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DEGRADATION OF BISPHENOL A AND PYRENE FROM HIGHWAY RETENTION BASIN WATER USING ULTRASOUND ENHANCED BY UV IRRADIATION

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Abstract

Due to the so-called road run-off, many various contaminants including Bisphenol A (BPA) and Pyrene (PYR) could enter the environment and retention basins. It was also suggested in the literature that their removal by using conventional treatment methods could be problematic, and modern techniques should be developed. In this study, the first attempt to remove BPA and PYR by using ultrasonication as a single process and with UV irradiation assistance was performed. The results showed that after 30 min of sonication, the degradation rate of BPA reached 92% while PYR was completely removed, however, after 1 min of the treatment degradation rate of BPA was significantly higher than PYR. In the study effect of pulsed ultrasound was also evaluated and it was found that its effectiveness in micropollutants removal could be higher than ultrasonication in continuous mode. Research revealed that the maximum removal rate of BPA and PYR was obtained during the ultrasonication process combined with UV irradiation-30 min of treatment resulted in 95% of BPA degradation. However, toxicity assessment showed that with an increase in the treatment time, an increase of toxic effects occurs. This phenomenon might be related to degradation of by-products formation which were identified in the study.

Keywords: **Retention basin water, Ultrasound, UV irradiation, Water treatment.**

1. INTRODUCTION

Recently, the development of analytical techniques has made it possible to identify many harmful pollutants in the aquatic environment. Particular attention should be paid to pollutants that are hardly biodegradable and widely spread in the aquatic environment, which could be a threat to human health, even in low concentrations such as pharmaceuticals and personal care products, pesticides, dyes, hormones, heavy metals and disinfection by-products [1, 2]. Serious health problems can be caused also by the presence of endocrine-disrupting compounds (EDCs) and polycyclic aromatic hydrocarbons (PAHs) in water [3, 4]. Due to the identification and detection of many EDCs in the environment, Bisphenol A (BPA) received much scientific and public attention [5]. BPA is the widely used name for 4’4-isopropylidenediphenol which is a colorless solid substance. It has a molecular weight equal to 228.29 g mol^{-1}, the chemical structure of $C_{15}H_{16}O_2$, and the log Kow value of BPA is 3.32 which indicates its low solubility in water [3, 6, 7]. BPA is one of the most popular EDCs and the most produced bisphenol all over the world. Moreover, the consumption of BPA in some areas is constantly growing because of its common use in polycarbonate, flame retardants, epoxy resins, and other polymer materials [8, 9]. Thus, BPA could be found in many household items such as plastic bottles, toys and CDs, thermal paper, books, paper towels, electronic devices, or even dog and human food [10–12]. It was estimated that more than 90% of people have detectable levels of BPA in their urine [13]. BPA exposure routes are gastrointestinal, respiratory, and dermal tract. Literature data show that adult person intakes from 0.03 to 0.07 μg per 1 kg body weight daily due to the diet [14]. According to Corrales et al. [15] BPA is ubiquitous in the environment and its concentration in wastewater treatment plant effluents could reach up to 370 μgL^{-1} while in surface water it could be 56 μgL^{-1}. Moreover, Ignatius et al. [16] reported that maximum concentrations of BPA were 0.2, 0.4, and 0.21 μgL^{-1} in tap, rain, and groundwater, respectively. It was proved that BPA could cause serious health issues such as reproductive and nervous system problems, or hazards for metabolic and immune function. Furthermore, exposure to BPA might be related to breast, prostate, and lung cancer [3, 17–19].

Polycyclic aromatic hydrocarbons (PAHs) are a group of hardly degradable substances which are also widely spread in the environment as a consequence of natural and anthropogenic processes such as fires, volcano eruptions, petroleum spills, tobacco smoking, and incomplete combustion of fuels. Furthermore, they are commercial use in the plastic and chemical industry, agriculture, pharmaceuticals, and photographic products [20–23]. In the environment, over 100 PAHs were already identified, however, according to U.S. Environmental Protection Agency (USEPA), 16 of them have the greatest impact on human health and the environment [24, 25]. One of the distinguished by USEPA substances is Pyrene (PYR). It is a colorless solid substance with a slight blue fluorescence. Its chemical formula is $C_{16}H_{10}$ and it contains four fused rings [6]. PYR is a persistent organic pollutant that gained scientific attention due to its high toxicity, mutability, high persistence to biodegradation, and ubiquitous in the environment. However PYR is not classified as cancerogenic for humans by International Agency Research on Cancer (IARC), literature data showed that as a result of certain processes it could be transformed into cancerogenic compounds (e.g. benzo(a)pyrene) [26–28]. Liu et al. [29] revealed that the concentration of PAHs in the surface drinking water reached 407 ngL^{-1} and the four-ring contaminants including PYR were dominant both in water and sediments. Moreover, Ma et al. [30] found that PYR concentration in soil and turfgrass could be one of the highest of PAHs.

