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Microwave-Assisted Chemical Process for Treatment of Hazardous Waste

Annual Report

by R. Varma, S. P. Nandi,
and D. C. Cleaveland



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**MICROWAVE-ASSISTED CHEMICAL PROCESS
FOR TREATMENT OF HAZARDOUS WASTE**

Project Number 23310

Annual Report

October 1987

R. Varma, S. P. Nandi, and D. C. Cleaveland,

Chemical Technology Division

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MICROWAVE-ASSISTED CHEMICAL PROCESS FOR TREATMENT OF HAZARDOUS WASTE

by

R. Varma, S. P. Nandi, and D. C. Cleaveland,

ABSTRACT

Microwave energy provides rapid *in situ* uniform heating and can be used to initiate chemical processes at moderate temperatures. The purpose of our work was to investigate the technical feasibility of microwave-assisted chemical processes for detoxification of liquid hazardous waste. Trichloroethylene, a major constituent of waste streams, was selected for this detoxification study. Experiments were performed (1) to investigate the oxidative degradation of trichloroethylene over active carbons (with and without catalysts) in air streams with microwave *in situ* heating and (2) to examine the feasibility of regenerating the used carbons. This study established that trichloroethylene in a vapor stream can be adsorbed on active carbon beds (loaded with Cu and Cr catalysts) at room temperature. When the bed is heated by microwave radiation to moderate temperatures ($<400^{\circ}\text{C}$) while a moist air stream is passed through it, the trichloroethylene is readily converted into less-noxious products such as HCl, CO, CO_2 and $\text{C}_2\text{H}_2\text{Cl}_2$. Conversion higher than 80% was observed. Furthermore, the used carbon bed can be conveniently regenerated by microwave heating while a moist- N_2 or moist-air stream is passed through the bed. The research serves as the groundwork for successful demonstration of a new technology for detoxifying hazardous organic wastes such as trichloroethylene, tetrachloroethylene, trichloroethane, and polychlorobiphenyls.

I. INTRODUCTION

A. Background

The major advantages that microwave heating has over thermal heating are: (1) it provides uniform *in situ* rapid heating, (2) the container vessel can remain at room temperature while the material in it is heated, and (3) chemical processes can usually be conducted at low temperatures.

Materials can be heated with microwave energy by placing them in a large cavity of an appropriate configuration, usually cubic or cylindrical. The use of large cavities permits the generation of a large number of resonant modes for any

particular exciting frequency (e.g., 2.45 GHz, or $\lambda = 12.236$ cm) under all loading conditions. This, in turn, allows efficient and uniform *in situ* heating of material bodies by microwaves, in sharp contrast to thermal heating.

The dielectric-loss parameter, ϵ'' , determines the microwave power density utilized in heating material bodies. Therefore, the dielectric-loss parameter and the specific heat of the material being irradiated solely determine the rate of temperature rise of the body being heated. The microwave heating is both material specific and frequency dependent. The principle of selective microwave heating is best seen by examining the general expressions for time rate of temperature rise (dT/dt) and the power dissipation (P) in a dielectric material:

$$\frac{dT}{dt} = 0.239 \times 10^{-6} P/c\rho \text{ (}^{\circ}\text{C/s)} \quad (1)$$

$$P = 55.63 \times 10^{-12} \nu E^2 \epsilon'' \text{ (W/m}^3\text{)} \quad (2)$$

where c represents the specific heat of the dielectric material, ρ is its specific gravity, ν is the microwave frequency, and E is the electric field intensity (V/cm). Since the heat energy can be absorbed *in situ* by reactants, the chemical reactions can usually be conducted at lower overall temperatures than they can with thermal heating processes. A variety of organic molecules and inorganic solids can be selectively energized and reacted using microwave energy.

B. Selection of Waste Stream

The hazardous defense waste (liquids or solids) presently in storage or being generated at various sites in the country can be broadly classified into two categories, each of which was the subject of a literature search: (1) organics (trichloroethylene, trichloroethane, tetrachloroethylene, polychlorobiphenyls, chlorobenzene, methylethyl ketone, etc.) and (2) heavy metals (Hg, Cd, Cr, Pb, Ra, Ba, Ni, etc.). The literature search was targeted at methods of treatment; particular attention was paid to the use of microwave radiation in the treatment of hazardous waste. Data were also obtained on the microwave equipment used in the food-processing industry. This information yielded the names of possible suppliers of microwave processing equipment.

A summary of the "Hazardous Waste Data Base" maintained by the Oak Ridge National Laboratory, Hazardous Defense Waste Program (ORNL-HDWP), was received and screened to select streams that could be suitable for microwave processing. From this screening, a preliminary selection of 12 streams was made. This selection was made from streams that are being generated; consequently, detailed characteristics of the streams are readily obtainable. Consideration also was given to the quantity of the stream being generated and the chemicals it

contained. The available data on these streams are given in Table 1, which is taken from the ORNL-HDWP data base. The ORNL-HDWP Office and the DOE Chicago Operations Office were contacted to obtain detailed analytical data on these streams.

Table 1. Characteristics of Hazardous and Mixed Wastes Generated at DOE Sites (from ORNL-HDWP data base)

Ref. No.	Office	Volume	Form	EPA No.	Constituents
816	Savannah	Undetermined	Liq.	F001	Tetrachloroethylene
824	"	9.5×10^6 L	Liq.	D002	Acids, metals, nitrate sulfates, uranium
831	"	Undetermined	Solid/Liq.	Undetermined	Oil, PCB
723	San Francisco	76,000 gal	Liq.	D007	Cr, Ni, Zn
1658	"	64,100 L	Liq.	F001	Acetone, methyl ethyl ketone, oil, isopropyl alcohol, Stoddard ^a solvent, trichloroethane, uranium-238
1125	Richland	4.6×10^6 L	Liq.	D002	Acetone, metals, HNO_3 , H_2SO_4 , NaOH, strontium-90, yttrium-90
1392	Oak Ridge	9000 L	Liq.	D001	Organics
1412	"	14,762 L	Liq.	D002	Organics
1504	"	6434 L	Liq.	F001	Solvents
1495	"	651,020 L	Liq.	Undetermined	Coolant
146	Albuquerque	2300 L	Liq.	D002	Black oxide, chromic acid, $\text{Cu}(\text{CN})_2$, Ni, NiCl_2 , NiSO_4
39	"	78,000 L	Liq.	F001	Solvents
831	Savannah	Undefined	Liq./Solid		Oil, PCB
736	San Francisco	28.3 tons	Solid		PCB

^aA petroleum distillate used mostly for dry cleaning.

