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10th INTERNATIONAL SYMPOSIUM ON GAS FLOW CHEMICAL LASERS

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05 SEPTEMBRE 1994

CO_2 Laser-Aided Waste Incineration

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ABSTRACT

Lasers are widely employed in laboratories and in certain industrial applications, notably for welding, cutting and surface treatment. This paper describes a new application, incineration, which appears warranted when the following features are required: high-temperature incineration (> 1500°C) with close-tolerance temperature control in an oxidizing medium while ensuring containment of toxic waste.

These criteria correspond to the application presented here. Following a brief theoretical introduction concerning the laser/surface interaction, the paper describes the incineration of graphite waste contaminated with alpha-emitting radionuclides. Process feasibility has been demonstrated on a nonradioactive prototype capable of incinerating 10 kg·h^{-1} using a 7 kW CO_2 laser. An industrial facility with the same capacity, designed to operate within the constraints of an alpha-tight glove box environment, is now at the project stage. Other types of applications with similar requirements may be considered.

Keywords: graphite, high-temperature incineration, radioactive alpha containment, CO_2 laser.

1. THE GRAPHITE CYCLE

By virtue of several ingenious mechanisms, minerals and living organisms develop and evolve using energy derived from a few sources. In this extraordinary natural system, humans have modified certain processes to satisfy real or imaginary needs.

Changes, however, require energy. Human activities have thus upset certain natural equilibria or kinetic processes by using energy to create "objects", in the broad sense of the term, that do not exist naturally. The return to a natural equilibrium, i.e. the destruction or disappearance of these objects, is generally slower than the production capacity. Hence the very real and very recent problem of waste.

One such new material is graphite. Considerable sums of human intelligence and energy have been mobilized to endow graphite with certain properties (hardness, conduction, resistance to oxidation and high temperatures) that constitute the utility of this material.

Because of these properties, however, its natural half-life is immensely long. This means that means must be developed to destroy it, restoring it to its natural, original form: carbon dioxide, from which it is produced at the conclusion of a process lasting billions of years (Figure 1).

Most known methods for burning graphite of this type (burners, fluidized bed furnaces, etc.) require prior crushing and fine milling – in some cases to 30 µm – to increase the graphite surface area in contact with oxygen and ensure an acceptable combustion rate under industrial conditions at standard incineration temperatures.

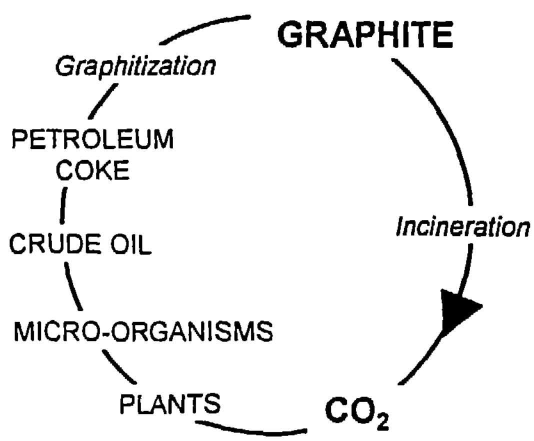

Figure 1. The graphite cycle

This procedure has a number of drawbacks:

- fine milling results in a substantial volume increase requiring very large storage facilities;
- the milling process generates wastes in the form of gas or liquid filters which must be treated as radioactive waste;
- milling is made more difficult by the hardness of nuclear-grade graphite;
- these operations may add impurities to the initially extremely pure graphite, and thus further increase the final ash volume.

Moreover, while combustion must be nearly ideal to minimize the final ash volume, the efficiency of burner-type combustion systems is not easy to maintain. This is attributable to the difficulty in controlling the flame temperature and the residence time of each particle inside the flame. A fluidized bed system provides greater control over the residence time, but contaminates the combustion gas with particles from the fluidized bed itself, and thus increases the ash volume.

The research work discussed here obviates these problems by burning the graphite blocks without crushing, using a powerful infrared laser.

2. GRAPHITE WASTE MATERIAL

This graphite is extremely pure, containing only 100 ppm of impurities and no volatile matter after successive graphitization treatments at 2700–3000°C in an electrically heated furnace.

