The ELIPSE Process: An Underwater Plasma Technology for Liquid Treatment - 16026

F.Lemont*, M.Marchand, M.Mabrouk, D.Milelli, JM.Baronnet
CEA, DEN, DTCD, SCDV, LPIC, F-30207 Bagnols sur Cèze, France
* florent.lemont@cea.fr

ABSTRACT

Radioactive liquid organic wastes are very various and produced in different quantities. Some of them are treated through specific processes when others are still waiting from outlet to be destroyed and stabilized. Their different composition (chlorinated, phosphate, sulfated, fluorinated, ...) make their treatment difficult in a same technology.

The ELIPSE process has been design to propose an innovative solution leading to a global treatment of what is usually called “orphan liquids”. This brand new technology involved a non-transferred plasma torch working under a water column ensuring the cooling, the filtration and the scrubbing of the gases coming from the combustion of the liquids. Furthermore, the water that ensures the global cooling of the system leads to low or to avoid the corrosion in a compact process. Only the nozzle of the torch may be affected by the corrosion and may become a wearing part of the system of treatment.

After giving a detailed description of the ELIPSE process, the present paper will focus on the results obtained for the treatment of such different liquids as tributyl phosphate, perfluorinated oil and trichloroethylene. In addition to obtaining destruction efficiency upper than 99%, the corrosion seems to have been significantly limited. These features lead to the first conclusion that the ELIPSE process could be a solution for the future to the problem of the hazardous liquid treatment, nuclear or not.

INTRODUCTION

To incinerate liquid waste with an organic fraction of nearly 100%, the oxidation potential of a plasma torch operating with oxygen was combined with the concept of a submerged plasma jet. The advantage of submerged thermal plasma treatment is the very high temperature obtained in an overall cold reactor, which limits corrosion problems; the high concentration of oxidizing radicals in the oxygen plasma and the intense UV radiation allow almost instantaneous combustion of the organic matter. Recombination reactions are minimized by rapid quenching. Turbulence induced in the water by the plasma jet ensures satisfactory gas-to-liquid transfer and trapping in solution of the waste mineral fraction liable to contain radionuclides. In this operating mode, the solution not only maintains the process at the ambient temperature but also ensures most of the functions of an off-gas treatment system: cooling, filtration and neutralization.

CONCEPT AND DESIGN

The concept of the ELIPSE process consists of implementing a plasma torch at the bottom of a reactor full of water. When organics are fed in the hearth of oxygen plasma, they burn in the nozzle of the torch and form off gases being quenched in the water.
In this way, water is used as off gas treatment: It cools, filters and scrubs the gases. Gas treatment is then replaced by a water treatment system involving a heat exchanger and a filtering system. The pollutants concentrating into the water are likely to precipitate depending on their concentration. They are then recovered thanks to the filter and can be driven toward a cementation or vitrification facility.
In this operating mode the solution not only ensures the gas treatment but maintains the process at the ambient temperature. This is an extraordinarily important and pioneering provision to prevent corrosion.
The concept of the process is shown in figure 1.

