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Component Analysis of Particulate Products in Electron Beam-Irradiated Xylene/Air Mixtures Using an Atmospheric Pressure Ionization Mass Spectrometer

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Particulate and gaseous organics are formed, as transient products, from dilute concentrations of xylene in air under electron beam irradiation; however, the components of the particulate organic products and the particle formation process are unknown. In the present study, the component analysis of particulate products were performed employing an atmospheric pressure ionization mass spectrometer (APIMS) based on a temperature-programmed desorption method to identify the components of the particles from *o*-xylene as a function of their gasification temperatures. Ions of 139–203 u with a peak interval of the m/z of 16 u, the same as the value of O^+, were observed as vaporized particle components at correspondent temperatures. The ions of 43, 47, 61, 97, 111, 135, and 139 u were detected as the vaporized components at 333 K. The ions of 155, 171, and 187 u were observed at 363 K, besides that of 203 u at 393 K. The ions of 392–421 u were also appreciable with peak heights of 10^{-2} order lower than those for the ions of 139–203 u at temperatures higher than 393 K. The gaseous products in irradiated *o*-xylene/air mixtures were also analyzed with the APIMS. The ions with m/z lower than 139 u were detected as gaseous products.

Particulate and gaseous organics are formed, as transient products, from dilute concentrations of aromatic hydrocarbons in air under electron beam (EB) irradiation.[1–3] Carbon dioxide and CO are exclusively produced as final products through further oxidation of gaseous organic products by irradiation.[3] The particulate organic products showed resistance against the oxidation reaction with free radicals relative to gaseous organic products. This characteristic seems to originate from either the condensed form of the particulate products in the gas phase or the chemical structure of particulate product components. For the purification of xylene/air mixtures, the particulate products should be oxidized into CO_2 in the gas phase; otherwise, they have to be removed from the gas phase with a filter after the EB treatment of aromatic hydrocarbon/air mixtures, because they accumulate in the respiratory organs and impact human health. The chemical structures of particulate product components and their vapor pressure values are necessary to clarify their chemical stabilities to oxidation and inhibit the formation of the particles with heating of air mixtures, respectively. Their vapor pressures also serve to understand the formation process of the product particles.

The chemical components of EB-induced particulate products from aromatic hydrocarbons, which are collected on a filter and dissolved in solution, have been analyzed by liquid chromatography, FT-IR spectrometry, and an organic acid analyzer.[1–3] Organics having no benzene ring, in other words nonaromatic hydrocarbons, were frequently identified as one of the particulate product components[2,3] by FT-IR spectrometry and an organic acid analyzer. The molecular weights of the particulate product components, analyzed by mass spectrometry, such as GC-MS and a LC-MS, are fundamental information to examine the chemical structures of the particulate product components, but difficult to find in other literature. For the analysis with GC-MS and LC-MS, the particulate products are generally collected and concentrated in an appropriate solution once, and the proper measurement conditions such as column materials, column temperature, and eluent should be selected based on the chemical properties of unknown substances. Mass spectrometry without separation process using columns and elements seem, accordingly, to be required for the measurement of the entire molecular weights of all the unknown substances like irradiation-induced particulate products. The analyses by mass spectrometry using a temperature-programmed desorption (TPD) method seem to be promising for the identification of the components of particles as one of the above methods. The temperatures for gasification of particulate product components and the molecular weights of vaporized particulate product components can be concurrently measured based on the mass spectrum at elevated temperatures. The analysis with a high-sensitivity mass spectrometer by a TPD method enables the examination of the molecular weights of trace amount components of the particulate products vaporized at lower temperatures. The molecular weights of the particulate product components can be, consequently, measured by such a mass spectrometer with considerable suppression of their pyrolysis.

An atmospheric pressure ionization mass spectrometer (APIMS) is suitable for such an analysis of extremely trace amounts of vaporized particulate products in inert carrier gases,[4–7] since it can analyze continuously trace substances at concentrations of ppb to ppt levels in atmospheric gases.