It is a well-known fact that water quality and the health of water organisms could be affected by the transportation system. During road runoff, a complex mixture of various contaminants could enter the environment including BPA and PYR [31]. Deshayes et al. [32] found that leaching from vehicle components such as plastic and metallic bodywork, seals, automobile optics, and tires contained up to about 3.5 μgL^{-1} of BPA. Moreover, Lamprea et al. [33] proved that BPA in runoff is also related to leaching from construction materials such a concrete and pipe made by PVC. Furthermore, PYR is also connected with transport because of vehicles, road dust, and abrasion of road surface [34]. It was verified by Nguyen et. al. [35] which proved that PYR concentration in runoff reached 8.77 ngL^{-1}. As a consequence of the above-mentioned processes, recently, it was discovered that both BPA and PYR are present in the water and sediments of retention basins [36, 37]. For instance, Girardin et al. [31] indicated that PYR concentration in the sediment of sedimentation pond could be the highest among PAHs.

Due to the fact, that efficiency of the conventional treatment methods might be not sufficient in the removal of the above-mentioned substances, modern techniques should be developed [9, 21]. An interesting alternative to the traditional treatment methods is the ultrasonication process which is considered as an effective technology in the removal of many harmful substances such as bacteria, algae, dyes, viruses, industrial chemicals, pharmaceuticals and personal care products (PPCPs), and hormones (Mahvi and Dehghani, 2005; Cameron et al., 2008; Huang et al., 2017; Vega et al., 2019; Yakout et al., 2019). Nowadays, there is also the tendency to combine the ultrasonication process with other advanced oxidation processes (AOPs) due to economic reasons [43, 44]. Recently, it was also found that ultrasonication can be effective when it is combined with membrane techniques because of the so-called fouling limitation [45–47]. According to literature data, ultrasonication could be effective also in BPA and PYR removal from the environment [4, 8, 9, 21, 48–53]. However, there is a lack of knowledge about the removal of BPA and PYR from the retention basins water matrices by using ultrasound, thus, the study is the first research on determining ultrasound (US) technology efficiency for BPA and PYR removal from highway retention basin. Furthermore, the effect of combined US and UV irradiation (US/UV) processes was determined. In this paper, to evaluate the effectiveness of the above-mentioned techniques and to determine generated by-products, gas chromatography (GC-MS) was used. Moreover, the potential toxic effect of the solution after treatment was evaluated by using Microtox ® bioassay.

1.1. Ultrasonication process principles

Currently, ultrasound is used in many different fields such as medicine, cleaning, soil remediation, desizing of cotton fabric, oil desulfurization, pipeline fouling removal, pelleting, treatment of sewage sludge, water disinfection, and wastewater treatment [54–60]. In the ultrasonication process, inaudible sound frequencies above 20 kHz are used to form the initial bubble which is growing until it violently collapses in the so-called compression phase of pressure oscillation. This phenomenon is described in the literature as acoustic cavitation [61]. Most of the researchers defined that during bubble collapse, local temperature and pressure can reach up to 5000 K and 100 MPa, however, there are some literature reports which indicated that these parameters can even rise to the order of 10000 K and 400 MPa, respectively (Flannigan and Suslick, 2005; Inoue et al., 2008; Lim et al., 2014; Tzanakis et al., 2016). Furthermore, during bubble implosion, shock waves, microjets, and shearing forces occur. This phenomenon is also accompanied by hydroxyl radicals, ozone, and hydrogen peroxide generation. So, the pollutants removal is connected with sonochemical and hydrodynamical effects [46, 66, 67]. Thermal decomposition of water and pollutants removal follow the equations [9, 68]:

$$H_2O + US \rightarrow H^\bullet + HO^\bullet \quad (1)$$

$$H^\bullet + HO^\bullet \rightarrow H_2O \quad (2)$$

$$H^\bullet + H^\bullet \rightarrow H_2 \quad (3)$$

$$HO^\bullet + HO^\bullet \rightarrow H_2O_2 \quad (4)$$

$$HO^\bullet + HO^\bullet \rightarrow H_2O + O \quad (5)$$

$$Pollutants + H_2O_2 \rightarrow Degradation\ products \quad (6)$$

$$Pollutants + HO^\bullet \rightarrow Degradation\ products \quad (7)$$

In general, there are two types of acoustic cavitation devices – ultrasonic horn in which the ultrasonic probe is directly immersed in the treated solution, and the ultrasonic bath where the transducer is located under the container with the solution. Generally, ultrasonic horns are characterized by the small diameter of the probe, so they can generate ultrasound with relatively high intensity. However, in this type of device, the ultrasound frequency is in most cases lower than in ultrasonic baths [44, 61]. Acoustic cavitation as a very complex process which depends on many factors, such as type of treated substance, temperature, the intensity of ultrasound, pH of the treated solution, frequency, surface tension, shape and volume of the reactor, etc. [69–75].