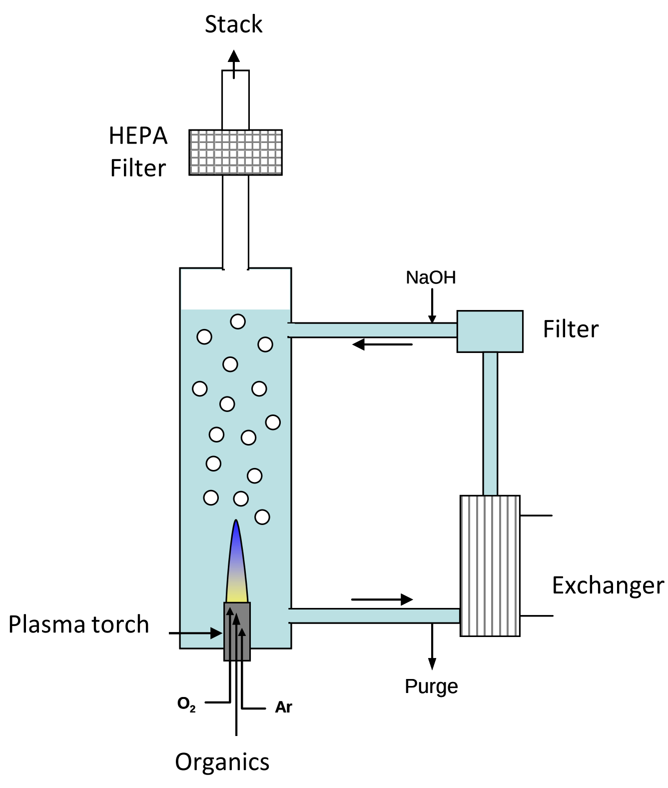

Figure 1: The ELIPSE process.

The design was validated by tests as shown in Figure 2 where one can see plasma working under a water column. A mockup has been built at the Marcoule center in order to assess the treatment of different kind of organics.

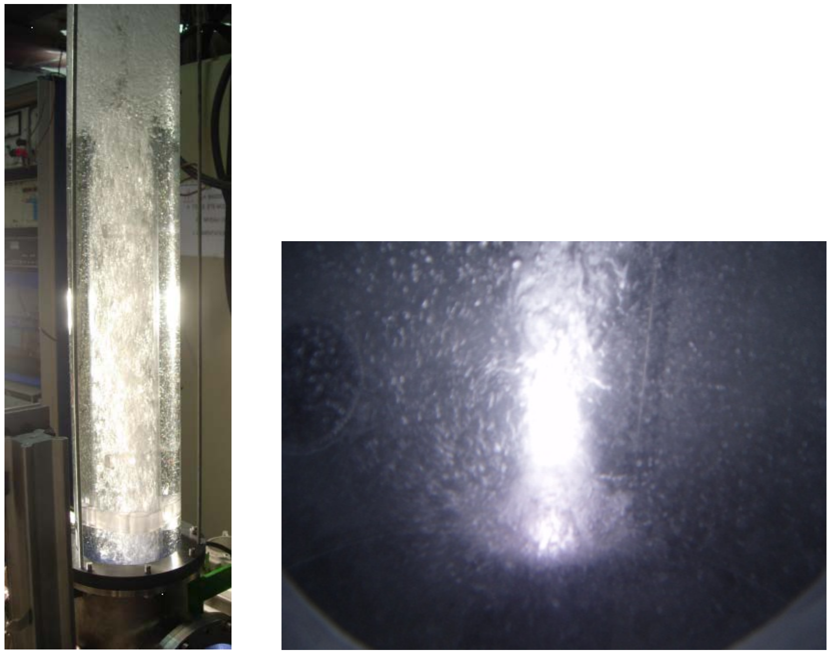

Figure 2: Working underwater plasma

The Reactor

The process is based on a non-transferred arc torch of an original design producing a plasma jet of oxygen. The plasma torch is disposed vertically to the base of a water-jacket reactor filled with water as described in figure 1. There is no specific recommendation for the water used to fill the reactor. Its pH must be adjusted around 7 as the treatment progresses.
Initially, the torch was ignited and the jet penetrated into the empty reactor for few second before it was filled with around 80L of water. Improvements have allowed to ignite the torch after it has been submerged what avoid an overheating of the empty reactor. The steam condenses and the non-condensable gas phase passing through the solution enters a condenser/demister to trap any residual water vapour and stop the water droplets carried over by priming.
Thermocouples indicate the temperature of the water in the reactor and at the condenser outlet. The solution temperature is controlled by a closed-circuit recirculation loop comprising a pump and a plate-type heat exchanger cooled by flowing water. The pH of the water is controlled thank to a probe and adjusted through soda addition. The mineral charges of the liquid are trap in a filter implemented in the middle of the loop.
All the reactor components (cathode, anode, reactor walls, condenser, ...) are cooled by separate and independent water circuits equipped with flowmeters and thermocouples to measure the heat loss in each circuit.