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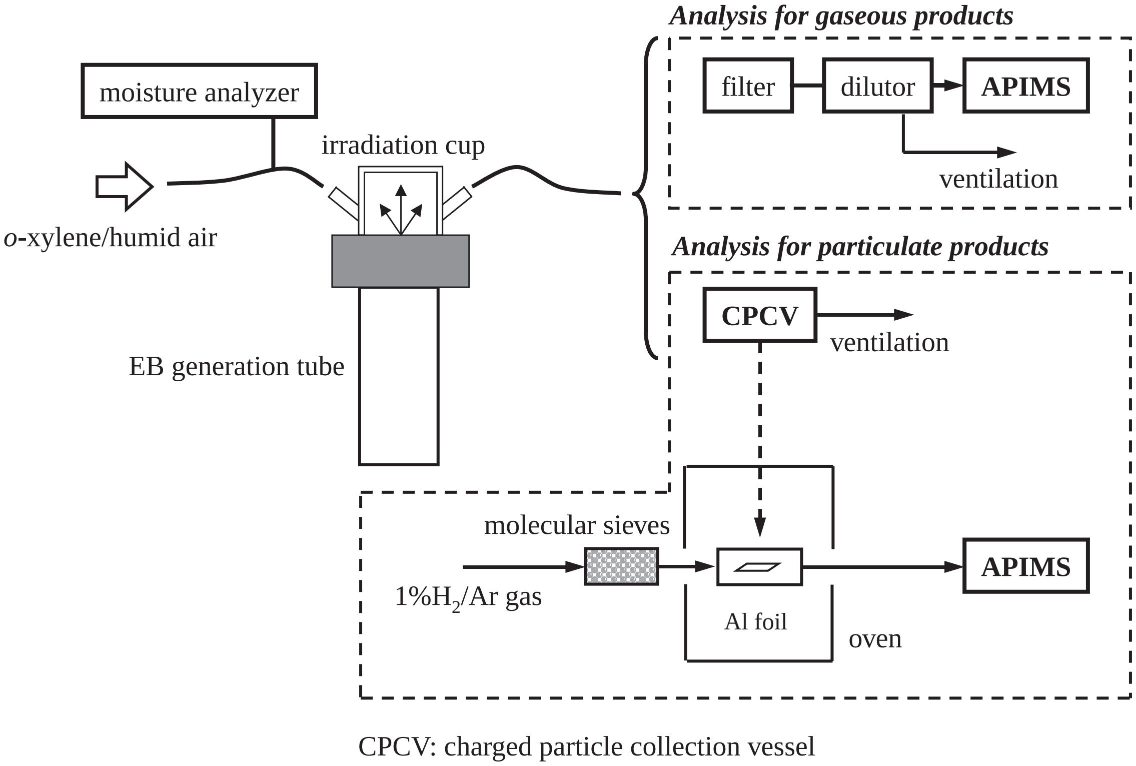

Fig. 1. Apparatus for the analysis of gaseous and particulate products from xylene/air mixtures with an APIMS.

The APIMS has been commonly used for the performance evaluation of the filters and gas purifiers found in the clean rooms of semi-conductor production processes.[8,9] Trace substances/base gas mixtures are ionized at an electric discharge ion source operated under atmospheric pressure and consequently form positive or negative ions in the gas mixture when a positive or negative voltage is applied to a needle electrode, respectively. Most of the unipolar ionized species in the gas mixtures are produced from inert carrier gas components. The chemicals even at extremely trace amounts are fully ionized with suppressing their fragmentation through charge-transfer reactions from ionized carrier gas components to trace substances after their frequent collisions.

In the present study, particulate and gaseous organic products were separately analyzed by the APIMS to identify each component with high sensitivities. Particulate products from *o*-xylene collected on an organic-free metal foil were vaporized in 1%v H_2/Ar gas mixtures by a TPD method and the resulting vaporized components of particulate products were analyzed by the APIMS through the ionization of them with less fragmentation in 1%-H_2/Ar gas mixtures to identify the components of the particles as a function of their gasification temperatures. The gaseous products in irradiated *o*-xylene/air mixtures after the removal of particulate products were also analyzed by the APIMS. The formation reactions of the particulate and gaseous products are discussed based on the detected component ions of particle and gaseous products.

Experimental

Preparation and EB Irradiation of Xylene/Air Mixtures. Air containing 5-ppmv *o*-xylene ($C_6H_4(CH_3)_2$) and 1.0×10^4 ppmv Milli-Q water, xylene/air mixtures, were prepared by dilution of synthetic air (Nippon Sanso Co., Ltd., Pure Air S, CO_2, CO, and CH_4: <0.05 ppmv, D.P.: <193 K) mixed with vaporized liquid xylene (>98%, Tokyo Kasei Co., Ltd.) and Milli-Q water, as shown in Fig. 1. The concentrations of xylene in the xylene/air mixtures before and after EB irradiation were evaluated by a gas chromatograph (Shimadzu Co., GC-8A) equipped with a FID and a packed column (GL Science Inc., BX-20 100/120, 2 mm I.D. × 3 m).

