulfides	mg Nm^{-3}	3.4	± 0.9	2.00-6.05
Temperature	°C	54	± 3.5	50.0-59.0

It can be observed that the main characteristics of the biogas are quite similar to that of MSW landfills with the sole exception of ammonia (much higher in MSW landfills due to the greater presence of organic nitrogen compounds in the waste).

For the objective of the present research, it is important to note that several VOCs were detected in the biogas, as potential precursors of PAH formation. In particular, toluene was found at the average concentration of 120.0 mg Nm^{-3}, hexane at 75.0 mg Nm^{-3}, trichloromethane at 55.5 mg Nm^{-3}, acetone at 43.5 mg Nm^{-3} and minor presences were also found for xylene (25.6 mg Nm^{-3}), isopropanol (20.2 mg Nm^{-3}), methyl-ethyl-ketone (18.0 mg Nm^{-3}) and methyl-isobutyl-ketone (14.5 mg Nm^{-3}).

3.2. Effects of biogas combustion on PAHs

Table 2 shows the main physical-chemical properties of the sixteen PAHs investigated. As known, PAHs are organic compounds with two or more aromatic rings in several structural configurations. Among them, those found in raw biogas are highlighted (grey background). Tables 3, 4, 5 and 6 show the results of flare combustion of the various PAHs. Each figure represents the PAHs with the same number of aromatic rings.

Tables 3 and 4 show that all PAHs with a relevant structural complexity (5 and 6 aromatic rings) are formed in combustion. This formation also affects three compounds such as benzo(g,h,i)perylene, indeno(1,2,3-cd)pyrene and dibenzo(a,h)anthracene not present in raw biogas.

However, the compound that shows the highest formation in combustion is benzo(a)anthracene, concentrations of which increase from the mean value of 6 ng Nm^{-3} to 71.5 ng Nm^{-3}; also remarkable is the increase of benzo(b)fluoranthene. Turning to PAHs with four aromatic rings (Table 5), the behavior appears controversial, since chrysene predominates the destructive effect, while for pyrene and fluoranthene a relevant formation occurs, up to over 70 ng m^{-3} for both compounds. Finally, Table 6 shows that the six PAHs with only two or three aromatic rings, are not only not present in the raw biogas, but they do not form in combustion.

In order to discuss the result shown in the tables, it is first necessary to consider that many chemical organic compounds are not completely destroyed during the combustion processes, even at temperatures as high as 1,000°C. Further, some organic compounds are synthesized in the combustion zone (and nearby) as a consequence of many secondary reactions. In a first decomposition step simple hydrocarbons and hydrocarbon radicals are formed in the gas phase (Figure. 1). These radicals are extremely reactive so that they give rise to several synthesis reactions, including cyclization. In this way, aliphatic hydrocarbons and aromatic hydrocarbons are formed which act as precursors to the formation of much more complex hydrocarbons, among which are the PAHs. Obviously the original presence of many VOCs in the raw biogas could be considered a more favorable condition for the synthesis of PAHs.

Table 2. Physico-chemical properties of PAHs. In grey background are PAHs detected in raw biogas (US-DHHS, 1995).

