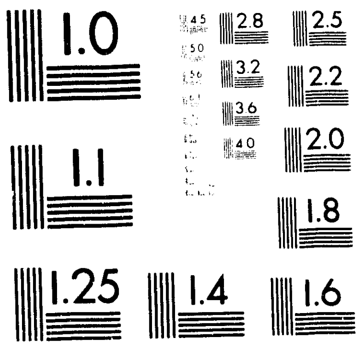
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UNVENTED THERMAL PROCESS FOR TREATMENT OF HAZARDOUS AND MIXED WASTES

by

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ABSTRACT

An Unvented Thermal Process is being developed that does not release gases during the thermal treatment operation. The main unit in the process is a fluidized-bed processor containing a bed of calcined limestone (CaO), which reacts with gases given off during oxidation of organic materials. Gases that will react with CaO include CO_2, SO_2, HCl, HBr, and other acid gases. Water vapor formed during the oxidation process is carried off with the fluidizing gas and is removed in a condenser. Oxygen is added to the remaining gas (mainly nitrogen), which is recirculated to the oxidizer. The most flexible arrangement of equipment involves separating the processor into two units: an oxidizer, which may be any of a variety of types including standard incinerators, and a carbon dioxide sorber. A key feature of a 15-cm fluidized-bed experimental unit now being prepared for experiments is the ceramic candle filters at the top of the bed, which retain the solid material in the bed while permitting the gases to leave. Lime is continuously added to this experimental unit and continuously withdrawn through an overflow pipe.

Several approaches have been considered for handling the spent lime, which is primarily calcium carbonate. It may be (a) mixed with cement-making materials and buried, (b) trucked to a cement manufacturer for inclusion in its products, or (c) transferred to a calciner and heated to 800-900°C to drive off the CO_2 and permit recycling of the lime. The third approach, which minimizes the volume of the final waste stream, divides the destruction of the organic material and the decomposition of the calcium carbonate into separate steps carried out in different equipment. Thus, the likelihood of the release of toxic materials would be greatly reduced from that sometimes associated with standard incinerators. The key factor in the acceptance of the process is believed to be the extent to which the unvented feature facilitates the process of obtaining an operating permit.

INTRODUCTION

When operated properly, incinerators can achieve high efficiency in destroying toxic organic materials in treating both hazardous wastes and mixed wastes (1-4). However, problems arise during improper operations or upset conditions, when dioxanes, furans, and other toxic materials may be released at dangerous levels. By means of careful design and operation, which are regulated by the Environmental Protection Agency, and various state and local agencies, dangerous emission levels can be avoided.

Satisfying the local public on all aspects concerning the operations of an incinerator is often more difficult than demonstrating to the scientific community and the appropriate agencies that the emission standards can be met. The public's main concern with incinerators is the risk of toxic organic emissions or radioactive emissions. The public perceives nearby incinerators to be a risk to health imposed by the government or industry, and also a factor tending to lower property values (4). As a result, the siting of incinerators has become very contentious and expensive (5). A process that does not produce emissions, such as the proposed Unvented Thermal Process described herein, may meet with greater public acceptance.

Work has been done previously to reduce or eliminate the emissions from incinerators (6,7). Stull and Golden at EG&G Rocky Flats have evaluated off-gas capture systems from a fluidized-bed oxidation unit (6). In the two systems evaluated, 85% of the off-gas from the fluidized bed oxidizer would be recycled with oxygen makeup, and the remaining 15% would be compressed to liquid CO_2, stored in tanks, sampled, and ultimately released. One process requires compression to 75 bar (1100 psia) at 16°C (60°F), and the other requires compression to 24 bar (350 psia) at -26°C (-15°F). Another approach was taken by Camp and Upadhye of Lawrence

Livermore National Laboratory (LLNL) (7). In their proposed process, the mixed waste could be destroyed in any one of many types of incinerators with an oxidant of oxygen and recycled CO_2. The off-gases would be directly quenched with water in a venturi scrubber. Acid gases would be removed by reacting with sodium hydroxide in a packed tower, and CO_2 would be removed in a second packed tower by contacting with calcium hydroxide. The remaining oxygen, CO_2, water vapor, and trace gases would be recycled to the waste destructor with makeup oxygen. The solid materials in the slurries exiting the packed towers would be consolidated to moist solids, which could be buried. The LLNL process would produce a large amount of solids for burial, which was viewed by them as a disadvantage.

A process is under development at Argonne National Laboratory (ANL) called the Unvented Thermal Process for treating mixed wastes that have accumulated at ANL over several decades. The type of mixed waste stored at ANL that has the largest volume is scintillating counting waste, which is stored in 200 drums of 55-gal. capacity. Each drum holds 2000-4000 vials of toluene-based scintillation fluid containing radioactive materials from a wide range of projects. The vials are packed in vermiculite, which also must be treated as a mixed waste. With the proper head-end step to separate the organic liquid from the balance of the materials, this waste inventory could be treated by the Unvented Thermal Process that we are developing. Also, the biological mixed waste at ANL could be treated by the proposed process, which would destroy the biological materials and separate of the inorganic low-level radioactive waste for burial.

