

CHAPTER 8

NUCLEAR REACTOR MATERIALS

Nuclear power reactors are characterized by the existence of high radiation fields and by the requirement that some materials possess certain properties with respect to neutron absorption. The radiation fields have a profound effect on the properties of many materials which, by design, must fulfill specific neutronic, structural and transport functions. Although all nuclear reactor materials must meet the specifications of strength and reliability, we will concern ourselves in this chapter primarily with those material aspects which are affected by various radiations and particles associated with nuclear fission and with those materials which fulfill a particular neutronic function.

8.1 RADIATION EFFECTS

In a previous chapter, we described the fission process and the properties of the various emitted radiations and particles. The appearance of these primary radiations and particles serves to initiate numerous and complex cascade effects in the materials. For example, atoms can be dislodged from their positions by the fission fragments while the emitted gamma ray may cause ionization and induce the production of x-rays. A simplified graphical representation of some of these radiations and processes is shown in Fig. 8.1.

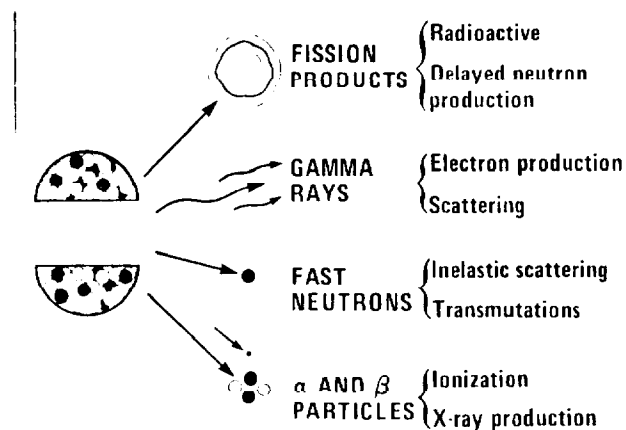


FIG. 8.1: Characterization of radiation sources associated with fission and their secondary effects.

The more fundamental aspects of radiation effects can be described on the atomic scale. In general, the effects of greatest interest can be described by the following groupings:

1. Vacancies or Knock-ons

This process identifies the removal of an atom from its position by the direct interaction of a high energy neutron or a fission fragment.

2. Interstitials

An interstitial is formed when an atom, which is knocked out from its position, comes to rest at some remote point.

3. Ionization

Ionization is caused by the removal of electrons from their electronic shells and has the effect of changing the chemical bonds of molecules.

4. Thermal Spikes

This term identifies localized high temperature domains caused by the deposition of energy from neutrons and fission fragments.

5. Impurity Atoms

The capture of neutrons and nuclear reactions induced by various radiations has the effect of transmuting an atom into an element which is foreign to the material.

Although the above are effects produced on the atomic and nuclear level, a sufficient number of these processes contribute to important macroscopic changes in the material. Some of the more important effects may be identified as follows:

1. Creep

This term is used to describe the continuous and unrecoverable change in material dimensions under constant load.

2. Void Formation

Under the action of a radiation field, microscopic bubbles and pores form in many metals with an accompanying decrease in material density and loss of strength.

3. Gas Formation

Some radiation and particle processes lead to the formation of atoms which, for the localized conditions of temperature and pressure, form a gas.

4. Mechanical Change

Many mechanical properties such as tensile strength, yield stress, thermal resistance, and volume, change considerably under the influence of a radiation field.

Extensive investigations have been carried out on nuclear radiation effects in materials. In some cases these studies have involved irradiation of materials by well-defined single-particle beams produced by accelerators or by radiations

emanating from radioactive isotopic sources. Other radiation effect studies have involved the use of radiation fields existing in a nuclear reactor. In these latter studies, the material specimen of interest would be placed directly in a reactor core and its properties examined after irradiation. Since it is neither possible nor in many cases necessary to identify in detail the radiation field, it is common to consider different types of irradiation conditions as the independent parameter.

The most direct parametric description of the effect of the neutron radiation field in a reactor on a non-fissile material is to consider simply the neutron exposure at the point of interest in the core. This neutron exposure is defined by

$$\text{Neutron exposure} = \iiint \phi(E,t) dE dt \approx \bar{\phi} \Delta t \text{ (neutrons/cm}^2\text{)}, \quad (8.1)$$

where Δt is the exposure time and $\phi(E,t)$ represents the neutron flux; $\bar{\phi}$ is the time and energy averaged neutron flux.

If the specimen of interest is fissile, that is a nuclear fuel, then the appropriate parameter to use should include the fission exposure or the linear power rating. The fission exposure is given in a form similar to the neutron exposure:

$$\text{Fission exposure} = \iiint \nu(E) \Sigma_f(E) \phi(E,t) dE dt \approx \bar{\nu} \bar{\Sigma}_f \bar{\phi} \Delta t \text{ (fissions/cm}^3\text{)}. \quad (8.2)$$

This represents the total number of fissions which have taken place in a unit volume. Here again we have reduced the exact integral expression by the equivalent averages; the symbols ν and Σ_f are used as previously defined.

The linear power rating represents the power produced per unit length (watts/cm) by a fuel element. This expression can be shown to be given by the integration of the thermal conductivity of the fuel, $k(T)$, over the temperature from the fuel surface, T_s , to the centre of the fuel, T_c . That is,

$$\text{Linear power rating} = \int_{T_s}^{T_c} k(T) dT \text{ (Watts/cm)}. \quad (8.3)$$

It has become widely accepted to use the symbol θ as the temperature variable and $\lambda(\theta)$ as the temperature dependent thermal conductivity of the fuel. The linear power rating is therefore more commonly represented by

$$\text{Linear power rating} = \int_{\theta_s}^{\theta_c} \lambda(\theta) d\theta \text{ (watts/cm)}. \quad (8.4)$$

The dimensional distinctions among the three parametric expressions for neutron exposure, fission exposure and power rating are important in a comparative assessment of irradiation effects.

8.2 STRUCTURAL MATERIALS

The in-core structural materials must, in addition to the usual structural characteristics, be viewed in the light of the following requirements:

1. Good Neutron Economy

Structural materials must minimize the parasitic absorption of neutrons. This requirement is dictated by the choice of natural uranium as the nuclear fuel and its attendant low concentration (0.72%) of fissile Uranium-235.

