

LA-UR-79-2558

MASTER

CONF-79204--1

TITLE: REALISTIC ASSESSMENT OF DIRECT RADIOLYSIS FOR SYNTHETIC FUELS PRODUCTION USING FUSION RADIATION SOURCES

AUTHOR(S): J. H. Pendergrass, L. A. Booth, F. T. Finch,
and T. G. Frank

SUBMITTED TO: 2nd Miami International Conference on Alternative Energy Sources, 10-13 December 1979, Miami Beach Florida

University of California

By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos Scientific Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.



LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

An Affirmative Action/Equal Opportunity Employer

REALISTIC ASSESSMENT OF DIRECT RADIOLYSIS
FOR SYNTHETIC FUELS PRODUCTION USING
FUSION RADIATION SOURCES

J. H. Pendergrass, L. A. Booth, F. T. Finch,
and T. G. Frank
University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87545 USA

The use of fission reactor radiations, including neutrons and gamma photons, but with emphasis on fission fragments which carry off ~ 80% of fission energy release, has been extensively investigated for direct radiolytic production of large volume chemicals. Potential products include synthetic fuels by water or carbon dioxide decomposition, fixed nitrogen from air, and ozone from oxygen. Interest in fission reactor radiolysis has largely vanished because of apparently insurmountable obstacles to simultaneous economical achievement of high efficiency in conversion of radiant energy to stored chemical energy of radiolysis products and acceptable levels of product radioactivity contamination. More recently, the use of fusion reactor radiations for direct radiolysis has been of interest. Fusion reactor radiations are apparently more suitable for radiolytic chemical production, but many obstacles to commercialization using fission radiation sources must also be faced with fusion sources, albeit to a lesser degree.

Because the conditions required for useful thermonuclear energy release are less extreme for deuterium-tritium (DT) fusion than for all other fusion reactions, at least the first few generations of fusion reactors are expected to burn deuterium and tritium. Up to ~ 80% of primary DT fusion energy release is represented by 14-MeV neutron kinetic energy, with the remainder carried off by x-ray photons and energetic ions. Further fusion reactor energy release results from exoergic reactions of fusion neutrons with reactor blanket, coolant, and structural materials. The highly penetrating nature of fusion neutrons permits utilization of a majority of fusion reactor energy release in blankets surrounding reactor cavities independent of cavity phenomenology. However, these highly penetrating neutrons also present problems. Many radiolysis reactions are conducted most efficiently in the gas phase, but gases have such small stopping powers for high-energy neutrons that either very thick blankets or high pressures, which generally reduce radiolysis efficiencies and impose undesirable reactor structural requirements, will be required to provide adequate neutron energy absorption. Tritium breeding in lithium-containing blankets surrounding fusion reactor cavities sufficient, or nearly sufficient, to close the tritium fuel cycle may be necessary for economic viability. Simultaneous tritium breeding, prevention of product contamination with tritium and other radioactive species

produced by neutron interactions with the radiolysis mixture and reactor structure, and use of a high percentage of total fusion reactor energy release for radiolysis appears to represent an extraordinarily difficult engineering problem with no apparent solutions in sight.

Radiolysis efficiency for a particular radiation and a particular radiolysis product can be expressed in terms of a G value, defined as the number of molecules of that product produced per 100 eV of radiant energy deposited in the reaction mixture. The ratio of experimental G values to maximum G values for endothermic reactions, corresponding to complete utilization of radiant energy, gives the efficiency of radiant energy utilization. Among the highest G values reported for H₂O decomposition to give H₂ and O₂, CO₂ decomposition to give CO and O₂, and HCl decomposition to give H₂ and Cl₂ are ~15, ~10, and ~8 respectively. The corresponding maximum G values are 40.3, 34.5, and 101.2, which result in respective radiolysis efficiencies of ~37%, ~29%, and ~8%. This suggests that only topping cycle applications or combined radiolytic-thermochemical cycle applications, for which HCl radiolysis has been proposed, can be economically viable. If this is the case, then thermal energy rejected from such cycles must be available at temperatures high enough to permit efficient further utilization.

