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CANADIAN HEAVY WATER PRODUCTION

by

A. DAHLINGER, W.E. LOCKERBY and H.K. RAE

**Paper IAEA-CN-36/183 presented at the IAEA International Conference on
Nuclear Power and its Fuel Cycle, Salzburg, Austria, 2-13 May 1977**

Chalk River Nuclear Laboratories

Chalk River, Ontario

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Production d'eau lourde au Canada*

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Résumé

Le présent mémoire passe en revue l'expérience acquise au Canada dans la production de l'eau lourde; il présente une projection des approvisionnements futurs en fonction de la demande en électricité prévue à long terme et il met en lumière les principaux domaines de perfectionnement qui forment l'essentiel de notre programme de recherche et de développement concernant les procédés de fabrication de l'eau lourde. Six usines d'eau lourde canadiennes ayant une capacité nominale totale de 4000 Mg/a sont actuellement en service ou en construction. Toutes ces échange d'eau et de sulfure d'hydrogène. Les premiers problèmes de fonctionnement ont été résolus et les facteurs de capacité annuels des usines canadiennes ont dépassé 70% tandis que les taux de production à court terme ont égalé les taux calculés. Voici quelques domaines où des perfectionnements seraient souhaitables: augmentation des taux de production en optimisant le contrôle des mousses afin d'accroître le rendement des plateaux perforés et d'obtenir de plus grands débits; réduction de l'inaptitude due aux dépôts de pyrite (FeS_2) et de soufre (entre 5 et 10%); amélioration du contrôle des circuits et optimisation des conditions de fonctionnement en ayant recours à des simulations mathématiques du profil détaillé du deutérium dans l'ensemble de chaque usine. D'autres procédés étudiés, qui semblent prometteurs, sont celui de l'échange eau-hydrogène et celui de l'échange hydrogène-amine. Même si ces nouveaux procédés en viennent à concurrencer sérieusement le procédé GS, il est probable que ce dernier restera le plus important au cours des 10 à 20 prochaines années. Par rapport à la demande en électricité à long terme, ce programme révèle un équilibre raisonnable entre les besoins en eau lourde et les approvisionnements disponibles. Enfin, le programme CANDU ne sera pas entravé par des pénuries d'eau lourde.

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ABSTRACT

The paper reviews Canadian experience in the production of heavy water, presents a long-term supply projection, relates this projection to the anticipated long-term electrical energy demand, and highlights principal areas for further improvement that form the bulk of our research and development program on heavy water processes. Six Canadian heavy-water plants with a total design capacity of 4000 Mg/a are in operation or under construction. All use the Girdler-Sulphide (GS) process, which is based on deuterium exchange between water and hydrogen sulphide. Early operating problems have been overcome and the plants have demonstrated annual capacity factors in excess of 70%, with short-term production rates equal to design rates. Areas for further improvement are: to increase production rates by optimizing the control of foaming to give both higher sieve tray efficiency and higher flow rates, to reduce the incapacity due to deposition of pyrite (FeS_2) and sulphur (between 5 and 10%), and to improve process control and optimization of operating conditions by the application of mathematical simulations of the detailed deuterium profile throughout each plant. Other processes being studied, which look potentially attractive, are the hydrogen-water exchange and the hydrogen-amine exchange. Even if they become successful competitors to the GS process, the latter is likely to remain the dominant production method for the next 10 to 20 years. This program, when related to the long-term electricity demand, indicates that heavy-water supply and demand are in reasonable balance and that the CANDU program will not be inhibited because of shortages of this commodity.

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Introduction

The purpose of this paper is to review Canadian experience in the production of heavy water, to relate the long-term projected supply and demand, and to highlight principal areas where we expect improvements as a result of our research and development program.

Table I

CANADIAN HEAVY WATER PLANTS

<u>Plant</u>	<u>Owner/Operator</u>	<u>Design Production Rate kg/h</u>	<u>Date of First Production</u>
Port Hawkesbury	AECL	48.3	1970
Bruce "A"	Ontario Hydro	96.6	1973
Glace Bay	AECL	54.4	1976
Bruce "B"	Ontario Hydro	96.6	1978
Bruce "D"	Ontario Hydro	96.6	1981
La Prade	AECL	94.6	1981

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IAEA-CN-36/180	AECL-5708	IAEA-CN-36/197	AECL-5713
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Heavy water is a vital component of the CANDU¹ system. The success of this system is dependent on a reliable supply of heavy water at an acceptable cost. There are now three heavy-water plants in operation in Canada and another three under construction. Table I lists these plants and notes the status and capacity of each. By 1982 Canada will have a total nominal capacity of 4,000 Mg/a.

