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Reducing of Pollutions Molecules in the Air by Photochemical Processing

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Abstract: The gas mixtures N₂, O₂, CO₂, SO₂, CHC, H₂, NO_x, Cl₂ are formed in atmosphere of modern industry centers with developing transport, chemical & energetic enterprisses. Cl₂ & NO₂ have a good adsorption in the region of 900nm. Atoms Cl & O, which are formed at the photochemical decay of Cl₂ & NO₂ molecules, respectively react rapidly with molecules of CH₄, H₂, CO, SO₂, NO_x & this bring to their degradation in air. The aim of our investigation was a study of kinetic regularities of pollution molecules degradation. **Keywords:** Degradation, Photochemical, Decay, Visible light, Pollution.

Introduction

This work concerns degradation of pollution molecules in the air, initiated by photochemical decay of Cl₂ under visible light (T. Ishii, 1999). Molecules Cl₂, NO₂ have a good adsorption in the region of $\Delta\lambda$ =300+600nm. The more important the CO₂ absorption, the hydroxide ions enhancing the CO₂ mass transfer from the gas phase to liquid phase, (A. Couvert, 2006). Formation of molecules HCl, ClO₂, CO₂, HOCl was assumed in the photolysis of gas mixtures air – H₂ – CH₄ – CO – Cl₂ under visible light (E.P. de Garmo, 1997).

Methodology

Experiments were carried out in static conditions in the glass cell at room temperatures &

total pressure was equal 10^5 pa. The analysis of substances was carried out by gas chromatography (Co, H₂, NO, SO₂, CH₄) and chemical titration (Cl₂, ClO₂) methods. Solar irradiation was used.

Rate of degradation reactions "W" of H₂, CO and CH₄ depend on the initial partial pressure of Cl₂, W-value of H₂, CO and CH₄ molecules increase by the rising of Cl₂ – pressure in initial mixtures (H.Yang, 2008). Most important reactions of Cl-atoms are given in table 1. Formation of molecules HCl, ClO₂, CO₂, HOCl was assumed in the photolysis of gas mixtures air – H₂ – CH₄ – CO – Cl₂ under visible light ($\lambda \geq$ 300nm) (J.Payamara, 2009 and E.J. Granite, 2005).

No.	Reactions	Rate constants, m ³ /s or cm ⁶ s ⁻¹
1.	$Cl + O_2 + M \longrightarrow ClOO + M$	1.7×10^{-33}
2.	$ClOO + M \longrightarrow Cl + O_2 + M$	$5.8 \times 10^{-9} \exp(-3580/\mathrm{T})$
3.	$K_{\infty} = \frac{K_1}{K_2}$	$2.95 \times 10^{-25} \exp(+3580/T)$
4.	$Cl + ClOO \longrightarrow Cl_2 + O_2$	1.6×10^{-10}
	\longrightarrow ClO + ClO	1.1×10^{-11}
5.	$ClO + CH_4 \longrightarrow Prod.$	$\leq 1.0 \times 10^{-12} \exp(-3700/\mathrm{T})$
6.	$ClO + H_2 \longrightarrow Prod.$	$\leq 1.0 \times 10^{-12} \exp(-4800/\mathrm{T})$
7.	$ClO + CO \longrightarrow Prod.$	$\leq 1.0 \times 10^{-12} \exp(-3700/\mathrm{T})$
8.	$ClO + ClO \longrightarrow Cl + ClOO$	$1.0 \times 10^{-12} \exp(-1238/T)$
	\longrightarrow Cl ₂ + O ₂	$5 \times 10^{-13} \exp(-1238/T)$
	$\longrightarrow OCl_2O + Cl$	$2.1 \times 10^{-12} \exp(-2200/\mathrm{T})$
9.	$O + OCIO \longrightarrow CIO + CIO$	5.9×10^{-11}
10.	$Cl + CH_4 \longrightarrow HCl + CH_3$	$1.3 \times 10^{-12} \exp(-1260/\mathrm{T})$
11.	$Cl + H_2 \longrightarrow HCl + H$	$3.5 \times 10^{-11} \exp(-2290/\mathrm{T})$
12.	$Cl + H_2O \longrightarrow HCl + O_2$	3×10^{-11}
13.	$ClO + HO_2 \longrightarrow HOCl + O_2$	2.0×10^{-13}
14.	$Cl + H_2O \longrightarrow HCl + O_2$	$1.8 \times 10^{-11} \exp(170/\mathrm{T})$
	\longrightarrow ClO + OH	$4.1 \times 10^{-11} \exp(-450/T)$
15.	$OH + HCl \longrightarrow Cl + H_2O$	6.6×10^{-13}

 Table 1. Main reactions of Cl-atoms(8).

Results

The formation more strong oxidant at photochemical oxidation of molecules Cl_2 was observed (T.Ishii, 1999). Total Cl-content in irradiated mixtures air – Cl_2 increase by rising of irradiation time. Probably, it is ClO_2 :

 $\begin{array}{ccc} hv & Cl_2 & \longrightarrow & 2 \ Cl \\ Cl + O & \longrightarrow & ClO_2 \\ Cl + ClO_2 & \longrightarrow & ClO \end{array}$

The formated atoms and radicate Cl, ClO rapidly react with pollutant molecules, such as CH₄,

CO and H₂. Experimental, results on the degradation of CH₄, CO and H₂ molecule in the photolysis of mixtures air - CO - CH₄ - Cl₂ are given in fig.1. Linearly decrease of CH₄ concentrations was observed. Carbon monoxide is formed in the CH₄ oxidation reactions, in part. Oxidation of CO molecules was observed, also, which bring to CO₂ formation. Oxidation reaction of CO molecules has a chain nature:

$$\begin{array}{ccc} \text{CO} + \text{CIO} & \longrightarrow & \text{CO}_2 + \text{CI.} \\ \text{Cl} + \text{O}_2 & \longrightarrow & \text{CIO}_2. \\ \text{CO} + \text{CIO}_2 & \longrightarrow & \text{CO}_2 + \text{CIO.} \end{array}$$



Fig.1.Change of CO, CH_{4 and} CO₂ concentrations on the time of irradiation by visible light

Kinetic curves of degradation molecules H_2 , CO and CH_4 are given in fig.2. As shown from this figure, the oxidation of Cl_2 molecules takes place after the H_2 , CO and CH_4 removal from gas mixture.

Conclusion

Stronger oxidant is formed at the photolysis of air – Cl_2 mixtures, probably it is ClO_2 dioxide chlorine. Linearly CH_4 removal and transformation of CO after some irradiation time takes place at the photolysis of air – Cl_2 – CO – CH_4 mixtures. CO and CO_2 are the products of methane & CO oxidation reactions.

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Fig2.Change of H_2 , CO and CH_4 and Cl_2 concentrations on the time of irradiation by visible light

Quantum yield of Cl – atoms is equal to1 at $\lambda \leq 480$ nm. Rates of H₂, CO and CH₄ removal processes depend on Cl₂ partial pressures and they increase by rising of P_{CL2}. The ClO₂ formation occurs after removal of H₂, CO and CH₄ molecules. In the consumption of CH₄, CO, H₂ molecules the following sequence is traced: CH₄ \longrightarrow H₂ + CO.

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