Significant influences on the efficiencies of radiolytic processes include (1) type of radiation, (2) radiation intensity or dose rate, (3) temperature and pressure, (4) radiolysis product concentrations, and (5) whether or not back reactions are inhibited by phenomena that separate excited reaction products until they are deactivated, by continuous removal of reaction products from the reaction mixture, or by prompt deactivation of reaction products with so-called scavengers. Some of these promote radiolytic production of desired substances, but others hinder their production. The listed G values were obtained under conditions that favor the desired radiolytic reactions, but which may also involve, (because radiolytic processes are still only imperfectly understood) hidden factors that hinder radiolysis and which if eliminated or counteracted might lead to higher G values.

The first influence is generally characterized in terms of the linear energy transfer or LET, the mean energy deposition density along a radiation track. Heavy charged particles have large LET values, e.g., ~3 to 25 eV/A for alpha particles of various energies and fission fragments (mean), whereas gamma and x-ray photons, beta particles, accelerated deuterons and neutrons exhibit small LET values, e.g., ~0.02 to 1.0 eV/A. For many radiolysis reactions, radiolytic efficiency increases with increase in LET value. Fusion neutrons interact with radiolysis mixtures and transfer energy in a variety of ways to readily convert neutron energy to a mixture primarily of high LET charged particle kinetic energy and low LET gamma photons. Considerable experimental evidence suggests that very-high-dose-rate, pulsed irradiation, characteristic of inertial confinement fusion can give substantially higher G

values than does steady irradiation, characteristic of magnetic confinement fusion, for the same time-averaged radiation fluxes. Although there are some apparent exceptions, G values generally decrease with increase in gaseous radiolysis mixture pressure, probably because of increased back reaction rates, but higher densities result in thinner blankets for gaseous radiolysis mixtures. Higher temperatures can result in higher G values for at least some radiolysis reactions. For example, G values for water vapor radiolysis jump sharply at about 250°C. However, theory suggests that it is a one-time effect only and that further increases in temperature will not lead to further sharp jumps. The highest G values are obtained when radiolysis product concentrations are low, corresponding to low back reaction rates. Back reactions can also be stimulated by radiation, so that steady state concentrations are often largely determined by the type and intensity of the radiation, rather than by ordinary thermodynamic considerations. On the other hand, low product concentrations can mean increased recycle of reactants and expensive product separations. Scavengers, such as HCl and HBr, that are simple, readily-reconstituted (required because of radiolysis of scavengers themselves), inorganic compounds substantially increase G values for hydrogen production by water radiolysis. Scavengers were used to obtain the listed maximum G values, but more effective scavengers may exist. Disproportionation of H₂ and O₂ between phases in liquid water-steam mixtures reportedly enhances G values for H₂ production by separation of radiolysis products, but the enhancement may be solely a result of the temperature effect previously discussed. On the other hand, a similar effect may be useful with other radiolysis systems. There is experimental evidence that inhibition of diffusion of dissociation products that occurs in critical regions due to vanishing of chemical potential driving forces for diffusion at critical points can significantly reduce rates of recombination of radiolysis products. Unfortunately, critical temperatures for many radiolysis systems of interest are rather low and efficient utilization of reject heat is thus precluded. Nonetheless, attractive radiolysis systems with higher critical points may be discovered. A principal reason for low G values for many radiolysis processes is that much of the radiant energy is transformed into molecular excitation energy short of that necessary to cause dissociation. Lasers could be used to selectively and efficiently introduce into the excited species the small additional amounts of energy required for dissociation.

Our, and other studies, of the economics of synthetic-fuel production by direct radiolysis, all of which ignore some of the technology problems, suggest that direct radiolysis cannot compete with other methods of synthetic-fuel production, including coal gasification and liquifaction, production of synthetic crude from tar sands and oil shales, and fusion-driven thermochemical and electrolytic hydrogen production. Principal barriers to commercialization of radiolytic processes include low G values and radioactivity contamination of products.