Four of the streams in Table 1 were considered for the development of detailed experimental detoxification schemes. The streams are Nos. 816, 723, 1658, and 146. Two of the streams (816 and 1658) primarily contain organics; the other two (723 and 146) primarily contain inorganics as hazardous constituents.

Of the different organic waste streams in the ORNL-HDWP data base, many contain chlorinated hydrocarbons: for example, the so-called "degreasing solvent" [a mixture of trichloroethylene (TCE) and trichloroethane]. Since TCE is a major organic constituent in many of the streams, it was selected for this detoxification study.

II. EXPERIMENTAL

For this study, we set up and tested a 600-W laboratory microwave reactor and then conducted experiments on oxidative degradation of a major hazardous organic waste contaminant, namely TCE. The objective of the experiments was to demonstrate the technical feasibility of the microwave process for detoxification of TCE in air streams. This demonstration could then constitute the basis for a future demonstration of the technology.

A microwave oven (CEM Model MDS-81 oven with 600-W output) was modified to energize a flowthrough reactor. The detoxification method chosen was to oxidize the TCE adsorbed on activated carbon by air in the presence of catalysts with microwave (2.45-GHz frequency) *in situ* heating. The experimental setup is shown in Fig. 1.

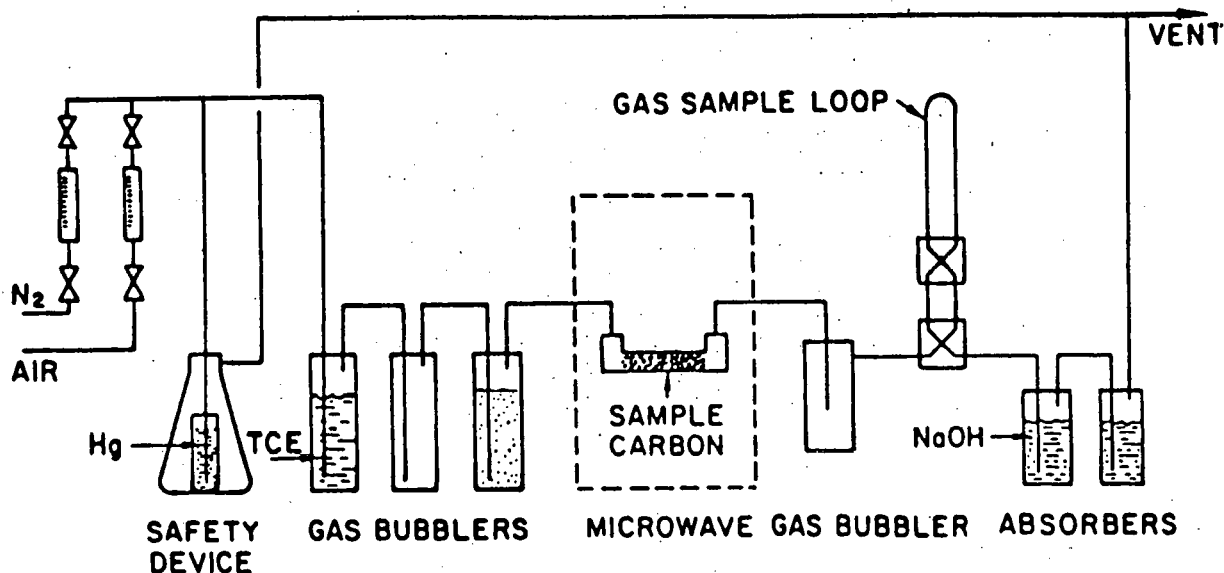


Fig. 1. Setup of Apparatus for Vapor-Phase Oxidation of Organics Using Microwave Energy

A known weight of active carbon (6 to 10 g) was placed in the sample tube, which was placed within the microwave oven. The sample tube was made of quartz and had a 12-mm ID, with standard joints at both ends. A quartz tube was used because of its transparency to microwave radiation and its high softening temperature. The tube was detachable and could be weighed when needed. The active carbon was initially dried in a nitrogen stream; at a 50% power setting, the sample drying was complete in 30 min. The dry weight of the carbon was noted. Trichloroethylene was placed in one of the bubblers (the third bubbler was packed with glass wool and worked as a demister), and nitrogen gas carried the vapors onto the active carbon. During the adsorption

step, the oven was not energized, and adsorption took place at ambient temperature (21-22°C). After the adsorption step, the sample tube was weighed, the weight of TCE adsorbed was noted, and the tube was placed back in the cavity. The flow of air was then started through the carbon bed; the oven was energized at the same time.

Active carbon beds with and without catalyst loading were used in the detoxification experiments. The active carbons were obtained from the Calgon Corp. The characteristics of these carbons are described in Table 2. Table 3 gives the results of ash analysis for the Calgon carbons.

Table 2. Characteristics of Active Carbons Used in Experiments

Name	Mesh	Source	Nitrogen BET Surface Area, m ² /g (dry)	Composition, wt %					
				Ash	Cr ₂ O ₃	CuO	C	H	N
Base Carbon	12 x 30	Coal	1050	8.6	-	-	90.3	0.65	0.55
FCA Calgon	12 x 30	Base carbon + catalyst	950	20.1	3.8	9.7	78.4	0.83	0.67
PCB Calgon ^a	12 x 30	Coconut shell	1100	3.2	-	-	-	-	-

^aThe sample was not analyzed in detail; it contains principally carbon and small amounts of hydrogen, nitrogen, and oxygen.

