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STUDY OF THE PROSPECTS FOR HEAVY WATER PRODUCTION VIA
LASER ISOTOPE SEPARATION

Final Report
for Period 1 April 1976 - 30 September 1976

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ABSTRACT

The rationale for "cheap" heavy water is discussed with emphasis on the economic, safety, and arms control implications of the widespread adoption of pressure-tube, heavy water moderated and cooled CANDU reactors, and variations thereof. Three classes of vibrational-photochemical laser processes are considered in detail, i.e., hydrogen halide-unsaturated hydrocarbon addition reactions, isotopically selective photoadsorption and photodesorption, and selective two-step molecular dissociation. General remarks are made concerning the economic viability of these techniques vis à vis hydrogen sulfide/water chemical exchange (the GS process).

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1.0 INTRODUCTION

This report develops in greater detail some of the ideas reported in the quarterly progress report for 1 April 1976 - 30 June 1976⁽¹⁾, and also describes some new areas of investigation. The report is organized as follows: in Section 2, we attempt to supply some motivation for this study by considering the distinctive feature of heavy water fission reactors in comparison with light water systems. In particular, we discuss relative economics and reactor safety, and comment on some implications for arms control of the widespread adoption of heavy water reactors. Section 3 is devoted to a discussion of the prospects for heavy water separation via infrared laser catalyzed chemical reactions in the gas phase. As a prototype, we consider in greater depth a system based on hydrogen halide additions to unsaturated hydrocarbons which was discussed briefly in⁽¹⁾. In Section 4, we present some preliminary ideas on isotopically selective photoadsorption and photodesorption with emphasis on the basic physical and chemical mechanisms involved. The final laser separation scheme to be analyzed in this report is the two-step dissociation of monodeuterated ammonia (Section 5). The two concluding sections are concerned with some of the basic economic facts of life for the use of lasers in heavy water production (Section 6), and a personal perspective on what has been accomplished in this field to date, and what remains to be done (Section 7).

2.0 WHY SEPARATE HEAVY WATER?

The basic rationale for this study that-"cheap" heavy water (D_2O) would be a good thing-is founded on the belief that heavy water fission reactors, such as the CANDU design developed in Canada, which uses D_2O as a neutron moderator and coolant (approximately 0.8 metric ton (mt) D_2O per megawatt of electrical capacity or 800 mt for a 1000 MWe reactor) have certain attractive features which merit consideration of these systems both in the U.S. and abroad. Since D_2O is expensive (the current price of \$120/kilogram is expected to escalate to ~\$200/kilogram by the end of the 1970's), it is "obvious"⁽²⁾ that if D_2O were "free" (relatively speaking), the current CANDU design (and advanced designs which would retain D_2O as the neutron moderator but might substitute thorium for natural uranium as fuel and/or an organic solvent for D_2O as coolant) would become a more attractive energy option. We recognize that deuterium is also projected for use as fuel for D-T and D-D controlled thermonuclear reactors (CTR). However, we emphasize the application in fission reactors because CTR reactor physics and technology have a long way to go before successful commercialization, and also because fusion deuterium requirements are relatively modest. (For example, a study at Brookhaven National Laboratory⁽³⁾ has concluded that if by the year 2020 all fossil fuels are replaced with synthetic fuels produced by D-D fusion reactors, and all electricity production is by CTR, the total amount of deuterium needed to supply all U.S. energy needs (estimated to be 500×10^{15} BTU's) would be only 1000 tons D_2 per year or 5000 tons D_2O (assuming 100% efficiency for the D-D fusion reaction.)

With the desirability of cheap D_2O as our basic premise we now turn to a consideration of the economics, safety, and arms control implications of heavy water reactors (HWR) vis à vis the light water reactors (LWR) used exclusively in the U.S. and extensively around the world. Before beginning this discussion, I feel that a caveat to the reader is in order. While it is possible to have a relatively objective discussion of the pros and cons of various laser isotope separation schemes, the same statement, unfortunately, cannot be made regarding either comparative reactor economics or safety. As regards safety, this statement will be almost a cliché to anyone familiar with the heated controversy surrounding the development of fission power, especially over the past few years. However, that the same can be said of economics might surprise those who feel this discipline to be (relatively) cut- and- dried.⁽⁴⁾ My own impression is that, depending on the values of such parameters as interest rates on reactor capital investment, and one's personal prejudices regarding the future costs of enriching uranium and heavy water and reprocessing spent fuel, either LWR or HWR can be shown to have an economic advantage over the other. In the following, I have attempted to present a balanced point-of-view, but the reader should be forewarned that these are complicated issues about which the author lays no claims to expertise.

A. Comparative Reactor Economics

We compare an enriched uranium fueled, light water moderated and cooled, pressure vessel reactor, to be concrete, a pressurized water reactor (PWR)⁽⁵⁾, with a natural uranium fueled, heavy water moderated and cooled, pressure tube reactor of the CANDU type.⁽⁶⁾ The conventional wisdom on this subject can be summarized as follows: because of the good neutron economy of the

D₂O moderator and coolant which permits the use of natural uranium fuel with no loss of U-235 in the tailings of an enrichment plant, and the pressure tube design which enables on-power fueling, the HWR has intrinsically lower fuel cycle costs than a LWR, and is also less sensitive than LWR to escalating prices for uranium ore and reprocessing ⁽⁷⁾, while being independent of rising uranium enrichment costs. On the other hand, this is balanced by the higher capital cost of a HWR, which is largely due to the D₂O inventory which must be present at reactor start-up (in contrast to uranium enrichment charges in a LWR which are spread over the life of the reactor), and the comparatively lower power density of an HWR. Table 1, adopted from Weinberg ⁽⁸⁾ presents this picture in somewhat greater detail, as of January 1975. (The assumed prices of uranium ore, separative work, and fuel reprocessing are already out of date; multiplication by a factor of two would probably present a more realistic estimate of the current situation.) Table 2, adopted from Benedict ⁽⁹⁾ presents the comparative fuel utilization and plutonium production picture in greater detail. We note that the assumed burnups have not achieved in practice; typical numbers are 8000 MWD/MT for the Pickering CANDU HWR, and 21,800 MWD/MT for the Zion I PWR in the U.S. Also, the annual makeup figures for the PWR assume uranium recycle (but no plutonium recycle). If reprocessing costs rise to a point at which uranium recycle is no longer economic, these figures should be modified as follows (details can be found in Appendix A); ST Natural U₃O₈ → ~197 and MT Sep. Work Units → ~123.2. Using these figures (and the theoretical burnups cited in (9), the approximate U₃O₈ and isotope separation requirements for 30 year operation of a 1000 MWe LWR and HWR are:

<u>Reactor Type</u>	<u>U₃O₈ Requirements</u>	<u>Cost of U₃O₈ at \$50/lb.</u>	<u>Isotope Sep. Req.</u>	<u>Cost of Isotope Sep. at \$100/kg SWU for U-235 and \$200/kg for D₂O</u>
LWR (once through)	6135 S.T.	~\$600 mil.	3700 MT S.W.U.	\$370 mil.
LWR (U-recycle)	5040 S.T.	~\$500 mil.	3800 MT S.W.U.	\$380 mil.
HWR (once through)	3840 S.T.	~\$380 mil.	~1000 MT D ₂ O (including makeup)	~\$200 mil.