Process Description

All of the Canadian plants use the Girdler-Sulfide (GS) process which is based on the deuterium exchange reaction between water and hydrogen sulfide. In this process, which is shown schematically in Figure 1, heavy water is concentrated by contacting water and hydrogen sulfide counter-currently in large towers. Feedwater enters a cold section of the tower and picks up deuterium from the gas stream. This enriched water flows through a hot section of the tower where its deuterium is transferred to the recirculating gas; this water then leaves with a deuterium content 20% less than the feed. Maximum enrichment occurs at the junction of the hot and cold zones from where an enriched stream is fed to a higher stage and replaced by a depleted stream from that stage. The process streams connecting one stage with the next higher stage may be gas, liquid, or both. The process is repeated through several stages until about 20% heavy water is reached. Final enrichment to reactor grade is achieved by water distillation.

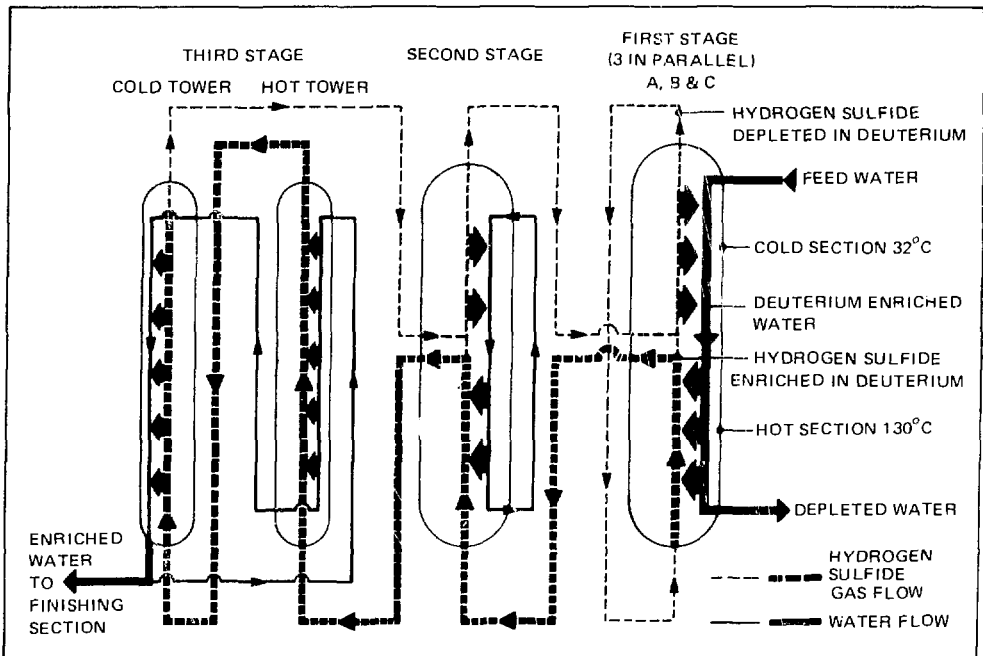


Fig.1. Schematic Flow Sheet of GS Enricher

¹Canada Deuterium Uranium

To enrich deuterium from its concentration of 0.015% in natural water to the reactor grade concentration of 99.8% requires an increase in the deuterium-to-hydrogen ratio by a factor of over 3 million. The plant volume and energy requirement are consequently large. The total energy consumption (expressed as equivalent thermal energy) is about 9 MWh/kg of heavy water.

Figure 2 is a view of the Port Hawkesbury plant showing the three parallel first-stage towers, a single second-stage tower and the cold/hot pair of the third stage. The first-stage towers are nearly 100 m tall and are amongst the largest pressure vessels in the world.

