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REPORT ON THE MANUFACTURE OF HEAVY WATER

Short Title of Document or Addressee of Letter

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~~REPORT ON~~ THE MANUFACTURE OF HEAVY WATER

by J. O. Maloney and H. S. Ray
September 18, 1945

The manufacture of plutonium is carried out in a chain-reacting pile by the absorption of slow neutrons in $_{92}U^{238}$. These neutrons are obtained from the fission of $_{92}U^{235}$. The neutrons from this fission, however, are fast neutrons and must be slowed down to suitable velocity in order to be absorbed to produce plutonium and to be effective in carrying on the chain reaction. This moderation of the velocity may be best achieved by collision of the neutrons with materials of nearly equivalent mass. Such materials as hydrogen, deuterium, beryllium, graphite, and heavy water (deuterium oxide) are effective moderators. Most other materials are unsuitable as moderators because they absorb the neutrons and stop the chain reaction. The problems of producing and using the various moderators have been subject to rather extensive study.

Considerable effort has been devoted to the production of heavy water, even though the piles at Hanford, Washington were moderated with graphite and only one experimental pile employing heavy water has been operated. Methods which have been extensively studied include distillation of hydrogen, catalytic exchange between hydrogen and water vapor, dual temperature exchange process using hydrogen sulfide and water, distillation from water, and electrolysis of water. In brief the principles of these processes are as follows:

1. Distillation of hydrogen. Since at the normal boiling point of hydrogen there is a marked difference in vapor pressure between H_2 and HD, and since these materials form perfect solutions, it is possible to separate them by fractional distillation.

2. Catalytic exchange between hydrogen and water vapor. It has been found that when a mixture of H_2, HD, and water vapor is passed over a suitable platinum or nickel catalyst, the deuterium transfers from HD to HDO. This HDO is then absorbed in water. By operation in a series of countercurrent towers with electrolysis of the water between towers the deuterium may be concentrated.

3. Dual temperature exchange process using hydrogen sulfide and water. In this process deuterium is stripped from a water stream by exchange with H_2S at the higher temperature. The deuterium is then removed from the H_2S stream by exchange with water at a lower temperature. By suitable adjustment of operating conditions an enriched water stream may be obtained.

4. Distillation of water. Since there is a small difference in vapor pressure between H_2O and HDO, separation may be achieved by fractional distillation of water.

5. Electrolysis of water. When water is electrolyzed there is a concentration of deuterium in the water. By operating on a countercurrent principle, a concentration of deuterium can be achieved.

Three of these processes have been used in the large scale commercial preparation of heavy water; namely, the distillation of water, catalytic exchange, and electrolysis. The electrolytic method is used only as an

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adjunct to the other two processes to obtain a final concentration of 99.8% D_2O. The Consolidated Mining and Smelting Company, Ltd., of Canada produces about 0.5 ton per month of heavy water by the catalytic exchange process, and E. I. du Pont de Nemours and Company produce about 1 ton per month by the distillation of water. Since the war has ended it has become necessary to evaluate more carefully the desirability of using heavy-water moderated piles. A major factor in such a decision is the cost of isolating the heavy water.

The purpose of this report is to record the present cost of preparing heavy water, to estimate the base cost that could be expected through improvements in the present processes, to estimate the cost of production by methods not yet operated on a full scale, and to draw any general conclusions which might assist in formulating a program for deuterium production.

Conclusions

1. The basic cost information on the four most attractive processes has been assembled and is attached.

2. The processes in the order of increasing cost per ton of heavy water are

Distillation of hydrogen	-	$127,000
Catalytic exchange	-	152,000
Dual temperature-H_2S	-	180,000
Distillation of water	-	570,000

The cost of the hydrogen distillation and the hydrogen sulfide process are estimates. These costs include plant investment charges of 20 per cent per year. The investment is composed of research and development costs, plant construction costs, and operating costs until the plant comes to equilibrium.

3. The cost of preparing heavy water by catalytic exchange can probably be reduced to about $106,000 per ton by decreasing the losses of water vapor. Saving in capital cost could probably be realized in the construction of a new plant; however, it was not possible to make an estimate of this at the present time.

4. At the present stage of development of the catalyst in the catalytic exchange process the hydrogen must be free from carbon monoxide and sulfur compounds. Furthermore water has to be decomposed to hydrogen and oxygen at each stage. Consequently this production method must be integrated with a plant making electrolytic hydrogen from water. The only suitable large scale source of such hydrogen in this continent known to the authors is in Canada. The economical development of a similar process in the United States does not appear feasible, since the production of hydrogen by other methods furnishes more than is needed.