With regard to U₃O₈ requirements, another important statistic is the projected natural uranium consumption of the thorium-burning CANDU: ~25 tons per 1000 MWe reactor per year, corresponding to a burnup of ~50,000 MWD/MT⁽¹⁰⁾.

My personal conjecture is that with the improvement of old techniques and development of new ones, the cost of both uranium and heavy water enrichment will decline, but that the cost of U₃O₈ might rise to a level that will make continued use of current LWR highly wasteful of uranium resources, but would not make the introduction of fast breeders imperative. This seems to me to be the major economic motivation for keeping a thermal "near-breeder" option, such as a thorium-fueled CANDU, viable in the U.S.

TABLE 1

COMPARATIVE COSTS OF NUCLEAR ELECTRICITY

Capital Cost (\$/KWe)	420	600
Capacity Factor (%)	70	80
Fuel Cycle (mills/KW hr.)*	2.8	1.1
Operating and Maintenance Costs	0.5	0.7
Total Costs (mills/KW hr.)		
at: 10% fixed charge	10.2	10.4
15% fixed charge	13.6	14.7
20% fixed charge	17.0	18.7

*Assumes: uranium ore (U_3O_8) at \$15 per pound; separative work for LWR fuel enrichment at \$50 per kilogram separative work unit; LWR fuel reprocessing at \$100 per kilogram; LWR fuel fabrication at \$75 per kilogram; LWR spend fuel shipping at \$10 per kilogram; conversion of U_3O_8 to UF_6 for LWR \$9.30 per gram; interest on LWR fuel cycle at 16.90% per year.

TABLE 2

COMPARISON OF FUEL CYCLE REQUIREMENTS
FOR 1000 MWe THERMAL REACTORS

(Enrichment Plant Tails at 0.2 w/o U-235)

<u>Reactor Type</u>	<u>PWR</u>	<u>HWR</u>
Thermal Efficiency, %	33	30
<u>Initial Charge</u>		
MT Uranium	79.3	151
w/o U-235	2.26	0.711
ST Natural U_3O_8	422	199
MT Sep. Work Units (SWU)	218	0.0
<u>Annual Makeup, 75% C.F.</u>		
Burnup, MWD/MT	32,600	9,600
MT Uranium	25.72	96.08
w/o U-235	3.21	0.711
ST Natural U_3O_8	159.3	125.5
MT Sep. Work Units	120	0.0
KG Fissile Pu Produced	161	220

C.F. = Capacity Factor

M.T. = Metric Tons, 1000 kilograms

MWD = Megawatt Days (Heat)

Pu = Plutonium

ST = Short Tons, 2000 pounds

B. Comparative Reactor Safety

A mini-perspective on the conventional wisdom in this area can be gleaned from the following comments by Hans Bethe, Henry Kendall of MIT and the Union of Concerned Scientists, Richard Webb, a nuclear engineer who has been active in the reactor safety debate, and W. G. Morrison, Assistant Director of Generation Projects, Ontario Hydro.

"The reactor which I like best is the heavy water reactor which has been developed by the Canadians.....It costs a little more to build because the heavy water is an expensive substance, but this will easily be repaid in the uranium economy. It is believed, in addition, that it is even safer than our present light water reactors." (11)

"Even reactors less vulnerable than American models to certain catastrophic accidents - CANDU for example - present dangers in the proliferation of nuclear weapons and in disposal of nuclear wastes." (12)

"Alternate Reactors. Of special interest is the CANDU reactor, which is widely in use in Canada. The reactor has several safety advantages; mainly, it does not require the successful operation of its emergency core cooling system to avoid a containment rupture in the event of a loss-of-coolant accident, because the reactor design is such that no fuel melting would occur. However, an autocatalytic power excursion accident would definitely occur during a loss-of-coolant accident, if a reactor SCRAM fails to occur, which would cause a core meltdown. (The CANDU has a backup SCRAM system.) It cannot be demonstrated theoretically that the containment would not be breached in this event." (13)

"The CANDU pressure-tube design confines the high-temperature coolant in the reactor to relatively small channels surrounded by cool moderator which would act as a heat sink in the event of a reactor accident. This distributed heat sink in the core is an important feature of the CANDU reactor, because the failure of emergency core cooling does not lead to a core meltdown as may be the case in the pressure-vessel concepts." (14)

What is alluded to by Bethe and Kendall, implied in Webb's remarks, and explicitly stated by Morrison, is the expectation that in the event of a loss-of-coolant accident (LOCA), the D_2O moderator will soak up the radioactive after-heat (15) from the un-cooled fuel rods, and thus prevent a core-meltdown even without the operation of an emergency core cooling system (ECCS). My personal reaction in first reading these remarks was that while this scenario constitutes an attractive hypothesis, it was far from obvious that events would take this course. As noted in reference (6), the fuel in a CANDU reactor is not in physical contact with the moderator, and heat transfer must be accomplished across an annular gap filled with an insulating gas. Thus the situation is not identical to that in a high temperature gas-cooled reactor (HTGR) where the fuel is in intimate contact with the graphite moderator. In this regard, the discussion by T. J. Thompson of the NRX accident is informative. (16) A few words of background. The NRX reactor was a D_2O moderated, H_2O cooled, research and testing reactor, using natural uranium fuel, which was capable of operating at power levels up to 30 MW. On December 12, 1952 there was a partial meltdown of the core caused by a power excursion due to control rod malfunctions and and safety circuit failures. The excess reactivity was quenched by dumping the D_2O moderator. Twenty three pressure tubes ruptured as a result of the accident. In several cases melting

of fuel in one pressure tube caused damage in an adjacent tube. According to Thompson, "Because of the fuel resistance to the escape of water, the steam pressure built up within the cooling section and at least two rods broke apart and the upper portions of their shielded sections leaped a foot or so into the air. The air-cooled rod (all rods were water-cooled with one exception) which was located in the circle of maximum flux did not damage the calandria tube as far as could be seen. The central parts of the aluminum sheaths melted and ran down the rod, congealing between the calandria tube and the rod. (The inner and outer sheaths of the pressure tubes were made of aluminum.) This aluminum formed a barrier on top of which molten uranium formed an ingot contained by the calandria tube. At this point, heat transfer between the calandria tube and heavy water apparently preserved the tube itself. Since there was no damage to the calandria tube in this case, it is evident that steam considerably influenced the course of the accident. Steam under pressure in the rods caused the initial rupturing of the outer sheaths."