2. Resistance to Radiation

High neutron and gamma radiation fields exist in the reactor core. The materials must retain their structural integrity over the required life time.

3. Compatibility with Heavy Water

The primary consideration is corrosion and the transport of corrosion products under the temperature and pressure conditions existing in the core.

Figure 8.2 shows a photograph of some of the material components as they exist in the reactor core. The horizontal tubes are the calandria tubes which separate the moderator from the coolant; these tubes contain the pressure tubes and the fuel bundles, Fig. 8.3. Note the penetration of the calandria tubes through the end shield at the right of this photograph. The vertical guide-tubes contain the several reactivity-control elements discussed in a preceding chapter. Also shown on this photograph is the calandria and its attachment to the end shield.

The core materials of particular interest are, therefore, those used for fuel sheathing, pressure and calandria tubes, control mechanism tubes and the calandria; the materials used in these components must be examined in the light of the above requirements.

Extensive investigations have been carried out on materials concerning their neutron economy, radiation resistance and compatibility with heavy water. It has been found that alloys of zirconium best meet these requirements. Other materials such as austenitic stainless steel (Type 304), zirconium-niobium alloys, aluminum, and silicon-carbide meet the desirable condition to a lesser extent; reasons of cost, availability and manufacturing limitations may, on occasion, dictate that some of these materials may be used in certain cases.

The only materials used for the fuel sheathing, pressure tubes, calandria tubes and the control mechanism tubes are the zirconium alloys identified by Zircaloy-2 and Zircaloy-4; austenitic stainless steel is used for the calandria. The composition of Zircaloy-2 and Zircaloy-4 is listed in Table 8.1; note that the

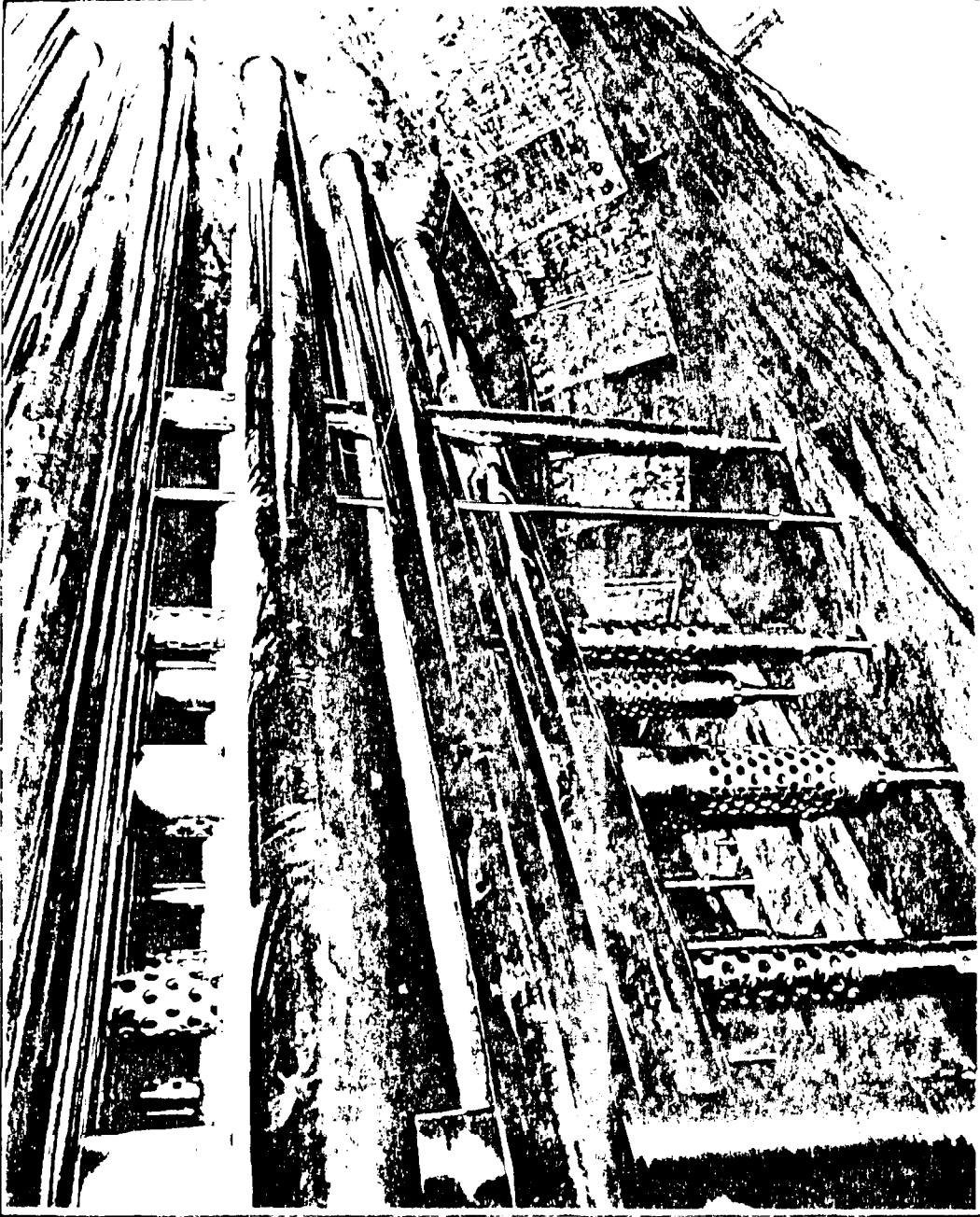


FIG. 8.2: Typical in-core reactivity mechanism guide tubes and calandria tubes.