PAHs	formula	n. rings	Mol.Wt	Melting Point (°C)	Vap. Pressure at 25°C (mm Hg)	Solubility in water at 25°C (µg/L)
Benzo(ghi)perylene	$C_{22}H_{12}$	6	276.4	273	$1.03 \cdot 10^{-10}$	0.26
indeno(1,2,3-cd)pyrene	$C_{22}H_{12}$	6	276.3	163.6	$1 \cdot 10^{-11}$ - $1 \cdot 10^{-6}$	6.2
Dibenz(a,h)anthracene	$C_{22}H_{14}$	5	278.3	262	$1 \cdot 10^{-10}$ (20°C)	0.5
Benzo(a)pyrene	$C_{20}H_{12}$	5	252.3	179	$5.6 \cdot 10^{-9}$	2.3
Benzo(k)fluoranthene	$C_{20}H_{12}$	5	252.3	215.7	$9.59 \cdot 10^{-11}$	0.76
Benzo(b)fluoranthene	$C_{20}H_{12}$	5	252.3	168.3	$5.0 \cdot 10^{-7}$ (20°C)	1.2
Benz(a)antracene	$C_{18}H_{12}$	5	228.3	158-162	$2.2 \cdot 10^{-8}$ (20°C)	10
Crysene	$C_{18}H_{12}$	4	228.3	255-256	$6.3 \cdot 10^{-7}$	2.8
Pyrene	$C_{16}H_{10}$	4	202.1	156	$2.5 \cdot 10^{-6}$	77
Fluoranthene	$C_{16}H_{10}$	4	202.3	108.8 (*)	$5.0 \cdot 10^{-6}$	200-260
Anthracene	$C_{14}H_{10}$	3	178.2	218	$1.7 \cdot 10^{-5}$	700 (*)
Phenanthrene	$C_{14}H_{10}$	3	178.2	100	$6.8 \cdot 10^{-4}$	1,200 (*)
Fluorene	$C_{13}H_{10}$	3	166.2	116-117	$3.2 \cdot 10^{-4}$ (20°C)	800
Acenaphthene	$C_{12}H_{10}$	3	154.2	95	$4.47 \cdot 10^{-3}$	1,930
Acenaphthylene	$C_{12}H_8$	3	152.2	92-93	$29 \cdot 10^{-3}$ (20°C)	3,930
Naphthalene	$C_{10}H_8$	2	128.2	80.2 (*)	$1.8 \cdot 10^{-2}$ (*)	30,600 (*)

(*) Mrozik et al. (2003).

All PAHs have a high melting point, so they are normally in a solid state and in biogas are as fine particulate.

Table 3. Effect of flare combustion on PAHs with 6 aromatic rings (concentrations expressed as mean, standard deviation and range min-max).

6 RINGS PAHs	Flare position	Concentration (ng Nm^{-3})		
		mean	sd	min-max
Benzo(g,h,i)perylene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	4.3	0.8	2.6 – 5.5
Indeno(1,2,3-cd)pyrene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	1.8	1.8	1.1 – 2.6

Table 4. Effect of flare combustion on PAHs with 5 aromatic rings (concentrations expressed as mean, standard deviation and range min-max).

5 RINGS PAHs	Flare position	Concentration (ng Nm^{-3})		
		mean	sd	min-max
Dibenz(a,h)anthracene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	2.0	0.8	0.9 - 3.1
Benzo(a)pyrene	in	1.8	0.6	1.1 - 3.6
	out	3.5	1.8	1.2 – 5.9
Benzo(k)fluoranthene	in	3.4	1.4	1.2 – 4.8
	out	9.0	2.8	4.7 – 13.3
Benzo(b)fluoranthene	in	4.1	1.6	1.8 – 6.3
	out	19.6	7.2	9.9 – 32.6
Benz(a)antracene	in	6.0	1.9	3.0 – 8.6
	out	71.5	13.5	54.0 – 98.0

Table 5. Effect of flare combustion on PAHs with 4 aromatic rings (concentrations expressed as mean, standard deviation and range min-max).

4 RINGS PAHs	Flare position	Concentration (ng Nm^{-3})		
		mean	sd	min-max
Chrysene	in	39.5	11.0	28.5 – 78.6
	out	21.1	6.6	11.5 – 33.6
Pyrene	in	2.3	0.6	1.2 – 4.5
	out	74.0	15.9	37.0 – 99.6
Fluoranthene	in	2.2	0.7	1.1 – 3.8
	out	78.6	21.2	46.2 – 107.0

Table 6. Effect of flare combustion on PAHs with 3 and 2 aromatic rings (concentrations expressed as mean, standard deviation and range min-max).

2 & 3 RINGS PAHs	Flare position	Concentration (ng Nm^{-3})		
		mean	sd	min-max
Anthracene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	< 0.1	< 0.1	< 0.1 - < 0.1
Phenanthrene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	< 0.1	< 0.1	< 0.1 - < 0.1
Fluorene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	< 0.1	< 0.1	< 0.1 - < 0.1
Acenaphthene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	< 0.1	< 0.1	< 0.1 - < 0.1
Acenaphthylene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	< 0.1	< 0.1	< 0.1 - < 0.1
Naphthalene	in	< 0.1	< 0.1	< 0.1 - < 0.1
	out	< 0.1	< 0.1	< 0.1 - < 0.1