The tentative conclusion which I believe can be drawn from the above is that, while the D_2O moderator is "too far away" to be completely effective as a heat sink, it may be possible by careful design to maintain the overall integrity of the pressure tube system in the event of a LOCA. While I have not been able to confirm this hypothesis by reference to the open literature, Mr. Eugene M. Yaremy, Branch Head, Safety Systems Branch of Atomic Energy of Canada Limited, has stated in a private communication to the author that the pressure tubes of the Bruce CANDU reactors, now under construction, are designed to slump against the calandria tubes in the event of a LOCA, thus facilitating heat transfer between the fuel and the D_2O moderator.

In this connection, it should also be noted that the Bruce reactors have no provision for moderator dump, and use ordinary water as emergency coolant in contrast to the Pickering reactors which are designed to dump the moderator, and use this D_2O as emergency coolant in the event of a LOCA. How effective the Bruce system would be in practice is hard to judge.

While the above barely scratches the surface of comparative reactor safety, further detailed discussion would, unfortunately, carry us too far afield. For further analysis of the safety-related characteristics of D_2O moderated and cooled pressure tube reactors and variations thereof, we recommend the comprehensive study of J. R. Dietrich.⁽¹⁷⁾

The basic conclusion we would like to leave with the reader is that there very well may be significant differences between a pressure-tube HWR and a pressure vessel LWR as far as certain classes of reactor accidents are concerned, but that in the absence of hard information in the open literature on the former system, comparable to that provided by WASH-1400⁽¹⁸⁾, one should be extremely wary of sweeping generalizations.

Finally, we take this opportunity to point out that the search for an improved heavy water process⁽¹⁹⁾, as a substitute for the existing method based on hydrogen sulfide/water chemical exchange (GS process) has a safety as well as an economic rationale. That is, hydrogen sulfide is highly toxic (maximum acceptable concentration of 20 ppm in air), (and very corrosive), and large quantities are required, e.g., a GS plant producing 400 metric tons per year of D_2O requires an inventory of approximately 600 metric tons of hydrogen sulphide⁽¹⁹⁾. Although stringent safety precautions are taken by the Canadian heavy water industry and the plants are sited in areas of low population density, the pos-

sibility of accidental releases of large amounts of hydrogen sulfide is a definite source of concern.

C. Arms Control Implications

The dangers inherent in a so-called "plutonium economy" have recently received widespread publicity in the technical and non-technical literature.⁽²⁰⁾ The concern is that the widespread use of plutonium as fuel in thermal and/or fast breeder reactors would inevitably lead to diversion of sufficient amounts of this material by "irresponsible" governments or terrorist groups to produce nuclear and radiological weapons. From this nuclear proliferation perspective, heavy water reactors of the CANDU type present a mixed picture.

The positive aspect is inherent in the fact that the CANDU system is economically viable operating on a once-through cycle. Thus, there would be little or no economic penalty involved in not reprocessing spent fuel, and since unprocessed fuel is extremely radioactive, this feature of the CANDU fuel cycle is highly attractive from an arms control point of view. Unfortunately, it is a fact of life that most of the existing and planned reactors are of the light water variety, and we have seen that there is considerable U-235 left in the spent fuel at projected burnups - ~0.83% vs. 0.711% in natural uranium. This latter fact forms the basis of an ingenious suggestion made recently by the United States Arms Control and Disarmament Agency (ACDA).⁽²¹⁾ In the proposed "tandem cycle" spent fuel from light water reactors would be used, without reprocessing, as fresh fuel in heavy water reactors. There are some obvious difficulties inherent in this approach, e.g., modifying the CANDU on-line fueling machines and pressure tubes to handle the hot, spent fuel from

the light water reactors, but ways can probably be found to circumvent the technical and political problems involved if arms control is perceived as deserving the highest priority.

The negative aspect is inherent in the use of on-line fueling for the CANDU vs. batch fueling in a LWR. That is, while one can be sure that no spent fuel is being taken out of a LWR while the reactor is on-line, this is not true for the CANDU, and continuous surveillance is necessary to prevent diversion.

3.0 VIBRATIONAL PHOTOCHEMISTRY

The potential of vibrational photochemistry for heavy water separation, and various specific processes were discussed in reference (1). Here we restrict ourselves to a consideration of a particular class of reactions, hydrogen halide additions to unsaturated hydrocarbons in the gas phase. As noted by Marling⁽²²⁾, there exist excellent coincidences between CO laser lines and the vibrational transitions of DCl, DBr, and DI. Moreover, activation energies for hydrogen halide addition to olefins fall into the range 15-40 Kcal/mole⁽²³⁾, and hence excitation of low-lying vibrational levels of the deuterated halides should result in addition reaction rates sufficient to beat both the deuterated halide/hydrogen halide and deuterated halide/olefin vibrational energy transfer reactions, if the added vibrational energy is effective in lowering the reaction activation energy. The theoretical prediction⁽²⁴⁾ that vibrational energy is generally more effective than translational energy in accelerating the rates of endothermic reactions, while the converse is true for exothermic reactions, has received its most striking confirmation in studies of diatomic molecule-atom exchange reactions, e.g.,^{(25), (26)}

and

The dogma regarding reactions such as (1) and (2) runs as follows⁽²⁷⁾: if the forward, exoergic reaction leads to products with large internal excitation, then microscopic reversibility implies that the rate

of the reverse, endoergic reaction will be enhanced when the energy is in the internal degrees of freedom rather than in relative translation. In reaction (1), the fraction of the total available energy becoming vibration in HF was approximately 57%, and the endoergic reaction rate constant was predicted to increase by orders of magnitude as energy is withdrawn from rotations and translation and transferred to HF vibration. In reaction (2), HCl ($v = 2$) reacted with Br 10^{11} times faster than HCl ($v = 0$).

