The Utilization of Fission Fragment Energy for the Fixation of Nitrogen

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The principles involved in designing a process for the production of fixed nitrogen by the direct use of fission fragment recoil energy are reviewed. The problems concerned with the radiation chemistry, development of fuel element, reactor design, and chemical process design are pointed out. Possible solutions to these problems incorporated in a complete plant design are presented. An economic evaluation, comparing the chemonuclear process with other conventional processes, is made. The conclusion is reached that at the present state of knowledge there does not seem to be any clear-cut advantage over conventional processes, even based on a nuclear economy. However moderate research efforts should continue for further evaluation of this process.

The availability of large sources of nuclear radiation resulting from the development of fission reactors in recent years has made it possible to consider the direct use of fission recoil energy for the economic production of industrial chemicals by so-called "chemonuclear" processes (*1, 2*). Radiation chemistry experiments (*3, 4, 5, 6, 7, 8*) have indicated the possibility of producing fixed nitrogen in the form of nitrogen dioxide from mixtures of nitrogen and oxygen. Although the technology is still in an early stage, the importance of fixed nitrogen has prompted a study of the problems involved in the development of this new technique and a comparison with other methods of production.

For any chemical product the total fission energy consumed in producing a unit quantity of chemical is

$$W = \frac{1{,}216}{G e_o M}$$

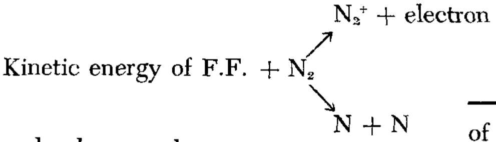
For producing nitrogen dioxide the power level of the fission fragment reactor is

$$P = \frac{2.21\,T}{G e_o}$$

For a given rate of production the reactor power level is fixed by a knowledge of G and e_o or the product of the two. Estimates of G value can be obtained from radiation chemistry experiments, while e_o depends on the type of fuel used.

RADIATION CHEMISTRY

Static and closed stirred-vessel experiments with pile radiation (neutron and gamma) and fissioning U-235 (*4, 8*) have indicated maximum G values ranging from 5 to 7.4 molecules nitrogen dioxide formed per 100 electron volts deposited in air at temperatures up to 400°F., pressures up to 1,000 lb./sq. in., and radiation intensities up to 300x10^6 rad./hr. (0.38 kw./lb. air). Nitrogen dioxide concentrations for these values as high as 11.2% are reported. The G value tends to increase with pressure and temperature. Briefly the mechanism for the homogeneous formation of nitrogen oxide and subsequently nitrogen dioxide from nitrogen and oxygen is believed to be as follows (*4*):



$$\text{Kinetic energy of F.F.} + N_2 \rightarrow \begin{cases} N_2^+ + \text{electron} \\ N + N \end{cases}$$

and subsequently

$$N + O_2 = NO + O$$

$$NO + 1/2\,O_2 = NO_2$$

On the assumption that only unionized atoms produce nitrogen a maximum G value of 9 can be expected. Because of the interaction of N with the products nitrogen oxide and nitrogen dioxide the kinetics of the system becomes extremely complex, and prediction of G values under various temperature, pressure, and intensity conditions, particularly in a flowing system comparable to that obtained in a power reactor, becomes very difficult. For establishing a firm process design, radiation chemistry experiments are needed at pressures up to 1,000 lb./sq. in., temperatures up to 1,100°F., concentrations of reactant gas varying from air composition down to less than 10% nitrogen in oxygen, and radiation intensities in the range of 200 to 2,500x10^6 rad./hr. (254 to 3,200 kw./lb.) for various residence times in the radiation field. The possible beneficial effects of energy transfer agents and catalysts also require investigation.

FUEL PROBLEM

Since the average range of the fission fragment in uranium containing solid fuel is in the order of 10 to 14 μ, the fuel must be so finely divided that at least one dimension is smaller than this range to permit a significant fraction of the fission fragments to escape the fuel. Fuel may take the form of thin plates, spheres, or rods in ordered or disordered arrays.

The necessity of maintaining a minimum fuel density for criticality purposes may cause loss of available energy owing to re-entry of the fission fragment into fuel particles spaced too closely to one another. The fraction of the total energy of fission deposited in the gas stream is a function of the product of three separate efficiencies:

$$e_o = e_{KE}\, e_{SG}\, e_{FG}$$

Expressions have been developed for evaluating various geometrical forms of fuel (*9, 2*). For ordered arrays of spheres, cylinders, or plates, where the distance between particles is larger than the fission fragment range in the process gas, re-entry cannot occur (e_{FG} = 1.0 or 100%), and the broken lines *F* and *G* in Figure 1 can be used for estimating this efficiency. The average fission fragment range in air under standard conditions is approximately 2.4 cm., while at 1,000 lb./sq. in. and 800°F. it is approximately 1 mm. The solid lines in Figure 1 show the efficiencies for spherical U_3O_8 dust particles in air at several values of dust loading, which is the weight ratio of solid fuel to gaseous reactant stream W_s/W_g.