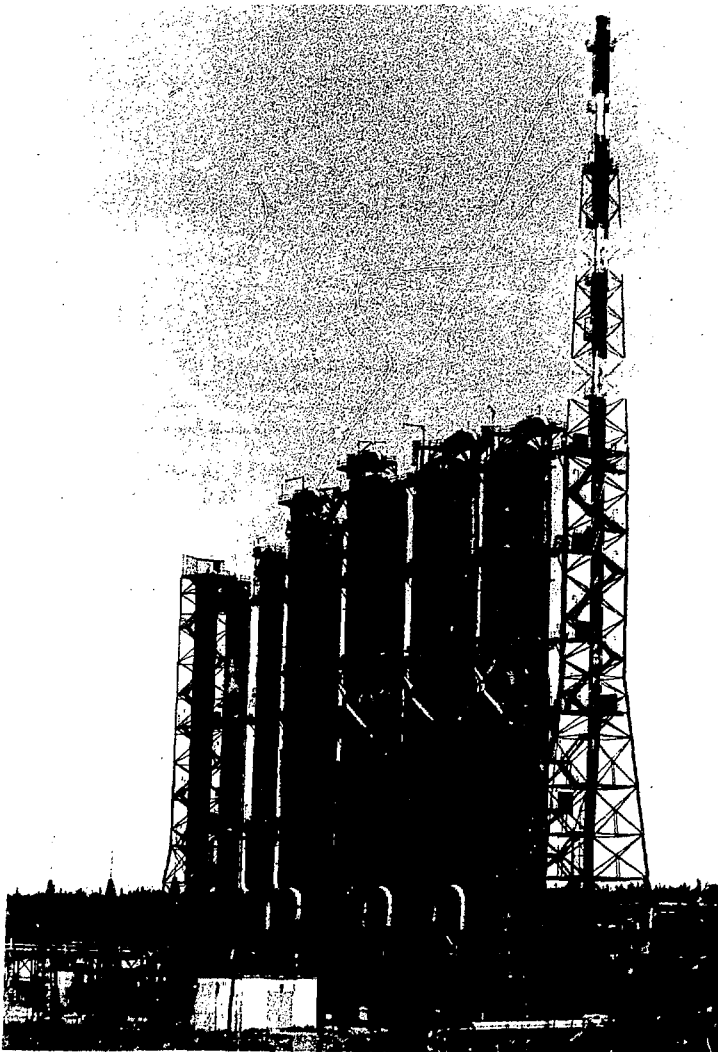


Fig.2. View of Port Hawkesbury Heavy Water Plant

Production Experience

Although the GS process was well established in the U.S.A. the transfer of technology and particularly the scale-up in tower size spawned a variety of early operating problems in the Canadian plants. However, the plants are now approaching mature operation. For example, the average production at the Bruce "A" plant over the first eight months of 1976 was 94% of the hourly design rate, and in fact equal to the nominal annual design capacity of 800 Mg/a.

Figure 3 shows the maturing experience of the Port Hawkesbury plant and the two units E1 and E2 which comprise the Bruce "A" plant. Each successive unit reached maturity more quickly than the previous one. This can be attributed, in part, to the fact that experience gained in commissioning one unit was applied to the next, and to a large research and development program.