To enhance the effectiveness of the ultrasonication process it could be combined with other AOPs (e.g. ozonation, hydrogen peroxide, UV irradiation) [44] or other diverse sonocatalysts and sonosensitizers can be used (e.g. TiO_2, ZnO, porphyrin, xanthone, curcumin, hypocrellin, etc.) [76]. UV irradiation is one of the most well-described method in micropollutants decomposition which depends on many parameters of treated solution and operational parameters such as appropriate wavelength and energy [77]. It was found to be effective in many pollutants removal, including BPA and PYR [78, 79]. However, AOPs could lead to the formation of intermediates which can be harmful to human health [80].

2. MATERIALS AND METHODS

2.1. Materials and reagents

The samples used in this study were collected from the retention basin located close to the highway characterized by a pH value of 7.3, turbidity equal to 0.55 NTU, and conductivity 370 μS/cm. These are the average values of three samples taken from the rim part of the basin, around 15 cm below the water surface. Then, separated solutions containing 1 mg L^{-1} of analytical grade BPA and PYR supplied by Sigma-Aldrich (Poznań, Poland) were prepared. In Table 1 properties of tested compounds are shown.

Table 1.
Chemical and physical properties of tested compounds [6]

Property	Value/description	
	BPA	PYR
Molecular formula	$C_{15}H_{16}O2$	C_6H_{10}
Molecular weight	228.29 g mol^{-1}	202.25 g mol^{-1}
Boiling point	360°C	404°C
Melting point	160°C	151°C
Solubility in water at 25°C	300 mg L^{-1}	0.135 mg L^{-1}
Octanol-water partition coefficient Log (K_{ow})	3.32	5.18

The turbidity of the sample was measured by the use of HI-93414-02 Turbidity meter by HANNA Instruments Inc. while the pH and conductivity was measured by using CPC – 505 device provided by Elmetron (Zabrze, Poland). The water quality was also assessed indirectly by total organic carbon (TOC) measurement using TOC – L Analyzer (Shimadzu, Kioto, Japan). Samples were treated in a 100 ml volume reactor by using ultrasound equipment (ultrasonic horn type) Sonics VCX 500 provided by Vibracell Sonics, Sonics and Materials Inc., (Newtown Connecticut, USA). It was characterized by a 20 kHz frequency and a maximum power of 500 W. In the study, a 13 mm diameter probe was used which can generate 114 μm maximum amplitude. During the second stage of the study, the ultrasonication process was assisted by UV irradiation. To carry out this experiment, a UV lamp provided by Heraeus (Hanau, Germany) with a power of 150 W and wavelengths equal to 313, 365, 405, 436, 546, and 578 nm was used. To ensure a uniform irradiation level, the treated samples were placed in the reactor 90 s after the UV lamp lightening up. To avoid overheating of the UV lamp and the treated solution, a cooling system with tap water as a flow medium was used. To provide mixing of the treated solution, the reactor was placed on the magnetic stirrer (Fig. 1). Both BPA and PYR degradation was tested at intensities of 9, 17, 26, and 43 W/cm^2 in the 1, 5, 10, 20, and 30 min sonication. In the study, two pulse ultrasound modes were also tested, namely R=4:2 (4 s on, and 2 s off mode) and R=2:4 (2 s on and 4 s off mode). Pulsed ultrasound experiments were carried out at the intensity of 43 W/cm^2 and 1, 5, 10, 20, 30 min sonication time.

2.2. Gas chromatography analysis

To evaluate the efficiency of the process and to identify by-products present in post-treated samples, GC-MS analysis was performed by using a 7890B gas chromatograph supplied by Perlan Technologies (Warsaw, Poland). To increase the chromatography