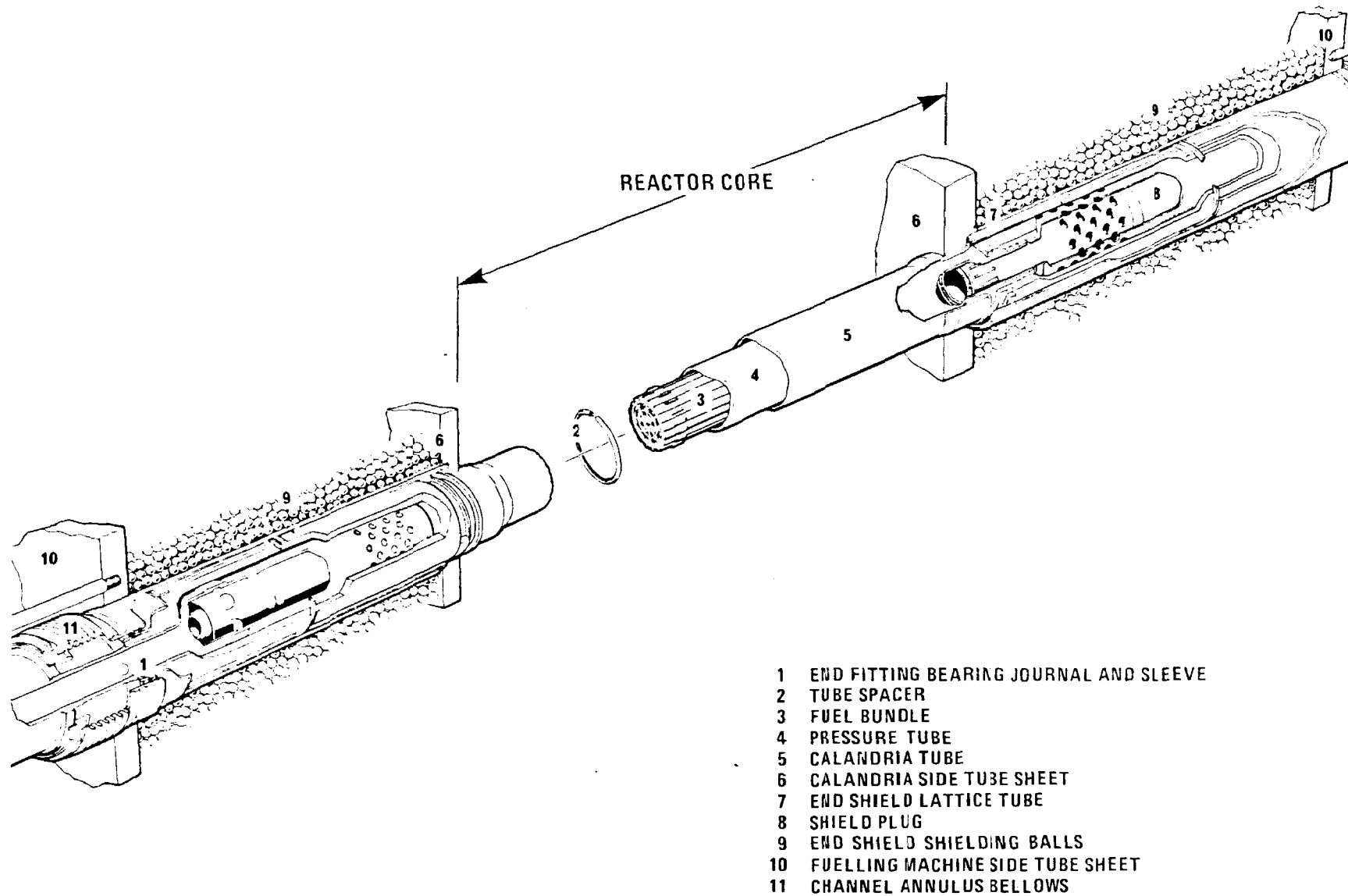


FIG. 8.3: Detail of a CANDU fuel channel.

difference between these two zirconium alloys is primarily in the absence of nickel in Zircaloy-4. This latter alloy has been found to possess a slight corrosion and hydrogen pick-up performance advantage over Zircaloy-4 in a CANDU reactor and is the preferred alloy particularly for fuel sheathing.

	Zircaloy-2	Zircaloy-4
Tin	1.20 - 1.70 wt%	1.20 - 1.70 wt%
Iron	0.07 - 0.20 wt%	0.18 - 0.24 wt%
Chromium	0.05 - 0.15 wt%	0.07 - 0.13 wt%
Nickel	0.03 - 0.08 wt%	-
Carbon	150 - 400 ppm	150 - 400 ppm
Oxygen	900 - 1400 ppm	900 - 1400 ppm
Zirconium	Balance	Balance

TABLE 8.1: Composition of Zircaloy-2 and Zircaloy-4; ppm stands for parts-per-million.

8.3 NUCLEAR FUEL

The nuclear fuel for a CANDU reactor has its beginning as a UO_2 powder produced from ore mined principally at Elliot Lake, Ontario, and Uranium City, Saskatchewan. This UO_2 powder is blended and pressed into cylindrical pellets about 1.5 cm in diameter and 2 cm in length. The polished pellets are subsequently placed end-to-end inside a zircaloy sheath some 50 cm in length. Several of these fuel elements are thereupon joined together by end plates and separated by zircaloy spacers. Fig. 8.4 shows several such fuel bundles associated with specific CANDU reactors.

A number of changes in nuclear fuel design have taken place since the first fuel charge was prepared for the NPD reactor in 1953. These first fuel elements were wire wrapped to provide space between the fuel sheath of adjacent fuel elements and to induce turbulence in the coolant to aid in heat transfer. The more recent fuel elements possess short bearing pads and spacers both of which are attached by brazing; the spacers are fastened at an angle to prevent interlocking. The diameter of the fuel elements and the number of pencils in a fuel bundle also changed with time as a better understanding of the effects of various parameters on performance was gained. Fig. 8.5 provides a graphical representation of the various fuel bundle cross sections.