Table 3. Analysis of Ash from Calgon Carbons

	Base Carbon, wt %	FCA, wt %
Al ₂ O ₃	31.2	10.2
CaO	3.0	1.1
Cr ₂ O ₃	0.1	19.0
CuO	0.3	48.7
Fe ₂ O ₃	10.6	3.5
MgO	1.2	0.4
Na ₂ O	1.5	0.5
SrO	0.4	0.1
TiO ₂	2.4	0.4
BaO	0.4	0.1
SiO ₂	45.2	15.1

As will be discussed later, the major products of reaction from microwave heating of active carbon (with and without catalyst) with preadsorbed TCE in air streams were identified by mass spectral analyses to be HCl, CO₂, CO, C₂H₂Cl₂, etc. The evolved-gas analysis system consisted of an ice trap (to remove any high boiling product), a gas sampling loop, and one bubbler containing 1N NaOH solution. Periodic gas samples for mass spectroscopic analysis were taken by adjusting the two four-way stopcocks without disturbing the flow of evolved gas. Hydrochloric acid produced in an experimental run from degradation of TCE was absorbed in the NaOH bubbler. The dissolved chloride solution was then precipitated and separated as AgCl and analyzed quantitatively. This step provided an independent integration of the total HCl produced during an experimental run.

Some effects of microwave radiation (frequency of 2.45 GHz) on active carbons not heretofore described were observed. When the carbon was irradiated by microwaves in various experimental runs, small sparks appeared throughout the bed. The intensity of the sparks decreased with time, but isolated sparks were still observed after heating for about 60 min. It was initially thought that the isolated sparks were caused by conducting minerals present in the carbon. Following acid washing of the coconut-shell carbon, a material with very low mineral content was obtained, but the sparking remained unaltered. It is now believed that small semi-crystalline regions exist in commercial active carbons and that these regions are centers for strong absorption of microwave radiation. It was one of our objectives to find means of eliminating or reducing sparking during microwave heating of the carbon bed to assure safe operations.

III. RESULTS AND DISCUSSION

A. Adsorption of TCE on Active Carbon

We determined the adsorption characteristics of TCE vapor on active carbon before engaging in experiments on microwave-assisted detoxification of TCE preadsorbed on carbon in air streams.

Trichloroethylene can be adsorbed on active carbons from the vapor phase, and the extent of this adsorption depends on the nature of the carbon (surface area and composition), the flow rate of the TCE stream, and the temperature of the bed. The adsorbed TCE can be desorbed by a gas stream (e.g., N₂ or N₂ + moisture) while the bed is simultaneously heated *in situ* by microwave radiation. Since the sorption/desorption characteristics of the carbon bed have potential application in the detoxification of TCE in an air stream originating either from TCE in a liquid state or in contaminated soil, we decided to experimentally investigate both the sorption and desorption of TCE. (The desorption of TCE and its concomitant detoxification are discussed in Sec. III. B.)

The sorption characteristics of TCE on two types of carbon beds (the Calgon FCA and PCB carbons, see Table 2) were investigated. The first type of carbon (Calgon FCA) contained Cu and Cr catalysts; the second carbon (Calgon PCB) used a standard coconut-shell charcoal with a similar surface area but contained no catalysts. Streams of nitrogen were saturated with TCE by bubbling the gas through liquid TCE at 22°C (vapor pressure of TCE, 7.7 kPa at 22°C). This stream was then passed through a column of glass wool to remove any mist before it was passed through the respective carbon beds for TCE loading. The flow was stopped periodically, and the column was weighed to determine the quantity of TCE that was adsorbed.

The weights gained by the respective carbon beds as a function of time are shown in Fig. 2. The catalyst-containing carbon was shown to have a slightly higher capacity for TCE adsorption than did the coconut-shell carbon. The linearity of the sorption isotherm indicates that the bed did not attain equilibrium in the time allowed for these measurements.

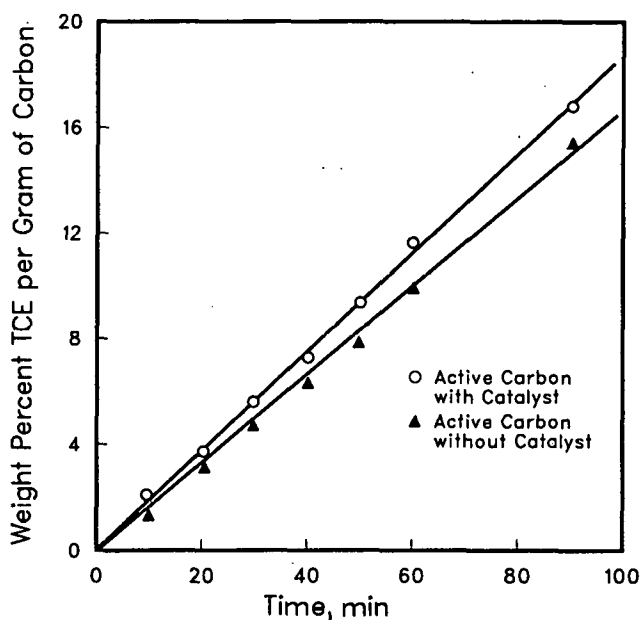


Fig. 2.

Adsorption of Trichloroethylene
by Active Carbons at 22°C

B. *Oxidative Degradation of TCE over Active Carbon*

We performed experiments on oxidative degradation of TCE passed through an active carbon under microwave heating, with dry and moist air flowing over the carbon bed. The setup shown in Fig. 1 was again used.

