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Conversion of emitted dimethyl sulfide into eco-friendly species using low-temperature atmospheric argon micro-plasma system

Hsin-Hung Chen[a], Chih-Chiang Weng[a,b], Jiunn-Der Liao[a,b,*], Liang-Ming Whang[c,d], Wei-Hung Kang[e]

[a] Department of Materials Science and Engineering, National Cheng Kung University, 1 University Road, Tainan City 70101, Taiwan
[b] Promotion Center for Global Materials Research, National Cheng Kung University, 1 University Road, Tainan City 70101, Taiwan
[c] Department of Environmental Engineering, National Cheng Kung University, 1 University Road, Tainan City 70101, Taiwan
[d] Research Center for Energy Technology and Strategy, National Cheng Kung University, 1 University Road, Tainan City 70101, Taiwan
[e] Chimei Innolux Corporation, Southern Taiwan Science Park, 3 Huansi Rd. Sec. 1, Tainan City 74147, Taiwan

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ABSTRACT

A custom-made atmospheric argon micro-plasma system was employed to dissociate dimethyl sulfide (DMS) into a non-foul-smelling species. The proposed system takes the advantages of low energy requirement and non-thermal process with a constant flow rate at ambient condition. In the experiments, the compositions of DMS/argon plasma, the residual gaseous phases, and solid precipitates were respectively characterized using an optical emission spectrometer, various gas-phase analyzers, and X-ray photoemission spectroscopy. For 400 ppm DMS introduced into argon plasma with two pairs of electrodes (90 W), a complete decomposition of DMS was achieved; the DMS became converted into excited species such as C*, C_2*, H*, and CH*. When gaseous products were taken away from the treatment area, the excited species tended to recombine and form stable compounds or species, which formed as solid particles and gaseous phases. The solid deposition was likely formed by the agglomeration of C-, H-, and S-containing species that became deposited on the quartz inner tube. For the residual gaseous phases, low-molecular-weight segments mostly recombined into relatively thermodynamic stable species, such as hydrogen, hydrogen sulfide, and carbon disulfide. The dissociation mechanism and treatment efficiency are discussed, and a treatment of converting DMS into H_2-, CS_2-, and H_2S-dominant by-products is proposed.

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1. Introduction

Volatile organic sulfur compounds (VOSCs) have recently been assigned important roles in the global sulfur cycle (a combination of biological, chemical, and geochemical processes). VOSCs have important environmental functions related to global warming, acid precipitation, and cloud formation. A major component of the VOSC group is dimethyl sulfide (DMS, CH_3–S–CH_3) [1]. DMS has an unpleasant smell with a low olfactory threshold (0.02–0.1 ppm).

A number of organic compounds, such as photoresists and solvents, are widely utilized in industry. The utilization of organic compounds creates a substantial amount of pollutant by-products that raise environmental concerns [2]. Volatile organic compounds (VOCs) and solvents, e.g., dimethyl sulfoxide (DMSO), are often necessary for thin-film transistor liquid crystal display (TFT-LCD) processing. DMSO is widely used as a solvent, lubricant, preservative, or stabilizer in a number of industrial, agricultural, and pharmaceutical applications [3,4]. Although DMSO itself has low toxicity, its main by-product from the reduction reaction, DMS, is volatile and noxious in nature. DMS particles, which have a long adhesion time, seriously affect the cleanliness of the semiconductor manufacturing process and dramatically reduce the production yield. DMS treatment is thus an environmental control issue.