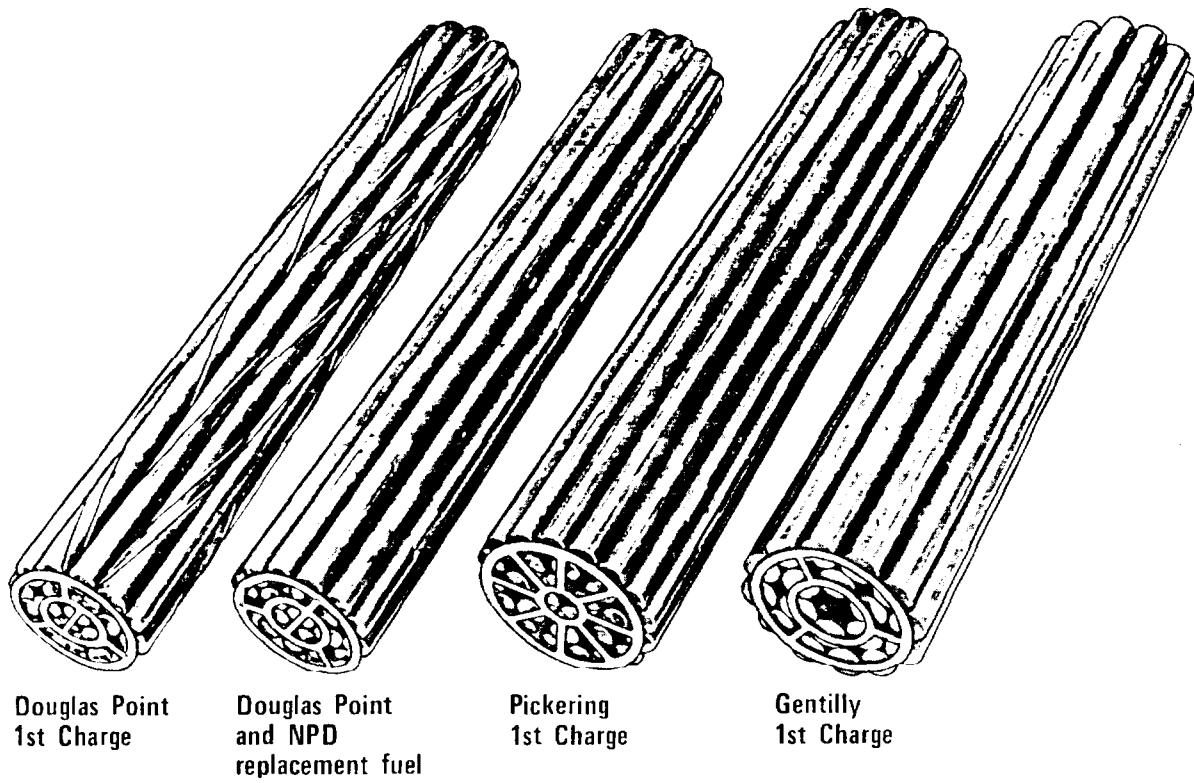


FIG. 8.4: Fuel bundles used in CANDU reactors.

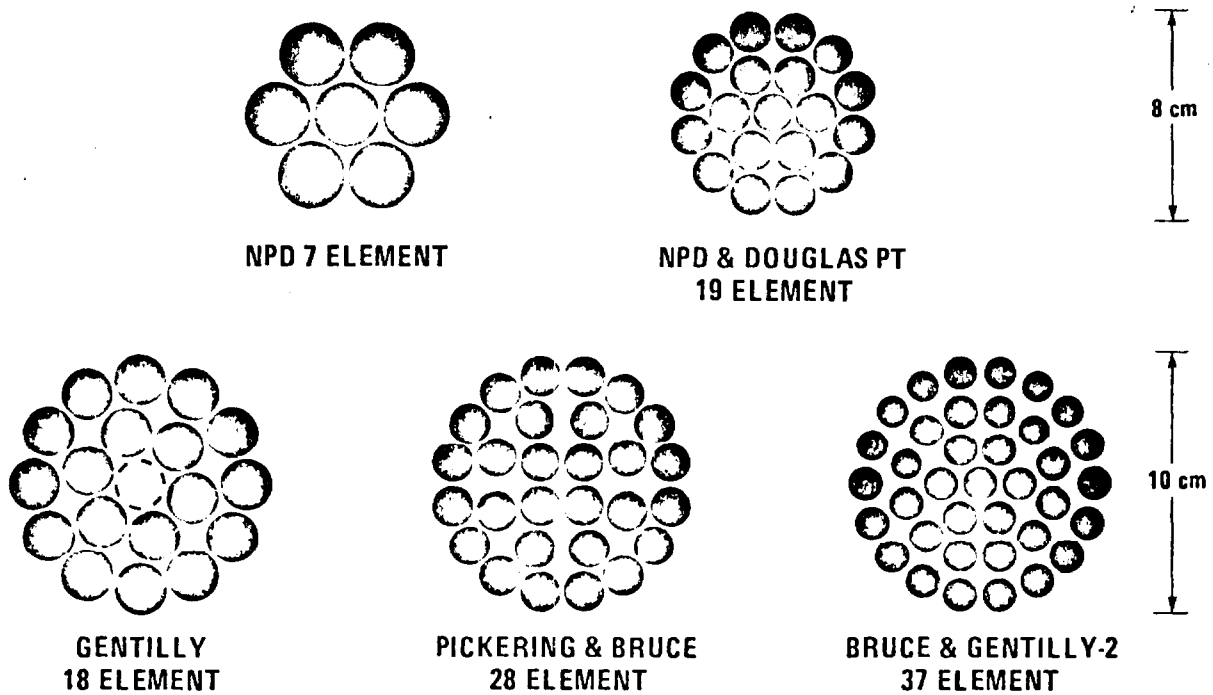


FIG. 8.5: CANDU fuel bundle cross sections

In addition to the fission process occurring in the nuclear fuel, there exist several additional and important processes. This includes processes such as swelling and cracking. In the photographs of Fig. 8.6 we show the effect of various central temperatures and heat ratings on irradiated UO_2 fuel pellets. Since UO_2 is a ceramic, it has a very low strength in tension and hence cracks during expansion and contraction.