In Run 107, TCE vapor mixed with moist air was passed through a bed of carbon containing a catalyst while the bed was irradiated with microwave radiation. The exit gas analysis for the run is shown in Table 4. The results

indicate no decomposition of TCE. It was evident from this observation that no evolution of HCl took place. We concluded that TCE does not undergo decomposition or oxidation if it is not preadsorbed on active carbon.

Table 4. Oxidation of Trichloroethylene (Run 107) in Moist Air Stream over Carbon with Cu and Cr Catalysts (Calgon Carbon FCA)

Oxidant: Moist Air, Flow Rate of 100 mL/min Power Setting: 20% (Max = 600 W, 2.45-GHz Frequency)

Componenta	Amount, vol %		
	5 min	15 min	30 min
H ₂ O	1.0 ± 0.3	2.5 ± 0.5	3.4 ± 0.7
N ₂	78.2 ± 1.6	78.1 ± 1.5	69.8 ± 1.4
CO	4.2 ± 0.4	4.6 ± 0.5	5.6 ± 0.6
O ₂	0.03 ± 0.01	0.03 ± 0.01	0.04 ± 0.01
HCl	< 0.01	< 0.01	< 0.01
Ar	0.58 ± 0.02	0.67 ± 0.02	0.83 ± 0.03
CO ₂	15.7 ± 0.5	16.7 ± 0.5	20.0 ± 0.6
C ₂ H ₂ Cl ₂	< 0.01	< 0.01	< 0.01
C ₂ HCl ₃	0.06 ± 0.03	0.3 ± 0.1	0.08 ± 0.04
(CH ₃) ₂ CO	0.2 ± 0.1	0.2 ± 0.1	< 0.1
Other	ND ^a	ND	ND

^aIn this table and Tables 5 and 7, ND = not determined.

The remaining runs (Nos. 101, 102, 105, 106, 114, 119, 127) were done with TCE preadsorbed on active carbon at room temperature (22°C). In these runs, liquid TCE was placed in one of the bubblers, and N₂ gas was bubbled through it at a flow rate of 100 mL/min; it was then passed for 30 min through the active carbon bed (~6-8 g of carbon packed loosely), which was contained in a quartz tube. After the adsorption step, the sample tube was weighed, and it was assumed that the increase in weight equaled the weight of TCE adsorbed. A flow of dry or moist air (i.e., air saturated with moisture by bubbling house air through a water bubbler at 25°C) was then started. Dry air was used for Runs 101 and 106, and moist air for Runs 102, 105, 114, 119, and 127. The bed was heated by microwave radiation in all cases at the 20% power level. The total reaction time was 60 min. The carbon bed was weighed again after reaction, and the used carbon was analyzed for chlorine. Gas samples were collected from the product gas stream during each run, to be analyzed by mass spectroscopy. A graphical integration of the HCl gas profile vs. time gave an estimate of total HCl formed in any particular reaction. The chloride formed in the NaOH absorber in each run was determined as AgCl to provide yet another estimate of total HCl produced.

Tables 5-7 give the results of mass spectral analyses for typical exit gases. Besides the observed formation of HCl, other chlorine-containing species such as $C_2H_2Cl_3$, and C_2HCl_3 were observed in the product gas stream. In a few experiments (e.g., Run 102), CH_3Cl was also detected. The other major products formed from oxidative degradation of TCE were CO_2 and CO.

Table 5. Oxidation of Trichloroethylene (Run 102) Preadsorbed on Carbon with Cu and Cr Catalyst (Calgon Carbon FCA)

Oxidant: Moist Air with Flow Rate of 100 mL/min
Power Setting: 20% (Max = 600 W, 2.45-GHz Frequency)

Components	Amount, vol %			
	4 min	10 min	30 min	60 min
H ₂ O	0.12 ± 0.06	0.06 ± 0.03	0.10 ± 0.05	0.06 ± 0.03
N ₂	38.9 ± 1.4	70.6 ± 1.8	76.2 ± 1.5	77.1 ± 1.5
CO	6.2 ± 0.6	0.7 ± 0.2	0.5 ± 0.5	0.5 ± 0.5
O ₂	0.05 ± 0.01	0.04 ± 0.01	0.04 ± 0.01	< 0.02
HCl	36.4 ± 3.6	7.1 ± 0.7	2.7 ± 0.3	0.01 ± 0.01
Ar	0.45 ± 0.02	0.8 ± 0.1	0.9 ± 0.1	1.0 ± 0.1
CO ₂	15.1 ± 0.8	19.8 ± 1.0	19.3 ± 1.0	20.8 ± 1.2
C ₂ H ₂ Cl ₂	1.1 ± 0.2	0.25 ± 0.02	0.08 ± 0.02	0.06 ± 0.02
C ₂ HCl ₃	0.4 ± 0.1	0.4 ± 0.1	0.07 ± 0.02	0.12 ± 0.03
NO _x ^a	0.6 ± 0.2	0.06 ± 0.03	0.03 ± 0.02	0.01 ± 0.01
C ₂ H ₂	0.2 ± 0.1	0.04 ± 0.02	0.02 ± 0.02	ND
(CH ₃) ₂ CO ^b	0.5 ± 0.2	0.2 ± 0.1	0.11 ± 0.05	0.3 ± 0.1
	c	c	c	

^aM/e 30 peak could be NOCl.

^bThe hydrocarbon traces reported in the previous samples include (CH₃)₂CO.

^cSmall peaks at M/e 73 and M/e 93 for spectra obtained at 4, 10, and 30 min suggest traces of chlorodimethyl silane or chlorotrimethyl silane (mass spectroscopy artifacts).

The data obtained from all detoxification runs were analyzed in terms of (1) initial TCE loading in the carbon bed, (2) calculated chlorine loading in the carbon bed, (3) percent of original chlorine loading in the bed that is retained after reaction (from chlorine analysis of the carbon bed), and (4) percent of original chlorine in gas phase as HCl, $C_2H_2Cl_2$, and C_2HCl_3 . The latter was determined from mass spectroscopic analysis and HCl analysis of the AgCl recovered. The results for this analysis are given in Table 8.