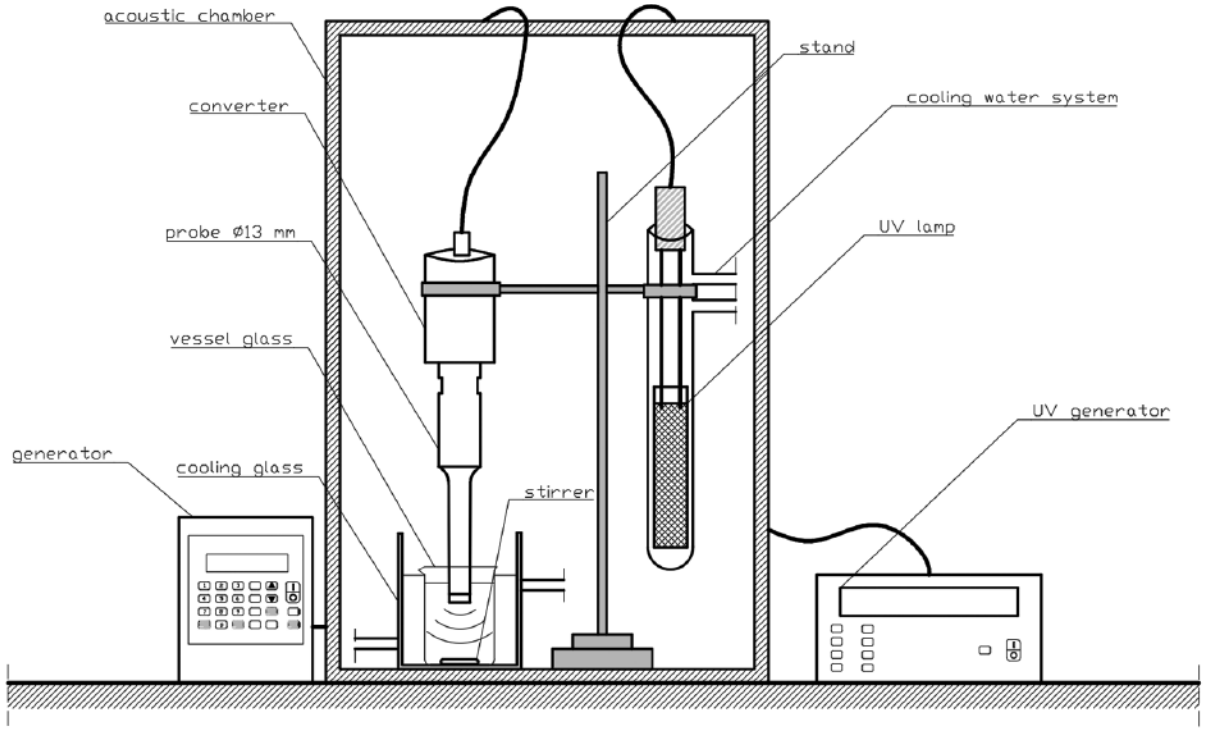

Figure 1.
Experimental setup used in the study

analysis accuracy, the solid-phase extraction (SPE) method was used. In this process, Supelclean^{TM} ENVI^{TM}-18 cartridges supplied by Sigma-Aldrich (Poznan, Poland) with an Octadecylsilane (C_{18}) as a cartridge bed were applied. In the conditioning and elution stage, 5 mL of methanol (MeOH), 5 mL of pure water (H_2O), and dichloromethane (DCM) were used, respectively. The SPE was carried out in 5-10 kPa of negative pressure, while the flow rate was set to 1 mL/min. During GC-MS analysis helium was used as a carrier gas and it flowed at a rate of 1.1 mL/min. The temperature of the ion source, trap, and injector was set to 230, 150, and 250°C, respectively. The capillary columns (0.25 µm thickness SLB^{TM} – 5 ms 30 m × 0.25 mm) were obtained from Sigma-Aldrich (Poznan, Poland). The degradation rate of the selected micropollutants from retention basin water was calculated according to the peak areas measurements which were compared with calibration curves estimation data. Limit of quantification LOQ and limit of detection LOD in GC-MS analysis was 8.5 µg L^{-1} and 0.05 ng L^{-1} for BPA solutions , while it was 6.2 µg L^{-1} and 0.03 ng L^{-1} for PYR solutions, respectively. It was evaluated that the recovery of BPA and PYR was qual to respectively 99 and 98%.

2.3. Toxicity assessment

To determine the toxicity of tested solutions, the Microtox ® test was used. The toxic potential was evaluated by an intensity of light emission of *Aliivibrio fischeri* which is highly sensitive to toxic micropollutants luminescent bacteria [1, 81]. The tests were performed according to Microtox Omni system procedure in the Microtox 500 analyser (Warsaw, Poland). Moreover, as described in [82, 83], the toxicity class of the treated solutions was determined.

2.4. Data analysis

Ultrasonic degradation of BPA and PYR from the samples, similarly like most of the other organic matter, followed pseudo-first kinetics order, expressed by equation [9]:

$$\ln\left(\frac{C}{C_0}\right) = -kt \tag{8}$$

where C is the concentration of pollutants at a given time t, C_0 is their initial concentration, and k represents the first-order reaction constant.

Based on the k value, the half-life of BPA and PYR $t_{1/2}$ was calculated as [52]:

$$t_{1/2} = \frac{0.693}{k} \tag{9}$$

In the study, acoustic pressure P_a was also determined by the following equation [84]:

$$P_a = \sqrt{2\rho c I} \tag{10}$$

where ρ is the water density (1000 kg/m^3), c is the sound speed in the water (1500 m/s), and I is the ultrasonic intensity described by acoustic power P per area of the transducer A.

3. RESULTS AND DISCUSSION

3.1. Effect of intensity and time on BPA and PYR degradation

Fig. 2 and Fig. 3 show the decrease in the concentration of BPA and PYR during ultrasound treatment at a constant frequency of 20 kHz.