Considerations of maximum efficiency and reactor design indicate the most useful range of fuel dimensions to be 1 to 10 μ and of fuel loadings 0.1 to 1.0 lb. fuel/lb. gas. Figure 1 shows that in this region deposition efficiencies, ranging from 45 to 81%, are possible with spherical U_3O_8 particles in a disordered array, a condition which can be obtained in circulating dust systems. Both ordered and disordered fixed arrays of cylinders or slabs 10 μ or less in diameter or thickness, can also yield efficiencies in this range. For equivalent fuel loadings diluents such as ferric oxide, silicon dioxide and fission fragments decrease the efficiency, but not significantly, in the range considered. Disordered arrays of cylinders in the form of uranium containing glass wool have been proposed as fuel (*4*). Under a pressure differential caused by resistance to fluid flow collapse of this type of fuel is possible, resulting in the lowering of

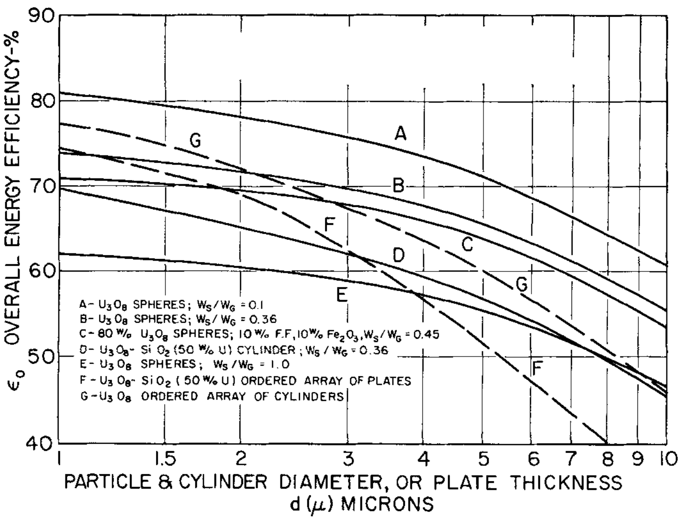


Fig. 1. Over-all fission energy deposition efficiency in air.

the deposition efficiency. Plated fuel elements have been made (3), but they lose half the fission fragment energy in the supporting substrate. Micron-thick ribbons or plates of U containing alloys supported on open grid screens in an ordered fixed array might be a solution to the problem of utilizing both sides of the plate. Fixed porous beds of uranium containing materials have been proposed (6). The high mass ratio of solid to gas would produce a low deposition efficiency, but it might be enhanced by surface energy transfer mechanisms. An ideal chemonuclear fuel would be a gaseous compound of uranium, but the halides, which are the only compounds of uranium with sufficient volatility, would be unstable in an oxidizing atmosphere. Other possibilities include contacting the reactant gas with fissioning solutions or suspensions of uranium in inert liquid media. In these cases loss of energy to the dispersing media must be considered.

Because of variations in the energy transfer mechanism along the path of the fission fragment experimental results are necessary to confirm the analytical values of efficiency for a specific fuel configuration (5).

In circulating uranium oxide dust systems it is predicted that opposing forces of agglomeration and attrition will cause the formation of an equilibrium dust particle size in the range of 1 to 10 μ, with an average size of 5 μ. The stability of such a fine particle circulating fuel system is still to be demonstrated. Requirements of chemical, mechanical, and thermal stability also place severe specification restrictions on micron-sized fixed fuel elements.

REACTOR DESIGN

Reactor designs include internally and externally moderated circulating and fixed fuel systems. Parametric studies were made for such factors as fuel loading, void fraction (reactant gas space) in reactor, total uranium inventory, moderator composition and size, and reactor power level.

Because of the relatively low nuclear-to-chemical energy conversion efficiency [at $G(NO_2) = 6$, maximum thermal energy converted = 2.1%] it is necessary to consider chemonuclear reactors that produce by-product power in addition to the chemical. For economic power generation, reactor effluent temperatures in the order of 800° to 1,100°F. are indicated. To maximize energy deposition efficiency higher reactant gas pressures are needed. Reactor vessel dimensions and the strength of construction materials at the elevated temperatures dictate pressures not much higher than about 1,000 lb./sq. in. The significant neutron absorption cross section of nitrogen (1.8 barns) makes it desirable, because of both the critical fuel inventory and nuclear safety aspects, to minimize the nitrogen concentration in the reactor. Fermiage theory including reflector savings approximations was used for the criticality estimates, assuming optimistically a 30% nitrogen–70% oxygen reactant gas composition.

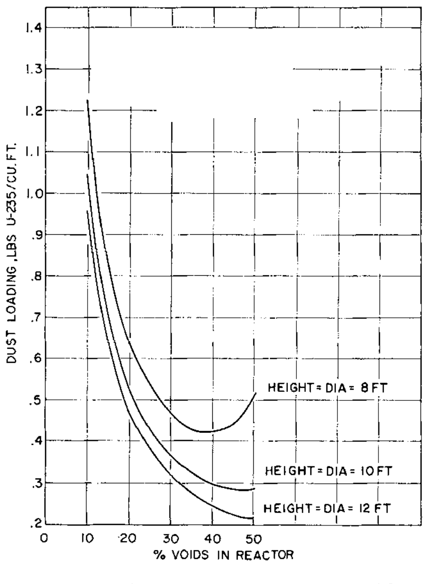


Fig. 2. Dust loading vs. percentage voids in reactor for 16-in. circular graphite moderator logs. U_3O_8 fuel, 50-cm. graphite reflector.