Chlorine material balances for all seven runs are on the low side. Why the material balance is not better is not completely understood. However, this result can be partially explained; the absorption of HCl from gas phase in NaOH may not be complete, at least during the initial test period when the liquid volume

Table 6. Oxidation of Trichloroethylene (Run 105) Preadsorbed on Carbon without Catalyst (Calgon Carbon PCB)

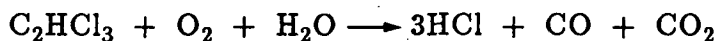
Oxidant: Moist Air with Flow Rate of 100 mL/min
Power Setting: 20% (Max = 600 W, 2.45-GHz Frequency)

Components	Amount, vol %					
	1 min	3 min	6 min	12 min	20 min	40 min
H ₂ O	0.18 ± 0.05	0.7 ± 0.2	1.0 ± 0.3	0.6 ± 0.2	0.9 ± 0.3	1.4 ± 0.3
N ₂	96.4 ± 1.0	77.4 ± 1.9	69.4 ± 1.7	72.7 ± 1.8	76.5 ± 1.9	71.3 ± 1.8
CO	trace	0.8 ± 0.4	7.5 ± 1.7	8.4 ± 1.8	3.0 ± 0.9	8.5 ± 0.7
O ₂	3.2	1.2 ± 0.1	0.09 ± 0.03	0.07 ± 0.02	0.04 ± 0.01	0.05 ± 0.01
HCl	< 0.1	9.6 ± 1.0	9.3 ± 1.0	5.0 ± 0.05	1.5 ± 0.2	0.13 ± .10
Ar	0.16 ± 0.01	0.14 ± 0.01	0.43 ± 0.01	0.74 ± 0.02	0.80 ± 0.04	0.80 ± 0.05
CO ₂	< 0.02	7.5 ± 0.04	9.3 ± 0.5	9.9 ± 0.5	13.4 ± 0.8	14.9 ± 1.0
C ₂ H ₂ Cl ₂	< 0.01	0.14 ± 0.04	0.25 ± 0.05	0.14 ± 0.04	< 0.10	< 0.10
C ₂ HCl ₃	< 0.01	1.5 ± 0.2	1.9 ± 0.2	1.9 ± 0.2	3.4 ± 0.4	2.7 ± 0.3
M/e = 30	trace	< 0.20	0.1	0.06	0.04 ± 0.02	< 0.01
C ₂ H ₂	trace	< 0.20	< 0.2	0.12	trace	trace
M/e = 73	trace	trace	trace	trace	trace	trace
M/e = 93	trace	trace	trace	trace	trace	trace
(CH ₃) ₂ CO	trace	trace	0.40	0.30	0.3 ± 0.1	0.1 ± 0.10

expansion is quite large, and some chlorine may be accounted for as chlorides of Cr and Cu, which were found to be deposited on the glass surface beyond the carbon bed.

The fraction of TCE converted during oxidative degradation in air streams was estimated in the following manner. Since the original TCE is converted into HCl gas and is also absorbed in carbon by reaction, the measure of the TCE converted is derived from the sum of the original chlorine loadings retained in the carbon and those in the gas phase (these appear as HCl and C₂H₂Cl₂ in Table 8). The total conversion of TCE into degradation products, on this basis, turns out to be greater than 80% in all cases. The oxidative degradation of TCE observed was highest with FCA carbon (i.e., catalyst-loaded active carbon) and with moist air.

The detoxification (breaking down to harmless products) of TCE preadsorbed on active carbon in a moist air stream under microwave heating can probably be described by



This reaction is complex because it involves, in part, catalysis, hydrogen abstraction, and thermal decomposition. Detection of one carbon atom molecule

Table 7. Oxidation of Trichloroethylene (Run 106) Preadsorbed on Carbon with Cu and Cr Catalyst (Calgon Carbon FCA)

Oxidant: House Air with Flow Rate of 100 mL/min
Power Setting: 20% (Max = 600 W, 2.45-GHz Frequency)

Components	Amount, vol %		
	2 min	5 min	10 min
H ₂ O	0.2 ± 0.1	0.2 ± 0.1	0.5 ± 0.2
N ₂	22.6 ± 1.1	57.9 ± 1.7	64.5 ± 1.3
CO	12.8 ± 1.1	4.2 ± 0.8	5.9 ± 1.0
O ₂	0.05 ± 0.01	0.04 ± 0.01	0.04 ± 0.01
HCl	41.1 ± 4.1	17.3 ± 1.7	5.4 ± 0.6
Ar	0.26 ± 0.02	0.67 ± 0.03	0.78 ± 0.03
CO ₂	16.3 ± 0.08	16.4 ± 0.8	19.9 ± 0.7
C ₂ H ₂ Cl ₂	2.4 ± 0.2	1.1 ± 0.1	0.7 ± 0.1
C ₂ HCl ₃	2.2 ± 0.2	1.7 ± 0.2	2.1 ± 0.2
M/e = 30	0.4 ± 0.2	< 0.1	< 0.1
C ₂ H ₂	0.8 ± 0.2	0.2 ± 0.1	< 0.1
CH ₃ Cl	0.3 ± 0.1	0.06 ± 0.03	0.05 ± 0.03
M/e = 73	< 0.1102	trace	ND
M/e = 93	< 0.3	trace	trace

such as CH₃Cl clearly suggests that the TCE molecule is thermally cleaved to produce chlorine, among other products.

C. Temperature of Carbon Bed during Microwave Heating

Since one advantage of microwave-assisted detoxification of TCE over the corresponding catalyzed oxidation of TCE conducted under thermal heating is its low-temperature operation, the temperature of the carbon beds during heating in the microwave oven (Model MDS-81) was measured. The active carbons, with and without the catalyst, have a high dielectric loss constant; consequently, they are expected to absorb microwave energy significantly.