The treatment of these wastes at the ANL site is a difficult problem because ANL is located near suburban residences. Installation of a conventional incinerator would be out of the question. To meet this need, we have conceived the Unvented Thermal Process, which does not release gases from the equipment during the thermal destruction of organic materials. The only products from the Unvented Process are liquid water and calcium carbonate. In treating the limited quantities of ANL waste, the calcium carbonate and water produced in the Unvented Process would be mixed with cement-making materials to produce cement. This solid material would be the only product from the process.

The Unvented Thermal Process is being patented and is discussed in more detail below.

DESCRIPTION OF PROCESS

The key feature of the Unvented Thermal Process (Fig. 1) for treating mixed and hazardous wastes is that it does not release gases during the thermal treatment operation. The main unit in this process is a fluidized-bed reactor containing a bed of calcined limestone (CaO), which reacts with gases given off during oxidation of organic materials. Gases that will react with CaO include CO_2, SO_2, HCl, HBr, and other acid gases. Water vapor formed during the oxidation process is carried off with the fluidizing gas and removed in a condenser. Oxygen is added to the remaining gas (mainly nitrogen), which is recirculated to the oxidizer. Thus, for most organic waste materials, there is no net production of gas by the reactions occurring within the equipment. A small amount of nitrogen might need to be collected as a result of (a) nitrogen impurity contained in the oxygen feed, (b) leakage of gas into the equipment through flanges, or (c) nitrogen released during the destruction of some organic compounds such as amines.

Insert Fig. 1 here)

The most flexible arrangement for the equipment involves separating the processor into two separate units: an oxidizer and a carbon dioxide sorber, as shown in Fig. 2. The oxidizer can be operated at high temperatures (700-900°C) to ensure complete oxidation, whereas the sorber can be operated at a lower temperature (500°C) appropriate for CO_2 sorption. As indicated by the equilibrium data in Fig. 3, $CaCO_3$ is stable at the operating conditions in the sorber, but $Ca(OH)_2$ is not. In this process, the oxidizer could be (a) a fluidized bed with a mixture of sand and lime for bed material, (b) a slagging combustor, or (c) various conventional incinerators. In all cases, the gas from the sorber is cooled to condense the water and recirculated to the oxidizer with sufficient oxygen to support combustion. Lime is continuously fed to the sorber bed and continuously withdrawn thorough an overflow pipe. The pressure at the outlet of the Fig. 1 circulating blower will be controlled at slightly less than atmospheric pressure to assure that any leaks in the system do not result in the escape of hazardous materials.

Insert Fig. 2 here)

Insert Fig. 3 here)

If all of the reaction products are converted to cement, as intended for the ANL wastes, the total volume of the waste material might be somewhat larger than the initial organic waste volume because of the conversion of the carbon in the organic material to $CaCO_3$. This moderate increase in volume is not important in treating the ANL wastes because the total waste volume is not large and more serious problems than the waste volume are solved by the new processing scheme. This approach may also be practical for treatment of certain mixed and hazardous wastes found at other sites.

For medical wastes and other organic wastes generated at moderate rates on a site, it may be inappropriate to convert the products of the Unvented Thermal Process to cement. The spent lime could be analyzed to demonstrate virtual complete destruction of the organic matter and then trucked to a cement manufacturer (which may also be the supplier of the lime) for inclusion in the products of that manufacturer.

Alternatively, the volume of the products from the Unvented Thermal Process and of the lime supplied as reactant may be greatly reduced yet retain the main features of the process. To reduce these volumes, spent lime (mainly calcium carbonate) would be calcined in a separate unit to decompose the $CaCO_3$, recover the lime, and release carbon dioxide (upper right-hand corner of Fig. 2). The most energy-efficient form of this process would be to transfer the hot bed material from the sorber to the calciner without cooling. To facilitate the reuse of the spent lime leaving the sorber, it would be advantageous to add lime to the oxidizer if acid gases such as SO_2, HCl, or HBr are being produced. This will result in the retention of sulfur and the halogens in the oxidizer ash. The source of the lime for the oxidizer would be a small side stream taken from the spent lime in the sorber. Thus, the only solid waste stream from the process would be the ash from the oxidizer. The volume of ash per unit mass of waste destroyed would be about the same as for a standard fluidized-bed incinerator to which limestone is added to retain the acid-gas anions with the ash.