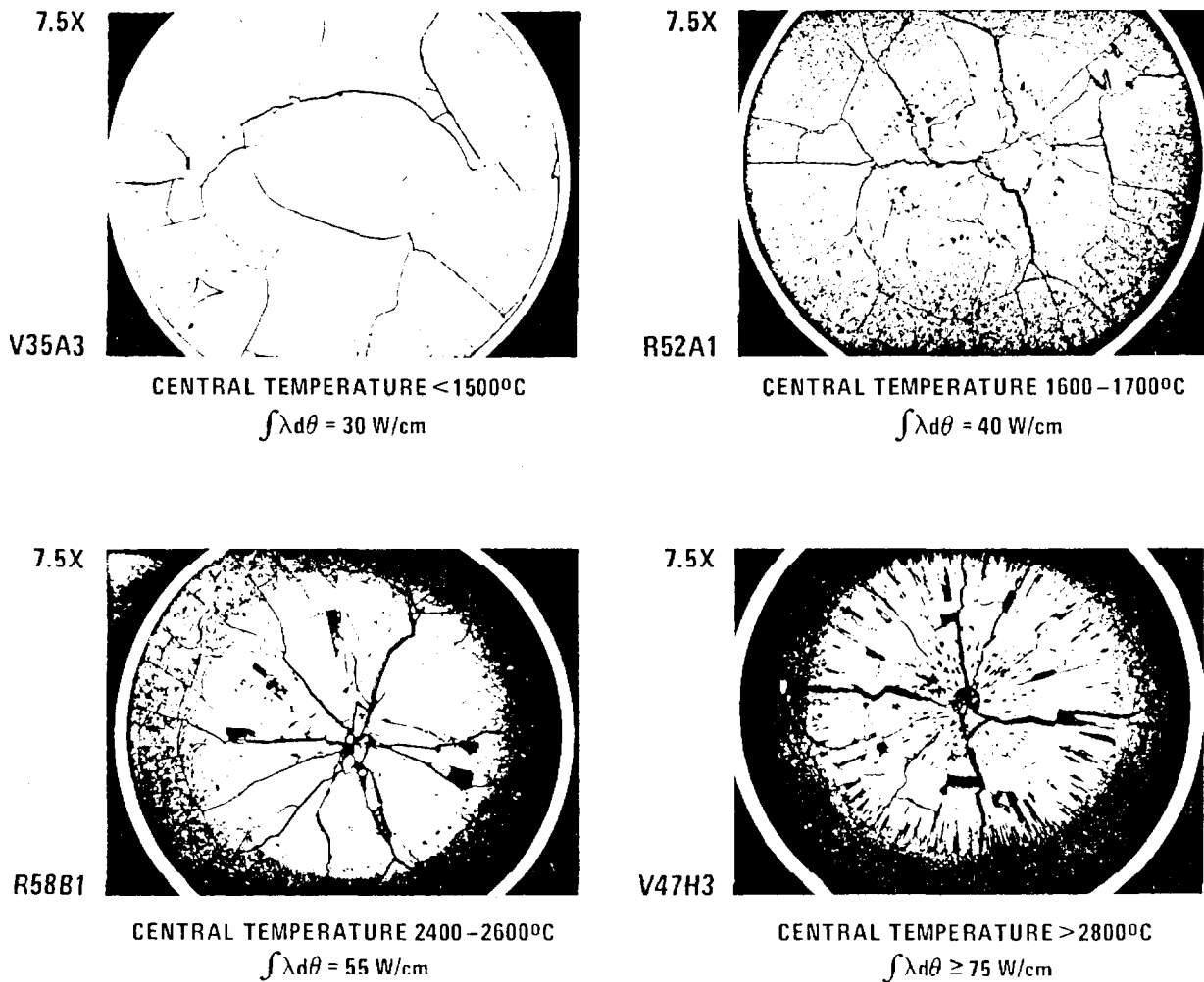


FIG. 8.6: Typical transverse cross section of irradiated UO_2 at Four Power Ratings.

It is known that some of the fission products occurring in the UO_2 fuel may appear as a gas. This creates a swelling in the fuel and has been found to contribute to a 0.7% change in the volume per 10^{20} fissions/cm³. The release of the fission

gas from the fuel is highly dependent upon the power rating or central temperatures. To accommodate this gas it is common practice to provide for a space in each fuel element.

Uranium dioxide (UO_2) has proven to be the most useful fuel for the current CANDU family of reactors. Other fuels have been studied and extensively tested. Uranium carbide fuel (UC) is known to provide a higher uranium density and seems particularly effective for organic cooled systems. The uranium-silicon alloy U_3Si has also been examined and found to possess adequate corrosion resistance. In anticipation of the potential use of recycled plutonium as a fissile fuel and thorium as a fertile fuel for the breeding of Uranium-233, fabrication and irradiation of $\text{UO}_2\text{-PuO}_2$ and $\text{ThO}_2\text{-UO}_2$ has been undertaken and has revealed no serious complications.

8.4 HEAVY WATER

We have previously indicated that heavy water (D_2O) is used as the moderator in all CANDU reactors and is the coolant in most of these. As indicated, the basis for preferring D_2O rather than light water (H_2O), or some other liquid coolant, rests largely in the excellent neutron-conservation characteristics of the deuterium nucleus. The microscopic absorption cross section for hydrogen (H) and for deuterium (D) corresponding to the 2200 m/s neutron speeds provide quantitative evidence to this significant distinction; hydrogen possesses an absorption cross section of 0.33 barns whereas deuterium has an absorption cross section of only 0.0005 barns. Thus, if a thermal neutron collides with a hydrogen, the likelihood that it will become absorbed is $0.332/0.0005 = 664$ times greater than if it were to collide with a deuterium nucleus. Heavy water therefore possesses a much better neutron economy than light water.

Another instructive demonstration of the relative superiority of heavy water over light water with respect to neutron economy is provided by the differences in mean-free-distances of neutron motion between their absorption in these two media. This parameter is readily calculated from the microscopic cross section, σ_a , and the atomic concentrations of the moderator, N:

$$\lambda_a = \frac{1}{\sigma_a N} = \frac{1}{\Sigma_a} \quad (8.5)$$

For pure H_2O and D_2O , these distances can be calculated to be given by 0.45 m and 301 m, respectively. That is, on the average, a thermal neutron will travel a total distance of less than 1/2 meters in pure water before it is absorbed, whereas in pure heavy water it will travel over 300 meters before absorption will occur. In part, this accounts for the requirement that heavy water reactor cores possess a much larger volume than light water reactor cores.

In addition to the above important neutronic properties of D_2O and H_2O , there are some small additional differences in some physical properties between heavy and

light water; these parameters are important in the analysis of thermal and transport properties. Some of these parameters are listed in Table 8.2.

Property	Light Water (H ₂ O)	Heavy Water (D ₂ O)
Density (gm/cm ³)	1.000	1.105
Freezing Point	0° (32°F)	3.82°C (39°F)
Boiling Point	100°C (212°F)	101.4°C (214.6°F)
Viscosity (centipoise)		
at 30°C	0.80	0.97
at 100°C	0.28	0.32
at 200°C	0.14	0.15

TABLE 8.2: Some properties of light water and heavy water.

Water, as it exists freely in nature, contains the molecular species H₂O, D₂O and HDO in equilibrium given by



If H₂O is removed then, by dissociation of 2 HDO, increasingly pure D₂O remains.