For an internally moderated circulating U_3O_8 dust reactor graphite is preferable as a moderator. The graphite logs are clad with 30 mils of stainless steel or silicon carbide to limit oxidation and erosion. Because of fast neutron leakage at higher void fractions increasing core voids increases critical inventory. This effect, combined with the increasing void space, causes the dust loading initially to decrease with increasing core voids and then to increase as shown in Figure 2. A practical core size is 8-ft. diam. x 8 ft. high with an additional 2 ft. of reflector thickness. A void fraction of 20% is a reasonable compromise between dust loading and critical inventory. At a dust loading of 0.8 lb. U_3O_8/cu. ft. (W_s/W_g = 0.36) the critical inventory is approximately 20 kg. U_3O_8 (93.5% U-235 enrichment). For particle sizes of 5μ the energy deposition efficiency could be 65.8%.

Designs at two levels of fixed nitrogen production and power are considered for technical and economic evaluation. One is a minimum sized reactor producing 200 tons/day of nitrogen dioxide (equivalent to 74 tons/day ammonia as fixed nitrogen), which, for the above conditions, requires a reactor power level of 112 mw. (thermal). The other is an optimum-sized power reactor of 700 mw. (thermal) capacity, capable of producing in the range of 1,200 to 1,300 tons/day nitrogen dioxide (460 tons/day ammonia). The conceptual design of the 112 mw. dust reactor is shown in Figure 3, and the design data for the system, including the power generation plant, are given in Table 1.

For the larger power reactor the required increase in the velocity of the circulating reactant gas (which is also the coolant) could cause excessive erosive wear of internal moderator logs. Therefore an externally moderated or cavity type of reactor having a minimum surface-to-volume ratio is desirable. Designs based on this concept and the work of Safonov (27) are found to be feasible (2).

Addition and withdrawal of fuel in the circulating dust systems can be made continuous, although control may be a problem. However in the fixed fuel reactors large amounts of excess fuel (two to three times the

Table 1. Dust Fueled, Internally Moderated, Chemonuclear Reactor Design Data

General	
Reactor thermal output, B.t.u./hr.	3.82×10^8
Reactor thermal output, mw.	112
Gross electrical output, mw.	40
Gross electrical efficiency, %	36
Net electrical output, mw.	34.3
Net over-all power efficiency, %	30.6
Chemical output, tons N_2O_4/day	200
Over-all deposition efficiency, %	65.8
Fuel	
Fuel composition	U_3O_8 dust
Average particle size, μ	5
Fuel enrichment, %	93.5
Dust loading, #/cu. ft.	0.8
Reactor core	
Nominal core height, ft.	8
Nominal core diameter, ft.	8
Moderator—16-in. diameter graphite logs clad in 30 mil s.s. or coated with SiC	
Moderator graphite weight, lb.	32,000
Reflector graphite weight, lb.	62,000
Critical mass, kg. U_3O_8	28.7
Volume fraction of nuclear materials in core	
U_3O_8	negligible
Graphite	0.80
Coolant	0.20
Reactivity coefficient at 1,000°F. $\Delta K/K/°C$.	-2.2×10^{-4}
Reactor vessel	
Design pressure, lb./sq. in. gauge	1,000
Design temperature, °F.	650
Diameter, O.D., ft.	14.5
Height, over-all, ft.	17
Wall thickness, in.	6
Material, vessel thermal barrier	SA-212
Gross weight of vessel, (including supports, graphite, thermal barrier, insulation, etc.) lb.	338,500
Primary cooling system	
Gas (nominal design)	30% N_2-70% O_2
Working pressure, lb./sq. in. gauge	1,000
Total flow, lb./sec. (through steam generators)	650
Reactor inlet temperature, °F.	550
Reactor outlet temperature, °F.	1,100
Gas velocity through core, ft./sec.	29
Gas velocity in piping, ft./sec.	100
Gas velocity through steam generator, ft./sec.	70
Circuit pressure drop, lb./sq. in.	34
Heat capacity of gas. B.t.u./# °F.	0.26
Steam generators	
Type	Once through
Number	2
Number of passes	2
Tube length per pass, ft.	19.5
Tube, I.D., in.	0.5
Number of tubes per exchanger	1,540
Tube area, sq. ft, per exchanger	7,900
Shell diameter, ft.	4
Gas inlet, temperature, °F.	1,100
Gas outlet, temperature, °F.	445
Feed water temperature, °F.	325
Steam temperature, °F.	1,000
Steam pressure, lb./sq. in.	1,000

critical mass) are needed to achieve an economic operating cycle. This excess reactivity necessitates many control rods; a more feasible design is to use a moderator which is moved up into the core as burn-up proceeds. The movable moderator reactor can be used with plated fuels or with filament type of fuels. An internally cooled double tube reactor design based on glass fiber fuel packing in the inner tube can also be designed (*1*).