From the above considerations, it is clear that insight into the effect of vibrational excitation of hydrogen halides on the addition reaction rate can be gleaned by looking at the data on the energy disposal of the reverse reactions, i.e., unimolecular hydrogen halide eliminations following chemical activation and UV photolysis. There is an extensive literature in this area; the experimental evidence is summarized by Berry⁽²⁸⁾ who concludes that all hydrogen halide elimination products acquire ~15-40% of the potential energy⁽²⁹⁾ available to the reactants (defined as the threshold energy for hydrogen halide elimination minus the endoergicity for final product formation) as vibrational energy. The remaining energy is channeled into hydrogen halide rotation, olefin product rotation and vibration and relative translational energy of the recoiling products. On the basis of this data, one may conjecture that vibrational excitation of the hydrogen halide will accelerate addition reactions to some extent, but that the dramatic rate enhancements characteristic of reactions such as (1) and (2) can not be expected. Tentative support for this conclusion is available from the work of Berry, Molina, and Pimentel on dichloroethylene photoelimination reactions⁽³⁰⁾, but more work remains to be done.

We turn now from photochemistry to process engineering. That is, assuming for the moment that the vibrationally catalyzed addition reaction works perfectly, what would the "back-end" of a plant using this process to produce heavy water look like? To provide a possible answer to this question, we elaborate on the block diagram of the prototype system discussed in reference (1), Figure 1, p. 3-4). In Figure 1 we have indicated the relative material flows in a plant in which the deuterated product (DClX) of the addition reaction is thermally dissociated to produce isotopically pure DCl and eventually D₂O through exchange with natural water. At the same time the HCl feedstock is redeuterated by exchange with natural water in the reflux tower. The material flows in the reflux tower assume equilibrium operation at 25° C. At this temperature the separation factor α for the exchange reaction

is

$$\alpha = \frac{\left(\frac{\text{D}}{\text{H}}\right)_{\text{H}_2\text{O}}}{\left(\frac{\text{D}}{\text{H}}\right)_{\text{HCl}}} = 2.5. \quad (31)$$

As stated in reference (1), the basic difficulty with the halogen halide-water systems is that they form constant boiling mixtures. In order to break the azeotrope⁽³²⁾ and decrease the solubility of the halide in water a common ion solute must be added. Thus for the HCl/H₂O system we would add NaCl. It is obviously important to know how much this use of NaCl will add to the cost of D₂O. In the absence of a detailed engineering design analysis we offer the following "back of the envelope" estimates based on the flows shown in Figure 1, and some comments.

Figure 1. Laser Addition - Reaction Separation Plant.

A saturated salt solution contains approximately 36 grams of NaCl per 100 grams H_2O . To obtain an upper bound on the amount of salt required we assume

$$\frac{30 \text{ gms NaCl}}{100 \text{ gms } H_2O} \equiv \frac{5.5 \text{ gms NaCl}}{\text{gm mole } H_2O}$$

Then, since the production of 0.5 gm mole D_2O (10 gm) requires a minimum feed of 3500 gm moles H_2O , we need to add 19,250 gms of salt. Assuming that salt in large quantities costs 1¢/lb, the cost of breaking the azeotrope with no NaCl recovery would be

$$\frac{19,250 \text{ gm}}{453 \text{ gm/lb}} \times \frac{\$0.01}{\text{lb}} \approx \frac{40¢}{10 \text{ gm } D_2O} = \frac{\$40}{\text{kg } D_2O}$$

This represents a lot of salt, e.g., for a heavy water plant producing 500 mt D_2O per year we would need

$$\frac{500 \text{ mt } D_2O}{\text{yr}} \times \frac{10^6 \text{ gm}}{\text{mt}} \times \frac{19,250 \text{ gm NaCl}}{10 \text{ gm } D_2O} \approx \frac{10^6 \text{ mt NaCl}}{\text{yr}}$$

To obviate the necessity of locating the heavy water plant near a salt mine and dumping all this salt into the feed water, it seems obvious that a much better route would be to recover the salt. To get an upper bound on the energy cost for this process, we assume it is equivalent to the heat of vaporization of water. Then one requires

$$\frac{3500 \text{ gm moles } H_2O \times \frac{18 \text{ gm}}{\text{gm mole}}}{453 \text{ gms/lb}} \times \frac{1000 \text{ B.T.U.}}{\text{lb}}$$

$\approx 140,000 \text{ B.T.U.}$ to evaporate water while making 10 gms of D_2O .

Assuming a heat cost of $\approx \frac{\$2}{10^6 \text{ B.T.U.}}$ gives an energy cost

$$\approx \frac{\$28}{\text{kg } D_2O}$$

This figure is probably too conservative for the following reasons:

- (1) If the NaCl concentration required to break the azeotrope is not too different from sea water, then one could use a standard desalination technique such as the Zarkhin process to recover the salt at a cost of $\sim \frac{\$1}{\text{m}^3 \text{H}_2\text{O}}$ or $\sim \$6/\text{kg D}_2\text{O}$.
- (2) Since H_2O is cheaper than NaCl it might be possible to use solar energy ponds to evaporate the water. The process cost would then be the cost of fresh water $\sim \frac{10\text{¢}}{\text{m}^3 \text{H}_2\text{O}}$ or $\sim 60\text{¢}/\text{kg D}_2\text{O}$.

This purpose of the foregoing exercise was to point out the importance of non-laser costs in considering the economics of laser isotope separation techniques. We discuss this point further in Section 5.

4.0 ISOTOPICALLY SELECTIVE PHOTOADSORPTION AND PHOTODESORPTION

Recent experimental and theoretical studies⁽³³⁾⁻⁽³⁶⁾ have raised the possibility that it may be feasible to separate isotopes of commercial interest, e.g., deuterium by vibrational excitation of the isotopic molecule of interest either before it collides with a surface (photoadsorption) or after it has already been desorbed (photodesorption). In this section, we review some of the relevant background material, sketch some of our own ideas, and suggest some experiments which should shed light on the possibilities for efficient isotope separation using these techniques.