The division of the (a) destruction of the organic material and (b) decomposition of the calcium carbonate into separate steps carried out in different equipment greatly reduces the likelihood of the release of toxic materials from that associated with standard incinerators. Although this assertion must be demonstration by experiments, we are confident of the result. Only gases enter the sorber, and these are almost entirely free of the hazardous organics fed to the oxidizer. Only a small fraction of the organic material that escapes the oxidizer will be retained on the hot spent lime released from the sorber. Most of the gas, including unreacted organics, will pass through the sorber and be recirculated to the oxidizer. To minimize the amount of organic material released from the calciner, it must be indirectly heated and thus involve no air or sweep gas addition. However, it may be practical to add a small stream of gas from the inert gas collection tank (which may be treated and sampled if desired) to dispose of the small amount of nitrogen entering the process, primarily as an impurity with the oxygen supply. Under these conditions of restricted gas addition, no gas is released from the calciner if the temperature inadvertently drops below the minimum calcining temperatures of 800-900°C. This approach results in long retention times at high temperatures for destroying the tiny fraction of organic gases which may be clinging to the spent lime. The demonstration that the reacted lime from the sorber bed contains virtually no organic material is an important feature of the experimental program because it would confirm this concept. The off-gas from the calciner as operated above, would be more than 99% carbon dioxide with small amounts of oxygen and nitrogen, even with the addition of gas from the inert gas collection tank as proposed above. Its volume per unit of waste destroyed would be about 10% of that from a conventional incinerator. The off-gas treatment system required for this carbon dioxide stream would consist of cooling and filtering equipment, and should be comparatively inexpensive.

If the waste to be treated contains hazardous metals or radioactive solids, it may be appropriate to vitrify the ash from the oxidizer. Alternatively, the oxidizer may be operated at an enriched oxygen level to produce a glassy slag directly. Operating at high oxygen levels is easily done in the Unvented Thermal Process because the oxidant added to the process is pure oxygen.

EXPERIMENTAL

An experimental program is underway at ANL to develop the Unvented Thermal Process. The experiments will be carried out with a single processing unit, as shown in Fig. 1. The main processing unit for these studies is a fluidized-bed reactor used previously for studying fluidized-bed combustion of coal (Fig. 4). This fluidized-bed reactor has an internal diameter of 15.2 cm (6-in.) and in the previous projects was equipped for continuous feed of limestone (for removal of sulfur) and continuous feed of powdered coal.

(Insert Fig. 4 here)

In the initial experiments on waste burning, the unit will be operated with once-through gas flow and with toluene fed as a model waste compound. The bed temperature will be raised to the ignition temperature by preheating the fluidizing/combustion air stream. Once the bed is at the ignition temperature, toluene will be injected into the fluidized bed, and the experimental conditions of bed temperature and the rates of toluene and lime feed will be established. The toluene and lime will be fed to the processor continuously, and bed material will continuously overflow into a closed collection vessel. Bed temperature will be maintained by controlling the toluene feed rate and by employing an in-bed heat exchanger. The planned range of test conditions is summarized in Table I.

Table I. Planned Test Conditions

Parameter	Value
Liquid feed	Toluene
Liquid feed rate, kg/h	0.20-0.36
Lime (CaO) feed rate, kg/h	1.5-2.3
CaO/C mole feed ratio	1.1-1.5
System pressure, bar	1.0
Bed temperature, °C	600-700
Fluidizing velocity, m/s	0.76-0.91

The objective of these initial tests will be to measure and verify basic process performance parameters, such as the efficiency of toluene destruction in a single pass through the fluidized bed, the extent of the conversion of CaO to $CaCO_3$ as a function of the toluene-to-lime feed ratio, and the CO_2 concentration in the off-gas. It will also be important to verify in these early tests that the reactive lime collected in the bed overflow is not contaminated by organic material.

Modifications to the facility have been designed and equipment procurements initiated to convert the existing fluidized-bed reactor to an Unvented Thermal Processor with gas recirculation. The modified equipment layout is illustrated in Fig. 5. A key component in the modified facility will be a candle filter assembly for filtering the dust from the combustion gases and returning the solids to the fluidized bed. Other components include a water condenser and condensate separator to remove water from the gas stream, a gas recycle blower, and oxygen addition equipment.

(Insert Fig. 5 here)

The present reactor system shows a slight tendency for the pressure to rise as a result of nitrogen contained as an impurity in the oxygen supply and in air leaking into the equipment through flanges. To control the pressure at the outlet of the circulating blower to slightly below atmospheric pressure, gas will be withdrawn by a compressor and stored at elevated pressure in a holding tank. This relatively small amount of gas may be analyzed and, if necessary, treated prior to release to the environment.

When the modifications have been completed, experiments will proceed using toluene and carbon tetrachloride as model compounds to demonstrate the destruction of organic materials without release of gases. Preliminary plans are underway for a pilot plant facility for treating both liquid and solids wastes that have been generated at ANL in a process having separate oxidizer and sorber, as shown in Fig. 2.

