Investigation of deposited material in an atmospheric pressure DC corona discharge in artificial air containing a trace of benzene, toluene or xylene

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1. INTRODUCTION

1.1 Background

According to the statistics of Japan[1], in 2009 about 156,000 tons of toxic chemical compounds were emitted to the atmosphere.

Aromatic compounds such as toluene and xylene occupy 75% of the emission.

Recently, many approaches for decomposing these substances using discharge plasma have been done, and the decomposition rate and efficiency are mainly discussed.

The decompositions of OA at the discharge are presented in the table below.

<table>
<thead>
<tr>
<th>Compound</th>
<th>Decomposition Rate</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toluene</td>
<td>95%</td>
<td>90%</td>
</tr>
<tr>
<td>Xylene</td>
<td>85%</td>
<td>80%</td>
</tr>
<tr>
<td>Benzene</td>
<td>75%</td>
<td>70%</td>
</tr>
<tr>
<td>others</td>
<td>60%</td>
<td>60%</td>
</tr>
</tbody>
</table>

1.2 Decomposition processes of plasma in the discharge plasma

Decomposition processes of plasma in the discharge plasma are presented in the figure below.

![Decomposition processes of plasma in the discharge plasma](image)

1.3 Recent works for the detailed analysis in the decomposition of aromatic compounds in the discharge plasma

Huang et al.[2] studied the decomposition of toluene in an atmospheric dielectric barrier discharge.

The decomposition rate of toluene in the discharge plasma is presented in the figure below.

![Decomposition rate of toluene in the discharge plasma](image)

Nagata et al.[3] studied the decomposition of benzene, toluene and xylene in an atmospheric DC corona discharge.

The decomposition rate of benzene, toluene and xylene in the discharge plasma is presented in the figure below.

![Decomposition rate of benzene, toluene and xylene in the discharge plasma](image)

1.4 Objective

To clarify the decomposition processes of benzene, toluene and xylene in detail

We minutely investigate gaseous by-products and deposited material from benzene, toluene and xylene in an atmospheric pressure DC corona discharge, and estimate the decomposition processes of benzene, toluene and xylene.

2. EXPERIMENTAL APPARATUS

2.1 Discharge chamber

A cylindrical discharge chamber, made of acrylic and nylon resin, is used. An inner diameter and a height of the chamber are 40 mm and 61 mm, respectively.

Electrodes: A multi-needle and a plate electrodes

- A multi-needle electrode consists of 18 clusters, each of which has 6 stainless-steel needles of 1.5 mm in diameter and 16 mm in length with 4 mm in interval.
- A plate electrode is made of stainless-steel plate with 150 mm in width and 55 mm in length, and cathode.
- A distance between the multi-needle electrode and the plate electrode is fixed at 15 mm.

2.2 Applied voltage and discharge current

- Applied voltage: +14.4 – 14.8 kV (DC)
- Discharge current: 1.0 – 1.1 mA
- Input power: 15 W
- Discharge duration: 30 min

2.3 Gas condition

- The mixture ratio of a background gas: N₂/O₂ = 80/20 (vol%)
- The initial concentration of C₆H₆, C₆H₅CH₃, and C₆H₅(CH₃)₂ = 800 ppm
- Gas flow rate: 3.0 L/min

2.4 Analyses of gaseous by-products and deposited material

- Gaseous by-products from benzene, toluene and xylene are identified by a Gas Chromatograph Mass Spectrometer (Shimadzu, GCMS-QP2010 Plus) and a Fourier Transform Infrared Spectrometer (Shimadzu, FTIR-8900) equipped with a gas cell (Infrared Analysis, 10-PX), which has an optical path length of 10 m.
- The transmittance of the deposited material on a Si wafer (10 mm × 90 mm) on the plate electrode is measured by the FTIR.

![Analyses of gaseous by-products and deposited material](image)

3. RESULTS & DISCUSSION

3.1 Infrared absorbance spectra with and without discharge

- The infrared absorbance spectra with and without discharge are presented in the figure below.

![Infrared absorbance spectra with and without discharge](image)

3.2 Chromatograms of sampled material

- The chromatograms of sampled material are presented in the figure below.

![Chromatograms of sampled material](image)

3.3 Concentrations of gaseous by-products

- The concentrations of gaseous by-products are presented in the table below.

<table>
<thead>
<tr>
<th>Material</th>
<th>Concentration (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toluene</td>
<td>0-150 (32.4 %)</td>
</tr>
<tr>
<td>Xylene</td>
<td>0-150 (32.4 %)</td>
</tr>
<tr>
<td>Benzene</td>
<td>0-150 (32.4 %)</td>
</tr>
</tbody>
</table>

3.4 Estimated decomposition processes of C₆H₆, C₆H₅CH₃, and C₆H₅(CH₃)₂

- The estimated decomposition processes of C₆H₆, C₆H₅CH₃, and C₆H₅(CH₃)₂ are presented in the figures below.

![Estimated decomposition processes of C₆H₆, C₆H₅CH₃, and C₆H₅(CH₃)₂](image)

4. CONCLUSION

Gaseous by-products and deposited material from benzene, toluene and xylene in an atmospheric pressure DC corona discharge are minutely investigated, and the decomposition processes of benzene, toluene and xylene are estimated.

- It is likely that phenyl radicals, benzylic radicals, methyl phenyl radicals and methyl benzylic radicals are produced from benzene, toluene and xylene at first, because the dissociation energy of C₆H₆, C₆H₅CH₃ and C₆H₅(CH₃)₂ is lower than the binding energy of an aromatic ring as shown in the table.
- Polyphenols, which can be produced by the polymerization of these radicals, deposit on the electrode, and the fragments from the cleavage of these radicals also deposit.
- In the gas phase, these radicals are mainly converted into CO₂, CO and HCOOH FAH, etc.
- Benzenyl radicals and methyl benzylic radicals react with O radicals[4] and benzaldehyde radicals[5] are produced from toluene and xylene, respectively.
- Further, benzylic radicals and methyl benzylic radicals are decomposed in the corona discharge, and fragments of these radicals are also converted into the gaseous products and the deposit.

![Disintegration energy of radicals](image)

- The disintegration energy of radicals is presented in the table below.

<table>
<thead>
<tr>
<th>Radical</th>
<th>Disintegration Energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C₆H₆</td>
<td>4.5 eV</td>
</tr>
<tr>
<td>C₆H₅CH₃</td>
<td>3.7 eV</td>
</tr>
<tr>
<td>C₆H₅(CH₃)₂</td>
<td>3.3 eV</td>
</tr>
</tbody>
</table>

- Methyl phenyl radicals and phenyl radicals are probably produced from the fragments of toluene and xylene containing the methyl group.
- In the deposited material, substituted benzene, polyenes, C=O, C=O-C and C-H, which are regarded as the fragments of benzene, toluene and xylene, are mainly contained.
- It is likely that phenyl radicals, benzylic radicals and methyl phenyl radicals are produced from benzene, toluene and xylene at first, and these radicals are converted into CO₂, CO, HCOOH, FAH, etc.