All three types of carbons (base, FCA, and PCB carbons from Calgon Corp.) were subjected to microwave irradiation at 2.45 GHz (maximum power = 600 W). The mass of carbon used in the quartz reactor tube was 6 to 10 g during the measurements.

The temperature was determined in the following manner. After 5 min of continuous microwave irradiation, the temperature of the carbon bed was measured while gas flow through the bed was maintained. This was done by switching off the microwave power after 5 min and quickly inserting a thermocouple.

Table 8. Chlorine Mass Balance Between Reactant and Products of Reaction from Trichloroethylene Oxidation Experiments Conducted with Microwave Heating (20% power level, air flow at 100 mL/min, and reaction time of 60 minutes)

Run No.	Fresh Carbon (unused)	Air Flow	Adsorption		Desorption/Detoxification					Chlorine Accounted For, %
			TCE ^a Loading (g/g) in Carbon	Calc. Cl Loading (g/g) in Carbon	Cl in ^b Carbon Bed (% of original Cl loading)	Chlorine in Gas Phase, ^b wt %				
						Cl as HCl (from AgCl)	Cl as HCl (gas anal.)	Cl as C ₂ H ₂ Cl ₂ (gas anal.)	Cl as C ₂ HCl ₃ (gas anal.)	
101	FCA	dry	0.217	0.176	37.3	41.3	45.5	4.1	3.9	90.8
106	FCA	dry	0.284	0.230	36.0	32.9	37.6	5.2	12.1	90.9
105	Base Carbon	moist	0.107	0.159	12.5	36.2	42.0	2.2	32.6	89.3
127	PCB	moist	0.180	0.146	8.2	36.5	41.8	5.1	29.2	84.3
102	FCA	moist	0.215	0.174	28.7	52.6	48.7	2.9	3.6	83.9
114	FCA	moist	0.268	0.217	29.2	30.9	37.5	6.1	10.6	84.3
119	FCA	moist	0.168	0.136	35.1	37.6	43.2	3.5	6.2	88

^aAs TCE initially preadsorbed on carbon bed.

^bChlorine retained in the bed after reaction and as HCl, C₂H₂Cl₂, and C₂HCl₃ species in gas stream.

The temperature was then read every 40 seconds. An extrapolation of the temperature-time plot gave an estimate of the bed temperature during irradiation.

All three types of carbon, when irradiated at the 50% power level in an N_2 stream at 100 mL/min, attained temperatures close to 425°C. At 20% power level in an N_2 stream, the temperature was observed to be ~330°C after 2 min of heating and remained so after longer heating periods.

The temperatures attained by active carbon beds during detoxification experiments in moist air streams were also measured. For FCA carbon with preadsorbed TCE, when subjected to microwave radiation in a dry-air stream at a flow rate of 100 mL/min through the carbon bed (~8 g) at 20% power level, the temperature was ~425°C; when moist air was used, the temperature was ~355°C. These results can be seen in Fig. 3. Routinely, the values recorded in moist air were 50-100°C lower than those recorded in dry air. This is a key element for safety of the waste-treatment process. If an air and steam mixture flows over the carbon bed, the danger of carbons bursting into flame from temperature overruns may not exist.

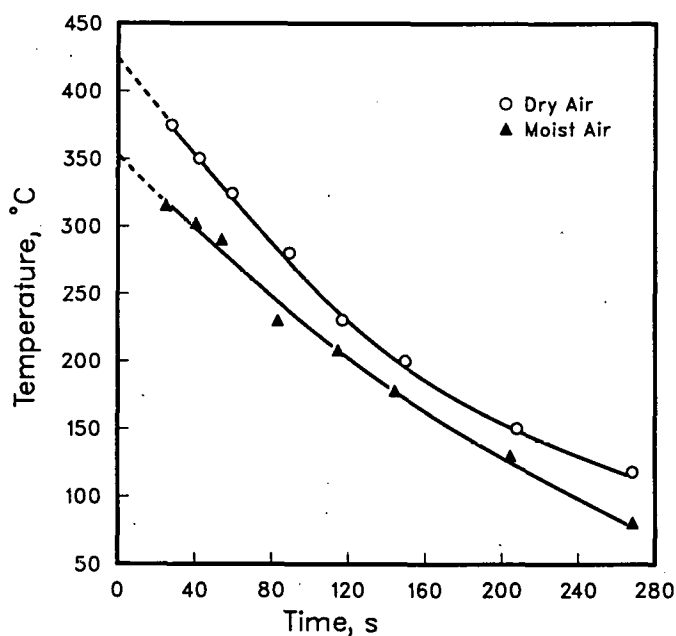


Fig. 3. Temperature of TCE Adsorbed on Fresh Carbon after Reaction with Dry and Moist Air at 100 mL/min for 5 min. (Thermocouple introduced into bed. Zero time is the time of power cut off to microwave oven. No gas flow during temperature measurement.)

D. Regeneration of Carbon

In order that the microwave-assisted detoxification process be cost effective, the active carbon (with or without catalyst loading), once used in detoxification of TCE, must be regenerated in the same system. Results from preliminary experiments suggest that this is feasible. During microwave-assisted TCE desorption, some TCE conversion to CO₂ and HCl was observed.

Desorption of TCE from the carbon bed was studied in four successive steps. In the first step, dry nitrogen was passed through the TCE loaded bed while the bed was subjected to microwave irradiation (2.45-GHz frequency, 25% power level). In the second step, dry nitrogen was passed through the bed, and the power was increased to the 50% level. In the third step, the nitrogen was presaturated with water at 22°C (partial pressure of 2.57 kPa) and passed through the bed with a power setting of 25%. In the fourth step, nitrogen was presaturated with water at 40°C (partial pressure of 7.19 kPa) with a power setting of 50%. The gas flow rate for the studies was maintained at 150 mL/min for 30 minutes.