MATERIAL AND HEAT BALANCES

To aid in assessing the practicality of the proposed process, a calculational program was developed with the use of Microsoft Excel software. The program evaluates the performance of the least complicated process (Fig. 1), which is also the configuration of our experimental unit. The program calculates the compositions and the content of the inlet and outlet gas streams and the inlet and outlet bed material streams. The calculation of the bed densities and volumes is based on our experience in using lime in the fluidized-bed reactor for coal-burning experiments. In those experiments, it was found that the lime particles obtained by calcining limestone have a calcium density of 0.024 g-mol/cm^3. The particles retain their size throughout the process while changing composition and density.

Some of the key parameters calculated from the computer analysis are shown in Table II. These sets of calculations were made for a liquid feed mixture of toluene-20 wt. % carbon tetrachloride. Both of these constituents of the feed mixture are contained in Argonne's mixed waste inventory, and it was of interest to determine how the process would handle chlorinated hydrocarbons, a common type of hazardous and mixed-waste constituent. The first two columns of values in Table II are for the experimental reactor, which has an inside diameter of 15.2 cm (6 in.). The results shown in the first column are for a feed rate just high enough to maintain the operating temperature of the reactor without cooling the bed. The calculations in the second column are for the maximum feed rate, which was taken to be that which required an oxygen concentration of 70% for the inlet fluidizing gas. The experimental reactor has a bed depth of only 91 cm (3 ft), which nearly coincides with the maximum practical amount of cooling tubing within the bed.

(Insert Table II here)

Calculations are shown for a large-scale reactor in the right-hand column of Table II. In scaling up the reactor, the program calculates the height of the expanded fluidized bed to be proportional to the 0.4 power of the diameter. For this reactor of 5 m-diameter, the height is 3.7 m. This configuration results in an outlet gas pressure of only 0.612 bar, indicating that the bed depth is near the practical limit. This deep bed results in a long retention time for the bed material, 68.5 minutes, which should facilitate achieving the assumed extent of lime reaction of 70%.

With the aid of the computer program, several charts were made to illustrate the effects of various parameters on the performance of the unvented processor. Figure 6 illustrates the difference between the amount of bed cooling required for treating toluene and carbon tetrachloride as a function of feedrate. It is apparent that the unvented processor is better adapted to processing large throughputs of chlorinated hydrocarbons than fuel-like materials such as toluene. The chlorinated hydrocarbons not only produce less heat but also require less oxygen.

(Insert Figure 6 here)

A problem in treating chlorinated hydrocarbons is the formation of calcium chloride, which may result in a tendency to particle agglomeration. Two solutions to this problem are proposed. One is to design a separate oxidizer and sorber, as shown in Fig. 2. The other is to blend the chlorinated hydrocarbon with other hydrocarbons to reduce the concentration of calcium chloride in the bed material. This approach is illustrated in Fig. 7, which shows that for a mixture of toluene-20 wt. % carbon tetrachloride, the bed material at equilibrium would contain only about 4% calcium chloride. Such a low concentration of calcium chloride is not likely to result in bed caking.

(Insert Figure 7 here)

The effect on the processor capacity of increasing the reactor diameter and of adding boiler tubes within the bed for cooling is illustrated in Fig. 8. For the assumed feed material, the feed rate increases by about an order of magnitude when bed cooling is provided; for the case of no bed cooling, the heat would be carried off with the fluidizing gas and through the reactor walls. The slight curvature in the line for no bed cooling is due to the proportionally higher rate of heat loss through the reactor walls for small reactors.

(Insert Fig. 8 here)

In future calculations, the study will include the use of separate oxidizers and sorbers and the sizing of the ancillary equipment. Also, transient analyses will be carried out to determine the time required to reach equilibrium conditions and the effects of various upset conditions.

TYPES OF WASTE SUITABLE FOR TREATMENT

Industrial and governmental installations produce many types of hazardous wastes requiring oxidation of organic material during treatment. Some liquid and solid wastes have very high heating values, while others require the addition of an auxiliary fuel to support combustion. Mixed wastes are particularly difficult to treat by standard incineration processes because the emissions may contain radioactive volatile matter or fine particles that escape filtration of the gaseous effluent. These problems are greatly alleviated by the Unvented Thermal Process, even for the version of the process where the spent lime is regenerated, because of the isolation of the lime during calcining from both the gases formed on destruction of the organic material and the ash which contains radioactive solids.

The need for pure oxygen as the oxidant for the Unvented Thermal Process is a cost factor that affects the types of wastes that can be economically treated. The combined effects of the cost of oxygen and the avoidance of emissions result in the following types of wastes being favored for treatment by the Unvented Treatment Process:

- Industrial, military, and DOE wastes containing chlorinated hydrocarbons. Such wastes have low oxygen requirements for destruction.

- Solid and liquid mixed wastes. These are particularly hazardous to destroy by standard incineration.

- Hospital wastes. These are generated near residential property, are not easily trucked away, and have only a moderate oxygen requirement.