The compact EB generator with a maximum accelerated voltage of 55 kV and maximum current of 0.5 mA was used as an ionizing radiation source for the air sample gases.[3] This EB generator consists of an EB generation tube, vacuum system, and control unit for a high voltage (H.V.) supply. The irradiation window (Ti, thickness: 2 µm) of the EB generation tube was covered by a stainless-steel irradiation cup (SUS-316L, I.D.: 46 mm and H.: 25 mm) equipped with inlet and outlet tubes (I.D.: 8 mm, O.D.: 9.5 mm). The xylene/air mixtures were introduced into the cup at a flow rate of 10 L min^{-1} and irradiated with 50 keV EBs. Electron beams were completely stopped in the air mixture volume. The temperature of the air mixture was maintained at 298 K during irradiation by the circulation of cooling water outside of the cup. The average absorbed dose per unit current in the air mixtures was estimated based on the chemical reaction of oxygen (O_2) under the same irradiation conditions.[10] The air mixture at a flow rate of 10.0 L min^{-1} was irradiated to doses of 0.5, 1, and 2 kGy by changing EB currents.

APIMS. The atmospheric pressure ionization mass spectrometer (API-MS, Nippon API Co., Ltd., PG-10) consists of a gas supply system, an ion source operated at atmospheric pressure, a differential pumping system that can dissociate cluster ions, and a single quadrupole mass spectrometer (ULVAC, Inc., MSQ-400).[5–7] In the present study, the ions were produced by a corona discharge at the ion source with the constant currents of 1 µA. The temperature of the ion source was kept at 393 K to prevent the adsorption of trace substances in gases. The condensable gaseous chemicals such as water in inert carrier gases might condensate around ions of both the carrier gases and chemicals to produce cluster ions due to the cooling of gas mixtures through an adiabatic expansion in the differential pumping system. The drift voltage of the system was fixed at 42 V to detect H_3O^+ as gaseous water in dry inert carrier gases through decomposition of water ion clusters of $H_3O^+(H_2O)_n$ ($n = 1$–3).[5,6] The quadrupole mass spectrometer can measure positive and negative ions with m/z values of 2–480 u at the minimum currents of 1×10^{-13} A. In the present study, all analysis of sample gases with the APIMS was performed under the condition for positive ion measurement. An electrical discharge at the ion source was made by a positive mode to produce positive ions, because ionization potentials and proton affinities of common organics are higher than those of inorganic gases. Both scanning m/z values and corresponding ion currents were recorded by a digital data logger (KEYENCE Co., GR-3500).

Analysis of Particulate Products with APIMS. Particulate products can be separated from EB-irradiated xylene/air mixtures using common polymer membrane filters for component analysis of only particles of the products. Gaseous organic products might be uptaken by collected particulate products on the filter while irradiated xylene/air mixtures are being filtered. Moreover, some organic chemicals may be generated from the filter material through its pyrolysis during a TDP process at elevated temperatures. The particulate products were collected on an organic-free metal foil (Al-foil) with less interaction between irradiated air mixtures and collected particles than that by filtration at membrane filters using the following method. Some population of the particles produced from xylene, 31–35 and 56–59% of the total number and volume concentrations, was positively and negatively charged under EB irradiation through the reaction of the particles of products and ions originated in air components.[11] The charged particles of particulate products can be separated from the gas phase in a precipitator equipped with two electrodes by applying an electric field between the two electrodes. The 5-ppmv xylene/air mixtures irradiated at doses of 1 and 2 kGy were accordingly introduced in the precipitator for 5 min at a flow rate of 10 L min^{-1} and the particles were collected on the Al-foil at one of the electrodes. The irradiated xylene/air mixtures were passed through the precipitator over the collected particulate products. The particles on the portion of the Al-foil having a size of 3 × 3 mm (carbon weight on this separated Al-foil was 1.5×10^{-2} mgC) were used as a sample to be vaporized in a TPD process. The sample was settled in the stainless-steel tube (SUS-316L, O.D.: 1/2″) installed in an oven. A commercially available 1%v-H_2/Ar gas mixture (Ar gas purities: >99.9995%, D.P.: <193 K) was used as an inert carrier gas for the ionization of vaporized organic substances with suppressing their fragmentation by the attachment of H^+ produced from the reaction of Ar^+ with H_2.[7] This carrier gas, after passing through a molecular sieve (GL Sciences Inc., 13X) packed column, was continuously introduced into the stainless-steel tube at a flow rate of 1 L min^{-1}. The stainless-steel tube (SUS-316L, O.D.: 1/8″), whose inner surface was electrically polished, was used for gas transportation from the column of molecular sieves to the sample gas supply of the APIMS. The temperature of the oven was elevated from 303 to 423 K with steps of 30 K and kept for 10 min at each step. After a TDP process, the particulate products remaining on the Al-foil were dissolved in Milli-Q water and the total concentrations of organic carbons were measured by a total organic carbon analyzer (Shimadzu Co., TOC-5000A). The connecting tube from the oven to the sample gas supply of the APIMS was maintained at 393 K. The particle components of the products having correspondent vapor pressures were vaporized one by one into the carrier gas at elevated temperatures step by step. The carrier gases containing vaporized particulate products were analyzed by the APIMS within two current ranges of 1×10^{-10}–1×10^{-8} A and 1×10^{-12}–1×10^{-10} A at temperatures of 303, 333, 363, 393, and 423 K.

