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exist for the publication of theoretical and experimental investigations of all aspects of the mechanics and thermodynamics of fluid-flow with special reference to fluid-flow machines

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poświęcone są publikacjom naukowym z zakresu teorii i badań doświadczalnych w dziedzinie mechaniki i termodynamiki przepływów, ze szczególnym uwzględnieniem problematyki maszyn przepływowych

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Wydawnictwo Instytutu Maszyn Przepływowych
Polskiej Akademii Nauk
ul. Gen. Józefa Fiszera 14, 80-952 Gdańsk, skr. poczt. 621,
☎ (0-58) 341-12-71 wew. 141, fax: (0-58) 341-61-44,
e-mail: esli@imp.gda.pl

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MARIUSZ JASIŃSKI, PIOTR SZCZUCKI, JERZY MIZERACZYK,
MIECZYSŁAW LUBAŃSKI, ZENON ZAKRZEWSKI[1]

Decomposition of chlorofluorocarbons using microwave torch plasma

Results of investigation of decomposition of chlorofluorocarbons CCl_2F_2 (CFC-12) and $CHClF_2$ (CFC-22) conducted in air flowing under atmospheric-pressure using a coaxial-line-based microwave torch plasma are presented. Concentrations of the chlorofluorocarbons in the flowing air were up to 10%. The decomposition efficiency of both CCl_2F_2 and $CHClF_2$ was almost 100%. This implies that the coaxial-line-based microwave torch plasma can be a useful tool for decomposition of highly-concentrated chlorofluorocarbons in air at atmospheric pressure.

1. Introduction

Chlorofluorocarbons and perfluorocarbons emitted to atmosphere cause environmental problems, such as the depletion of the ozone layer, the greenhouse effect etc. Efficient methods for removing the stored chlorofluorocarbons or the ones being emitted by industry are strongly required. Conventional methods, e.g. adsorption, absorption, catalytic combustion seem not to be efficient enough. Recently, thermal and nonthermal plasma methods involving electrical discharges have been examined as a possible method for efficient decomposition of chlorofluorocarbons into at least less harmful compounds.

In this investigation the usefulness of a microwave torch developed for processing of chemical compounds [1] was discussed, which was tested during decomposition of chlorofluorocarbons CCl_2F_2 and $CHClF_2$ in their mixtures with air. For this purpose the coaxial-line-based microwave plasma generator that ensures better operation stability and lower energy cost of decomposition of volatile organic compounds ($CHCl_3$, CCl_4, $C_6H_5CH_3$ and CH_4) than the waveguide-based torch structure [2,3] was employed.

[1]Centre for Plasma and Laser Engineering, Institute of Fluid Flow Machinery, Polish Academy of Sciences, Fiszera 14, 80-952 Gdańsk, Poland

2. Experimental set-up

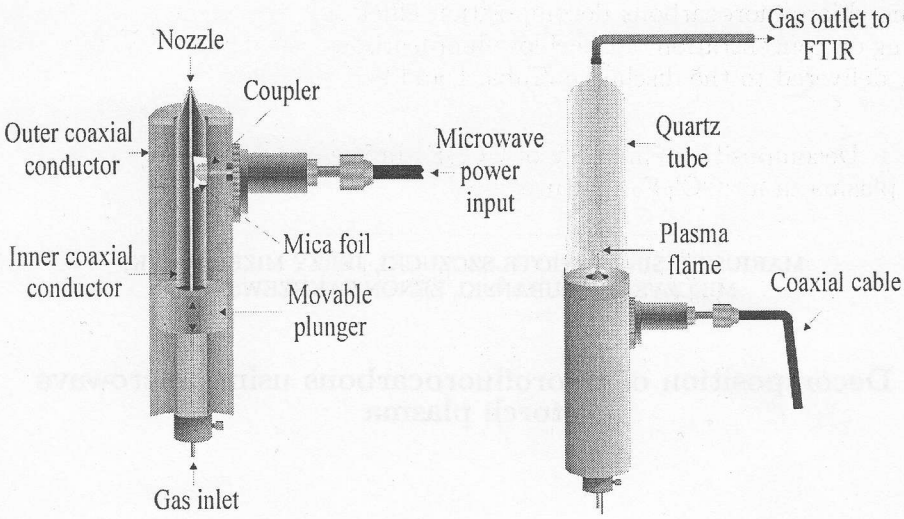

Fig. 1. Coaxial-line-based microwave torch plasma generator and plasma reactor for processing gas mixture.