MANUFACTURING AND OPERATING COSTS

It is anticipated that equipment and installation costs for an Unvented Thermal Process will be comparable to the cost of other advanced technology processes for treatment and disposal of hazard or mixed wastes. The oxidizer and sorber can be combined into one unit, a fluidized bed containing lime, for destruction of liquid combustible wastes. For most other wastes, including all solids and sludges and liquids having high chlorine, bromine, or sulfur content, the oxidizer and sorber would be constructed as two separate units.

The oxidizer and sorber, or the combined unit with these functions, will be constructed primarily of carbon steel and lined with refractory materials, as is typically done in the construction of commercially available boilers and furnaces. Fluidized-bed combustors are currently used for converting waste to energy while fully complying with applicable regulations. Suppliers of these combustors typically develop system designs and specifications for the fabrication of components and procurement of auxiliary equipment. Worldwide sources for commercial fluidized-bed combustion equipment are also available for supplying the equipment for the Unvented Thermal Process.

For erection of a facility for the Unvented Thermal Process, the carbon dioxide sorber, water condenser, and recirculation equipment would be installed instead of the extensive exhaust treatment system of a conventional incinerator. For the version of the process in which the lime is regenerated and recycled on site, the off-gas from the calcining unit would have only 10% of the volume of the off-gas from a conventional incinerator and would require only cooling and filtration to remove solids. On-line monitoring, sampling, and analytical costs are likely to be lower for the Unvented Processor than for an incinerator.

The oxygen requirements for the combustor is known to be a major cost item. A preliminary analysis indicates that the cost of the oxygen will be $50 to $300 per ton of waste, depending on the oxygen requirement of the particular waste treated. The oxygen cost will be offset to some extent by the higher throughput available for an oxidizer if supplied with gas at high oxygen concentration.

CONCLUSIONS

Much experimental work remains to be done to establish the kinetics of CO_2 sorption for actual processing conditions and to establish the feasibility of recycling the spent lime to achieve low waste volumes. Work is also needed to demonstrate that virtually no organic material is given off with the CO_2 on calcining the lime. Our experiences with fluidized-bed coal combustion and the favorable reaction conditions indicate that the experimental results are likely to be positive.

Various market niches in the treatment of hazardous wastes and mixed wastes could be filled by versions of the Unvented Thermal Process that we propose. The key factor in the acceptance of the process is the extent to which the unvented feature facilitates the process of obtaining an operating permit for an actual installation.

ACKNOWLEDGMENTS

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Table II. Calculated Performance of Unvented Processor

System Parameters	Experimental Reactor: No Bed Cooling	Experimental Reactor: Maximum Rate	Large Scale Reactor Maximum Rate
Reactor internal diameter, cm	15.2	15.2	500
Control temperature, °C	700	700	700
Feed Composition, wt %			
Toluene	80	80	80
Carbon tetrachloride	20	20	20
Feed rate, kg/h	0.991	5.84	6,320
Oxygen reaction requirement, g-moles/min	1.31	7.73	8,360
Bed Material Parameters			
Expanded bed height, cm	91	91	370
Volume of boiler tubing within bed, liters	0	2.61	3,180
Ca density of particles, g-moles/cc	0.024	0.024	0.024
Percent CaO reacted	70	70	70
Feed rate, kg/h	5.14	30.2	32,700
Discharge Rate, kg/h	7.98	47.0	50,900
Bed Retention time, min	104	14.9	68.5
Inlet Gas Conditions			
Temperature, °C	40	40	40
Pressure (absolute), bars	0.988	0.988	0.988
Oxygen concentration, vol %	20.2	70.0	70.0
Linear flow rate at reactor temp., ft/sec	3.29	2.25	3.08
Fraction of carbon dioxide reacted, %	90	90	90
Outlet Gas Conditions			
Temperature, °C	700	700	700
Pressure (absolute), bars	0.846	0.846	0.612
Oxygen concentration, vol. %	10	10	10
Linear flow rate at reactor temp., ft/sec	3.29	2.25	3.08
Reactor Heat Balance, (25°C basis), kW			
Heat entering with fluidized gas	0.091	0.094	102
Heat from burning toluene mixture	8.93	52.5	56,800
Heat from lime reactions			
Reaction of CO_2 from toluene	3.0	17.7	19,200
Reaction of CCl_4	0.3	1.73	1,870
Heat leaving with fluidizing gas	4.05	3.1	3,370
Heat leaving with bed material	1.56	9.18	9,950
Heat losses through reactor wall	6.67	6.67	78.3
Heat removed by cooling coil, kW	0	53	64,600