The Torch

As described in figure 3, the torch, with gas-vortex stabilization of the arc column, is made of a tungsten cathode, a copper anode and an intermediate diaphragm. The cathode tungsten tip is protected by a stream of argon. Oxygen, the main gas, is injected downstream the diaphragm. The change in diameter inside the nozzle anode creates a zone of recirculation of the flow, colder thus more electrically resistive, fixing the anodic spot behind the ledge [1]. At around 200A, the voltage is about 250V meaning the effective power dissipated is about 30kW.

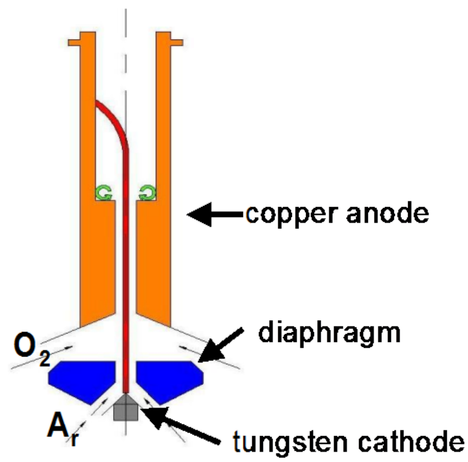

Figure 3: Schematic view of the torch.

Combustion Zone

The liquid is injected into a stage attached to the anode outlet as described in figure 4. If we assume that the dissociation and oxidation reactions are rapid enough to ensure that thermodynamic equilibrium is reached at the outlet of the injection stage, the amount of CO should be high at a temperature ranging between 3000 and 4000K. If the gases are directly driven toward the solution, the quenching rate estimated by NV.Alekseev [2] as around $2.10^7 K.s^{-1}$ is fast enough to ensure that the gas composition does not change from the equilibrium what has been confirmed through experiments.
Measurements during the treatment of 3L/h of TBP/dodecan have shown that quenched gases contained about 10 %(vol) of CO.
In order to decrease the CO content, a dilution stage using water injection was design and implemented downstream the liquid injection as described in figure 4. Measurements done during similar treatment that those mentioned above have shown CO content less than 0.2% (vol). In order to upgrade the process it has been decided to inject water coming

from the reactor. This option has the advantage of passing the solution several times through the plasma ensuring a better destruction of any residual organic compounds.
Finally a last stage of cooling has been added to increase residence time before quenching, to allow controlled cooling of the mixture. This stage is optional and could be removed if required.

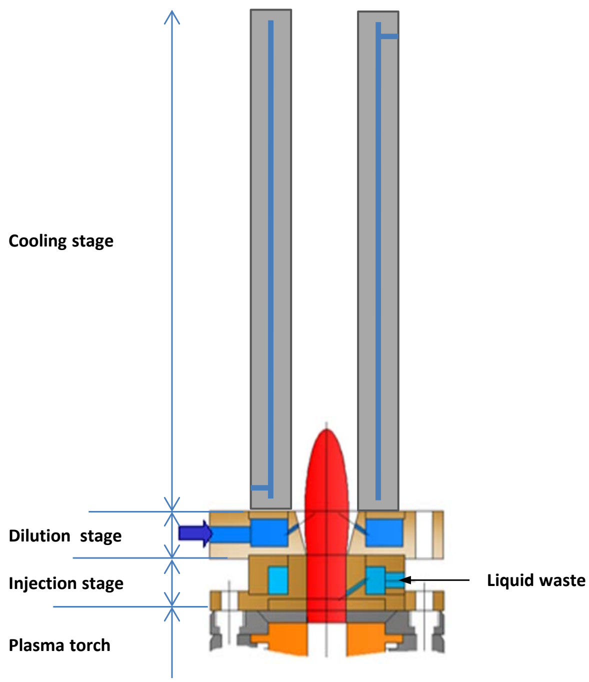

Figure 4: Schematic view of the combustion zone.