The experimental set-up (Fig. 1) used in this investigation consisted of a coaxial-line-based microwave plasma generator, plasma reactor, gas supplying system and Fourier Transform Infrared (FTIR) spectrophotometer. The plasma was generated in the form of a "plasma flame" within a quartz reactor. Microwave power, of about 70-100 W was supplied to the torch system through a standard 50 Ω coaxial cable with the use of a coupler (microwave antenna). There was a mica foil between the coupler and the inner coaxial conductor. The mica foil improved the microwave coupling by increasing the capacity between the coupler and inner coaxial conductor. The microwave mode conversion and impedance-matching functions were achieved with a system of impedance matching which consisted of a conventional waveguide-to-coaxial line transition using the intrinsic tuning element in the form of a movable plunger. The movable plunger could minimize reflecting power. The microwave field in the inner coaxial conductor is focused in a small volume resulting in a high microwave field near by the tip, terminating the inner conductor of the coaxial line air duct. The operating gas (a mixture of synthetic air (80% N_2 + 20% O_2) and a chlorofluorocarbon) flowed at a rate of 1-2 l/min along the inner conductor of the coaxial line section and exited through a nozzle at its conical tip. Initial concentrations of CCl_2F_2 and $CHClF_2$ in their mixtures with air were 2-8% and 2-10%, respectively. Compositions of gas before and after the plasma processing were determined using the FTIR spectrophotometer operating in the range of 4400-1000 cm^{-1}.

3. Results

The chlorofluorocarbons decomposition efficiency was about 50 – 100%, depending on concentration of the chlorofluorocarbons, gas flow rate and microwave power delivered to the discharge [Tabs. 1 and 2].

Table 1. Decomposition efficiency of CCl_2F_2 in the coaxial-line-based microwave torch plasma in air:CCl_2F_2 mixture

Initial concentration of CCl_2F_2 in air	Microwave power	Flow rate	CCl_2F_2 decomposition efficiency
%	W	l/min	%
2.0	100	1	90.6
	100	2	62.9
	90	2	59.4
	80	1	88.8
4.0	100	1	79.2
	80	1	66.9
8.0	100	1	78.6
	90	1	73.5

Table 2. Decomposition efficiency of $CHClF_2$ in the coaxial-line based microwave torch plasma in air:$CHClF_2$ mixture

Initial concentration of $CHClF_2$ in air	Microwave power	Flow rate	$CHClF_2$ decomposition efficiency
%	W	l/min	%
2.0	100	1	100.0
	100	2	53.0
	85	2	43.1
	75	1	100.0
4.0	100	1	97.5
	100	2	46.8
	80	1	87.5
8.0	100	1	91.1
	95	1	86.5
10.0	100	1	88.5

The direct products of CCl_2F_2 and $CHClF_2$ decomposition observed in spectra

were CO, CO_2, phosgene $COCl_2$ and carbonyl fluoride COF_2 (Fig. 2 and 3). Moreover, Cl_2 and F_2 (which cannot be observed by FTIR) were found in the exit gas when both chlorofluorocarbons were processed (using paper wetted with KI and starch solution that changes colour in the presence of Cl_2 and F_2). Due to the presence of air in the processed mixture, the decomposition of chlorofluorocarbons was accompanied by the production of NO, NO_2 and N_2O_4. By contrast to the results of Sekiguchi at all. [4], HF and HCl were not detected in our experiment. Phosgene ($COCl_2$) and carbonyl fluoride (COF_2) could be easily removed from the exiting gas by passing it through a washing bottle filled with water, or recovered from the exiting gas by condensing it in a cooling system having the temperature below 9^0C and -83^0C, respectively. Moreover, phosgene, carbonyl fluorine and other chlorine and fluorine compounds (Cl_2, F_2, HCl and HF) could be removed by passing it through the fluidized particles CaO [4], activated carbon [5,6], zeolite filter [6] or $CaCO_3$ [7]. It can be seen in Fig. 3a H_2O because $CHClF_2$ used in our experiment was polluted by water.