A. Background

To establish a point of reference, we quote some introductory material from a book by Young and Crowell⁽³⁷⁾:

"Adsorption of a gas on a solid is a spontaneous process and is therefore accompanied by a decrease in the free energy of the system. Since the process involves loss of degrees of freedom of the gas, in passing from the free gas to the adsorbed film, there is also a decrease in entropy. It follows from the equation

$$\Delta F = \Delta H - T\Delta S$$

that the adsorption process must always be exothermic. This is true regardless of the nature of the forces involved."

"Adsorption processes may be classified as physical or chemical, depending on the nature of the forces involved. Physical adsorption, also termed van der Waals adsorption, is caused by molecular interaction forces; the formation of a physically adsorbed layer may be likened to the condensation of a vapor to form a liquid. On the other hand, chemisorption, involves transfer of electrons between the solid (or adsorbent) and the gas (or adsorbate). The process

essentially involves the formation of a chemical compound between the adsorbate and the outermost layer of adsorbent atoms."

"The heat of physisorption is of the same order of magnitude as the heat of liquefaction of the adsorbate and is rarely more than twice or three times as large, whereas the heat of chemisorption is of the same order as that of the corresponding bulk chemical reaction. A physically adsorbed layer may be removed by reducing the pressure, at the temperature at which adsorption took place. the removal of a chemisorbed layer, however, often requires much more rugged conditions."

"Under suitable conditions of temperature and pressure, i.e., close to those required for liquefaction, physically adsorbed layers several molecular diameters in thickness are frequently found. In contrast, chemisorption is complete once a monomolecular layer is built up, although physisorption may occur on top of the chemisorbed monolayer."

The relevance of these remarks to the subject at hand is as follows:

(1) The potential of vibrational catalysis in physisorption is clear, i.e., typical activation energies for physisorption (~ 1 Kcal/mole) are comparable to or less than vibrational energies of diatomic and simple polyatomic molecules such as DCl, HDO, HDS, and NH_2D which are potential candidates for deuterium separation (e.g., $\text{DCl } (v = 1) \equiv \sim 5.6$ Kcal/mole). Thus, absorption of a single IR quantum can be expected to have a pronounced effect on adsorption and desorption rates, while the photodesorption of chemisorbed species suffers from the same problem as vibrational photochemistry in the gas phase, i.e., significant enhancement of the reaction rate usually requires the absorption of many IR quanta. The situation with regard to photoadsorption of chemisorbed species is less clear since there are

many systems in which chemisorption involves an activation energy barrier which may be overcome by absorption of a single IR quantum.

(2) If the experimental conditions favor the formation of multi layers, either physisorbed layers on top of a primary chemisorbed layer, as occurs, e.g., for water adsorbed on amorphous silica, or a thin film of physisorbed material, surface scrambling may degrade the isotopic selectivity "induced" by IR absorption. We illustrate this point with two examples:

a. Water on Amorphous Silica ⁽³⁸⁾

From studies of the hydration and dehydration of silica, it is fairly certain that the surface is ordinarily covered with a chemisorbed monolayer of hydroxyl groups formed in the reaction

This layer of chemisorbed water attracts additional water which is physically adsorbed to the hydroxyl groups via hydrogen bonding, e.g., ⁽³⁹⁾

(Dotted lines represent hydrogen bonds)

We stress that the mechanism represented by Equation (5), i.e., that pairs of hydroxyls are favored adsorption sites for water, is an attractive hypothesis. There are many other possibilities, e.g., the hydroxyl groups may be involved in hydrogen bonding among

themselves. For sake of argument we suppose Equation (5) represents the state of affairs, and concentrate on the one in 3500 hydrogen-bonded HDO molecules. The basic question is: what will happen if the O-D stretching vibration is excited by an IR laser? Even an educated guess requires a knowledge of the differential heat of adsorption. The figure cited in reference (36), 15 Kcal/mole, characterizes the adsorption of water at 50% average on synthetic Na-faujasite, a zeolite (natural hydrated silicate of calcium and aluminum). It represents mainly the interactions of the lone electron pairs of the oxygen atoms of the water with the hydroxyl groups on the surface and with the zeolite cations. If, as assumed in reference 36, the O-D bond can be selectively excited at $3.8\mu \simeq 7.5$ Kcal/mole, three things might happen: the HDO molecule may desorb, the vibrational energy may be transferred to the "local environment", i.e., the surface hydroxyl groups, or the absorbed energy may be sufficient to overcome the barrier to surface migration, and the HDO molecule may hop around the surface, becoming deactivated in the process. The latter two processes would, of course, lead to a loss in isotopic selectivity. Much experimental and theoretical work remains to be done to clarify this situation.

b. Variations on the "Basov Mechanism"

In reference (33), Basov et al. have proposed a simple model for vibrationally catalyzed adsorption or condensation whose essential features can be summarized as follows: a vibrationally excited gas molecule falls on the surface of an already-condensed (adsorbed) phase. It is assumed that the molecule sticks to the surface, and because of very efficient vibrational relaxation in liquids, there is an "instantaneous" transfer of this energy into local heating. The subsequent temperature rise is governed by the Green's function for the heat equation

$$T(r, t) = \frac{\Delta E_v}{4 c \rho (\pi X t)^{3/2}} \exp \left(\frac{-r^2}{4 X t} \right). \quad (6)$$

Here ΔE_v is the vibrational energy deposited by the laser, and c , ρ , X are the specific heat, density, and diffusion constant of the condensate (adsorbent), respectively. In the Basov model the irradiated molecule quickly evaporates (desorbs) due to the rise in temperature; in order to find the activation energy E_a which characterizes this process, they assume that the probability of the molecule leaving the surface is characterized by the expression

$$P(\Delta E_v) = \int_0^\infty \frac{1}{d} \sqrt{\frac{kT}{2\pi m}} \exp \left(\frac{-q}{kT} \right) dt, \quad (7)$$

where m is the molecular mass, d , the intermolecular distance, and q the heat of condensation (adsorption). By substituting the expression for T given by Equation (6) - neglecting the spatial variation - into Equation (7), and using the ansatz $P(E_a) \simeq 1$, E_a is found to be on the order of 1 eV for typical values of the parameters, e.g., $q \sim 1$ Kcal/mole, $X \simeq 10^{-3}$ cm²/sec, and $e \simeq 1$ gm/cm³. In this formulation, where the spatial variation is ignored, one cannot estimate the number of molecules in the local environment surrounding the "hot" molecule which also get off the surface due to having their temperatures raised above the minimum required for evaporation (desorption). To obtain an order-of-magnitude estimate of the quantity - which characterizes the isotopic selectivity in this model - we assume that a minimum temperature $T_{\min}$, corresponding to the heat of vaporization, is required for desorption. Then, using Equation (6) it is straightforward to show that the maximum surface area which desorbs is given by

$$(\pi r^2)_{\max} = \frac{3}{2e} \left(\frac{\Delta E_v}{c \rho T_m} \right)^{2/3}. \quad (8)$$