All concepts investigated have feasible critical masses in the range of 20 to 40 kg. U-235. The main problem in the reactor design is safety. In the circulating dust systems fluid instabilities causing changes in the dust concentration through the system could result in dangerous oscillations of the reactor power level. The dust system may however have a safe self-limiting negative prompt coefficient of reactivity owing to simultaneous expulsion of fuel and process nitrogen in the event of a pressure loss or temperature rise. The fixed fuel system has a potentially dangerous positive prompt reactivity coefficient because of the possible loss of nitrogen from the reactor without loss of fuel. Concepts for dealing with the safety problem include the use of nuclear or mechanical safety devices, reduction of nitrogen concentration, and pressure loss delay systems, but these require additional analysis. A chemonuclear reactor producing carbon monoxide from carbon dioxide (as a step toward nitrogen fixation) could be more easily designed to give a safe fixed fuel system.

PROCESS DESIGN

For the fixed fuel reactor the process design includes extraction of thermal energy from the reactor for delivery to a steam generator or heat exchanger, separation and recovery of the nitrogen dioxide produced, and separation of contaminating activity from the liquid and gaseous plant effluents. For the circulating dust system shown in Figure 4 the process includes in addition separation and recovery of fuel dust.

The separation of nitrogen dioxide, if present in sufficient concentration, can best be accomplished by direct condensation. The energy and mass balance indicates an increase of 0.5 to 1% in nitrogen dioxide concentration in a single pass through the reactor. For product separation by condensation at 2°C. the minimum nitrogen dioxide concentration level with a system pressure of 1,000 lb./sq. in. is 1.65%; therefore the process gas must be recycled to build up product concentration. Some radiation chemistry experiments indicate that the nitrogen dioxide concentration can be built up. For the present estimates a 10% nitrogen dioxide reactant effluent concentration is optimistically assumed. Although thermal dissociation of nitrogen dioxide to nitric oxide and oxygen is significant at reactor temperatures, it is not a problem in the process because the rate of association at condenser conditions is high. Other recovery schemes, such as adsorption on silica gel (*11*), would increase the cost. Recovery and absorption in water would introduce a severe corrosion problem in the reactor circuit, which can otherwise be constructed of inexpensive nonalloy steel.

The severest processing problem is removal of contaminating activity from plant effluent streams. The contaminants may be classified as follows:

1. Argon-41 from neutron activation of argon-40 in the atmosphere. Gases are usually released for dilution through a stack of sufficient height to reduce the activity to permissible concentrations at ground level (*15*). The amount of argon-41 produced in a single-pass process is too great for release through a reasonably sized stack; therefore low temperature fractionation of air for argon removal and a recycle flow sheet are necessary. The air separation plant separates and removes nitrogen so that a mass balance is maintained for producing nitrogen dioxide in the recycle process (nitrogen/oxygen in feed = 0.5).

2. Volatile fission product activity: iodine, bromine, xenon, and krypton. The halogens can be removed in a column containing silver nitrate coated packing (*14*), with a decontamination factor (D.F.) as high as 10^4. If argon has been removed from the feed gas, the volume taken for purge can be small, and storage under pressure is possible for decay of the shorter lived gaseous activity. Maximum permissible concentrations (MPC) of activity in air based on recommendations of the National Committee on Radiation Protection (*NCRP*) (*16*) can be used to compute the required stack height for release to the atmospheric environment. For the smaller reactor a stack height of 225 ft. is required.

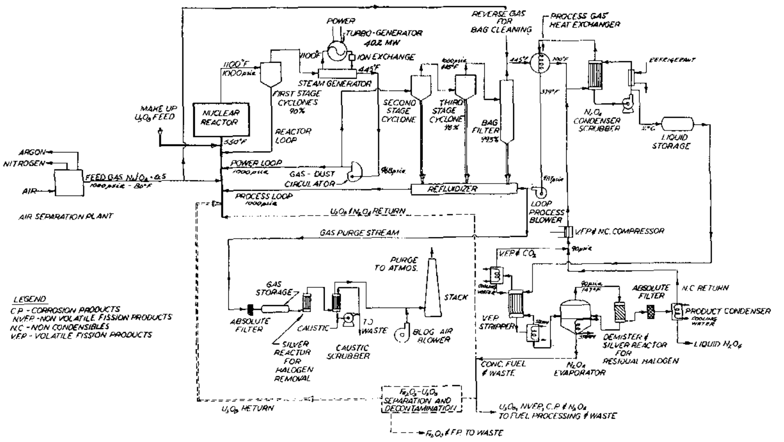


Fig. 4. Production of fixed nitrogen by chemonuclear process, circulating dust fuel.

