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FEASIBILITY OF A LIQUID PHASE CHEMONUCLEAR OZONE PROCESS

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MASTER

At the Chemonuclear Workshop held at Brookhaven National Laboratory in November 1962 it was pointed out that the production of ozone from oxygen by a liquid phase fission fragment chemonuclear process appeared to have reasonable economic potential. Before any judgment regarding the desirability of pursuing a liquid phase chemonuclear ozone development program could be made, a definitive study of a reactor and chemical separation system based on present-day cryogenic and ozone technology was needed. The major objective of the present study was to establish a reasonable plant design in order to be able to make such a judgment.

The first consideration was to estimate the primary production rate of ozone using fission fragments. The maximum theoretical reaction yield for the formation of ozone from oxygen is a "G" value of 68 molecules per 100 ev based on heat of reaction data. This ideal "G" value would be attainable if all the energy were utilized in producing the ozone. The most recent calculated gas phase "G" value reported by Magee is in the range of 12 to 19 based on detailed experimental and theoretical reaction kinetics data. Experimentally observed "G" values for Co-60 gamma irradiation of liquid oxygen have ranged from 6 to 15. Ozone concentrations of 8 w/o were experimentally attained and equilibrium concentrations of 13 w/o were estimated from the data. No experimental "G" value has been determined yet for irradiation of liquid oxygen with fission fragments. The authors felt a "G" value of 15 to be reasonable for this study.

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The reactor system chosen, though unusual in having liquid oxygen as its moderator, is formally not much different from the class of boiling water reactors, since the operating pressure and temperature were selected so as to allow boiling of the oxygen-ozone mixture. Liquid oxygen is fed into the reactor vessel and passed through the core. Some of the oxygen is partially converted into ozone. Unreacted oxygen removes fission heat by vaporization. The oxygen vapor is condensed external to the reactor vessel and recycled. A bleed stream from the reactor vessel containing the equilibrium oxygen-ozone mixture provides the feed solution to the chemical separation system. The plant process flow sheet indicating the basic unit operations is shown on Slide I - air separation plant for liquid oxygen supply; refrigeration system for condensing oxygen vapor; reactor system; chemical separation system; and fission product ozone solution storage and disposal system.

Based on a recent market study of ozone users the chemonuclear plant production rate was set at 130 tons per day. This required that the reactor be operated at a thermal power level of 25 Mw and at a fluid temperature of 109°K, which is the saturation point for a 10 w/o ozone solution at 5 atm.

The fuel elements are fixed solids arranged in a packed bed in a cylindrical vessel. The reactor fuel material is 20 w/o dispersion of U-235 in palladium ribbon 1 micron thick. Material of this composition has been fabricated with a thickness of 1.3 microns. Presently, uranium-palladium foils are being irradiated in the BNL Research Reactor and the Materials Test Reactor. (Slide 2) The fuel foils might be manufactured in a honeycomb array, possibly an inch in height and stacked in the reactor in some orderly array similar to packed towers in the chemical process industry. Sample fuel elements

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have been fabricated with micron size foils ($2-3\mu$) into honeycomb arrangements. For such thin foils, the experimentally-observed fission fragment energy deposition efficiency in the surrounding medium is about 70%. For the assumed "G" value of 15, this means that roughly 16% of the total reactor power is consumed in forming the chemical product, the remaining energy being removed by thermal heating of the ozone-oxygen solution.

The reactor dimensions were selected from a core parameter study using a modified age-diffusion theory. Some of the core parameter study results are shown on slides.

Effect of void fraction not as significant as would be expected in standard BWR. Design void fraction 3%. The minimum in the curve is quite broad, but requires cores too large to be practical. The reference core has a diameter of 9.1 feet, a height of 13.1 feet, an initial U-235 mass of 20 kg, and is reflected radially by 2.3 feet of graphite. The initial neutron multiplication constant (K_{eff}) is 1.09 which is sufficient for a core reactivity lifetime of 100 days. The average U-235 burn-up is 13 a/o (or 1.3% of total U + Pd atoms). Under operating conditions, the void coefficient of reactivity is -0.95 fractional change in K_{eff} per void fraction change. The thermal neutron lifetime of about 3 milliseconds is attractive for control purposes.

The liquid oxygen supply facility is of a conventional design and size. Air separation plants in the 100 ton per day range are presently operating with a reasonably good safety record.

The chemical process system selected consists of phase separation units. Initial non-volatile fission product removal is by means of impingement-type filters (i.e., stainless steel wire mesh) assuming that the major non-volatiles are in the form of insoluble oxides. The second stage of fission product removal and ozone product concentration is accomplished by distillation. In

this regard, I should say a few words on the potential hazard of a distillation separation. A prime safety consideration in air separation plants is hydrocarbon contaminants in the atmosphere which could accumulate to detonation levels in the nitrogen-oxygen distillation column. Use of silica gel beds and operational procedures such as periodic bleeding of the column re-boiler section have apparently solved this problem. Experimental detonation observations of high liquid ozone-oxygen solutions indicate the acceptable concentration level of hydrocarbons to be orders of magnitude lower than with liquid oxygen alone. If hydrocarbon contamination was the sole cause for the past history of ozone explosions, elimination of these impurities should permit ozone distillation. Admittedly, more experimental data is needed before final acceptance of distillation as an ozone-oxygen separation mechanism. For this study a double-column design was utilized, which is common in air separation plants.

Assuming that the liquid ozone would be used exclusively for fuel propulsion in the space field or equivalent application in which human ingestion is not a consideration, the radioactivity level in the product ozone need only be low enough to permit handling without any external radiation hazard. A safe radiation limit for liquids is about 10^{-3} μ c per cc for gross fission products, which for this design concept means a required total product decontamination factor of about 10^9 . The mechanical filters should provide a DF of 10^2 to 10^3 . Hence, the distillation column would have to accomplish a fission product separation factor of 10^6 to 10^7 which is not unreasonable.

With regard to the refrigeration system to condense the oxygen vapor from the reactor, liquid nitrogen appeared to be a suitable coolant.

Presently, the most efficient refrigeration system design is based on Keesom's nitrogen cascade. The energy requirements (work of compression) for the Keesom cascade utilized as a closed refrigeration cycle is about 0.44 kwhr per kg of liquid nitrogen, as compared to the ideal thermodynamic reversible energy requirement of 0.21 kwhr per kg. Using a Keesom refrigeration cycle for the proposed system would result in a compressor power requirement of 200 electrical Mw. This is about four times larger than the power required for producing ozone by the conventional electric discharge process. It should be noted that with the present-day conventional process a minimum of 100 ozonators would be required for a 130 ton per day ozone production plant.

The total plant investment cost is estimated to be in the range of \$100,000,000 with approximately 90% of the cost attributed to the nitrogen refrigeration system. This results in a product cost of approximately 50 cents per pound, as compared to the conventional method product cost of about 15 to 20 cents per pound. However, the chemonuclear product is pure liquid ozone whereas the conventional process product is an oxygen-ozone gas mixture of 3 - 5 w/o ozone so that costs are not directly comparable.

It seems clear that the large capital investment associated with the refrigeration compressors in the design presented here will have to be reduced if low ozone costs are to be obtained. Other system designs demanding less refrigeration are, for example, a gas phase chemonuclear reactor. This will be considered in future studies.

November 19, 1963

CHEMONUCLEAR OZONE PROCESS
FLOW SHEET

Folded metal assembly of 0.25 mil foil.