For example, assuming $c \simeq 1 \text{ cal/gm}^\circ \text{C}$, $\rho \simeq 1 \text{ gm/cm}^3$, $kT_{\min} \simeq 10 \text{ Kcal/mole}$ (heat of vaporization of water), and $\Delta E_v \simeq 3800 \text{ cm}^{-1}$ corresponding to the symmetric stretch of HDO, then $(\pi r^2)_{\max} \simeq 10^{-14} \text{ cm}^2$, and taking the "size" of a water molecule on the surface $\simeq 10 \text{ \AA}^2$, we find that approximately one in ten molecules leaving the surface is deuterated. Of course, this model is very crude, but it does indicate how one may incorporate surface scrambling effects in a more realistic theory.

B. Qualitative Theoretical Considerations

The quantum-theory of gas atom-clean surface collisions at thermal equilibrium (quantum theory of thermal accommodation), dates back to the work of Lennard-Jones and co-workers in the 1930's. In a long series of papers⁽⁴⁰⁾, they have developed a model in which an incoming gas atom interacts with a single surface atom via a Morse potential. Using first order perturbation theory, they calculate the probability of a transition in which the gas atom gives up its kinetic energy to a single lattice phonon, and is thus accommodated on the surface. This basic implication of the Lennard-Jones model, i.e., that the amount of kinetic energy which can be accommodated by the lattice is limited to essentially the Debye energy, has been criticized by Zwanzig⁽⁴¹⁾ who points out that if the mass of the gas atom is comparable to the mass of a surface atom, the interaction is no longer weak, and large amounts of energy (many phonons) can be transferred from a gas to a solid. Using a simple classical model in which the solid is a one-dimensional semi-infinite lattice, and the force constant which characterizes the gas atom-lattice interaction is equal to that of the lat-

tice, Zwanzig calculates that all gas atoms having kinetic energies less than $\sim 25 \times$ the potential well depth will be trapped (accommodated) if the gas atom/surface atom mass ratio is unity, while the critical kinetic energy for trapping is on the order of the well depth when this ratio is one-half. Zwanzig's model has since been generalized by a number of investigators⁽⁴²⁾, however, what is of most concern to us here is the applicability of his basic insight concerning the efficacy of energy transfer in atom-surface collisions to the determination of the probability of deactivation of vibrationally excited molecules upon collision with surfaces. Both Black *et al.*⁽⁴³⁾ and Egorov *et al.*⁽⁴⁴⁾ have recently reported results on the deactivation of N_2 ($v=1$ at 2331 cm^{-1}) on glass; in addition, the former reference includes a table of deactivation data for a variety of diatomic and simple polyatomic gases, and both provide simple theoretical models of the deactivation process. The essential conceptual feature of these models is that deactivation is considered to be a two-stage process, in which accommodation of the translation degree of freedom of the molecule is followed by (irreversible) transfer of its vibrational energy to the surface. The probability of the primary "sticking" process is assumed to be independent of vibrational excitation. On this basis, it is concluded that the small values of the wall deactivation probability, $\gamma \simeq 5 \times 10^{-4} - 2 \times 10^{-3}$, is due primarily to the large difference in energy between the vibrational energy of the molecule and the lattice modes of the substrate. In contrast to these small values of γ for N_2 , the deactivation probability for CO_2 ($v_2=1$ at 667 cm^{-1} and $v_3=1$ at 2349 cm^{-1}) on a variety of surfaces has been measured to be in the range, $\gamma \simeq 0.18 - 0.40$.⁽⁴⁵⁾⁻⁽⁴⁷⁾ In ruminating on this data, the following questions come readily to mind:

1. Why is the γ for CO_2 ($v_2 = 1$) and CO_2 ($v_3 = 1$) essentially identical?
2. Why is γ for CO_2 ($v_3 = 1$) the same on a wide variety of surfaces, e.g., pyrex, teflon, mylar, and brass?
3. Why is γ for N_2 ($v = 1$) so much smaller than that of CO_2 ($v_3 = 1$) and HCl ($v = 1$ at 2886 cm^{-1}) and N_2O ($v_3 = 1$) at 2224 cm^{-1})⁽⁴⁷⁾?

We feel that a key element of the answer to these questions has to do with the extent of surface coverage, θ . In the experiments on N_2 , the coverage was small, $\theta \lesssim 10^{-3}$, and as noted by Black et al., so is the probability of vibrational-vibrational (V-V) energy transfer. On the other hand, we conjecture that in the experiments on CO_2 , HCl , and N_2O , the coverage was high, and the adsorbed molecular layer(s) provided a very effective "impedance match" for V-V transfer to the surface. This, in analogy to the implication from Zwanzig's work that the energy transfer between gas atoms and atoms of equal mass adsorbed on a surface should be much more effective than energy transfer between the gas and a bare surface. The hypothesis of a surface film also provides an explanation for lack of dependence of γ on surface type. We note that an alternate explanation of vibrational energy transfer at gas-solid surfaces has been suggested by Hunter⁽⁴⁸⁾. Hunter's is a "bare-surface" theory in which vibrational-translational (V-T energy transfer) takes place during surface migration of the adsorbed molecule. He has been able to obtain agreement with his experimental results on CO_2 and N_2O deactivation by assuming that, although the gases were excited in their v_3 modes, because of fast intramolecular equilibration effectively all the molecules hitting the surface were in their lowest energy (v_2) mode. Further experimental work which carefully takes into account the nature of the surface and the vibrational state of the molecule is needed to test the importance of

this mechanism; we consider some experiments which are relevant to the possibility of isotope separation in the next section. We conclude this discussion by remarking on the "richness" of the molecule-surface interaction. Assume a vibrationally excited molecule is incident on a surface. The molecule may be specularly reflected, with or without losing its vibrational excitation. If the molecule sticks, it can give up its energy by V-V transfer to adsorbed molecules and/or optical phonons, or by V-T transfer to the local environment or in surface migration. Similar remarks apply to possible interactions following the vibrational excitation of a molecule which is already adsorbed on a surface.