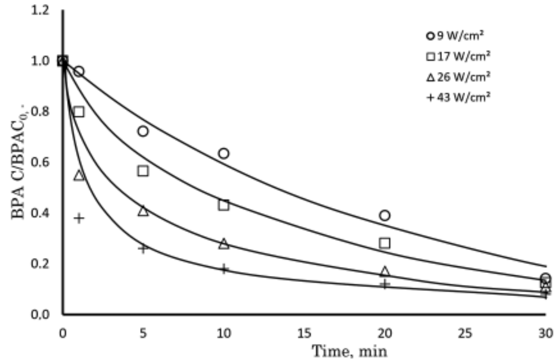

Figure 2.
Ultrasound intensity influence on BPA degradation (9, 17, 26 and 43 W/cm^2)

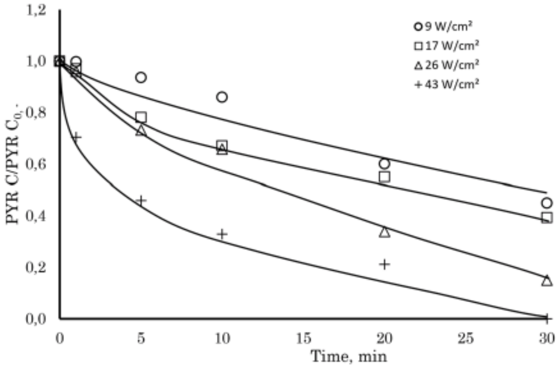

Figure 3.
Ultrasound intensity influence on PYR degradation (9, 17, 26 and 43 W/cm^2)

The degradation rate of mentioned compounds was proportional to the irradiation time and ultrasound intensity. Thus maximum reduction of BPA was achieved during 30 min sonication at the intensity of 43 W/cm^2 and it was 92%. PYR in the same conditions of the process was completely removed from the solution. However, during 1 min of ultrasonication at the highest intensity degradation rate of BPA was significantly higher than PYR and it was 62 and 30%, respectively. Moreover, results showed that ultrasound treatment can be effective in BPA removal even at the lowest intensity. At 9 W/cm^2 degradation rate after 30 min of sonication was 86%. The turbidity and TOC value of the treated solution increased after the sonication process. It might be explained by the formation of BPA and PYR intermediates or the cavitational erosion of the sonotrode used in the study. Obtained data showed also that contaminants degradation followed the first kinetic order. The calculated degradation rate constants of BPA were 0.053, 0.110, 0.213, and 0.320 respectively for intensities of 9, 17, 26, and 43 W/cm^2, while the acoustic pressure varied from 0.52 to 1.14 MPa. Degradation rate constant for PYR were 0.018, 0.036, 0.052 and 0.21, respectively. Increasing the ultrasound intensity, the half-life time of contaminants degradation decreased which indicates an improvement in the process efficiency. Calculated parameters are shown in table 2.

Table 2.
Parameters of the ultrasonication process

Compound (-)	I (W/cm^2)	P (W)	P_a (MPa)	k (1/min)	$t_{(1/2)}$ (min)
BPA	9	12	0.52	0.053	13
	17	22	0.71	0.110	6
	26	35	1.02	0.213	3
	43	57	1.14	0.320	2
PYR	9	12	0.52	0.018	39
	17	22	0.71	0.036	19
	26	35	1.02	0.052	13
	43	57	1.14	0.21	3

I – ultrasound intensity, P – ultrasound power, P_a – acoustic pressure, k – first order reaction constant, $t_{(1/2)}$ – half-life of degradation reaction

Similarly, David [4] in the case of PYR obtained a constant rate equal to 0.029 during ultrasonication at 20 kHz frequency and 6.1 W/cm^2 intensity. Moreover, similar results were found by Manariotis et al. [52] (0.021 at the frequency of 862 and 1142 kHz). Zhang et al. achieved a rate constant of BPA degradation from 0.0307 to 0.1910 at 3.0 W/cm^2.

3.2. Effect of pulsed ultrasound

Pulse ultrasound occurs, while the ultrasound irradiation is intermitted by so-called silent cycles of specific duration. Literature data showed, that it could be an effective method for micropollutants removal. The results of the pulse ultrasound influence on BPA and PYR degradation are shown in Fig. 4 and Fig. 5.

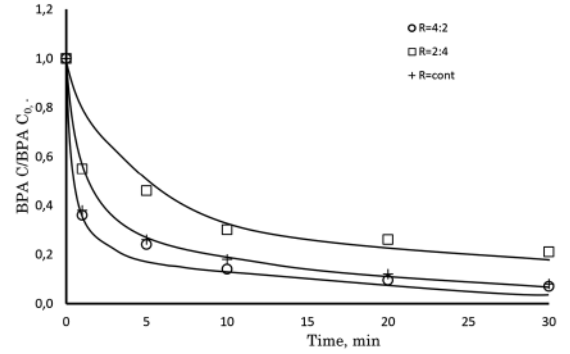

Figure 4.
Ultrasound influence on BPA degradation at pulse ultrasonication modes (R=4:2, R=2:4) and continuous mode (R=cont)