The energy yield of decomposition of both chlorofluorocarbons, shown as a function of specific energy density in Fig. 4, ranged from 40-210 g/kWh, depending on the concentration of chlorofluorocarbons, gas flow rate and microwave power delivered to the discharge. These results are superior to those when other plasma methods (e.g. gliding arc or corona discharge) were employed for chlorofluorocarbons decompositions [8-10].

The specific energy density obtained by the authors of this paper was between $0.7 - 1.7$ kWh/Nm^3.

4. Conclusions

The results of this investigation show that CCl_2F_2 and $CHClF_2$ can be almost completely decomposed in the microwave torch plasma. The direct products of the chlorofluorocarbons decomposition are CO, CO_2, $COCl_2$, COF_2, Cl_2 and F_2. Unfortunately, the decomposition of the chlorofluorocarbons by the microwave plasma torch in air is accompanied with the production of nitrogen oxides.

The energy yield of the decomposition of the chlorofluorocarbons by the microwave plasma torch is better than those when other plasma methods were employed.

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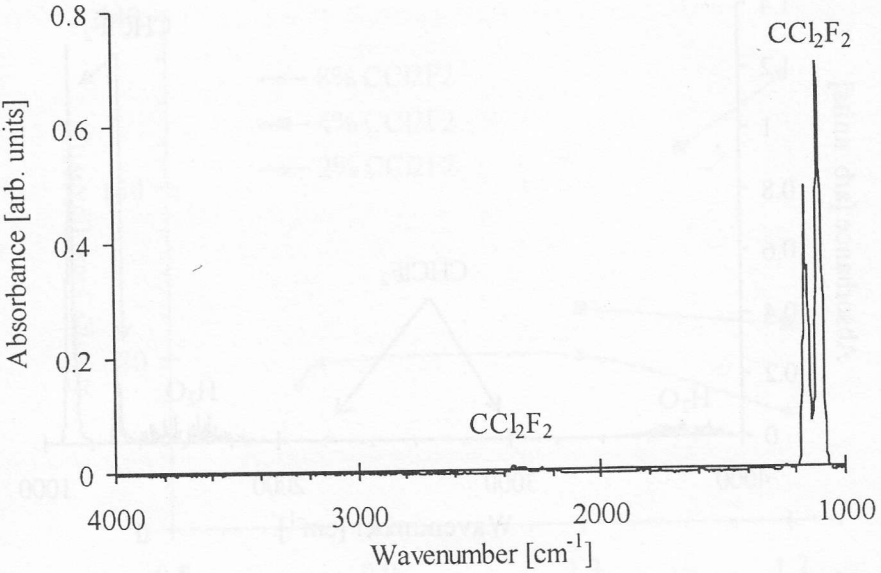
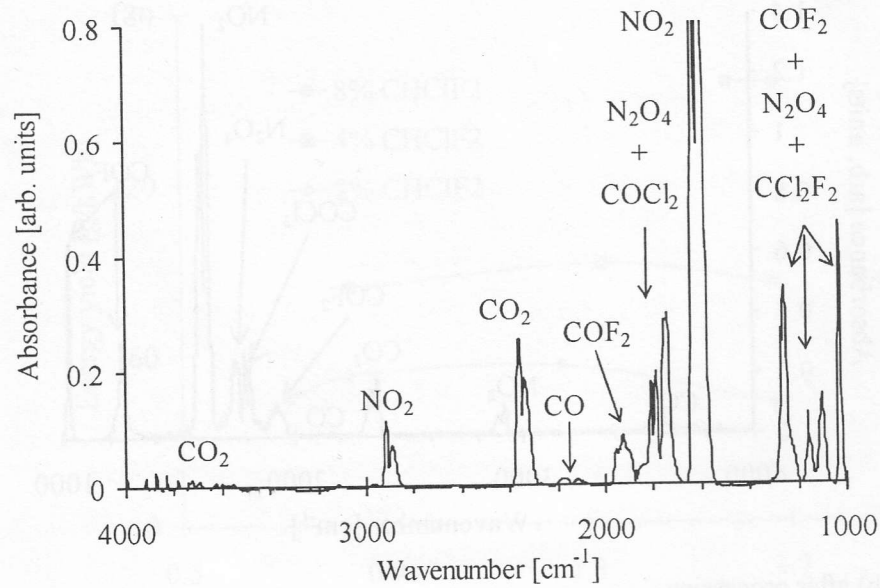
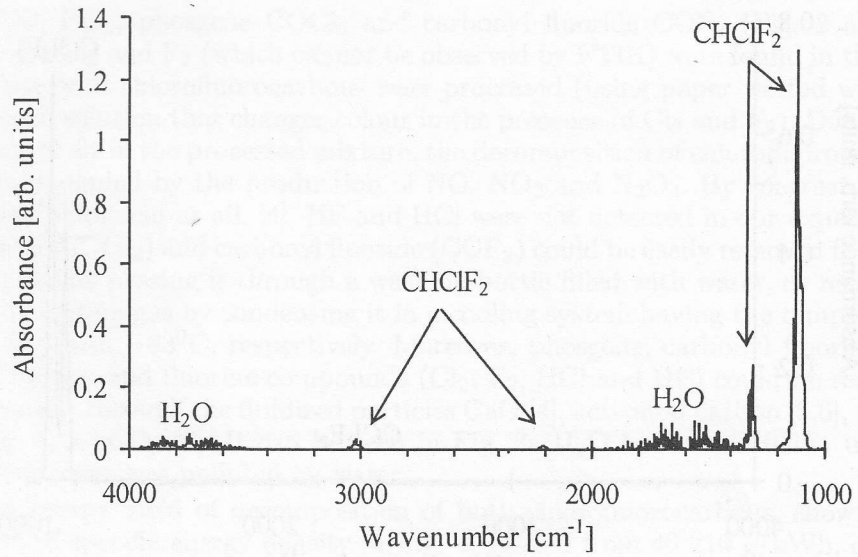