At low temperatures, the material porosity has an appreciable effect on combustion reactions since it increases the internal surface area in contact with the oxidant gas. This effect is attenuated when the reaction occurs at high temperatures, as most of the exchanges take place on the outer surface where the reaction rate is several orders of magnitude higher than at the material core.[1] To ensure fast combustion reaction kinetics, the graphite is therefore heated to high temperatures; investigations have shown that the effect of the porosity is then negligible.

Its main optical property is to absorb incident radiation over most of the electromagnetic spectrum. At temperatures below 2000°C, it exhibits the following characteristics:

- monochromatic emissivity (10.6 µm): approx. 1
- mean spectral emissivity: approx. 0.9
- hemispherical spectral emissivity: constant to 70° from normal incidence

Its thermal conductivity diminishes constantly as the temperature rises above 300 K. The heat capacity increases with the temperature and stabilizes at 2000 $J \cdot kg^{-1}K^{-1}$ above 1500 K; the characteristic value is 711 $J \cdot kg^{-1}K^{-1}$ at 300 K.

3. LABORATORY EXPERIMENTS

As shown in Figure 2, the laser beam was directed by a set of mirrors and a converging lens to obtain a given interaction surface area depending on the distance from the sample to the focal point of the optical system. The power density or flux (in $W \cdot cm^{-2}$) determined the heating rate. A 5 kW industrial laser with adjustable power and an initial beam diameter of 35 mm was used.

The cylindrical 750 g sample was placed on a *Sartorius* piezoresistive balance with 0.01 g precision, connected to a microcomputer in order to monitor the mass loss in time. The temperature of the laser-heated surface was monitored by a *Maurer* radiation absorption pyrometer calibrated for the 0.8–1.1 µm range with a response time of a few milliseconds, and corroborated by a disappearing-filament pyrometer that did not require emissivity correction.