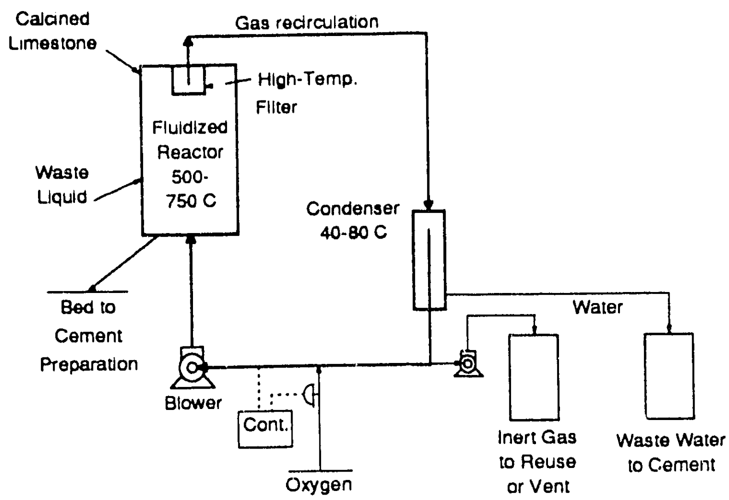

Fig. 1. Unvented Thermal Process with oxidation and carbon dioxide sorption combined in one unit.

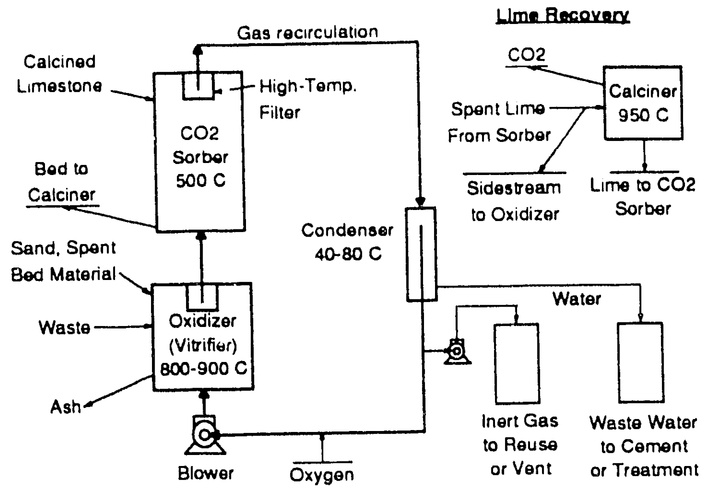

Fig. 2. Unvented Thermal Process with separate oxidation and sorption units and provision for lime recycle.

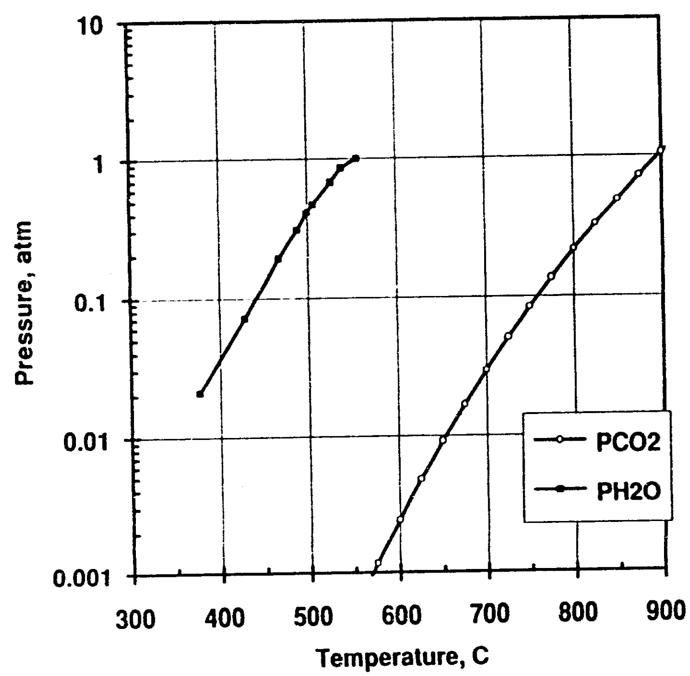

Fig. 3. Equilibrium gas pressures for calcium carbonate (CO_2) and calcium hydroxide (H_2O). Data from International Critical Tables.

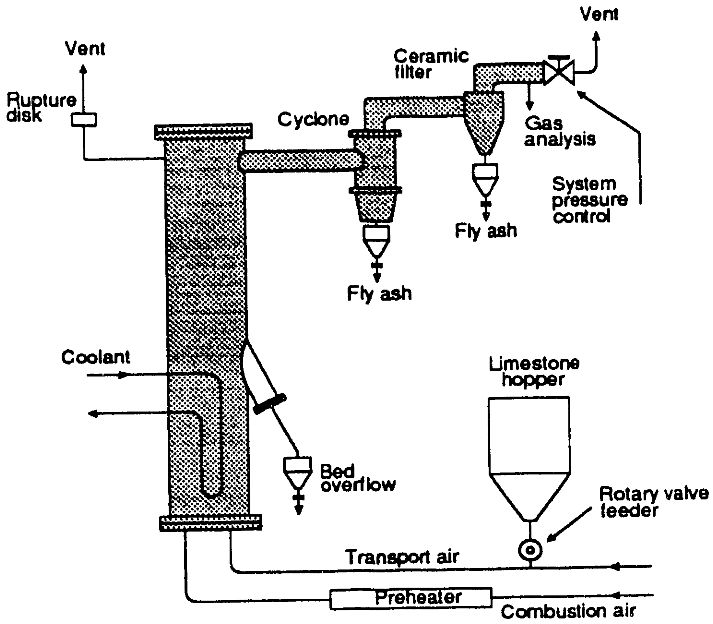

Fig. 4. Experimental fluidized-bed processor with once-through gas flow.