C. Experiments

To test the relevance of these ideas to isotope separation we propose the following (conceptually) simple experiments:

a. DCl/HCl

We excite DCl ($v=1$) in the gas phase in a gas-solid system having the natural (D/H) concentration, and measure what comes off the surface as a function of coverage. We do a similar experiment exciting HCl ($v=1$) instead. Because of the large isotope shift, we expect DCl ($v=1$) $\rightarrow$ HCl ($v=0$) V-V transfer to be relatively inefficient.

b. HCl³⁵/HCl³⁷

In the gas phase there is very efficient V-V transfer between these isotopes. Again, we excite one isotope in the gas phase and see what comes off.

c. ¹²CO₂/¹³CO₂ or HDO/H₂O or NH₂D/NH₃

Similar to a. and b., but here we attempt to probe the influence of the polyatomic nature of the molecules.

5.0 SELECTIVE TWO-STEP (STS) PHOTODISSOCIATION OF NH_2D

Nitrogen isotope separation by selective two-step photodissociation of ammonia was first accomplished by Ambartsumian *et al.*⁽⁴⁹⁾. They irradiated a 1:1 mixture of $^{14}\text{NH}_3$ and $^{15}\text{NH}_3$ with the pressure broadened P (16) line of a CO_2 laser and ultraviolet light at 220.5nm, thus inducing the sequential transitions $(\tilde{X}, v'' = 0) \rightarrow (\tilde{X}, v'' = 1)$ and $(\tilde{X}, v'' = 1) \rightarrow (\tilde{A}, v' = 0)$ of $^{15}\text{NH}_3$, and enriching ^{15}N via pre-dissociation from $\tilde{A}$. (The vibrational transitions are in the v_2 mode.) For NH_2D , sufficient spectroscopic and photochemical information is not available (at least to the author) to predict with assurance the probability of efficient deuterium separation using the STS photodissociation technique. Therefore, we confine ourselves to the following observations:

1. A CO_2 laser can be tuned to several v_2 transitions in NH_2D ⁽⁵⁰⁾, and relevant information concerning ground state vibration-inversion-rotation energy levels is available⁽⁵¹⁾.
2. Because of the comparatively large $\text{NH}_2\text{D}/\text{NH}_3$ isotope shift, e.g., $(0^- \rightarrow 1^+) \simeq 874 \text{ cm}^{-1}$ in NH_2D and $\simeq 932 \text{ cm}^{-1}$ in NH_3 , loss of selectivity due to V-V energy transfer should be slow compared with $^{15}\text{NH}_3/^{14}\text{NH}_3$.
3. As emphasized recently by Letokhov and Moore⁽⁵²⁾, a necessary condition for STS photodissociation is that the shift of the electronic absorption band due to infrared absorption be much greater than the red wing edge of this band. Douglas has investigated predissociation in the $\tilde{A}-\tilde{X}$ bands of ND_3 ⁽⁵³⁾; his work shows that the bands are well separated, and there is an initial decrease of diffuseness in going from 0-0 to 0-1, after which the band width remains fairly constant. The situation with regard to NH_2D is not known, and more data on UV spectroscopy of this molecule is needed.

4. Ambartzumian et al.⁽⁴⁹⁾ have hypothesized that the process of photochemical decomposition of ammonia into nitrogen and hydrogen comes about as a result of a series of radical reactions. In analogy with their work, we conjecture that predissociation of NH_2D might lead to the following:

or a "net" of: $2 \text{NH}_2\text{D} \rightarrow 2 \text{HD} + \text{H}_2 + \text{N}_2$.

5. Turning for a moment to the "back-end" of the separation cycle, ammonia is attractive for reflux because the $\text{NH}_3/\text{H}_2\text{O}$ exchange equilibrium constant is essentially unity, and, unlike formaldehyde, no catalyst is required. However, like formaldehyde, ammonia is highly soluble in water, and therefore a stripping section would be necessary to remove the ammonia from the water before discharging it.

6. Besides ammonia, there are other candidates for deuterium enrichment via STS photodissociation; we mention, in particular, the study by Jensen and Lyman⁽⁵⁴⁾ of the "natural choice", i.e., H_2O .

Finally, a few words about multiple-photon dissociation processes. We have previously discussed⁽¹⁾ the work of Welge and co-workers⁽⁵⁵⁾ and Koren et al.⁽⁵⁶⁾ on high intensity CO_2 laser dissociation of NH_3 and HDCO , respectively. While we would agree that their intrinsic interest (and possible applicability to uranium isotope separation) justifies the efforts of the many groups working in this area, it is important to note that a factor of $\sim 10^4$ improvement in currently attainable quantum efficiencies will be necessary to make these schemes economically viable for deuterium separation.

6.0 ECONOMICS

In terms of research dollars, man-hours, publications in scientific journals and the popular press, etc., etc., laser isotope separation (LIS) of deuterium is a poor relation of uranium, especially in the U.S. As we pointed out in reference (1), this is, to a certain extent, justified by the economic facts of life. That is, using current technologies U-235 is much more expensive than D_2O , and thus the constraints on LIS of D_2O are correspondingly more severe⁽⁵⁷⁾. While favorable projections of the operating and capital costs of laser separated U-235 have been presented by ERDA and Exxon-Nuclear, detailed process engineering calculations in support of these estimates are not available in the open literature. This is not surprising, since both the atomic and molecular U-235 separation processes are still in the laboratory stage, and much basic physics and chemistry, not to mention development work, remains to be done before an educated guess for, let us say, the unit cost of laser separative work, can be made. The much smaller laser deuterium separation community is in much the same boat. Most of the effort in this area has been devoted to an investigation of the usefulness of formaldehyde photopredissociation in the UV⁽⁵⁸⁾. Although this work does not fall within the scope of this study, we note that a necessary condition for its economic viability is the development of tunable, narrow linewidth ($\Delta\lambda \lesssim 0.01 \text{ \AA}$) efficient ($\gtrsim 1\%$) UV lasers of high average power ($\gtrsim 1$ kilowatt per ton of D_2O per year). For example, Marling⁽⁵⁸⁾ calculates that the laser energy cost of this process, assuming a 1% efficient laser, a single-step enrichment factor of 3300 and the availability of electrical power at 10 mills per kwh, is approximately \$25/kg D_2O . Since current single stage enrichments and efficiencies are at

least an order of magnitude smaller, and 10 mills per kwh electricity is a pleasant memory, much remains to be done. Beyond this, the "back-end" of this process is also non-trivial, e.g., we have previously mentioned the difficulties involved in refluxing depleted formaldehyde by exchange with water⁽¹⁾, and the necessity of including a stripping section to remove formaldehyde from water before discharge. While one is tempted to agree with Edward Teller's assertion that (because of the large isotope shifts) "it should be possible to separate deuterium with one's bare hands", to do so economically is a formidable challenge.