3. Nonvolatile fission products: barium, cesium, strontium, etc. A major fraction of the nonvolatiles will probably be carried along with the reactant gas stream and will act as a dust. Removal at higher temperatures and pressures can be effected by glass fiber-asbestos filters of the absolute type which are designed to give removal factors of 10^4 down to particle sizes of 0.3 μ (*14*). The fiber filters, although highly efficient, pose a difficult problem of maintenance (cleaning and replacement) in a radiation field. It is expected that a significant fraction of the nonvolatile activity not removed by filters will be entrained in the condensed liquid nitrogen dioxide. One week's storage will decay most of the shorter lived activity (half lives of 8 hr. or less). After having been stripped of any dissolved volatile activity, the liquid may be decontaminated by evaporation leaving a residue of nonvolatile activity. Based on the NCRP total body MPC values for drinking water under continuous exposure, decontamination factors of at least 10^7 to 10^8 are required for decontamination of the liquid nitrogen tetroxide product sent to the evaporators. Based on work with evaporators operating in conjunction with demisters at low boil-up rates (*15*), such D.F.'s may be attainable, but they have yet to be demonstrated.

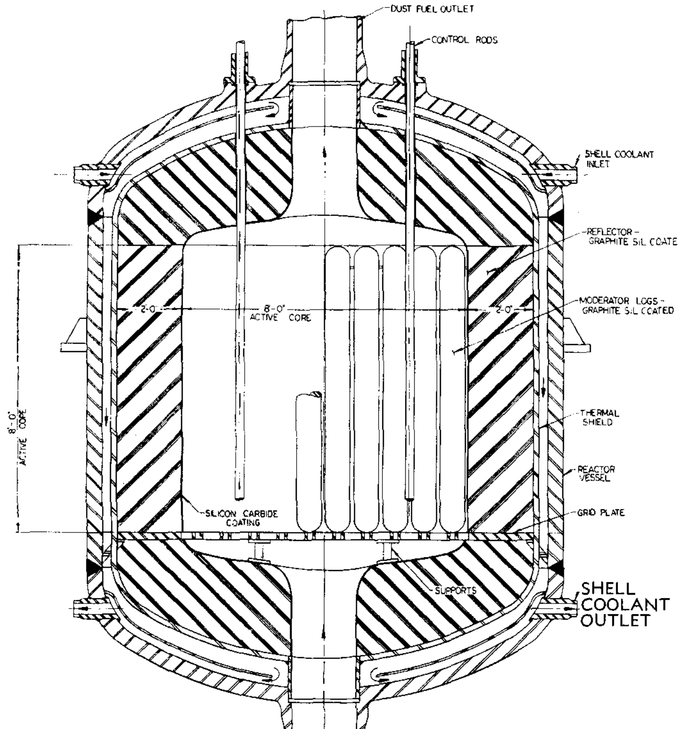


Fig. 3. Conceptual design, 112-mw. dust fueled, internally moderated chemonuclear reactor.

4. Carbon-14 from the neutron activation of nitrogen according to the N-14 (*n*, *p*) C-14 reaction. The contamination due to C-14 is not significant. About 1.5 lb. of C-14 as carbon dioxide are formed per 100,000 tons of liquid nitrogen dioxide. This contributes little to the total activity and can either be removed as an insoluble carbonate or distilled with the volatile fission products for release to the atmosphere.

5. Activated corrosion and erosion products. These are due mainly to iron-55 and iron-59. Although the mass of erosion products could be large in the case of the circulating dust system, the activity is of much less importance than that of the nonvolatile contaminants. Activation of equipment external to the reactor by delayed neutrons carried by the volatile halogens needs examination, especially in connection with maintenance problems.

6. Uranium contamination. The U_3O_8 alpha contamination is more important in the case of the circulating dust system, where large quantities of fuel dust require separation from the process gas; however estimates show that under the process removal conditions assumed this activity is small compared with that of the fission products.

Decontamination in the circulating dust process depends primarily on the reliable performance of at least three stages of cyclone dust separators operating on dust particle sizes (10 μ or less) at the lower limit of their dust separation ability. For the high degree of recovery required the cyclone separators must be followed by continuous bag filters. While bag filters operating under elevated pressure and temperature conditions are known, there is no experience with them in a damaging radiation field.

The dust loadings and circulation rates are comparable to those in fluid catalytic petroleum cracking plants (*12*), but the particle sizes are much smaller. Optimistic estimates (1 mil/yr.) indicate that the erosion and corrosion product formation may be ten times the fission fragment formation rate. To control dilution of the dust fuel by erosion products, it is necessary to separate ferric oxide from U_3O_8. This may be accomplished along with fission fragment removal in a solvent extraction reprocessing operation (*28*). The fuel requires cooling for 50 days prior to processing to prevent damage to the solvent, which increases inventory and fuel processing costs.

Protection from intense radiation requires shielding (*2*) with the equipment being placed in a concrete canyon. A third containment shell may be necessary as an additional factor of safety against release of contaminating activity.

ECONOMIC EVALUATION

Energy Utilization

Fixed nitrogen can be produced by several alternate processes, with a nuclear fuel economy assumed. These are compared on an energy utilization basis in Table 2. In the Wisconsin process (Case III) a high temperature nuclear reactor would replace the gas heated pebble furnace used. The thermal energy required, computed on the basis of heating air, is at worst twice that of the most optimistic estimate for the chemonuclear process. However if the formidable problem of finding nuclear reactor construction materials to withstand the high temperatures could be solved, the process would be simple, and with an indirect heating cycle the product would be uncontaminated.