DMS can be typically treated by catalytic incineration [5]. However, this consumes a lot of energy and generates secondary pollution. For example, wet scrubbers have a high efficiency, but require costly oxidizing chemicals (e.g., ClO_2). Potentially, they can produce chlorinated hydrocarbons if not properly controlled [6]. Thermal oxidation implies high energy consumption for greatly diluted effluents. Adsorption processes using activated carbon are non-destructive techniques and result in the use of expensive secondary treatment processes [7]. Plasma torch at atmospheric pressure is a promising alternative to conventional thermal combustion processes; it requires no electrodes and produces high electron, ion, and radical densities. The decomposition of halogenated VOCs with thermal plasma sources has been performed [8]. The maximum gas temperature of a plasma torch may reach ≈4000 K [8] and achieve a complete decomposition of VOCs or

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* Corresponding author at: Department of Materials Science and Engineering, National Cheng Kung University, 1 University Road, Tainan City 70101, Taiwan. Tel.: +886 6 2757575x62971; fax: +886 6 2346290.
E-mail address: jdliao@mail.ncku.edu.tw (J.-D. Liao).

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gaseous sulfide compounds with an energy density of ≈1000 J/cm^3 [9]. Thermal plasma is effective in decomposing dense VOCs due to its high temperature; however, it is still not economical for the decomposition of dilute VOCs.

Low-pressure plasma has been applied to reduce VOSCs such as carbon disulfide, methanethiol, and DMS due to its relatively low energy consumption and high treatment efficiency [10,11]. VOSCs can be decomposed at room temperature using a radio-frequency (RF) argon plasma reactor under low pressure. The decomposition efficiency increases with increasing applied power and oxygen content [12]. However, low-pressure plasma systems usually require an expensive vacuum apparatus for batch treatment.

Non-thermal atmospheric plasma systems operating at high voltage and low current are a promising alternative for the treatment of VOSCs. The systems are usually based on dielectric barrier discharge (DBD) or pulsed corona discharge (PCD) [13]. The former technique has some practical advantages, such as being a relatively low-temperature process and having high decomposition efficiency for the decomposition of VOCs [14]. Due to low energy efficiency, the combination of DBD and catalysts is frequently applied for decomposing VOCs [15]. Nevertheless, the application of catalysts may lead to secondary pollution. PCD has been used for the decomposition of gaseous sulfide compounds [7,16,17]. Removal and destruction efficiencies of ≈99% can be obtained with power levels ranging from 0.1 to 0.3 J/cm^3, which are competitive with those obtained using existing technologies. However, PCD reactors use a wire-in-cylinder design; an inner electrode is usually used in the apparatus. The attachment of reactive gases onto the inner electrode tends to corrode it, reducing the efficiency of the reactor with operation time.

Atmospheric-pressure micro-plasma technology, e.g., micro-plasma reactors based on capillary electrode discharge (CPE), has remarkable stability, which can be explained by Paschen's Law [18], and low energy consumption for the study of VOC destruction [19]. CPE uses capillary jet mode, which produces jets of high-intensity plasma at atmospheric pressure. Maximum VOC destruction efficiencies of 80–100% can be obtained with an energy density of 2 J/cm^3 for most compounds studied. The decomposition of several prototypical aliphatic (e.g., ethylene, heptane, octane) and aromatic (e.g., benzene, toluene, ethylbenzene, xylene) compounds as well as ammonia has been studied [18,20].

Micro-plasma technology has a low breakdown voltage and a low energy requirement with high output plasma density. Table 1 of Appendix A shows a comparison of plasma parameters for various atmospheric plasma sources. To prevent the PCD-type electrode from corroding, a custom-made RF-driven micro-plasma system, which is a type of micro-plasma jet containing two outer electrodes, is applied for the decomposition of DMS. A RF-driven plasma jet maintains non-thermal properties by preventing glow-to-arc transitions via an oscillating electric field and the inclusion of rare gases in the reactor process stream to prevent discharge instabilities [21]. In the experiment, argon, which has a relatively low breakdown voltage and is an atomic gas, was employed as the buffer gas. A probable mechanism of DMS decomposition as a function of treatment time at atmospheric pressure is presented.

