CHAPTER 5

B. MATERIALS IN THERMALHYDRAULICS LOOP OF CANDU REACTORS

R.G. Fleck
Ontario Hydro Research Division

ABSTRACT

Key issues related to the performance of components in the primary heat transport system of CANDU reactors are presented. A brief description is provided of the rationale for the selection of materials for specific components. Irradiation enhanced deformation of pressure tubes and calandria tubes is described along with an outline of the deformation mechanisms and the important metallurgical parameters. The factors which resulted in pressure tube leaks in Pickering and Bruce reactors are discussed and a brief description of the delayed hydride cracking mechanism is provided. Fuel bundle performance has been satisfactory; however potential failure of cladding could occur by stress corrosion cracking and this is discussed. The performance of steam generator tubing is presented and possible failure mechanisms are outlined.

5 .1 Introduction

The materials aspects to be discussed are concerned with the performance of the most critical reactor core components and also steam generator tubes. The rationale for selection of materials and their performance under the operating conditions of pressure, temperature and irradiation will be presented.

5 .2 Reactor Core Materials

CANDU reactors use natural uranium as fuel and heavy water as moderator and coolant. Structural components must have a low capture cross section to enhance neutron economy in addition to corrosion resistance in water and adequate tensile strength and ductility. Zirconium is the only practical material which can satisfy the above requirements and consequently the structural components in the core of CANDU reactors are fabricated from the zirconium alloys, Zircaloy 2 and 4 and Zr-2.5 wt% Nb (Table 1).

5 .3 Pressure Tubes

Pickering Generating Station 'A' (PGSA) was the first commercial CANDU power station and since it was commissioned in the early 1970's the most significant concerns have been related to pressure tubes. In particular two aspects will be discussed: elongation and cracking. Several review papers have been written on these topics/1,2,3,4,5,6/.

Elongation: to accommodate pressure tube elongation the end-fitting assembly was designed to move out of the reactor (Figure 1). The initial yoke-nut-gaps were set at about 6 mm as elongation was only anticipated to occur by thermal expansion plus thermal creep. Currently pressure tubes elongate at about 4 mm/7000 EFPH (Effective Full Power Hours) and consequently extensive in-service maintenance has been required to adjust yoke-nut-gaps and ensure that the end-fittings remain on their bearings (Figure 1). The higher than anticipated elongation rates occur because the fast (E>1 MeV) neutron flux accelerates the thermal creep rate and also results in irradiation growth/7,8/. Irradiation growth is a dimensional change which occurs in anisotropic metals without the application of a stress and does not result in a density change of the metal. The extent of deformation by irradiation creep and growth is dependent on the metallurgical structure of the pressure tube which, in turn, is dependent on the fabrication process

Pressure tubes are produced by an extrusion and cold drawing process (Figure 2) and a review of pressure tube production has been presented by Ells and Evans/9/. The extrusion process develops a pronounced crystallographic texture in the tubes/10/ and also elongates the grains. The cold-drawing primarily increases the dislocation density which contributes to the tensile strength of the tubes. Pressure tubes produced by the above process have demonstrated good corrosion resistance and uniform tensile properties. Various studies on pressure tube elongation have indicated that the important metallurgical variables are texture, dislocation density and grain shape/11,12/. Figure 3 describes the relevant operating conditions of a Pickering pressure tube. The wall thickness of the pressure tube depends on the alloy used. There are two alloys which have been used. Pickering Units 1 and 2 have Zircaloy pressure tubes whereas Units 3 and 4 have Zr-2.5 wt% Nb pressure tubes. The basic reason for the change from Zircaloy to Zr-2.5 wt% Nb was to take advantage of the higher UTS value of the Zr-2.5 wt% Nb. With the increased strength it was possible to reduce the wall thickness of the pressure tubes which has resulted in better fuel economy in Units 3 and 4 as opposed to Units 1 and 2. The metallurgical parameters which influence in-reactor deformation are:

a) Texture - zirconium has an hcp crystal structure (Figure 4) and studies on irradiation growth indicate that an expansion occurs in the a-type directions and a contraction occurs in the c-type directions/13/. In pressure tubes the c-axes are preferentially aligned in the transverse direction (Figure 5). Irradiation growth, therefore, results in an increase in length and a decrease in diameter of the tube.
Deformation by creep depends on the stress assisted motion of dislocations. For zirconium dislocation motion mainly results in deformation in the a-type directions (Figure 4); such directions are considered "soft" wheras the c-type directions are considered to be "hard". The hoop stress is the largest stress which acts on the pressure tubes and consequently is the most favourable direction for creep. Internally pressurized tubes of an anisotropic material will deform under creep conditions to give an increase in diameter with either a positive or negative change in length. For pressure tubes deformation by creep results in the expected increase in diameter as well as an increase in length.