The Birkeland-Eyde electric discharge process (Case IV) can derive its electrical power from a nuclear power reactor. When one uses the demonstrated power requirement (*17*), and a reasonable power cycle efficiency, the energy requirement increases to from three to six times that of the chemonuclear. Modern plasma technology may bring an improvement in this case, although thermodynamics predicts a maximum yield of 13% nitrogen oxide by volume at 3,500°K., which would decrease the requirement only by a factor of 10 at most.

The conventional catalytic ammonia process with hydrogen generated in efficient electrolytic cells, which derives electricity from a nuclear power reactor (Case V), yields a thermal energy requirement as low as in the best chemonuclear case. Furthermore an uncontaminated product, which can be the readily marketable ammonium nitrate, can be produced with a safely designed reactor system and a demonstrated conventional process.

Case VI is a unique dual cycle system employing fission recoil for nitrogen dioxide and the excess power for producing ammonia via electrolytic hydrogen. Under optimum conditions

TABLE 2. COMPARISON OF THERMAL EFFICIENCIES FOR VARIOUS METHODS OF NITROGEN FIXATION

For a nuclear fuel economy

	Case	Heat of reaction ΔH at 25° Kcal.	Over-all thermal energy efficiency	G, value	Thermal energy requirement kw.-hr. thermal/lb. fixed nitrogen (N)	(Ref.)	Advantage	Disadvantage
I	For stoichiometric Reaction-theoretical $1/2N_2 + O_2 = NO_2$	+8.1	100%	284	0.305	(*20*)		
II	Direct chemonuclear process F.F.							
	(1) $G = 7.4$ and $\epsilon = 0.80$		2.1	7.4	14.7	(*6*)		1 Contaminated product 2 High investment cost 3 Not demonstrated on an engineering scale
	(2) $G = 6.0$ and $\epsilon^o{}_o = 0.50$		1.1	6.0	29.0	(*4*)		
III	Wisconsin process-thermal Direct or indirect nuclear heat at 2,400°K.–2% NO conc. by vol.		1.0		31.0	(*18*)	1 Clean product if indirect heating is used.	1 Difficult construction material problem 2 Contaminated product by direct heating in a neutron field 3 Not demonstrated on an engineering scale in a nuclear plant
IV	Birkeland Eyde-electric discharge Nuclear power–35% efficient 1.2% NO conc. by vol.		0.4		83.5	(*17*)	1 Clean product 2 Safe reactor 3 Demonstrated on an engineering scale	1 High energy requirement but has potential
V	Single-purpose power plant with conventional plants							
	Nuclear power 35% efficient							1 High investment cost
	Electrolytic $3/2H_2O = 3/2H_2 + 3/4O_2$ 70% efficient	+102.5				(*19*)	1 Clean product 2 Can produce NH_4NO_3 3 Safe reactor 4 Demonstrated on an engineering scale	
	Catalytic $3/2H_2 + 1/2N_2 = NH_3$	−11.0						
	Oxidation $NH_3 + 5/4O_2 = NO + 3/2H_2O$	−54.3						
	Oxidation $NO + 1/2O_2 = NO_2$	−13.5	2.0		15.6			
VI	Dual cycle, chemonuclear NO_2 and nuclear power with conventional plant							
	(1) $G = 7.4$ and $\epsilon_o = 0.80$		4.0		7.6		1 Highest efficiency 2 Can produce NH_4NO_3	1 Contaminated product 2 Not demonstrated on an engineering scale 3 High investment cost
	Hydrogen efficiency = 35% × 70% = 24.5%							
	(2) $G = 6.0$ and $\epsilon_o = 0.50$		3.0		10.1			
	Hydrogen efficiency = 24.5%							
VII	Single purpose chemonuclear CO and conventional plant							
	Chemonuclear $3/2CO_2 = 3/2CO + 3/4O_2$ $G = 9$ $\epsilon_o = 0.8$	+101.5				(*21*)	1 Safer reactor	1 Contaminated product 2 Inhibitor required 3 Not demonstrated on an engineering scale
	Water gas $3/2CO + 3/2H_2O = 3/2CO_2 + 3/2H_2$	−15.9				(*19*)	2 Can produce NH_4NO_3	
	Catalytic $3/2H_2 + 1/2N_2 = NH_3$	−11.0						
	Oxidation $NH_3 + 5/4O_2 = NO + 3/2H_2O$	−54.3						
	Oxidation $NO + 1/2O_2 = NO_2$	−13.5	1.7		18.1			

Note: Conventional ammonia plant using natural gas has a thermal energy equivalent of 5.93 kw.-hr.(*t*)/lb. N for raw material and process heat, and 1.65 for compression or a total of 7.58 kw.-hr.(*t*)/lb. N.