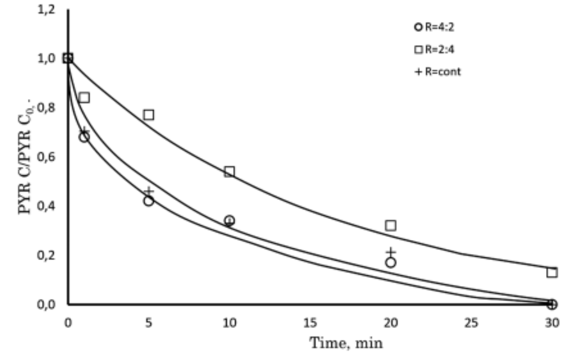

Figure 5.
Ultrasound influence on PYR degradation at pulse ultrasonication modes (R=4:2, R=2:4) and continuous mode (R=cont)

Research revealed that the highest pollutants removal efficiency was obtained in R = 4:2 mode. During 30 min of ultrasonication, BPA degradation degree reached 93%, while PYR was completely removed from the solution, however, the difference in pollutants removal efficiency between continuous and R = 4:2 mode was not significant. In general, during sonication (on-time) bubbles are generated and they are growing in the acoustic cycles while during the silent cycle, bubbles as a result of shrinkage by dissolution could actively cavitate through the next on-cycle. Thus, there is time for diffusion of the contaminants to the cavitation bubbles, and the diffusion coefficient is inverse proportional to the molecular weight of the pollutant [38, 85].

However, the silent cycle may be related also to bubbles disappearance due to buoyancy force or dissolution [86]. It was proved in the study that ultrasound working in R=2:4 pulse mode had the lowest efficiency of BPA and PYR removal. It can be caused by too long off-cycle and as a consequence, sonochemical effects disappearance [87]. In mentioned mode, the degradation rate of these pollutants after 30 min sonication was respectively 79 and 87% which was 13% lower compared to the continuous mode.

3.3. Effect of UV irradiation

In the last stage of the study effect of ultrasound and UV irradiation (US/UV) in BPA and PYR removal as a combined process was tested. The results showed that this method had the highest efficiency of the treatment (Fig. 6, Fig. 7).

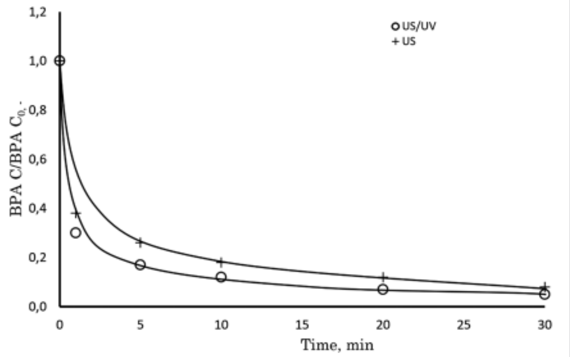

Figure 6.
The degradation rate of BPA in US/UV and US process (intensity of 43 W/cm^2)

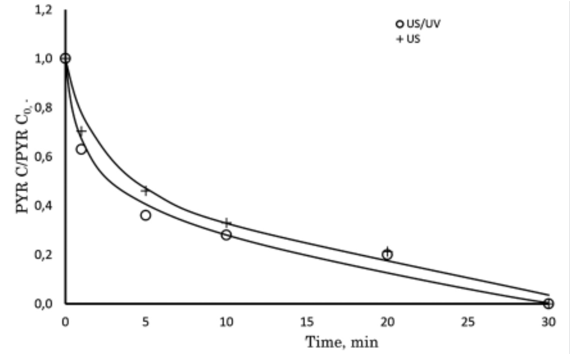

Figure 7.
The degradation rate of PYR in US/UV and US process (Intensity of 43 W/cm^2)

Samples were treated at the intensity of 43 W/cm^2 and the ultrasonic equipment was set to continuous mode. As a consequence of the US/UV process, after 30 min of treatment 95% of BPA was removed from the solution which was the highest BPA degradation rate obtained in this study. After 1 min. of the treatment, BPA was degraded at the level of 70% which was 8% more effective than continuous ultrasound used as a single process at the same intensity. Similarly, UV irradiation enhanced the reduction rate of PYR and at 1 min. of the treatment its reduction rate was 37%, thus the improvement of 7% was achieved (comparing to ultrasound treatment in continuous mode). Moreover, after 30 min PYR was eliminated from the solution. The constant rate was equal to 0.230 and 0.401 1/min for PYR and BPA, respectively. Although removal of BPA and PYR during the US/ UV process was at a high level, the turbidity also increased as previously described. Moreover, total organic carbon (TOC) reduction in the post-treated samples was not observed which might be further evidence for the formation of by-products in the solution.