Analysis of Gaseous Products with the APIMS. The particles of the products from irradiated xylene/air mixtures with sizes larger than 10 nm could be separated using Fluoropore membrane filters (Millipore Co., FGLP04700), which have a pore size of 0.2 µm.[3] The irradiated xylene/air mixtures after passing this filter were regarded as the irradiated air containing only gaseous products. The particle-free irradiated xylene/air mixtures were diluted with N_2 gas (>99.9995%, D.P.: <193 K) 10 times. A portion of such diluted irradiated air mixtures was continuously introduced into the sample gas supply of the APIMS at a flow rate of 3 L min^{-1} with aspirating these air mixtures by a rotary pump equipped with a mass flow controller downstream of the outlet of this supply system. The xylene/air mixtures before and after irradiation were analyzed within current ranges of 1×10^{-10}–1×10^{-8} A and 1×10^{-12}–1×10^{-10} A by the APIMS.

Results and Discussion

Particulate Products. The particulate products vaporized into a 1%v-H_2/Ar carrier gas at temperatures of 303–423 K were continuously analyzed with the APIMS for m/z of 2–480 u with currents of 1×10^{-10}–1×10^{-8} A and m/z of 300–480 u with currents of 1×10^{-12}–1×10^{-10} A. The mass spectra for m/z of 2–480 are shown in Figs. 2a–2e at temperatures of 303, 333, 363, 393, and 423 K, respectively, for the vaporized particulate products in xylene/air mixtures irradiated at a dose of 2 kGy. The mass spectrum for only the carrier gas was identical to that of the carrier gas passing over the collected particulate products at a temperature of 303 K. This suggests that hardly any of the components are vaporized into the carrier gas at a temperature of 303 K. The ions of 19, 29, 41, and 81 u shown in Fig. 2a are produced in the carrier gas from a trace of H_2O and the components of the carrier gas and seem to be H_3O^+ and N_2H^+, ArH^+ and Ar_2H^+, respectively. The ions of 43, 47, 61, 97, 111, 135, and 139 u observed at 333 K in Fig. 2b were detected as the vaporized components of particulate products besides the ions from Ar and H_2O. The mass spectrum at 363 K shows the additional peaks at the ions of 153, 155, 171, and 187 u. The ion of 203 u also appeared at 393 and 423 K. The mass spectra for vaporized particulate products at a dose of 1 kGy had the peaks at the same m/z as those at a dose of 2 kGy at each temperature. The concentrations of particulate products increased at doses of 1 to 2 kGy with keeping the same components. After these TPD processes, the organic carbon concentrations of particulate products on the Al-foil were reduced to 12–15% of those before the process. The TPD process at temperatures to 424 K enabled the vaporization of 85–88% of the collected particulate products.

The ions of 139–203 u have the constant peak interval of m/z of 16 u and this interval corresponds to the m/z value of O^+. The FT-IR analysis of collected particulate products shows that the particulate products are not retaining a benzene