2. Experimental

2.1. Micro-plasma system with the mixture of ca DMS in argon

A schematic diagram of the experimental setup is shown in Fig. 1(a). The setup mainly consists of a micro-plasma reactor, a DMS feeding system, and a 13.56 MHz RF power supply (maximum input: 325 W, ACG-3B, MKS *ENI Corp.*, Rochester, USA) with a matching network (MW-5DM11, *ENI Corp.*, Rochester, USA). The micro-plasma system is a type of micro-plasma jet system, which contains a quartz tube as the gas channel and a dielectric layer. The outer and inner diameters of the tube are ≈3.0 and ≈1.5 mm, respectively. To prevent the reactant gas from becoming attached to the inner electrode, which may reduce efficiency, two outer electrodes are designated for the dissociation of DMS in argon.

Copper tape pasted on the outer surface of a capillary quartz tube (width: 6.4 mm; thickness: 0.06 mm, *Electron Microscopy Sciences*, PA, USA) was employed as the electrode. One of the electrodes near the gas inlet was connected to the output of the RF power supply; the other electrode near the gas outlet was connected to the ground (configuration is shown in Fig. 1(b); this type of configuration is denoted as "one pair of electrodes" hereafter). The micro-plasma was stably ignited and maintained; the gap between the two electrodes was kept at ≈3.0 mm with the applied power was 40 W. For this gap, the effective treatment volume was ≈27.7 mm^3. Therefore, the average treatment time per DMS molecule was ≈6.7×10^{-4} s for a flow rate of 2500 sccm. Under this condition, the maximum concentration of DMS which can be added into argon micro-plasma was ≈400 ppm. Commercially available DMS (99.9%, CAS number: 75-18-3, *Merck*, Germany) was utilized in the experiment. To obtain a suitable concentration (i.e., ≈400 ppm) of DMS, high-purity argon (99.99%, *Yun-Hai Co., Ltd.*, Taiwan) was used as both the buffer gas and the carrier gas to pass through a DMS container; the DMS was kept at −78 °C in a cooling system with alcohol as the media. The dissociation mechanism of DMS under these conditions is discussed in Section 3.4. To determine the influence of treatment time and input power, series of two pairs of electrodes (this type of configuration is denoted as "two pairs of electrodes" hereafter), as shown in Fig. 1(c), was designated for the decomposition of DMS. The reaction time, up to 1.34×10^{-3} s, of the DMS sample was expected to be doubled compared to that of one pair of electrodes.

2.2. Analysis of residual gas after plasma treatment

Before plasma treatment, 400 ppm DMS/argon was collected as the control. After igniting 400 ppm DMS/argon plasma and lasting for 10 min (i.e., until the plasma became stable), the residual gas was collected using a gas sampler and a 700 ml sampling bag (232-945A, *SKC Inc.*, PA, USA) from the sampling exit, as indicated by the single asterisk in Fig. 1(a).

The variation of sulfide with plasma treatment was estimated using a gas chromatography/flame photometric detector (GC-FPD, GC-2014, *Shimadzu Corp.*, Japan) combined with a GS-Q column (30 m × 0.53 m), which is typically employed to analyze the compositional change of sulfide and phosphide. A gas chromatography/thermal conductivity detector (GC-TCD) was employed to examine the by-products derived from hydrogen or hydrocarbon molecules. A Fourier transform infrared spectrometer (FTIR, Nexus 470, *Thermo Nicolet*, USA) combined with a transmittance-type 9.6 m gas cell was applied to analyze the gaseous compositions after micro-plasma treatment.

2.3. Optical diagnosis of excited species

The excited species in argon and DMS/argon micro-plasma were detected using a single monochromator (SpectraPro 2300i, *Princeton Instruments*, New Jersey, USA) equipped with a charge-coupled detector (CCD, 1340 × 100 pixels, NTE: 100B, *Princeton Instruments*, New Jersey, USA). The CCD resolution is ≈0.14 nm. The head of the optical fiber used for detecting light emissions was placed side-on ≈12 mm away from the discharge zone between the two electrodes to record the real-time emission spectrum with one pair of electrodes (40 W), as shown in Fig. 1(a). The focal length of the monochromator is ≈300 mm. Two gratings were used: 200–500 nm (1200 g/mm) and 500–1100 nm

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