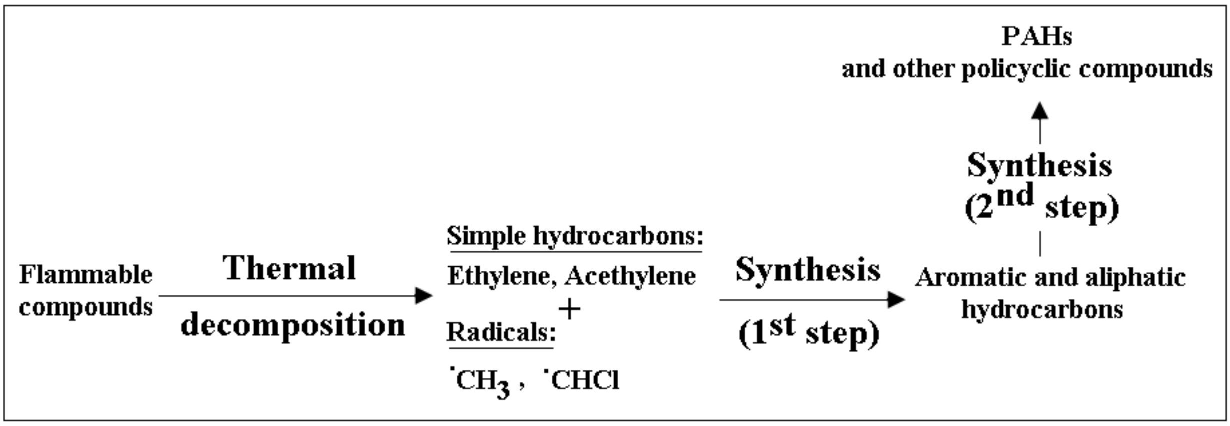

Figure 1. Pathway for PAH formation in a combustion chamber.

The results of this experience demonstrte that at 1,000°C and long residence time in combustion, all synthesis reactions lead to the formation of 4- to 6-ring PAHs. The only exception is chrysene, which undergoes significant destruction. This aspect requires further investigation to be fully understood. It is interesting to note that chrysene also differs from the other PAHs with high numbers of rings, for its better biodegradability (Urbini et al., 2014).The total lack of formation of PAHs with only 2 and 3 aromatic rings could be reasonably explained by the fact that the synthesis of high molecular weight PAHs is largely determined by condensation and dimerization reactions of the most simple PAHs (Siegmann and Sattler, 2000).

4. CONCLUSION

This study has proven that PAHs with a relevant structural complexity (5 and 6 aromatic rings) are formed in an enclosed flare combustion, operating at 1,000°C and 3.6 s residence time. This formation also produces compounds such as benzo(g,h,i)perylene, indeno(1,2,3-cd)pyrene and dibenzo(a,h)anthracene not present in raw biogas. However, the compound that has shown the highest formation in combustion is benzo(a)anthracene, the concentrations of which increase from the mean value of 6 ng Nm^{-3} to 71.5 ng Nm^{-3}; the increase of benzo(b)fluoranthene is also remarkable. PAHs with four aromatic rings have shown controversial behavior, as chrysene has undergone a noticeable destruction (this aspect requires further investigation to be fully understood), while pyrene and fluoranthene have increased their concentrations to over 70 ng m^{-3} for both compounds. Instead, PAHs with only two or three aromatic rings, are not only not present in the raw biogas, but they do not form in combustion. The pathway for the synthesis of new PAHs can be explained as a consequence of many secondary reactions. In a first decomposition step, simple hydrocarbons and hydrocarbon radicals are formed in the gas phase. These radicals are extremely reactive, so that they give rise to several synthesis reactions, including cyclization. In this way, aliphatic hydrocarbons are formed, as well as aromatic hydrocarbons, which act as precursors of the formation of much more complex hydrocarbons, among which are the PAHs. The presence of many VOCs in the raw biogas could be considered a more favorable condition for the synthesis of PAHs. The total lack of formation of PAHs with only 2 and 3 aromatic rings could be reasonably explained by the fact that the synthesis of high molecular weight PAHs is largely determined by condensation and dimerization reactions of the simplest PAHs.

5. ACKNOWLEDGMENTS

This research was supported by the ARF (Atheneum Research Funds) of Sapienza University.

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