7.0 CONCLUSION

This study represents a preliminary assessment of the prospects for heavy water production via laser isotope separation. We believe that a "case" for cheap heavy water has been established, and several potentially rewarding areas for further research have been identified. One should bear in mind that, like gaseous diffusion, the G-S process for separating D_2O is a well-developed technology, and its current dominant position is soundly based⁽⁵⁹⁾. Nevertheless, we feel that the toxicity, corrosiveness, and energy-intensity of the GS process justifies a continuing effort to develop new methods, and that any possibility for dramatic decrease in the cost of D_2O may well lie in the development of new and existing laser techniques.

APPENDIX A. URANIUM FEED AND SEPARATIVE WORK REQUIREMENTS
OF A PWR ASSUMING NO URANIUM RECYCLE

The 159.3 ST of Natural U_3O_8 required for annual makeup in a PWR are first converted to 121.91 MT of UF_6 containing 0.711% U-235 to serve as feed for the enrichment process. Assuming uranium recycle, 24 MT of uranium containing 0.83% U-235 is available as UF_6 after reprocessing of the spent fuel. Using material balance⁽⁶⁰⁾, we find that the feed-to-product ratio, F/P, characterizing the fresh fuel and recycle fuel streams are ~ 5.89 and 4.76 , respectively. Then, of the total enrichment plant output of 25.72 MT of uranium enriched to 3.21%, $24/4.76 = 5.04$ MT comes from reprocessed fuel. If uranium is not recycled, then $5.89 \times 5.04 \simeq 29.69$ MT of UF_6 or $\simeq 37.33$ ST of additional U_3O_8 would be required.

The total annual amount of separative work assuming uranium recycle is 120 MT SWU. Of this, ~ 99.2 MT SWU is required for the fresh fuel ($S/P \simeq 4.8$), and $120 - 99.2 = 20.8$ for the reprocessed fuel. On the other hand, if there is no recycle the amount of separative work required for the additional is $\sim 4.8 \times 5 = 24$ MT SWU.

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6. Although D_2O has a very small neutron capture cross section as compared with H_2O , its slowing-down length is about twice as great, and this would dictate the need for a larger, thicker, and costlier pressure vessel than ordinary water reactors of comparable output. The Canadians instead have evolved an ingenious alternate design in which the fuel and coolant is contained in a system of individual pressure tubes (390 in each of the 540 MWe Pickering reactors) which is separate from the D_2O moderator. The latter is contained in a cylindrical steel vessel, called the calandria, with the pressure tubes through it parallel to its axis, which is horizontal. The pressure tubes are located concentrically inside the calandria tubes, their outside diameter being about 1.5 cm less than the inside diameter of the calandria tubes. The sealed annulus between the two tubes is filled with nitrogen gas as a thermal insulation between the high pressure (1280 psia), high temperature coolant ($249^{\circ}C$ at inlet and $293^{\circ}C$ at outlet), and the atmospheric pressure, low temperature ($68^{\circ}C$ maximum) moderator. This separation of moderator and coolant is a basic feature of the CANDU design. The safety implications of this arrangement will be discussed in Section 2.0-B.
7. Although a HWR produces more plutonium per kilogram of natural uranium consumed (see Table II) than a LWR, there is at present no economic incentive to recover this plutonium and the small amount of U-235 ($\sim 0.2\%$) in the spent fuel. Thus the CANDU system operates efficiently on a "once-through" cycle. This has important political implications (see Section 2.0-C). On the other hand, LWR have a high U-235 content ($\sim 0.83\%$) in their spent fuel, and therefore

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54. R. J. Jensen and J. L. Lyman, "Laser Induced Recovery of Deuterium or Tritium from Water", Proc. Second European Elec.-Opt. Conf., Montreux, Switzerland, April 1974.
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56. G. Koren, V. P. Oppenheim, D. Tal, M. Okon, and R. Weil, Appl. Phys. Lett. 29, 40 (1976).
57. The cost of enriched uranium is given by

$$C_P = C_F \frac{F}{P} + C_S \frac{S}{P}$$

where C_P is the unit cost of uranium containing x_P weight fraction U-235, C_F is the unit cost of natural uranium in

the form of UF_6 , C_S is the unit cost of separative work (SWU), and F/P , S/P are the feed-to-product and separative work-to-product ratios. For a gaseous diffusion plant producing reactor grade ($x_p = 3\%$) uranium with a tails assay of 0.2% from natural uranium feed, $F/P = 5.479$ and $S/P = 4.306$ (see reference (9), p. 159), and assuming, $C_F \approx \$100/\text{kg U}$ and $C_S = \$100/\text{kg SWU}$, $C_P \approx \$978.5/\text{kg 3\% U-235}$ or $\$32.60/\text{gm U-235} \approx \$7665/\text{mole U-235}$.

On the other hand, D_2O at $\$200/\text{kg}$ is equivalent to $\$2/\text{mole D}$. (The numbers for C_F , C_S , and D_2O are my "1980" projections.)

58. See J. B. Marling, "Isotope Enrichment of Oxygen-17, Oxygen-18, Carbon-13 and Deuterium by Ion Laser Induced Formaldehyde Photo-Predissociation", Preprint UCRL-78317, June 1976 for a comprehensive discussion of the American work in this area. The parallel Canadian work of J. Vanderleeden and co-workers at Ontario Hydro has not yet been reported in the literature.
59. Over the years many alternate techniques have been considered. See e.g., the report of F. T. Barr and W. P. Drews, Chem. Eng. Progress 56, 49 (1960), which surveys 98 (!) potential processes, and the more recent study by A. I. Miller and H. K. Rae, "The Search for an Improved Heavy Water Process", Chemistry in Canada 27, 25 (1975). While the GS process is relatively energy intensive, requiring ~ 0.65 MW(e) and 6 MW(th) per kilogram of product, the fact that the process heat requirements fall within the capability of nuclear steam has thus far minimized its cost escalation.
60. See reference (9), p. 159.