a) before processing

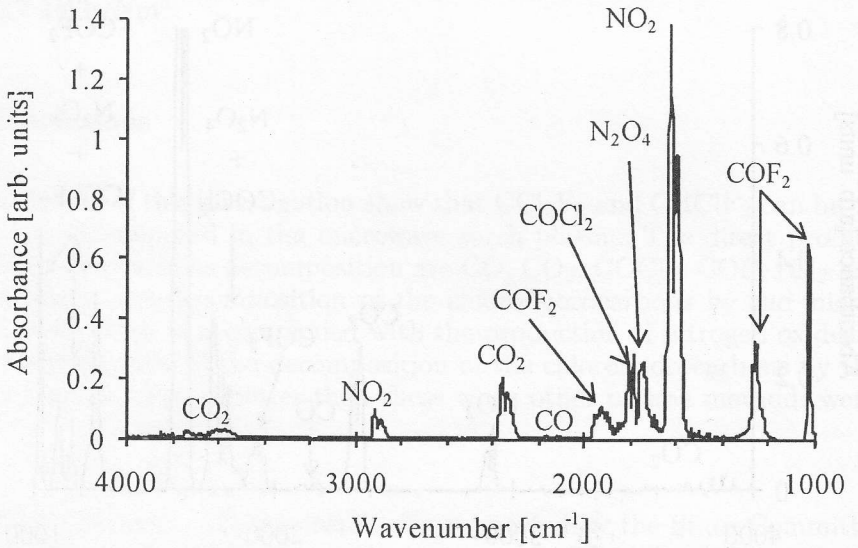


b) after processing

Fig. 2. FTIR spectra of air:CCl_2F_2 (2%) mixture before (a) and after (b) coaxial-line-based microwave torch plasma processing. Microwave power – 100 W, gas flow rate – 1 l/min.



a) before processing



b) after processing

Fig. 3. FTIR spectra of air:$CHClF_2$ (2%) mixture before (a) and after (b) coaxial-line-based microwave torch plasma processing. Microwave power – 100 W, gas flow rate – 1 l/min.