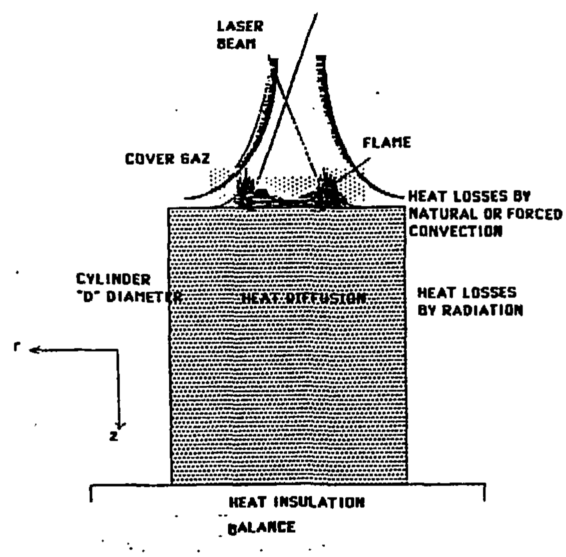

Figure 2. Experimental configuration

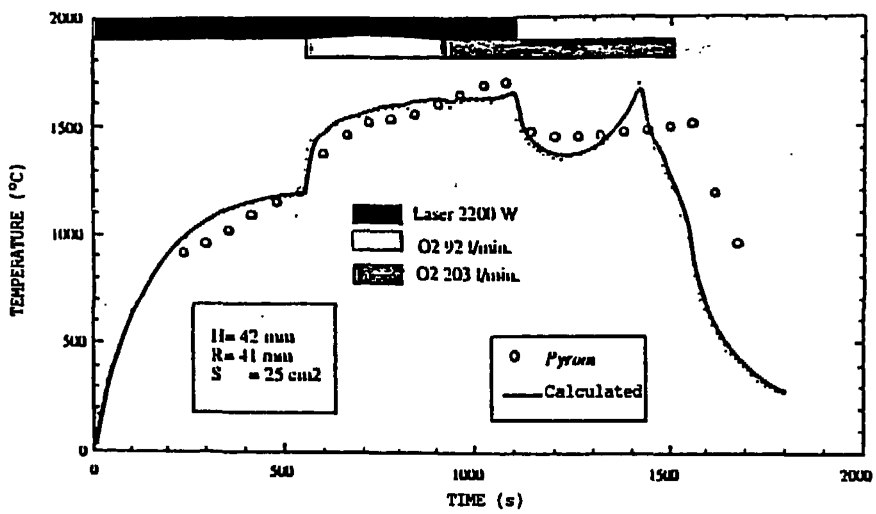

Figure 3. Temperature plot for test 3B

All the experiments[2] followed the same procedure. The sample was first heated by the laser beam to a temperature of 1100–1200°C in the illuminated zone. The oxidant gas was then supplied, causing spontaneous ignition of a bluish flame indicating combustion (i.e. oxidation of CO to form CO_2). Typical test measurements are shown in Figure 3.

The most favorable result obtained was a combustion rate of about 1 $kg \cdot h^{-1}$ with an oxygen flow rate of 12 $m^3 \cdot h^{-1}$, a laser flux of 100 $W \cdot cm^{-2}$ and an illuminated surface area of 25 cm^2 (i.e. 0.12 $kg \cdot s^{-1} m^{-2}$). Self-sustained combustion, in which the chemical reaction continued even without laser energy, was obtained under these conditions. The heat released by combustion was sufficient to maintain the temperature above 1200°C to offset the thermal losses.

4. NUMERICAL MODEL

The model solves the energy equation for the boundary conditions, allowing for variations in the physical properties of the graphite:

$$\rho C_p \frac{dT}{dt} = \mathrm{div}\left(-k\ \overline{\mathrm{grad}\ T}\right)$$

Boundary conditions included the following (refer to Figure 2):

- upper (heated) face: laser energy input, exothermic combustion reaction, forced convection losses in the cover gas and high-temperature radiation losses;
- side faces: low-temperature radiation losses and natural convection losses;
- lower face: zero flux condition (sample placed on insulating material).

The energy equation in cylindrical geometry is:

$$\rho C_p(T)\frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(K(T)r\frac{\partial T}{\partial r}\right) + \frac{\partial}{\partial z}\left(K(T)r\frac{\partial T}{\partial z}\right)$$

The equation is therefore solved in a variable bounded region.

The computer algorithm uses a finite differences method with implicit alternating directions to solve the equation in a plane comprising a regular 41 × 41-point mesh. The plane is an axial cross section of the cylinder, delimited by the cylinder centerline, the side face and the upper and lower faces (Figure 4). The code allows for the heat transfer mechanisms involved, and provides the temperatures at every mesh point together with the oxidation rate at the geometric boundary. Figure 3 illustrates the correlation between calculated and experimental values for one test (3B).

The variations in the power retention with the combustion power during the test (Figure 5) correspond to the different phases of the process:[3]

- The graphite brick was initially heated: the power retention was near the laser power, but diminished rapidly due to losses caused by the temperature rise.
- When oxygen was supplied, the combustion power sharply increased to nearly 5 kW and the instantaneous power retention increased accordingly by about 60% of the combustion power (C → CO reaction) + 40% of the CO → CO_2 reaction.
- When the laser was switched off, the instantaneous power retention suddenly became largely negative, reversing the heat flux and cooling the graphite block. The combustion power remained stable, however, since the surface temperature remained very high and oxygen was still supplied.
- After about 550 seconds, the instantaneous power retention became positive again due to gains from the incineration reaction and lower radiation and convection losses.
- The instantaneous power retention stabilized at a near-zero value characteristic of self-sustained combustion.