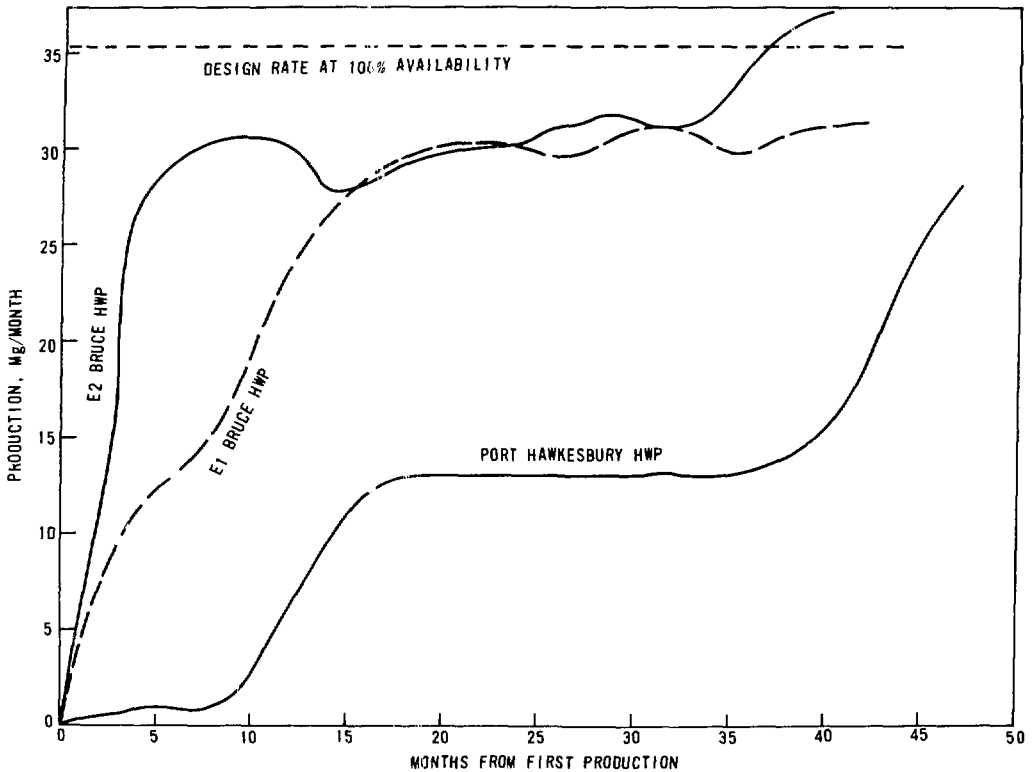


Fig.3. Maturing Experience of Heavy Water Plants

Projected Supply and Demand

The Canadian heavy-water supply involves:

- Ontario Hydro, a Crown corporation of the Province of Ontario which is responsible for the acquisition and operation of heavy-water plants to satisfy the demands of all CANDU reactors in the province.

- Atomic Energy of Canada Limited (AECL), a federal Crown corporation which is responsible for the acquisition and operation of heavy-water plants to satisfy the demands of CANDU reactors built in provinces of Canada other than Ontario and in foreign countries.

Figure 4 shows the projected demand for D₂O for CANDU reactors until the year 2000. The projections, which have been smoothed, are based on forecasts of installed nuclear generating capacity in Canada by the federal government's Department of Energy, Mines and Resources, and in foreign countries by AECL in conjunction with several federal government departments. The projection assumes that an initial inventory of 0.85 Mg per MWe of generating capacity will be required and that operating reactors will require make-up at the rate of 1% of inventory per annum to replace net losses of D₂O. An operating reserve of 800 Mg (AECL and Ontario Hydro together) is also included. This reserve is available to deal with acute situations:

- (a) in operating reactors whereby major loss or downgrading of D₂O may occur,
- (b) in heavy-water plants whereby a plant may be out of service for an extended period.