3.4. Toxicity assessment and degradation by-products

As a consequence of reaction between $HO^{\bullet}$ and other highly-reactive species with the micropollutants, some decomposition by-products could be generated. The chromatographic analysis of the post-treated samples revealed the occurrence of several intermediates. It was found that after 30 min of sonication the solution containing BPA at the maximum intensity, 4-[2-(4-hydroxyphenyl)propan-2-yl]benzene-1,2-diol, 4,4'-isopropylidenebiscatechol, 4'-hydroxyacetophenone, and 4-hydroxybenzoic acid were generated. Torres et al. 2008 [51] also confirmed the formation of 4-[2-(4-hydroxyphenyl)propan-2-yl]benzene-1,2-diol and 4'-hydroxyacetophenone in samples subjected to the sonication process of BPA water solutions. In addition they identified also other intermediates with molecular weights ranged from 242 to 150 g mol^{-1}. The formation of these by-products of BPA decomposition has also been demonstrated in previous studies conducted by [88–90]. Moreover, chromatographic analysis of PYR containing samples after the same time of ultrasonic treatment also revealed the occurrence of some by-products, namely 1-hydroxypyrene, 3-phenanthrenol, 9-phenanthrenol, and 1,4-benzoquinone. It can be concluded that the identified products resulted mainly from the hydroxylation of their parent compounds.

As shown in Table 3, the number of identified compounds was proportional to the sonication time.

Table 3.
BPA and PRN intermediates identified in samples after ultrasonication

Compound	Intermediate	Process time, min				
		1	5	10	20	30
BPA	4-[2-(4-hydroxyphenyl)propan-2-yl]benzene-1,2-diol	-	-	-	+	+
	4,4'-isopropylidenebiscatechol	-	-	-	+	+
	4'-hydroxyacetophenone	-	-	-	-	+
	4-hydroxybenzoic acid	-	-	-	-	+
PYR	1-hydroxypyrene	-	-	+	+	+
	3-phenanthrenol	-	-	-	+	+
	9-phenanthrenol	-	-	-	+	+
	1,4-benzoquinone	-	-	-	-	+

(+) – presence of the compound in the sample, (-) – no presence of the compound in the sample

Noteworthy, during US/UV as an integrated process conducted on BPA removal, additionally 4-(2-hydroxy-2-propanyl) phenol and 1,4-benzoquinone were identified as decomposition by-products. Similarly, in the PYR solution two more intermediates were found, namely, 4-phenanthrenol and 1,2-benzenediol. Furthermore, the generation of the intermediates was faster in the US/UV treatment than in ultrasonication used as a single process (Table 4). The intermediates identified in samples after the US/UV process were generated in the same way like those noted in solutions after single ultrasonication. The applied analytical procedure did not allow for the identification of compounds formed in the second step of compound decomposition. During this second step the opening of the aromatic ring by breaking the bonds between carbon atoms (C-C) should be observed. Although in the case of PYR the presence of compounds with a lower number of aromatic rings such as 3-phenanthrenol, 4-phenanthrenol, 9-phenanthrenol, 1,2-benzenediol, 1,4-benzoquinone was noted. In the Fig. 8 and Fig. 9 proposed pathways of BPA and PYR decomposition during UV/US are shown.

Table 4.
BPA and PRN intermediates identified in samples after ultrasonication supported by UV light

Compound	Intermediate	Process time, min				
		1	5	10	20	30
BPA	4-[2-(4-hydroxyphenyl)propan-2-yl]benzene-1,2-diol	-	+	+	+	+
	4,4'-isopropylidenebiscatechol	-	+	+	+	+
	4-(2-hydroxy-2-propanyl) phenol	-	-	+	+	+
	4'-hydroxyacetophenone	-	-	+	+	+
	4-hydroxybenzoic acid	-	-	-	+	+
	1,4-benzoquinone	-	-	+	+	+
PYR	1-hydroxypyrene	-	-	+	+	+
	3-phenanthrenol	-	-	-	-	+
	4-phenanthrenol	-	-	-	-	+
	9-phenanthrenol	-	-	-	+	+
	1,2-benzenediol	-	-	-	-	+
	1,4-benzoquinone	-	-	-	+	+

(+) – presence of the compound in the sample, (-) – no presence of the compound in the sample

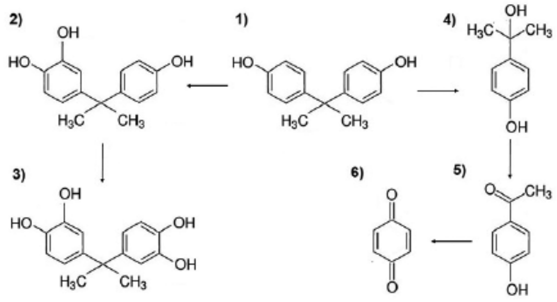

Figure 8.
Pathway of generation of BPA decomposition by-product (where: 1) BPA,
2) 4-[2-(4-hydroxyphenyl)propan-2-yl]benzene-1,2-diol,
3) 4,4'-isopropylidenebiscatechol,
4) 4-(2-hydroxy-2-propanyl) phenol, 5) 4'-hydroxyacetophenone, 6) 1,4-benzoquinone)