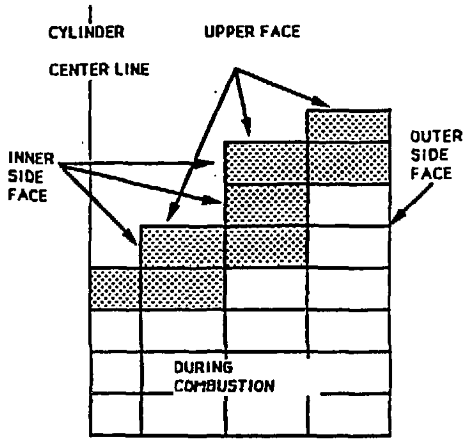

Figure 4. Sample geometry

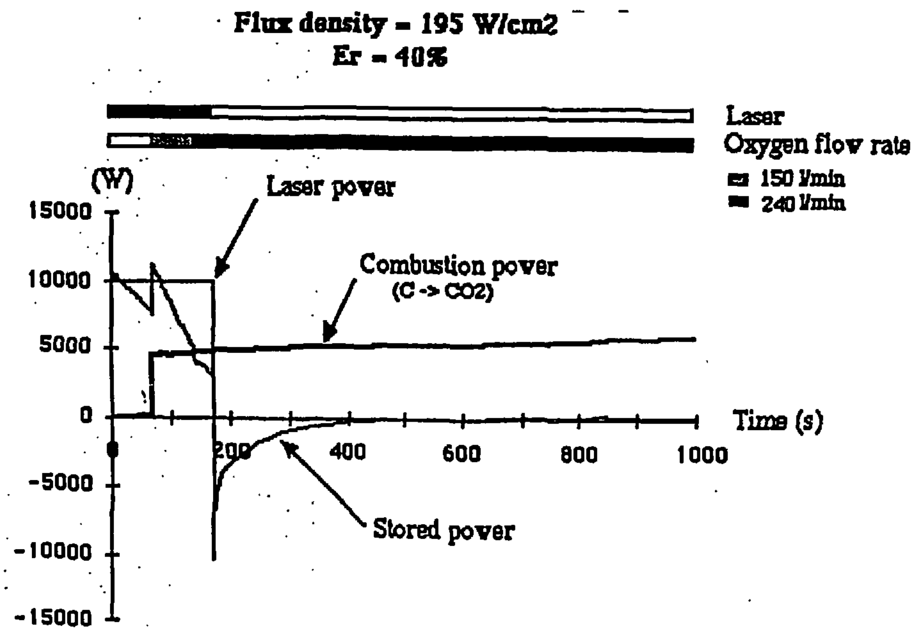

Figure 5. Self-sustained combustion on graphite (flux density 195 W·cm^{-2})

5. PILOT-SCALE FURNACE TESTING

In the light of the previous experiments, a furnace was specially designed and built for pilot-scale testing (Figure 6). It consists of a semicylindrical stainless steel shell polished on the inner surface to reflect the radiation from the hot graphite, and covered with mineral wool. An opening is provided on the top for the laser beam. The hearth is protected by a layer of refractory bricks. Oxygen is supplied by four cylindrical injectors 9 mm in diameter set obliquely along one side of the shell and directed toward the longitudinal centerline of the furnace.

Combustion products are exhausted through a 30 cm diameter duct to a stack outside the furnace room by means of a mobile electrically-driven centrifugal extraction blower at a rate of 2500 to 5000 $Nm^3{\cdot}h^{-1}$. A small-diameter flexible tube connects a sampling tap to a gas analyzer. The blower operating temperature is limited to 80°C by the manufacturer; a Venturi water spray system was therefore added in the flow stream before the blower in order to lower the exhaust gas temperature.

The surface temperature in the laser beam impact zone is the primary temperature reference. It is monitored by a *Maurer* infrared pyrometer operating in the 800–1100 nm band at temperatures between 500 and 2000°C.