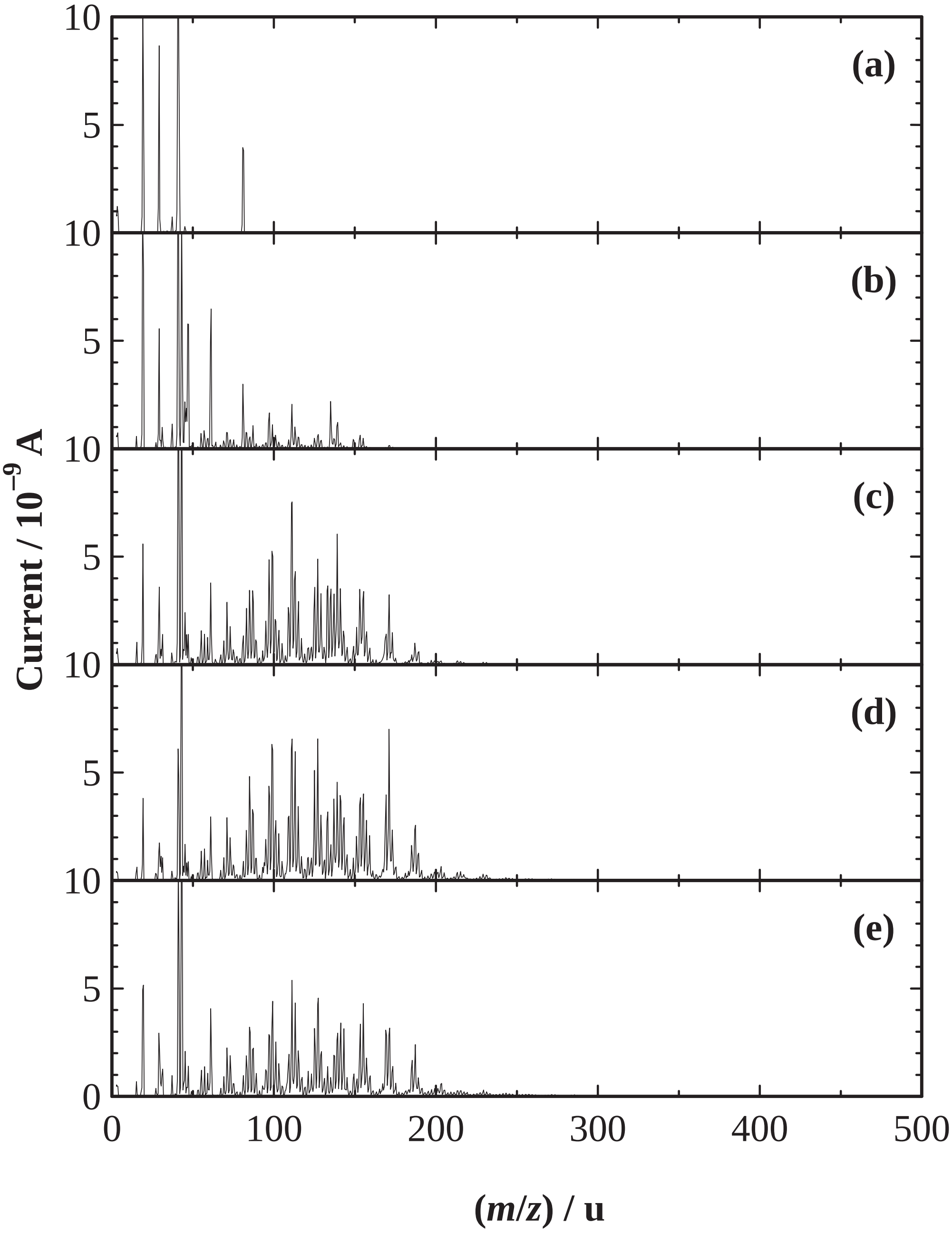

Fig. 2. Mass spectra of particle components vaporized at different temperatures as a function of m/z values (a: 303, b: 333, c: 363, d: 393, and e: 423 K). m/z range: 2–480 u, current range: 1×10^{-10}–1×10^{-8} A.

ring and are composed of alkyl acid and alkyl aldehyde having chemical bonds of C–C, C–H, and O–H.[3] The ions of 155, 171, 187, and 203 u are presumably produced from the ions of 139 u with uptaking O atoms. The chemical structure of the ion of 139 u showed an alkyl aldehyde or alkyl acid having unsaturated bonds. For instance, one of the expected chemicals having such a chemical structure is $OHC–C(CH_3)=C(CH_3)–CH=CH–CHOH^+$. The particulate products were highly oxidation-resistant relative to gaseous products, which is suggested from the carbon-based yields of products as a function of dose.[3] The oxidation-resistant characteristics of particulate products may attribute to the chemical structures in which their oxidation are undertaken without the break of carbon bonds.