TABLE 3. PRELIMINARY ECONOMIC COMPARISON OF CHEMONUCLEAR PLANTS FOR FIXED NITROGEN—1960

Fuel type Case Reactor type Purpose	Circulating dust C-1 Internal moderated SN + SP dual		Circulating dust C-3 External moderated LN + LP dual		Fixed F-3 Internal moderated LN + LS dual		Fixed Power LN single		Fixed Dual cycle Internal moderated LN single		Conventional chemical plant	
1. Nitrogen capacity, tons N_2O_4/D	200		1,330		1,180		1,180		2,600		1,180	
equivalent, tons NH_4NO_3/D							(975)		(2,260)		(975)	
2. Power capacity, mw.(*e*)	34.3		216		2,280,000††		0		0			
Power capacity, mw.(*t*)	112		700		700		516		700			
3. Fuel and fuel loading, lb./cu. ft.	5μ-0.8		5μ-0.36		3μ-ribbon		Optimum		3μ-ribbon			
4. Deposition efficiency, design—%	65.8		70		62		nuclear		62			
max.—%	65.8		70		62		power plant		62			
5. Power or steam efficiency, %	36		36		95		35		36			
6. Mass of fuel—critical—kg. U_3O_8	28.7		43		39		10 mills/kw.-hr.*		39			
In reactor	29.2		44		95		power		95			
In plant	93		223		95				95			
Total inventory	555		1,520		285				285			
7. Capital investment	$/millions	%	$/millions	%	$/millions	%	$/millions	%	$/millions	%		
Reactor plant	33.7	72	73.5	73	52.8	57	54.0	62	72.5	48.0	—	
Chemical plant	12.9	28	27.5	27	40.5	43	32.8	38	77.2	52.0	26.2	
Total	46.6	100	101.0	100	93.3	100	86.8	100	149.7	100	26.2	
Reactor—$/kw.(*t*)	300		105		75		105		104			
Chemical—$/annual ton N_2O_4	215		69		115		93		99		74	
8. Production cost												
Chemical basis—$/ton N_2O_4—%	$/ton	%	$/ton	%	$/ton	%	$/ton	%	$/ton	%	$/ton	%
1. Fuel and fuel cycle	45.80	23.8	20.70	32.5	15.05	24.2	36.60†	65.5	6.85	16.6	7.85	25.5
1. Utilities, labor, maintenance	27.10	14.0	4.10	6.5	4.60	7.4	1.90	3.4	2.60	6.3	3.75	12.2
3. Depreciation, L. and F. tax, ins.	119.60	62.2	38.80	61.0	42.60	68.4	17.60**	31.1	31.85	77.1	19.20	62.3
Gross manufacturing cost incl. return on investment	192.50	100.0	63.60	100.0	62.25	100.0	56.10	100.0	41.30	100.0	30.80	100.0
By-product credit—power at 7 mills/kw.-hr.	43.10	22.4	29.35	46.0								
steam at $0.50 mm.B.t.u.					25.40	40.0	—	—	—	—	—	—
Ar at $0.02/cu. ft.												
Net production cost—$/ton N_2O_4	149.40		34.25		36.85		56.10		41.30		30.80	
equivalent—$/ton NH_4NO_3							(64.50)		(47.50)		(35.40)	
Ratio to market price	1.15		0.26		0.28		0.43		0.32		0.22	
Ratio to conv. mfg. cost	3.2		1.1		1.2		1.9		1.3		1.00	
Distance from conv. plant to compete at 6%/100 miles, in miles	3,700		200		350		1,500		500		0	
Payout time, yrs.	7.8		1.8		2.0		2.7		1.6		0.8	

* Estimated cost of power from a conventional nuclear power plant.
† Includes all costs for a.c. power.
** Depreciation only for chemical plant including conversion to d.c.
†† Steam capacity, lb./hr.

SN = small nitrogen capacity.
LN = large nitrogen capacity.
LP = large power capacity.
LS = large steam capacity.

the energy requirement decreases to about half that of the chemonuclear case. This complex requires a high capital investment but has the advantage of being a single-purpose plant producing a marketable fertilizer with no other gross raw material requirement than uranium, air and water.

One other possibility is the production of carbon monoxide from carbon dioxide by a chemonuclear process (Case VII). The carbon monoxide after separation is used to form hydrogen by the well-known water-gas reaction, which is then used for catalytic nitrogen fixation. For high *G* values inhibitors are required in the system (*21*), and under optimum conditions a thermal energy requirement only slightly higher than that of the direct chemonuclear process results. An advantage of this system is that a safer (negative prompt reactivity coefficient) nuclear reactor can be attained.

Cost

Preliminary capital investment and production cost estimates have been attempted for a number of fixed and circulating fuel, single and dual-purpose chemonuclear plants, and are compared on the same basis with those of conventional plants with natural gas. Cost estimation procedures for proposed nuclear power and nuclear fuel reprocessing plants have been used (*23, 24, 25*).

Table 3 summarizes for several representative cases (*2*) some of the pertinent design information, reactor and chemical plant costs, and gross and net production costs. Two levels of production, 200 tons/day nitrogen dioxide and 1,180 to 1,330 ton/day, were examined for each of the following cases: single-purpose nitrogen production plants, dual-purpose nitrogen plus smaller and larger by-product plants (C-1 and C-3), smaller and larger dual-purpose nitrogen plus by-product steam plants (F-3), nuclear-powered conventional plants, dual-cycle plants, and conventional plants (*22, 26*).

For convenience all estimates were made on a nitrogen dioxide basis. Since nitric acid is the principal product in the conventional process, cost comparisons are relative and not absolute, with a penalty assessed against the conventional process. Recently a market for nitrogen dioxide has developed owing to government emphasis on usage as a rocket oxidant material.