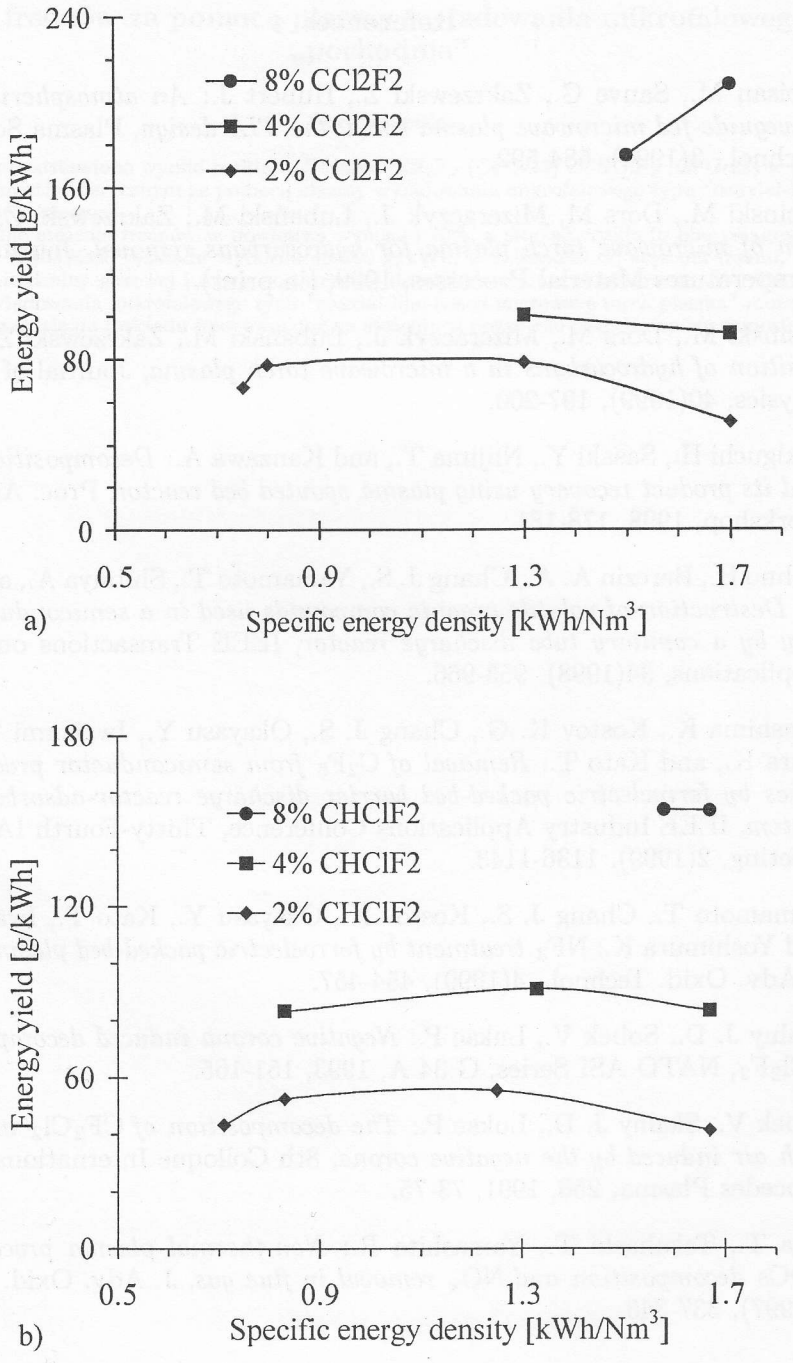


Fig. 4. Energy yield of the decomposition of CCl_2F_2 (a) and $CHClF_2$ (b) as a function of specific energy density in the coaxial-line-based microwave torch plasma processing.

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Rozkład freonów za pomocą plazmy wyładowania mikrofalowego typu „pochodnia"

Streszczenie

W pracy przedstawiono wyniki rozkładu freonów CCl_2F_2 (CFC-12) i $CHClF_2$ (CFC-22) w powietrzu pod ciśnieniem atmosferycznym za pomocą plazmy wyładowania mikrofalowego typu "coaxial-line-based microwave torch plasma".

Maksymalne stężenie freonów w powietrzu wynosiło 10%, a stopień rozkładu obu osiągnął wartość bliską 100%. Wydajność rozkładu wynosiła 40-210 g/kWh, w zależności od stężenia freonu, natężenia przepływu mieszaniny gazowej i mocy dostarczonej do wyładowania mikrofalowego. Wyniki wskazują, że plazma wyładowania mikrofalowego typu "coaxial-line-based microwave torch plasma" może stanowić przydatne narzędzie do rozkładu freonów o dużym stężeniu w powietrzu pod ciśnieniem atmosferycznym.