The concentration of deuterium in water is an important parameter in a heavy water separation process; even a small increase in initial concentration can lead to significant cost reductions. This concentration is known to vary from a low of about 135×10^{-6} parts in rain water to a high of 155×10^{-6} parts in sea water. That is

$$130 \text{ ppm} \approx \frac{\text{D}}{\text{D} + \text{H}} \approx 154 \text{ ppm} , \quad (8.7)$$

where the ppm means parts-per-million. The variability of this ratio for Canadian waters at several locations is shown in Fig. 8.7. It is of interest to note here the effect of natural fractionation by evaporation and precipitation. The Great Lakes cause some enrichment in deuterium by evaporation as suggested by the differences in thermodynamic properties between H₂O and D₂O; the concentration in the St. Lawrence River at Cornwall, Ontario is 149 ppm whereas the values for the rivers feeding the Great Lakes is about 144 ppm.

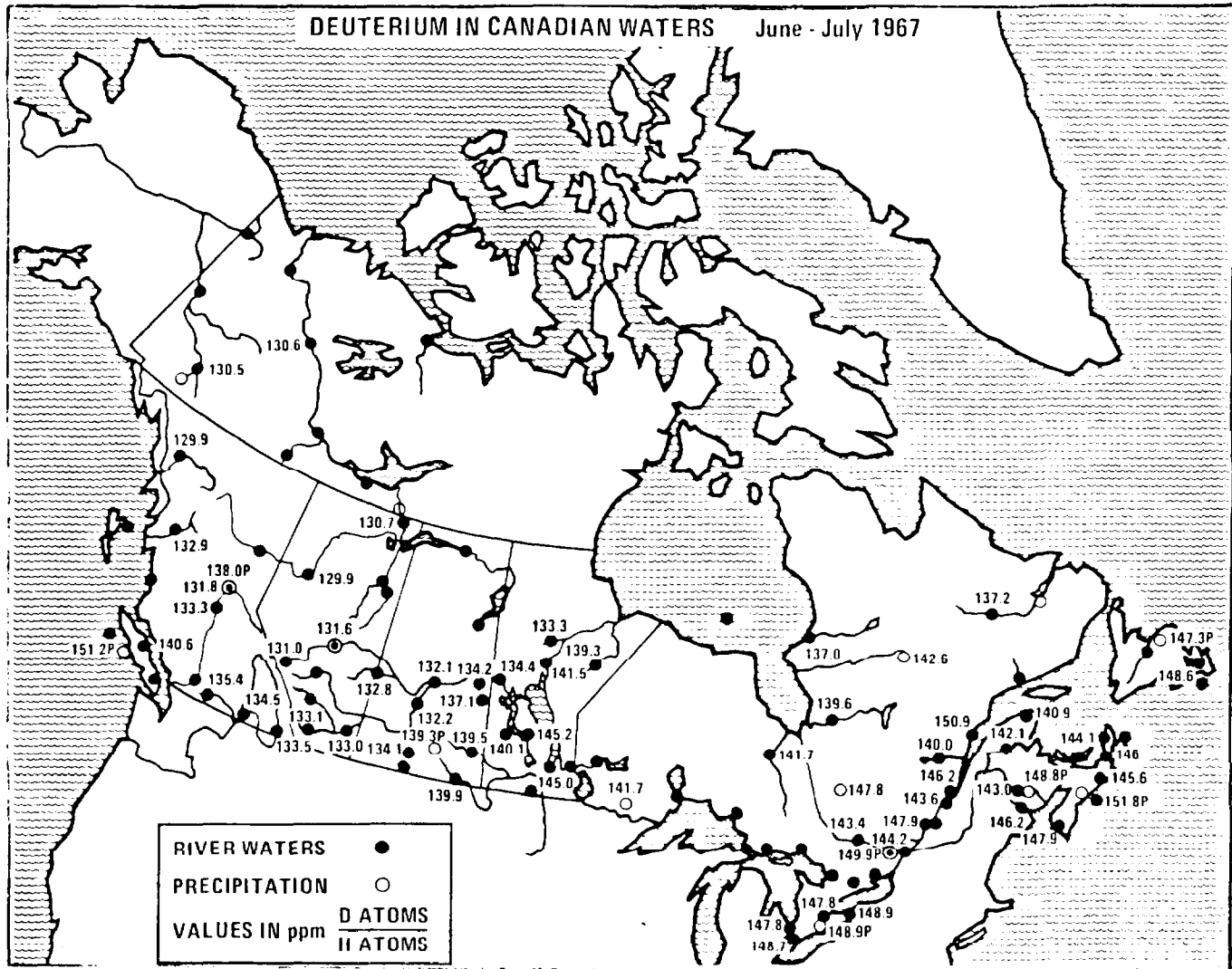


FIG. 8.7: Variation of heavy water content in Canadian waters.

In excess of one hundred chemical and atomic separation processes for extracting heavy water from natural water have been suggested and many of these have been extensively studied. The one process which has been found to be both technically feasible and economically viable is the dual-temperature $\text{H}_2\text{O}-\text{H}_2\text{S}$ exchange process. This process is based on the isotopic exchange property that the reaction



proceeds in the forward direction at low temperatures and in the reverse direction at a high temperature. Thus, deuterium can be transferred between water and hydrogen sulphide by selectively changing the temperature of a water and hydrogen sulphide gas mixture.