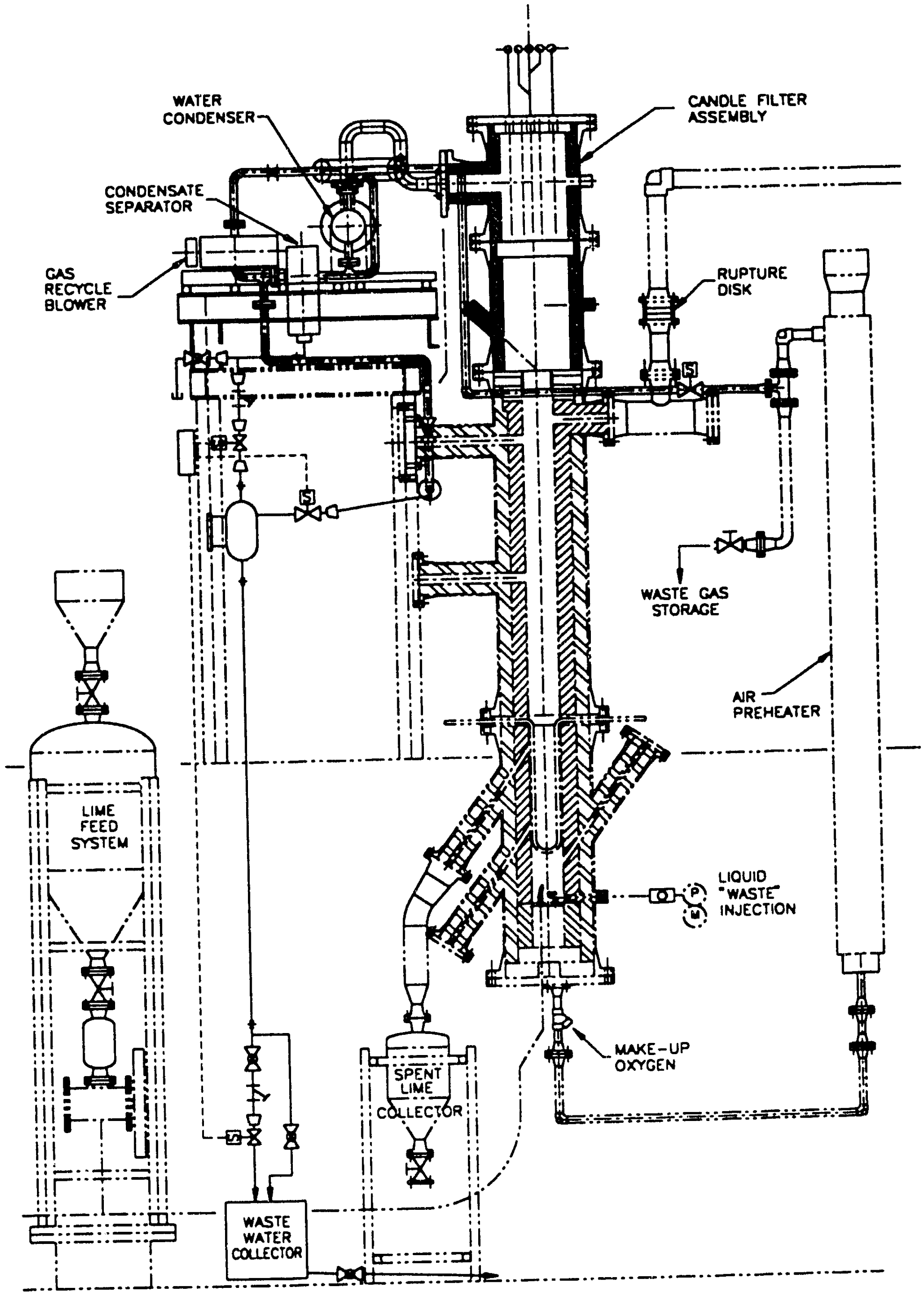

Fig. 5. Experimental fluidized-bed processor designed for gas recirculation as in Fig. 1.