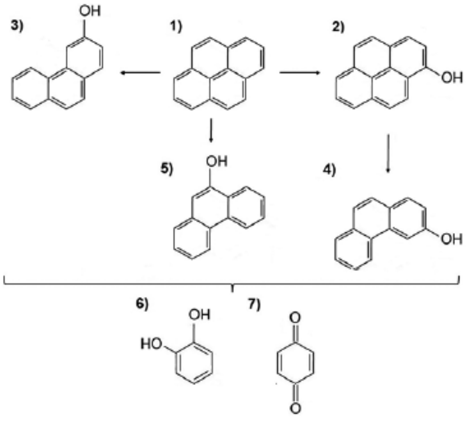

Figure 9.
Pathway of generation of PRN decomposition by-product (where: 1) PRN,
2) 1-hydroxypyrene, 3) 3-phenanthrenol,
4) 4-phenanthrenol, 5) 9-phenanthrenol,
6) 1,2-benzenediol, 7) 1,4-benzoquinone)

Performed Microtox® assessment proved that both ultrasonication and UV irradiation treatment of BPA and PYR were related to a significant increase in toxicity which might be a result of not complete mineralization of the pollutants. The toxic effect varied from 8 to 60% and from 10 to 74% for BPA and PYR, respectively. It is known that toxicity analysis could be an indirect way to evaluate the oxidation intermediates formation [1], thus increase of this parameter could be explained by the occurrence of oxidation by-products listed in Table 4. Obtained results were classified into the four classes of solution toxicity, described in [82, 83]. As shown in Fig. 10 and Fig. 11 after 30 min of the treatment, BPA and PYR solutions change the toxicity characteristics from non-toxic to toxic. As mentioned before, the increase in toxicity is associated with the formation of compounds with potentially toxic effects on living organisms. For example, the presence of phenanthrenols in PYR post-treated solutions increases their toxicity, which was confirmed by [91]. Furthermore, benzoquine, which was identified in post-treated solutions of BPA and PYR is also related toxic effects on organisms [92]. It is a recognized human carcinogen, which can form highly reactive to harmful metabolites [93]. The results are also consistent with the turbidity, TOC, and conductivity values which increased after treatment. After 30 min of US/UV treatment of solution containing BPA at maximum intensity turbidity increased from 0.7 to 6.0 NTU, conductivity from 313 to 342 μS/cm and TOC from 291 to 300 mg L^{-1}, while in case of solution containing PYR it was respectively from 9.4 to 16.2 NTU, from 386 to 407 μS/cm, and from 395 to 432 mg L^{-1}.

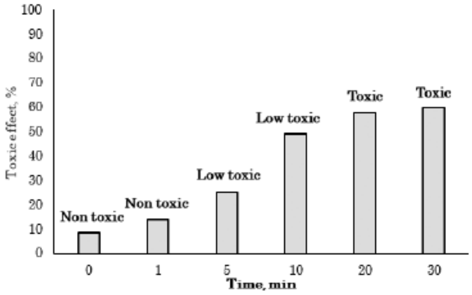

Figure 10.
Toxicity of the samples after US/UV treatment for BPA removal

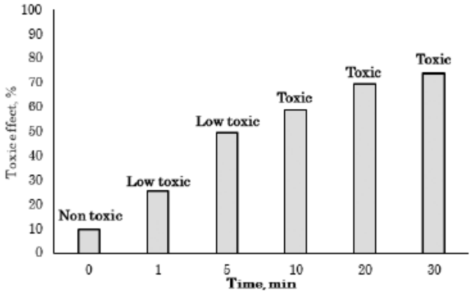

Figure 11.
Toxicity of the samples after US/UV treatment for PYR removal

Similarly, the increase of the toxicity after the treatment by advanced oxidation processes was observed in [1, 81].

4. SUMMARY

To sum up, in this study first attempt on BPA and PYR from highway retention basin removal by using ultrasound treatment was performed. These substances could be found in that water matrices due to e.g. leaching from construction materials, pipes, car elements, and as a result of so-called road run-off. In the experiments, the effect of continuous and pulsed ultrasound in BPA and PYR reduction was examined as well as the influence of ultrasound assisted by UV irradiation. It can be concluded that pulsed ultrasound in R=4:2 mode enhanced the removal efficiency due to increased time of the sonochemical effects. The effect of ultrasound treatment was improved also by UV irradiation assistance. As a result, the degradation efficiency of BPA and PYR was as follows: US/UV > US (R=4:2) > US (set to continuous mode). However, in the post-treated samples, there was no reduction in turbidity, TOC, and toxicity. This relation might be caused by the generation of some by-products which were recognized in the research. Obtained results showed that despite the high degradation rate of the BPA and PYR, water still might be harmful to human health and intermediates identification is a very important issue. In general, ultrasound is considered as a perspective technology, especially combined with other advanced oxidation processes, however, its further development is still needed among others to scale it up and use industrially.

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ABBREVIATIONS

BPA – Bisphenol A
PYR – Pyrene
EDCs – Endocrine disrupting compounds
PAHs – Polycyclic aromatic hydrocarbons
IARC – International Agency Research on Cancer
AOPs – Advanced oxidation processes
US – ultrasound
GC-MS – gas chromatography

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