The mass spectra for m/z of 300–480 u measured with currents of 1×10^{-12}–1×10^{-10} A are shown in Figs. 3a–3d at temperatures of 303, 363, 393, and 423 K, respectively. No ions were observed in the mass spectra at temperatures of 303–363 K (Figs. 3a and 3b). The ion of 392 u appeared in the spectra at temperatures higher than 393 K. The ions of 413 and 421 u were appreciable in the spectra besides that of 392 u at 423 K. These ions disappeared from the spectra when the particulate products deposited on the Al-foil were removed from the heated tube. The ions of 392, 413, and 421 u are accordingly regarded as one of the particle components of the products from xylene. The substances that are fully oxidized with retaining carbon–carbon bonds are speculated to

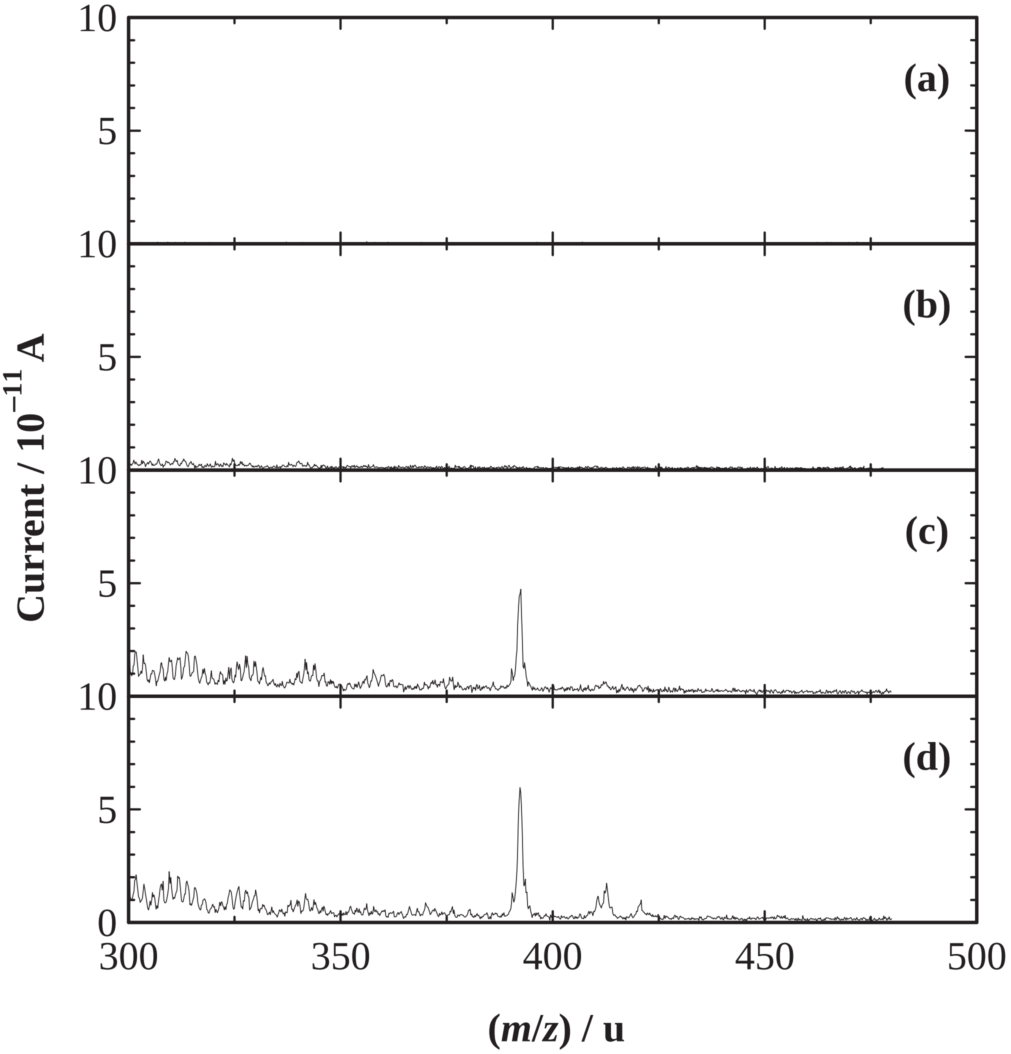

Fig. 3. Mass spectra of particle components vaporized at different temperatures as a function of m/z values (a: 303, b: 363, c: 393, and d: 423 K). m/z range: 300–480 u, current range: 1×10^{-12}–1×10^{-10} A.

be HOOC–C(OH)(COOH)–C(OH)(COOH)–$C(OH)_2C(OH)_2$–COOH ($C_8H_{10}O_{14} = 330$ u). Existence of the ions having m/z values higher than 330 u suggest that some components of the particles of products should result from the reactions between two radiation-induced product molecules (for example, dimerization). Such dimerization reactions might occur on the surface of the particles consisting of 139–203 u components through condensation. On the other hand, these heavier ions of 392–421 u seem to be extremely low vapor-pressurize substances because of their peak heights lower than those of ions of 139–203 u. Such heavier substances accordingly might be produced in the gas phase and subsequently form the nucleus of the particles through condensation (homogeneous-heteromolecular nucleation[12]).