During the desorption/detoxification step, the weight loss of the bed was determined after each step, and any HCl generated was determined by monitoring the chloride content of a 1.0 N NaOH solution that was used to scrub the exit gases. The results of the four-step study, shown in Table 9, indicate that HCl is

Table 9. TCE Desorption/Detoxification Characteristics of Active Carbon Beds^a

Treatment	Active Carbon with Catalyst		Active Carbon without Catalyst	
	TCE Mass Desorbed, wt %	TCE Converted to HCl, wt %	TCE Mass Desorbed, wt %	TCE Converted to HCl, wt %
I Dry nitrogen flow, 25% power for 30 min	50.9	14.4	63.9	16.2
II Dry nitrogen flow, 50% power for 30 min	9.9	1.7	22.2	4.9
III Moist nitrogen flow, ^b 25% power for 30 min	3.4	9.5	6.2	21.3
IV Moist nitrogen flow, ^c 50% power for 30 min	15.8	14.3	2.6	8.2
Total	80.0	39.9	94.9	50.6

^aThe amounts of TCE initially adsorbed were 0.168 g and 0.154 g on the catalyst-free and catalyst-containing beds, respectively.

^bVapor pressure is 2.57 kPa.

^cVapor pressure is 7.19 kPa

formed in each step, even in dry nitrogen. The presence of HCl in the exit gas indicates that TCE is being, in part, converted to HCl. Since the active carbons have only ~0.6% hydrogen and the total HCl produced far exceeds this amount, the TCE molecule must be decomposing to liberate HCl and free chlorine. This free chlorine probably reacts with the carbon bed since the chlorine in carbon is difficult to remove.

The results in Table 9 also indicate that, under *in situ* heating by microwave radiation, TCE and the detoxification products are more readily desorbed from a catalyst-free bed than from one containing a catalyst. Separate experiments indicated that chlorine retention in the reacted or used carbon adversely affected the detoxification or oxidative capacity of carbon. This effect is shown in the curves of Fig. 4 and the results of Table 10. It is not easy to remove the last traces of chlorine from used carbon. However, simple boiling with water reduces the chlorine content from 6.2 to 1.5 wt %. Passing water vapor through the used carbon bed (under microwave irradiation) removed only one-third of the chlorine. This may be due to the low concentration of water vapor (determined by saturated vapor pressure at 25°C) in the air stream. If we use a higher concentration of water vapor (e.g., steam plus air), we anticipate that regeneration of carbon would be more successful.

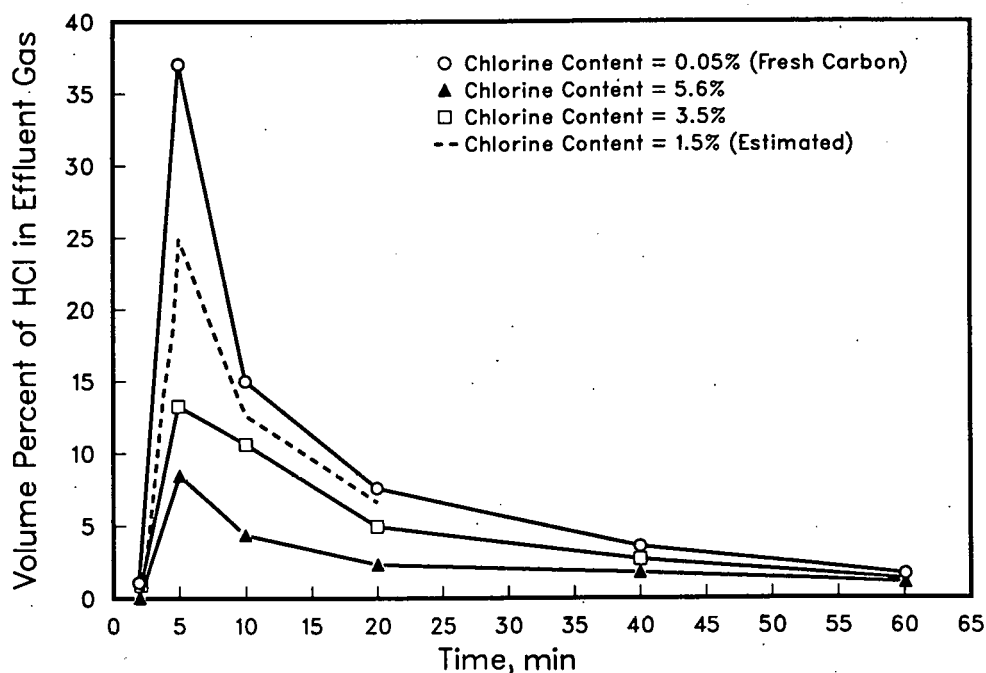


Fig. 4. Effect of Chlorine Content of Carbon on the Decomposition of TCE

Table 10. Moist Air Oxidation of TCE Adsorbed on Used Carbon
(FCA Variety)^a under Microwave Heating at 20% Power
Level for 60 Minutes with Moist Air Flow

Run No.	Initial Chlorine, wt %	TCE Adsorbed, g/g	Total Chlorine Loading, ^b g/g	Chlorine Content as % Total Chlorine Loading					Chlorine Accounted For, %
				Cl in Carbon Bed	Cl in HCl as AgCl ^c	Cl in HCl ^c (from gas anal.)	Cl in C ₂ H ₂ Cl ₂ (from gas anal.)	Cl in C ₂ HCl ₃ (from gas anal.)	
116	5.1	0.198	0.212	26.2	16.6	19.7	4.7	27.3	77.9
118	3.5	0.268	0.252	20.7	21.3	19.3	3.3	36.6	79.9
121	2.5	0.201	0.187	31.4	31.9	36.5	4.2	16.3	88.6

^aVariable chlorine content obtained by prior treatment (moist N₂ for 30 min at 20% power, washing in 90°C water).

^bEquals initial chlorine in carbon plus chlorine in TCE.

^cChlorine in HCl from gas analysis arbitrarily chosen for mass balance.