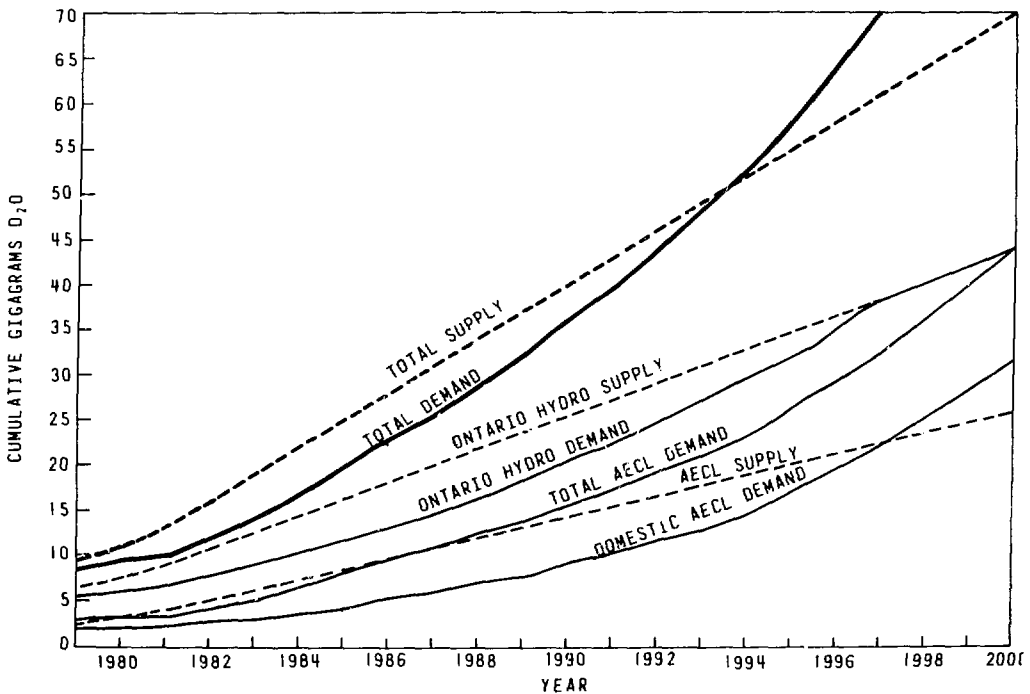


Fig.4. Heavy Water Supply and Demand

Also shown in Figure 4 is the projected supply of D₂O for Canada as a whole, and for Ontario Hydro and AECL separately. The projected production is from plants now operating or under construction and is based on the conservative assumption that average production rate is 70% of the design or

instantaneous production rate. This de-rating of design capacity is necessary to allow for scheduled and unscheduled maintenance, disruptions in steam and power supplies and other factors.

The fact that in reality the demand curve is a series of steps rather than a smooth curve is allowed for by having supply slightly greater than demand (Fig. 4).

Because the nuclear industry is still a young industry with long lead times, it is difficult to forecast accurately the number of reactors which will exist 10 to 20 years from now. The lead time for new heavy-water plants is also long - from 7 to 8 years from the decision to build to the in-service date.

Two points should be noted from Figure 4; first, that heavy-water supply is not a limiting factor in the decision to opt for a CANDU system in the near future; the second, that the cross-over point of the supply and demand curves is far enough in the future to provide time to react to an upturn in demand.

The capital investment in heavy-water plants represents about 5% of the nuclear plant capital investment which it will service. The portion of the capital cost of a new CANDU generating station which is attributed to heavy water has remained at 15-20% and is expected to stay in this range.

Research and Development

A strong industry-wide research and development program has helped to successfully bring Canada's large heavy-water plants through a developmental phase and is now providing a basis for significant gains in production rate in existing plants as well as for improved design for future plants. The three major thrusts of the program are mathematical modeling of plant performance, corrosion and solids deposition, and tray performance.

Complex mathematical models have been developed to simulate the steady-state deuterium profile through each plant and have proved extremely valuable in optimizing plant production. They have also been used to develop alternative flowsheets for future plants which may offer up to 10% more output for the same capital investment. The models are being extended to describe dynamic performance of the plants; this will lead to improved process control.

Nearly one-half of plant shutdown time results from either corrosion problems or from the need to remove deposits of iron sulfide and, to a lesser extent, sulfur from process equipment. A detailed knowledge of the chemistry of the iron-sulfur-water system is being acquired and the factors controlling carbon steel corrosion and deposition are being studied. There is also a continuing program to improve the understanding of all facets of localized corrosion of the high-strength alloys used in the aggressive chemical environment of heavy-water plants.

The deuterium transfer between the gas and liquid in the towers of a heavy-water plant takes place on sieve trays. Figure 5 shows schematically how the gas and liquid are mixed and then separated again. The gas flows through a large number of holes in the tray, mixes with the liquid to form a froth, then disengages from the liquid. Water is supported on the tray by the pressure-drop of the gas passing through the holes. It flows across the

tray, over a weir and, via a downcomer, to the tray below. While one requires a high froth height to provide a large volume of mixed liquid and gas to promote deuterium transfer, it is also necessary to have enough space for the gas and liquid to separate. Similarly, as the liquid flows through the downcomer, there must be sufficient residence time for bubbles to disengage. If there is too much foaming it is necessary to restrict flow rates.