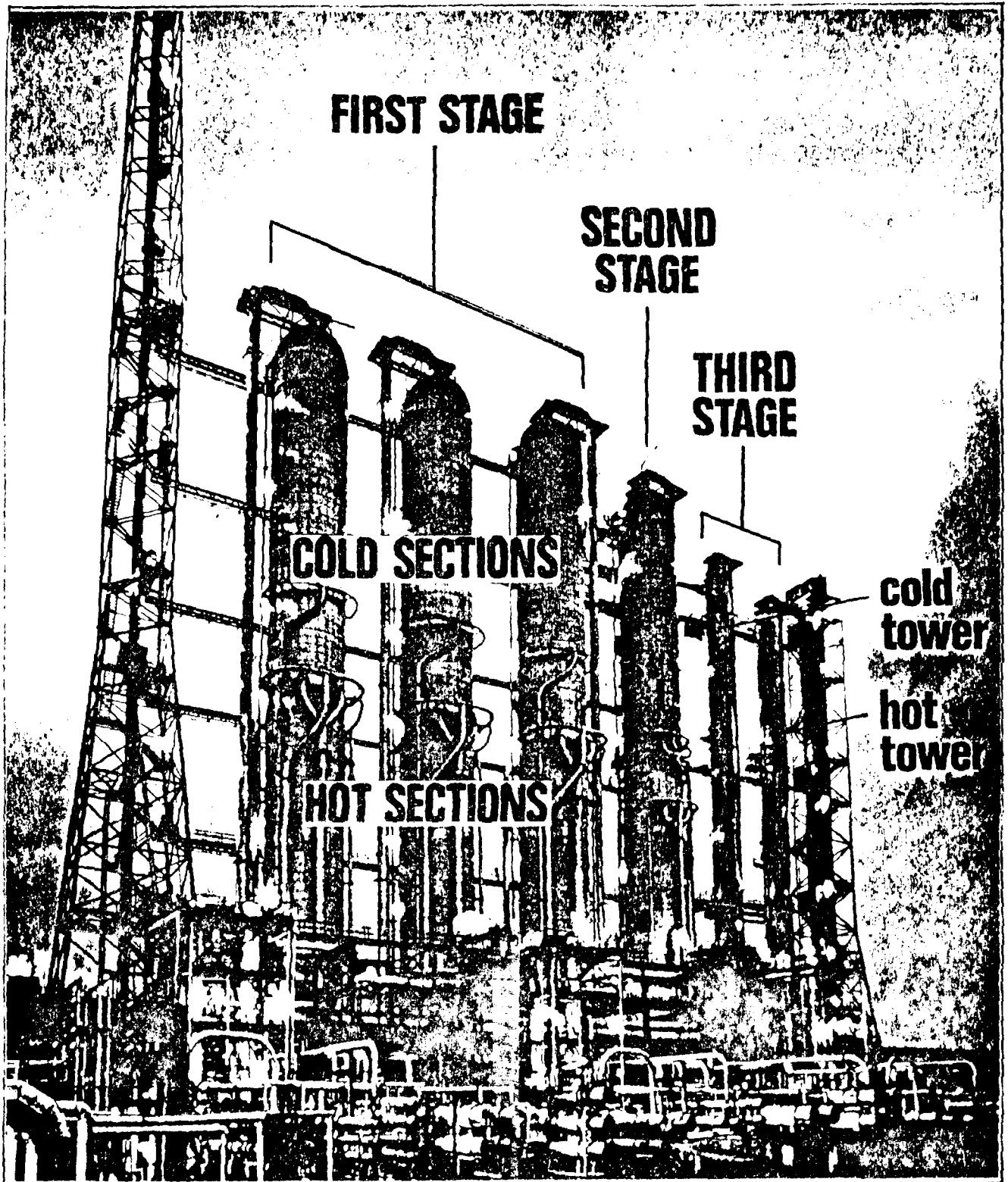


FIG. 8.8: Heavy water production plant at Port Hawkesbury, N.S.

The H₂O-H₂S dual-temperature process for the production of heavy water in kiloton quantities occurs in chemical separation plants characterized by several tall exchange towers. Fig. 8.8 shows such a facility and also identifies the various sections and stages. We will describe the exchange processes occurring in such a plant with the aid of Fig. 8.9.

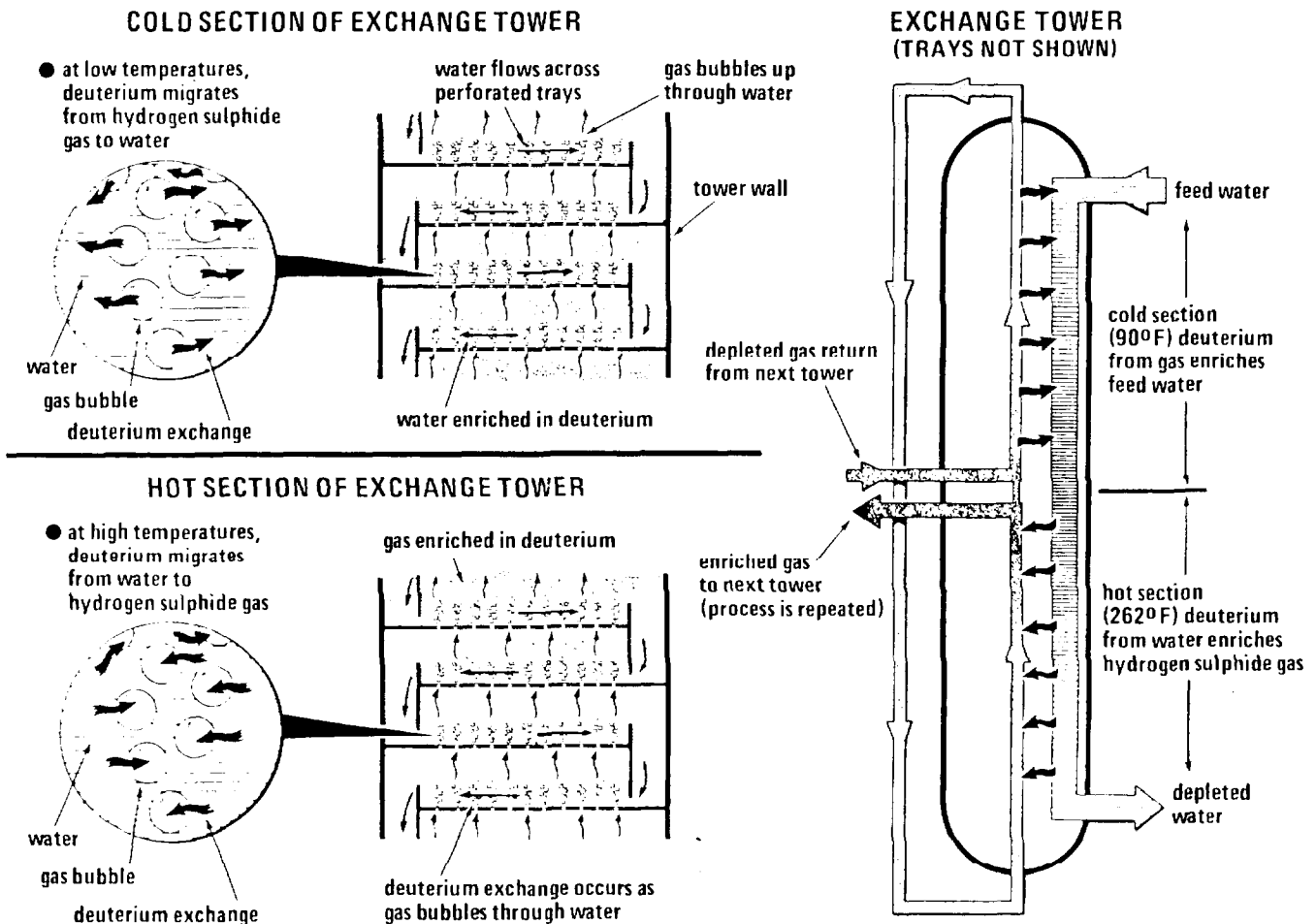


FIG. 8.9: Exchange process in the production of heavy water by the H₂O-H₂S dual-temperature process.