Total annual fixed charges of 14 and 19% were respectively used for the reactor and chemical plant investments including shielding. A by-product credit of 7 mills/kw.-hr. is taken for power, $0.50/million B.t.u. for steam, and $0.02/std. cu. ft. for argon production up to 10% of the present total U.S. consumption only. The conventional plant was assumed to be located in the midwest, with natural gas at $0.40/million B.t.u. and to be equipped with electrically-driven compressors with power at 7 mills/kw.-hr. For the cases supplying nuclear power to conventional plants, nuclear power was assumed at 17 and 10 mills/kw.-hr. for the smaller and larger plants, respectively (*23, 29*).

In comparing fixed and circulating fuel plants of the same capacity there is no significant difference in capital investment or net production cost because the smaller investment cost for the latter is offset by lower fuel and fuel cycle costs for the former.

Although for the set of conditions chosen none of the plants examined shows competitive status, it is of interest that extrapolation at a given nitrogen capacity of 1180 tons/day and increasing the power-producing level to 350 mw. (e), [1,000 mw. (t)], indicates a competitive situation at a *G* value no higher than 3.7 or at a deposition efficiency no higher than 38% with a *G* value of 6. For a given nitrogen capacity increasing the *G* value tends to decrease the net production cost, but for dual-purpose plants the rate of decrease diminishes rapidly because less credit can be taken for by-product power. A *G* value of 7.5, energy deposition of 70%, a production level of 1,330 tons/day nitrogen dioxide, and reactor power level of 560 mw.(t) are needed to reach a break-even point in today's market.

It should be emphasized that the economics of dual-purpose plants depends as strongly on the balance in market demand for both power and nitrogen at a single-plant location and the interest and attitude of both the chemical and power industries in a venture of this type as on variations in *G* value or energy deposition efficiency.

For a fixed capacity increasing the *G* value in single-purpose chemonuclear plants does not cause costs to converge rapidly toward conventional costs because of the increased unit plant costs in smaller, more efficient chemonuclear plants.

Figure 5 shows production cost as a function of capacity for the various types of plants. The net production cost for smaller chemonuclear plants is more than three times as high as for present conventional plants. Larger dual purpose plants (power or steam) show net production costs that are about 10 to 20% higher than conventional and tend to approach conventional costs rapidly with increasing capacity.

The production cost for a nuclear power-based conventional plant is about twice as high as for conventional plants at both lower and higher capacities and does not converge rapidly because of the increased capital investment and lack of by-product power credit. The higher cost must be balanced against the advantages of an independent single-purpose plant, a product requiring no proof of safe usage, and a plant design requiring no additional development outlay beyond that for developing an economical nuclear power reactor.

The single-purpose, dual-cycle complex has been compared with other nuclear processes at the same reactor power level, which in effect doubles the production rate. The production of 2,600 tons/day nitrogen dioxide is approximately equivalent in nitrogen capacity to that of a 1,000 tons/day ammonia plant which is about the largest in existence (*19*). Compared with conventional plants the cost is still about 30% higher because of the large investment in this complex.

CONCLUSIONS

There is no inherent technological barrier against the large-scale production of fixed nitrogen by a fission recoil process. However because of the many technical problems requiring solution the development can be costly and time consuming. At the present state of knowledge there does not seem to be any clear-cut advantage over conventional processes, even based on a nuclear economy, and therefore a large-scale development effort does not seem warranted now. Nevertheless because of the importance of fixed nitrogen and the possibility that a favorable situation may arise in the future, moderate research efforts should continue in radiation chemistry, fuel development, decontamination problems, and studies aimed at evolving a safe chemonuclear reactor concept so that further evaluation can be properly performed.

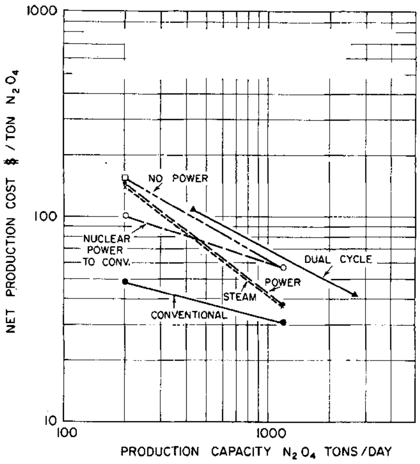


Fig. 5. Comparison of net production cost for fixed nitrogen by chemonuclear and conventional processes.

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NOTATION

W = energy consumed in forming the product, kw.-hr./lb.
G = radiation yield, molecules of product formed per 100 electron volts of energy deposited in the reactant stream
e_o = over-all energy deposition efficiency-fraction of total fission energy deposited in the reactant stream
M = molecular weight of product
P = power level of nuclear reactor in megawatts
T = production rate of nitrogen dioxide in tons/day
e_{KE} = fraction of fission energy released as kinetic energy = 170 mev./200 mev./fission = 0.85
e_{SG} = the self-absorption efficiency or the fraction of kinetic energy escaping the fuel
e_{EG} = the nonre-entry efficiency or the fraction of kinetic energy which remains in the gas stream

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