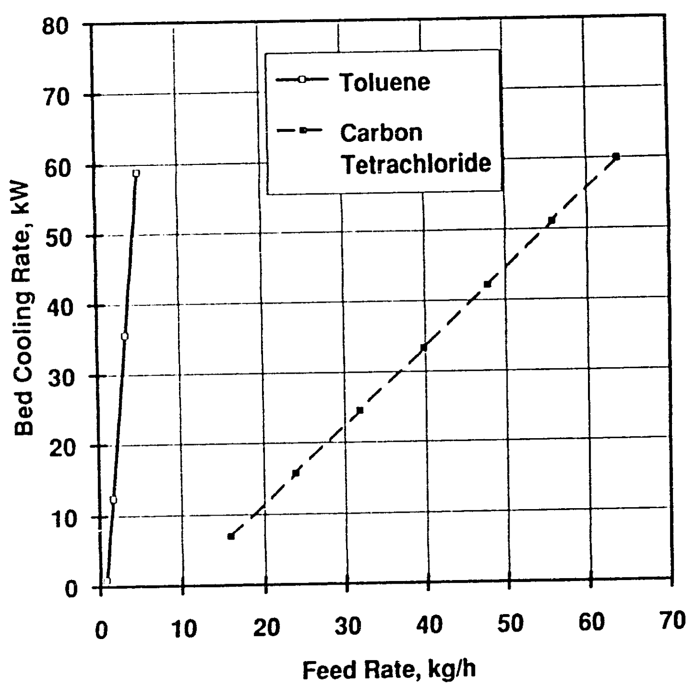

Fig. 6. Calculated cooling rate required for experimental Processor. Reactor diameter: . 15.2 cm; lime reaction: 70%; outlet oxygen concentration: 10%.

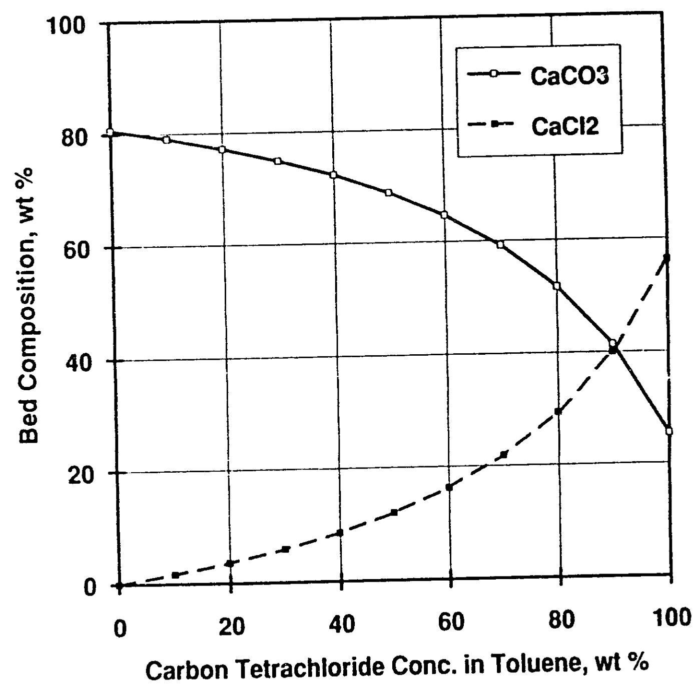

Fig. 7. Calculated effect of carbon tetrachloride in the waste feed material on the composition of discharged bed material. Extent of lime reaction: 70%.

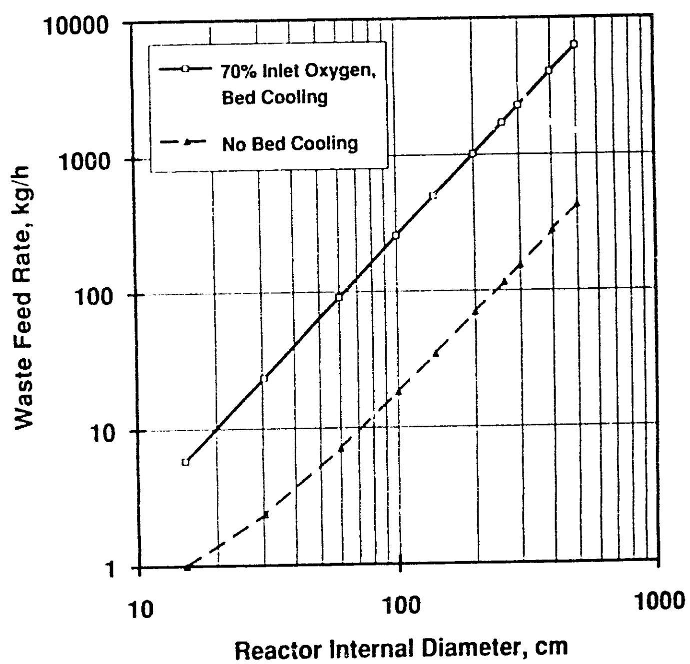

Fig. 8. Scale-up of unvented processor. Liquid waste feed mixture: toluene - 20 wt% carbon tetrachloride; temperature: 700°C; outlet oxygen concentration: 10%.

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