IV. DESIGN OF 6-kW MICROWAVE REACTOR

A microwave heating assembly was designed, procured, and fabricated. This assembly consists of (1) a microwave generator (Cobler Electronics) capable of providing 6-kW adjustable power at 2.45 GHz with 16-kW power input, (2) a microwave cavity (2 x 2 x 4 ft) fabricated from cold-rolled steel and equipped with a hinged door with a choke-screen/glass window, (3) an infrared temperature sensor operable at a 250-800°C temperature range, (4) quartz tubing, which has 48-mm OD and 72-cm long sections with quartz standard taper joints, (5) the required connectors, tuners, wave guide sections, flanges, and (6) a microwave power meter. The design of the assembly is shown in Fig. 5. The facility is not yet operational in the laboratory because of funding cuts. The installation and testing of the assembly will be conducted as soon as funds become available.

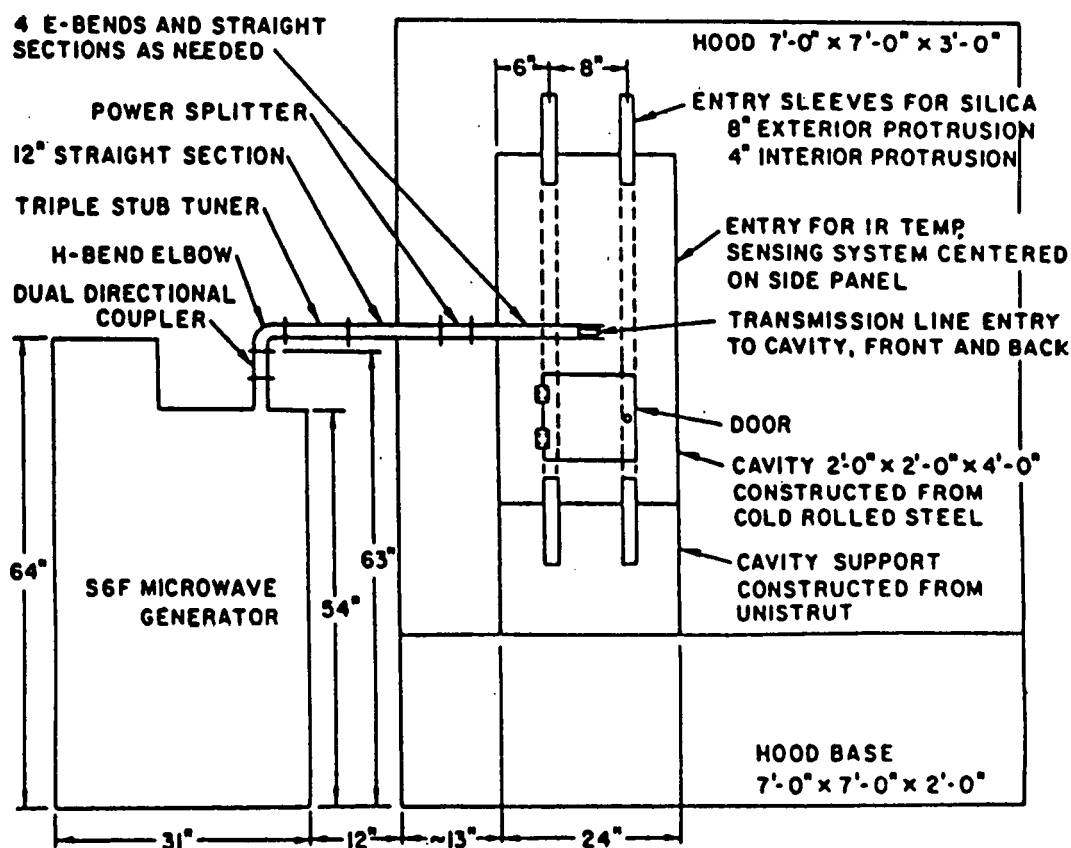


Fig. 5. Microwave Heating Assembly

V. CONCLUSIONS

Our experimental study has established that TCE in a vapor stream can be adsorbed on an active carbon bed (loaded with Cu and Cr catalysts) of adequate length at room temperature. On heating the bed with microwave radiation to

moderate temperatures ($<400^{\circ}\text{C}$) while a moist air stream is passed through it, the TCE is readily detoxified, i.e., converted into HCl , CO , CO_2 , and $\text{C}_2\text{H}_2\text{Cl}_2$. Conversion higher than 80 wt % has been observed.

The mass flow rate of oxygen should be such that the exit streams do not contain any free oxygen. This precaution will assure safety and avoid fire hazards in any temperature overruns. Also, if the moisture content in the air stream is kept high (e.g., 20%), the risks for temperature overruns leading to fire are avoided.

Chlorine removal from the active carbon is critical to the regeneration of used carbon. Results from preliminary experiments have suggested that regeneration of used active carbon is possible. The used carbon can be conveniently regenerated by microwave heating while moist N_2 (or better still, steam mixed with an air stream) is passed through the bed. Several Japanese patents¹⁻⁴ report that active carbons can be regenerated using microwave radiation in the frequency range of 0.9 to 4 GHz. The temperature attained during regeneration is claimed to be in the range from 450 to 550°C.

Our research has laid the groundwork for a successful demonstration of new technologies for detoxifying liquid hazardous organic wastes such as TCE, tetrachloroethylene, trichloroethane, and polychlorobiphenyls. The proposed new technology may serve the dual purpose of detoxifying the TCE and regenerating the carbons. Our preliminary cost calculations show that the TCE removal cost from vapor will be ~\$1.20 per pound of TCE, and the carbon regeneration cost will be ~\$0.18 per pound of carbon for the proposed process.

Another concept for detoxifying halohydrocarbons would utilize microwave-assisted fluidized-bed combustion (using suitable ceramic, e.g., yttria pellets or granules) at 600-700°C in a single-step flow process. This is a cost-effective technology worth considering.

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