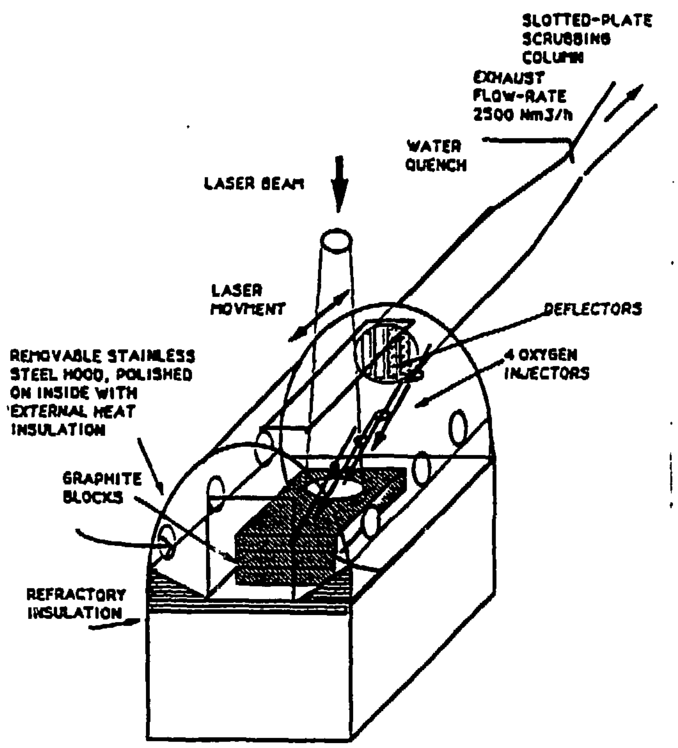

Figure 6. Pilot laser furnace schematic

After heating for 5 minutes with a 7 kW laser, combustion is initiated with oxygen injection and propagated by moving the laser beam over the stack of graphite blocks. Combustion rates of 14 $kg{\cdot}h^{-1}$ are easily obtained on a stack of fresh graphite bricks 20 cm on a side weighing 1900 g. A few fine particles ejected from the combustion zone burn without difficulty in the laser beam.

6. PROPOSED APPLICATION TO INCINERATION OF GRAPHITE WASTE CONTAMINATED BY ALPHA-EMITTERS

The objective is to incinerate contaminated graphite molds to recover the radionuclides and diminish the graphite volume. The method adopted is laser-heated calcining of the graphite.

6.1 Combustion Chamber

The furnace (Figure 7) includes a vee-shaped crucible at the bottom to accommodate the graphite waste, and an arch with a top opening for the laser beam and an air stream to provide dynamic containment. The combustion chamber is fully lined with refractory materials providing sufficient thermal protection to maintain a temperature of approximately 30°C in the glove box containing the furnace. The chamber also includes an oxygen feed line.

6.2 Laser Optical System

The 7 kW CO_2 laser will be installed in a room adjacent to the cell containing the furnace glove box. The laser optical system includes a beam transport system comprising several water-cooled mirrors directing the beam to the furnace glove box, two

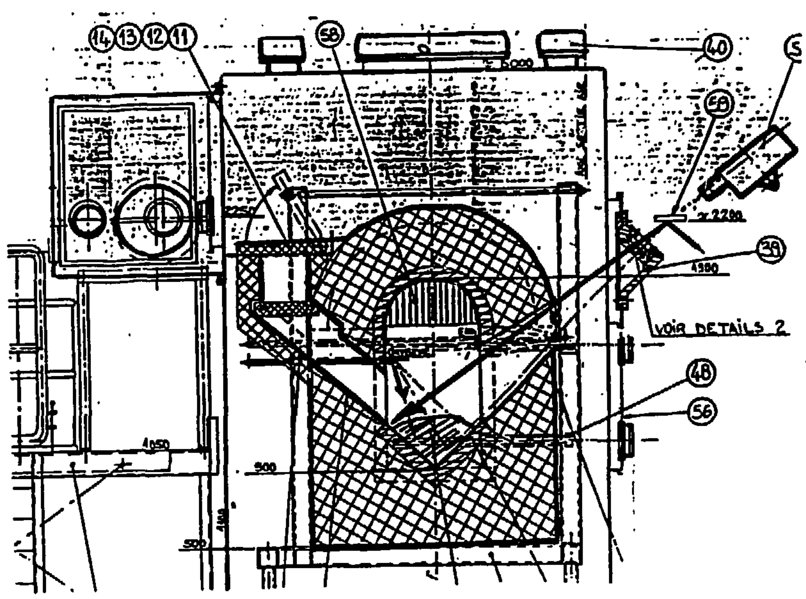

Figure 7. Laser incineration facility for graphite

ZnSe windows in the glove box allowing the beam to traverse the wall without alteration, and a motor-driven water-cooled mirror outside the furnace glove box capable of deflecting the beam to any point on the graphite inside the glove box.

6.3 Glove Boxes

Two glove boxes will be used in this facility: one containing the calcining furnace, and the second containing recovery vessels for ashes from the reusable off-gas system filter. Both glove boxes will provide Class 2 containment, with fabricated structures equipped with viewports and suitable transfer provisions (bag ports, DPTE flanges, etc.). The glove boxes will be ventilated via the building glove box exhaust system.