5. The cost of preparing heavy water by water distillation can probably be reduced to about $255,000 per ton by improved sealing of the plates in the large towers.

6. The operating cost/ton of D_2O exclusive of plant investment charges for the two processes now in operation are

Catalytic exchange	$ 52,000
Distillation of water -	$355,000

7. The decisions to employ the catalytic exchange and water distillation processes for commercial production were justified in the light of the critical material shortages and the uncertainties existing at the time decisions had to be made.

8. If existing compressors from closed down ammonia plants could be made available for the hydrogen distillation process, the operating charges would be reduced by 15 to 20 per cent.

9. The principal uncertainties in the hydrogen distillation process were

1. The amount of solid nitrogen deposition in the equipment.
2. The behavior of materials of construction at 20° K.

10. The principal uncertainty in the hydrogen sulfide process was the corrosion rate of materials of construction.

Recommendations

1. It is recommended that this report be used as a guide if additional facilities for the production of heavy water are planned.

2. It is recommended that the hydrogen distillation method be given primary consideration if additional production of deuterium up to 10 tons/month is required. For quantities greater than 10 tons/month the dual temperature hydrogen sulfide method should receive primary consideration.

3. It is recommended that some organization be made responsible for following all developments in the production of heavy water and for investigating the uncertainties which exist in the more attractive but undeveloped processes.

4. It is recommended that consideration be given to obtaining the hydrogen for the hydrogen distillation from either the electrolysis of water or the natural gas-steam reaction. There exist enough plants in the United States operating on the natural gas-steam reaction to furnish between 5 and 10 tons of D_2O per month.

5. It is recommended that the seized German information on heavy water manufacture be released to the individuals working on heavy water isolation.

6. It is recommended that the modifications of the existing and proposed plants, as detailed in the forthcoming engineering report on the deuterium separation methods, be considered if additional facilities are planned.

Economic Considerations in the Isolation of Deuterium by Four Methods

	Distillation of Hydrogen plus Electrolytic Finishing	Catalytic Exchange plus Electrolytic Finishing	Dual Temperature Method-Hydrogen Sulfide Process plus Electrolytic Finishing	Distillation of H_2 plus Electrolytic Finishing
	(Estimated)		(Estimated)	
COST				
Research and Development				
To date	$ 42,000	$ 470,000	$ 25,000	$ 65,000
To complete, total	$ 300,000	Completed	$ 200,000	Completed
Investment in Plant				
Total	$12,000,000 (1 or 2 plants)	$2,350,000 (1 plant)	$30,600,000 (1 plant)	$14,500,000 (3 plants)
Investment/ton	$ 5,000,000	$4,700,000	$6,120,000	$13,700,000
Operating Cost/ ton				
Present) includes fixed	-	$ 152,000	-	$ 570,000
Potential) charges	$ 127,000	$ 106,000	$ 180,000	$ 255,000
Present, exclusive of fixed charges	-	$ 52,000	-	$ 355,000
PRODUCTION				
Tons/month, present	-	0.5	-	1.06
Tons/month, potential	2.5	0.725	5.00	2.40
PERCENT RECOVERY OF D_2O IN FEED				
Present	-	43	-	2.5
Potential	90 +	62	17	5.2
PERSONNEL				
Research and Development (man-months)				
To date	60	630	70	125
To complete, total	300	Completed	240	Completed
Construction (man-months)				
To date	None	2250 (1 plant) Completed	None	13,750 (3 plants) Completed
To complete	10,000 (1 or 2 plants)		33,500 (1 plant)	
Per ton of product	4,000	4500	6,700	12,900
Operation				
Man-months/month	90	30	110	90
Man-months/ton	36	60	24	84
TIME TO COME TO EQUILIBRIUM, DAYS	0.20	240	60	75-100

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Distribution of Effort and Wages for Research and Development in the Isolation of Deuterium by Four Methods

Type of Service	Estimates of Additional Expenditures for Proposed Processes						Approximations for Processes Now Used					
	Hydrogen Distillation			Hydrogen Sulfide			Catalytic Exchange			Water Distillation		
	man-months	rate/month	amount	man-months	rate/month	amount	man-months	rate/month	amount	man-months	rate/month	amount
Director	25	$600	$ 15,000	24	$600	$14,400	24	$600	$14,400	10	$400	$ 4,000
Assistant Directors	50	500	25,000	24	400	9,600	72	400	28,800			
Research Men	225	330	74,300	192	300	57,600	432	300	129,600	115	300	34,500
Others	---		5,000				102	200	20,400			
TOTAL	300		$120,000	240		$81,600	630		$193,000	125		$38,000

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