Gaseous Products. The irradiated xylene/air mixtures after removal of particulate products by a filter were analyzed with the APIMS for m/z values of 2–480 u with currents of 1×10^{-10}–1×10^{-8} A and m/z of 300–480 u with currents of 1×10^{-12}–1×10^{-10} A. The analysis of non-irradiated xylene/air mixtures was also performed to compare the radiation-induced products with the electric discharge-induced products at the ion source of the APIMS. The mass spectra for non-irradiated xylene/air mixtures and the xylene/air mixtures irradiated to doses of 1 and 2 kGy are shown in Figs. 4a–4c for m/z values of 2–480 u with currents of 1×10^{-10}–1×10^{-8} A.

The ions of 19, 30, 37, 55, 73, 91, and 106 u are detected as non-irradiated xylene/air mixtures, as shown in Fig. 4a. These ions are produced from xylene/air mixtures through ionization and dissociation by electric discharges at the ion source. The ions of 19, 37, and 55 u with higher peak heights and that of 73 u with lower peak heights are regarded as water ion clusters of H_3O^+, $H_3O(H_2O)^+$, $H_3O(H_2O)_2{}^+$, and $H_3O(H_2O)_3{}^+$, respectively. The formation of water ion clusters of $H_3O^+(H_2O)_n$ ($n = 1$–3) suggests that water vapor condenses around H_3O^+ and produced such water ion clusters through an ion-induced nucleation due to the cooling of gas mixtures through an adiabatic expansion in the differential pumping system of the APIMS, even by applying a drift voltage of 42 V in the presence of 1%v H_2O. The water ion clusters of $H_3O^+(H_2O)_n$

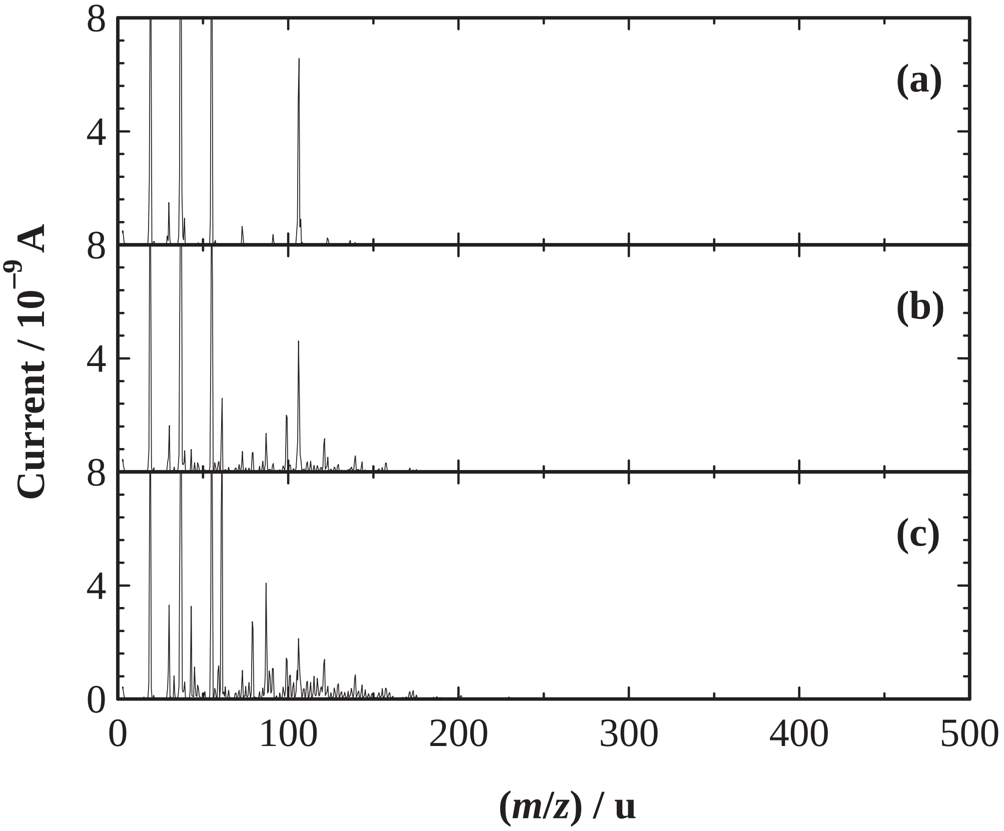

Fig. 4. Mass spectra of gaseous products in irradiated xylene/air mixtures as a function of m/z values (a: without irradiation, b: 1 kGy, and c: 2 kGy). m/z range: 2–480 u, current range: 1×10^{-10}–1×10^{-8} A.