The Mockup

The whole mockup implemented at the Marcoule Research Center is shown in figure 5. It has been design for a throughput of around 3L.h^{-1}. Depending on the experimental results, this objective may be increased and optimized. The reactor equipped with several windows appears in the foreground of the picture. The background shows the closed-circuit recirculation loop including the filter, the heat exchanger, the different pumps, and the neutralizer.

Figure 5: ELIPSE mockup

Process Evaluation with Treatment of TBP / Dodecan

Experiments have been performed using TBP/ dodecane (30 – 70 vol %) solution with a feed rate of 3L.h^{-1}. The CO content did not exceed 0.17 %, while the CO_2 content of the off-gas stream remained relatively constant at about 23%: this value corresponds to the complete conversion of carbon present in the waste into CO_2 in gas form.
Hence the combustion efficiency is about 99.3 %. At the end of the test, the TOC in solution was measured: 4.67 g compared with 1797 g in the feed stream. The destruction efficiency was therefore 99.7 %.
In addition, the solution samples were analyzed by ion chromatography and using the Hach_Lange cuvette test system to determine the total phosphorus concentration.
These results show that the measured quantity phosphorus can be estimated at 81.7 g, whereas 83.4 g were introduced. The phosphorus capture yield is thus 98 %.
After testing, the reactor and components were removed for inspection. While stainless steel behavior was satisfactory in the solution rich in phosphoric acid, copper surfaces were strongly attacked—especially the external hard-soldered joint of the injection stage. The copper stages were replaced by Inconel 600 stages: this overcame the corrosion problem.

Three Additional Evaluations

One very big issue is the ability of the process to destroy very different liquid. How efficient is the treatment? What is about corrosion?
To answer these questions, three experiments have been carried out with the ELIPSE process into which the copper parts were changed for cooled Inconel parts. The feed rates have been lowered to 2L.h^{-1} in a first step:

- TBP/Dodecan: Treatment efficiency of 99.90% with a phosphorus capture of 98%. Inspection of the different parts of the process showed a very good behavior of the new Inconel nozzle without corrosion.

- Trichlorethylene: Treatment efficiency of 99.96% with a chlorine capture of 99%. Inspection of the different parts of the process showed a very good behavior of the new Inconel nozzle without corrosion.

- Perfluorinated oil: Treatment efficiency of 99.8% with a fluorine capture of 94.1%. Inspection of the different parts of the process showed a very good behavior of the new Inconel nozzle without corrosion.

CONCLUSION

The work presented an overview of the scientific and technological progression leading to the design of a submerged plasma process for the treatment of organic liquids. After demonstrating that cooling the gas before it penetrates into the reactor guarantees a composition with an acceptable CO/CO_2 output ratio, tests were conducted to evaluate the destruction of a TBP/dodecane mixture.
The tests substantiated the process efficiency: the TOC destruction efficiency always exceeds 99 % regardless of the type of waste, and the trapping yields for inorganic materials such as chlorine are near 100 %, within the measurement uncertainty margin. Only fluorine shows a lower capture that has to be investigated.
The very good behavior of the process regarding treatment efficiency and maybe above all the lack of corrosion opens considerable perspectives for the destruction of various liquids. We can then already conclude that the process now being developed has very strong potential. Versatility, compactness and robustness are the terms that best describe it.

REFERENCES

1. Zhukov MF(2007) Thermal plasma torches. Cambridge International Science Publishing, Cambridge.
2. Alekseev NV, Samokhin AV, Agafonov KN, Tsvetov YV. Oxidation of organic compounds in water under injection of thermal plasma jet. 14th International Symposium of Plasma Chemistry, Prague, Czech Republic, 1999 August 2-6