As indicated previously, the essential design consideration is the provision of a gas-water mixture at two separate temperatures. This two-phase mixture is produced by permitting water to flow over a set of vertically stacked perforated trays in the towers. Hydrogen sulphide gas passes through the perforations and

mixes with the water. The upper section of the first and second stage towers are called cold sections where the liquid-gas mixture exists at about 32°C (90°F) while the lower hot sections permit the two-phase mixture to exist at about 128°C (262°F).

In each tower, hydrogen sulphide flows up while water flows down. In the cold section of the tower the chemical exchange process favours the migration of deuterium from the gas to the liquid; this process is further aided because the gas has already been enriched in deuterium in its preceding passage through the lower hot section of the tower where deuterium is transferred from the liquid to the gas. This temperature difference between the top and lower section sets up a D_2O gradient in the water. Typically, it varies from a feed of about 145 ppm at the feed water inlet to about 600 ppm at the cold-hot junction and about 120 ppm in the waste.

Small streams of the deuterium-rich hydrogen sulphide gas are extracted from the middle of each tower and passed on to a second and third stage. After the third stage, water becomes enriched in deuterium to some 25%. With such concentrations, it now becomes economical to use vacuum distillation to produce the reactor grade heavy water consisting of 99.75% D_2O . A flow chart of a typical process is shown in Fig. 8.10.

The use of hydrogen sulphide in a heavy water production plant involves a number of special considerations. Although this gas is colourless, it has an odour similar to that of rotten eggs. H_2S will burn in air to form sulphur dioxide and water while mixing it with water leads to the formation of sulfuric acid. Most significantly, hydrogen sulphide is a toxic substance. Hence, considerable precautions, such as wearing of masks by certain plant workers, must be exercised.

The early availability and use of heavy water in Canada represents a fascinating study.

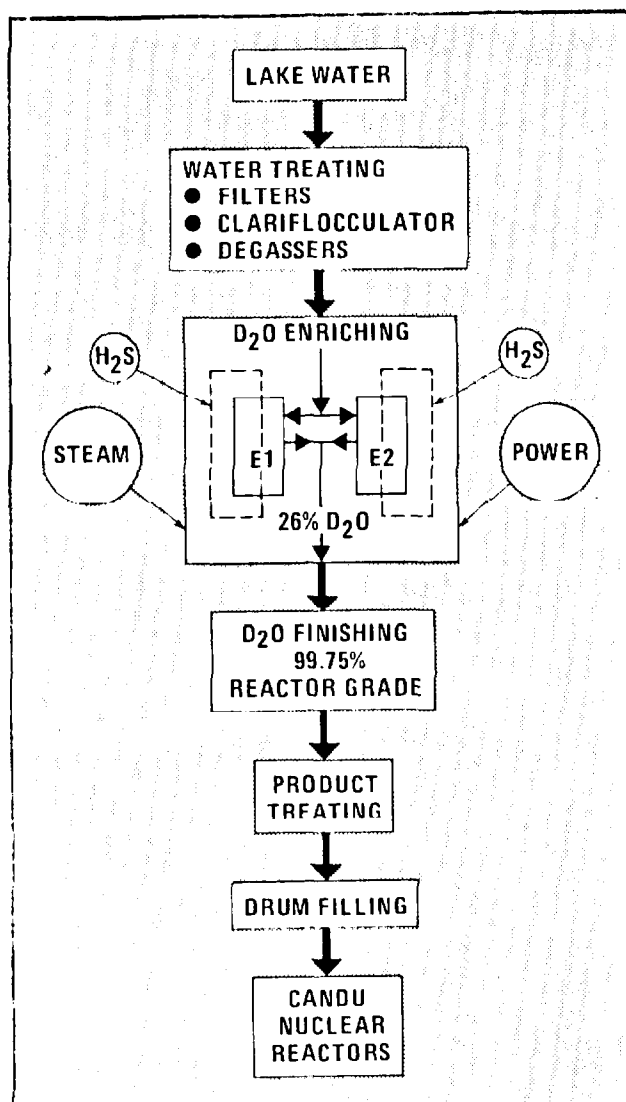


FIG. 8.10: Flow chart of heavy water production.

The first production of water with a high concentration of deuterium occurred at the Consolidated Mining and Smelting Company in Trail, British Columbia, in 1943. Here, heavy water was obtained as a by-product from a process involving the production of electrolytic hydrogen. The yield was small and was earmarked for the U.S.A. nuclear energy program. Also in 1943, some 200 kg of high purity water arrived in Canada from England in connection with the joint Canada-Britain-U.S.A. nuclear energy program. This quantity of precious water had travelled far; it was initially spirited away from its origin in Norway ahead of the advancing war front and transported to France and subsequently, for similar reasons, taken to England and finally found its way to Canada. A group of nuclear scientists became attached to this rare quantity of heavy water in order to study the neutron migration properties in this liquid; several of these scientists also came to Canada.

Canada's current heavy water production program is centered at three locations: Glace Bay, N.S., Port Hawkesbury, N.S. and Kincardine (Bruce), Ontario. A fourth location for a heavy water production plant is near the Gentilly nuclear power station in Quebec, Table 8.3.

Name and Owner	Completion Date	Capacity
Port Hawkesbury, Nova Scotia Canadian General Electric Company	1970	400 ton/year
Bruce (three units), Ontario Ontario Hydro Electric Power Commission	1971	3 @ 800 ton/year
Glace Bay, Nova Scotia Atomic Energy of Canada Limited	1976 (expected)	400 ton/year
La Prade, Quebec Atomic Energy of Canada Limited	1978 (expected)	800 ton/year

TABLE 8.3: Heavy water production plants in Canada.