($n = 1$–3) were also observed in the analysis of EB-irradiated humid gases with a differential mobility analyzer equipped with a Faraday cup electrometer without a differential pumping system.[13] The ion of 30 u is produced from the ionization of NO and the ions of 91 and 106 u are from the ionization of xylene. The ionization of a xylene molecule with the APIMS exclusively leads to the formation of the ion of 106 u with suppressing their fragmentation during ionization, although the ionization by a common GC-MS (EI ionization) leads to the formation of the ion of 91 u with the highest peak height through fragmentation associating with that of 106 u with the second-highest peak height.[14] The peak height at 106-u ion induced from xylene decreased to 70 and 32% of its initial peak height at doses of 1 and 2 kGy, respectively, and these degradation ratios of xylene were almost the same as those obtained by GC-FID in the previous work.[3]

The ions of 61, 73, 79, 87, 99, 121, and 139 u were observed as products by oxidation of xylene as shown in Fig. 4b. The peak heights at these ions at a dose of 2 kGy become higher than those at a dose of 1 kGy. These ions should be produced from xylene/air mixtures by EB irradiation. According to their m/z values, the ions of 61, 73, 87, and 121 u are presumably CH_3COOHH^+, $C_2H_3COOHH^+$, $C_3H_5COOHH^+$, and $C_6H_4(CH_3)CHOH^+$, respectively. The ion of 139 u is detected as one of the particle components of the products. This result suggests that these chemicals stay in gaseous and particulate forms in the irradiated xylene/air mixtures. The ions of m/z values lower than 139 u are exclusively contained in gaseous form in the irradiated air mixtures.

The peaks of ions were not observed in the mass spectra of 300–480 u with currents of 1×10^{-12}–1×10^{-10} A. This result suggests that the ions of 392–421 u exclusively stayed in the particles of the products; otherwise, these ions are hardly ionized due to exclusive ionization of other gaseous products in irradiated xylene/air mixtures.

Conclusion

The particles of products from xylene/air mixtures irradiated by a 50-keV electron beam (EB) were collected on an organic-free metal foil (an Al-foil) by electric fields, and vaporized in 1%v-H_2/Ar gas mixtures by a temperature-programmed desorption (TPD) method in the temperature range of 303–423 K. Such gas mixtures were analyzed with the atmospheric pressure ionization mass spectrometer (APIMS) through ionization of the vaporized substances with less fragmentation to identify the particle components of the products at elevated temperatures. The gaseous products in irradiated xylene/air mixtures were also analyzed with the APIMS. The ions of 43, 47, 61, 97, 111, 135, and 139 u were detected at temperatures higher than 333 K as vaporized components of the particulate products. The ions of 153, 155, 171, and 187 u were additionally detected at 363 K and the ion of 203 u also appeared at temperatures higher than 393 K. The TPD process at temperatures to 424 K enabled the vaporization of 85–88% of the collected particulate products.

Ions of 139–203 u with the interval of the m/z of 16 u, the same as the value of O^+, were observed as vaporized particle components correspondent to elevated temperatures. The ions of 155, 171, 187, and 203 u were presumably produced from the ions of 139 u with uptaking O atoms. The highly oxidation-resistant characteristics of particulate products could attribute to the chemical structures since their oxidations are undertaken without the break of carbon bonds. The ion of 392 u was also detected with peak heights lower than the above ions of 139–203 u at temperatures higher than 393 K. Such a substance having heavier m/z values would result from the reactions between two radiation-induced product molecules on the surface of the particles consisting of 139–203 u components. Otherwise, the substance is presumably one of the nucleus substances in the particles of products after formation in the gas phase. For the analysis of gaseous components in the irradiated air mixtures, the ions of m/z lower than 139 u are exclusively contained in gaseous form in the irradiated xylene/air mixtures. The particulate products from xylene/air mixtures are well soluble in water, although the components of them are comprised of heavier molecules such as components with m/z values of 139–203 u and 392–421 u. Therefore, the O atoms stay as hydrophilic groups such as –OH and –COOH in their molecules.

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