6.4 Operating Principles

Graphite will be place in the incinerator and continuously illuminated with a CO_2 laser until it reaches the self-ignition temperature of 1800°C. Combustion will occur in an oxygen-rich atmosphere, with oxygen supplied locally to the graphite surface by 4 nozzles. At the self-ignition temperature, combustion will be self-sustaining for some time. The graphite surface illuminated by the laser will be destroyed, with propagation beyond the illuminated zone.

The incinerator is designed to burn alpha-contaminated graphite at a rate of 10 $kg \cdot h^{-1}$, 6 hours a day, 5 days a week. Once a week, ashes from the furnace and the reusable process filter will be recovered and transferred to a counting system.

6.5 Gaseous, Liquid and Solid Wastes

The incineration off-gases will be removed, cooled to 150°C and filtered across a preliminary reusable HEPA filter, then two additional HEPA filters prior to release via the exhaust stack. The nominal release rate will be approximately 2640 $Nm^3 \cdot h^{-1}$.

The graphite incineration ashes will be recovered by suction from the furnace into a flask, which will be removed from the glove box inside a double vinyl wrapper.

6.6 Operating Procedure

6.6.1 Startup

The starting procedure involves loading the furnace and starting the laser system. The contaminated graphite will be supplied inside two sealed vinyl bags inside a PVC-lined drum. Each bag will contain about 10 kg of graphite. The drum and liner will be opened in the cell. One section of the furnace glove box is designed to hold 5 or 6 waste bags, introduced through a bag port. One or two bags will then be opened and dumped into the furnace feed chute, leading to the vee-shaped crucible in the combustion chamber.

6.6.2 Normal Operation

Once the process rate of 10 $kg \cdot h^{-1}$ is reached, the laser may be shut off if the reaction is self-sustained. The normal operating principle is thus to monitor and control the combustion, using a camera and software to determine the temperature profile. The operator decides when to switch on or off the laser beam depending on profile variations. When 9 or 10 kg of graphite have been incinerated, the furnace may be resupplied while combustion of the remaining graphite continues. At the end of the day the laser and oxygen supply are shut off and the furnace remains hot to facilitate startup operations the following day.

6.6.3 Shutdown and Cleaning

The incinerator and process filter must be cleaned at weekly intervals. Graphite combustion is terminated on Friday and the furnace allowed to cool down over the weekend before cleaning on Monday. The ashes are vacuumed directly into flasks placed inside the glove box via bag ports. The flasks are then enclosed in double vinyl wrappers and placed in a sealed container for counting and subsequent chemical processing to recover the radionuclides. The filter is cleaned, and the particles drop into a recovery pan. The filter glove box includes a vacuum cleaner identical with the one in the furnace glove box. Particle matter is thus vacuumed directly into specific flasks, which are then removed from the glove box via a bag port and transferred to the counting station in a sealed container.

7. CONCLUSION

Laboratory experimentation and subsequent pilot operation have result in an incineration project for contaminated graphite waste with a capacity of 10 $kg \cdot h^{-1}$. The process presents a number of advantages:

- Prior milling of the waste material is unnecessary.
- Variable processing capacity: 1 – 150 $kg \cdot h^{-1}$.
- Temperature control by sweeping laser beam.
- Steel materials acceptable.
- Oxidizing medium.
- Flexible modular operation.
- Simple, compact facility.
- High-quality incineration monitoring by infrared imaging.
- Quick extinguishing and reinitiating of combustion.
- No oxidant required.
- Heat source (laser) totally independent of furnace.

There is no doubt that other applications of the process will be developed.

8. REFERENCES

1. *Les Carbones*, Le Groupe Français d'Etude du Carbone. Masson: Paris (1965).
2. J.R. Costes, P. Caminat and P. Bournot. *Continuous CO_2 Laser-Controlled Combustion of Waste Graphite Blocks at Pilot Scale*, Incineration '91: Knoxville (May 1991).
3. J.R. Costes, N. Barbannaud and P. Caminat. *Advanced Powerful CO_2 Laser-Controlled Incineration of Waste Graphite (II)*, Incineration '92: Albuquerque (May 1992).