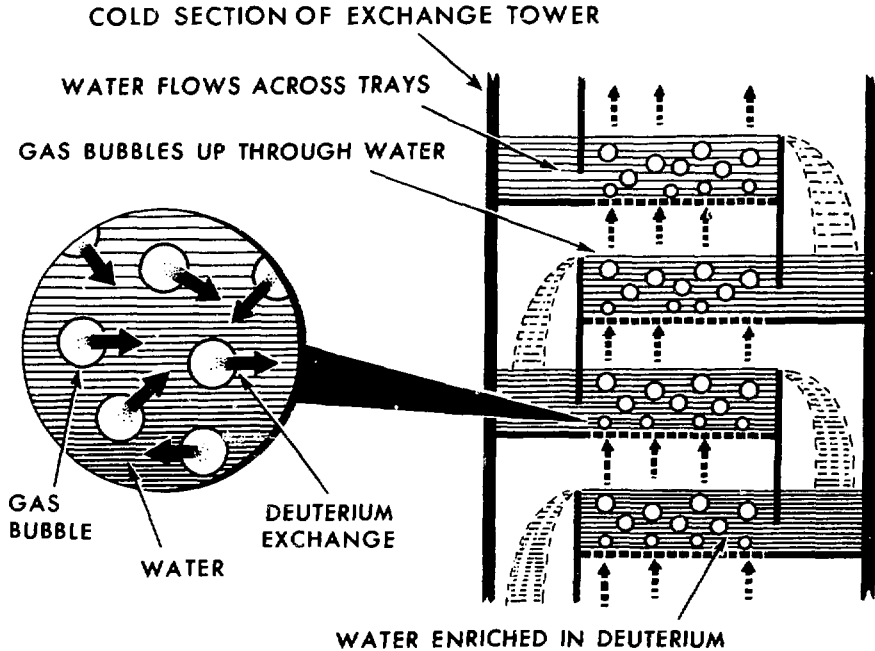


Fig.5. Schematic Sieve Tray Operation

The deuterium transfer rate is related to the liquid flow rate and the tray efficiency which, in turn, depends on the tray design and frothing characteristics of the gas-liquid mixture. The latter can be controlled within limits by the addition of anti-foam agents. It is estimated that deuterium extraction rates can be further increased by up to 10% by optimizing tray performance and froth characteristics. A pilot plant at AECL's Chalk River Nuclear Laboratories and a specially instrumented production tower at the Bruce plant support a program aimed at improving deuterium transfer rates on trays. The large production tower with extensive instrumentation, which is part of a broad collaborative program between Ontario Hydro and AECL, ensures that experimental effects observed in the pilot plant can be duplicated in a production plant. One observation is that a change in the type of anti-foam agent can increase production by as much as 5%.

AECL has evaluated a large number of separation methods for heavy water production. The only potentially attractive water-based process other than the GS process is the water-hydrogen exchange process employing a platinum catalyst. AECL has a small basic research program aimed at improving catalyst performance to the point where this process could become economic. Water-hydrogen exchange combined with water electrolysis in future energy

systems may be economic even at current levels of catalyst activity. Plant sizes, however, would likely be small compared to today's GS plants.

A process for recovering deuterium from natural gas has been proposed by Gulf Research and Development. Because the energy consumption is expected to be high, twice that for the GS process, it is unlikely that this process will be exploited.

Hydrogen-based processes that are attractive include hydrogen distillation, ammonia-hydrogen exchange and amine-hydrogen exchange. A disadvantage of these plants is that their size is limited by the size of hydrogen streams available today. For example, a 1000 Mg per diem ammonia plant can support a heavy water production of only 70 Mg per annum. At this scale of operation these processes are barely competitive with the GS process. We believe that the amine-hydrogen is the more attractive of the hydrogen-based processes and have done extensive development work on it. AECL has also collaborated with Sulzer Bros. with the objective of exploiting this process. As larger hydrogen streams become available, the amine-hydrogen process should become significantly cheaper than the GS process.

In summary, new processes, particularly those based on hydrogen, may be successfully introduced in competition with the GS process but will not have a large impact on heavy-water supply for the next 10 - 20 years.

Conclusion

The GS process has been successfully adapted to meet Canada's large heavy-water requirements. Heavy-water production has become a major industry in Canada and, in terms of committed capacity, represents about 90% of the world total. The experience accumulated to date provides a firm underpinning for the CANDU program, both domestically and for foreign markets. Supply and demand for heavy water are in reasonable balance and there is adequate lead time available for the construction of the plants, as yet uncommitted, which will be required in the future.

Research and development have been important factors in helping heavy-water plants to overcome early operating problems and continue to play a role in the optimizing of plant operations. Though new processes are the subject of research and are being exploited to a limited extent abroad, the GS process will remain the dominant heavy water production method for the next 20 years.



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