b) Dislocation Density - the dislocation density can be considered directly related to the level of cold work. Irradiation growth studies on Zircaloy cold-worked to different levels illustrates that the extent of growth is directly related to the level of cold work (Figure 6). Although pressure tubes are stress relieved (autoclaved) after cold drawing the dislocation density is still high/14/ and it is expected that pressure tubes will deform by irradiation grwoth at a steady rate throughout the life of the reactor.

The dependence of irradiation creep on the dislocation density/cold work is not as strong as the growth dependence. However, there is a slight increase in creep rate as a result of increases in the dislocation density/cold work/ 15,16/.

c) Grain Shape - the elongation characteristics of the Zircaloy pressure tubes in PGSA Units 1 and 2 are different than those of the Zr-2.5 wt% Nb tubes in Units 3 and 4. The average elongation rates are the same but the tube-to-tube variation is much greater in Units 1 and 2 than in Units 3 and 4. Examination of material from "fast" and "slow" elongating Zircaloy tubes indicated a difference in grain shape. The "fast" tubes had highly elongated grains (Figure 7), whereas the slow tubes tended to have an equiaxed grain shape/2/. Theoretically, the grain shape is expected to influence the irradiation growth component - elongated grains in effect increase the extent of anisotropic deformation in the pressure tubes. For Zr-2.5 wt% Nb pressure tubes the grain shape is one of highly elongated grains (Figure 8) and to change the grain shape a post-extrusion anneal would have to be implemented. Consequently, the grain shape of Zr-2.5 wt% Nb tubes is consistently one which promotes irradiation growth in the axial direction of the tube.

In summary, the analysis of pressure tube elongation indicates that irradiation growth is the main factor which contributes to elongation/11/. A development program has been undertaken to produce pressure tubes with a metallurgical structure that would result in a reduction of the in-service elongation rate/17/.

5.4 Pressure Tube Cracking

The only long-term shutdowns of CANDU reactors related to pressure tubes were due to crack formation in some tubes in Pickering 'A', Units 3 and 4. Cracks were indicated because of leakage of the coolant into the gas annuli between the pressure tube and the calandria tube. Leakage was first detected in Unit 3 in August 1974 when the reactor was being returned to power. A total of 17 tubes were replaced in this unit. In May 1975 similar leaks were detected in Unit 4 and a total of 52 pressure tubes were replaced. A further pressure tube leak occurred in 1982 in the Unit 2 reactor at Bruce generating station.

Examination of the tubes removed as a result of leakage revealed that cracks had formed and grown by a delayed hydride cracking mechanism/4,18/. The influence of zirconium hydrides on the ductility of pressure tubes had been extensively investigated prior to the development of the pressure tube production route. Zirconium hydrides precipitate as platelets in the metal and the ductility depnds on the orientation of the hydrides relative to the principal stress. For tensile tests the ductility is severely reduced if the normals of the platelets are parallel to the tensile stress. However, if the platelet normals are perpendicular to the applied stress the ductility is not reduced. Pressure tubes are produced such that the hydride platelets are in the circumferential axial plane and consequently their normals are perpendicular to the hoop and axial stresses. At room temperature the terminal solid solubility (TSS) of hydrogen in zirconium is less than 5 ppm and "as received" pressure tubes contain typically 12 ppm of hydrogen and so contain hydrides in the as-received condition.

The cracks which occurred in the above-mentioned reactors were a result of hydride re-orientation into the radial-axial plane of the pressure tube. The re-orientation occurred locally in an area close to the rolled joint and was attributed to high residual stresses introduced as a result of over-extension of the rolling tool during pressure tube installation/14/ (Figure 9). The delayed hydride cracking mechanism occurred as follows: hydrides preferentially precipitated in areas of high stress such as found at the tip of a small surface flaw grew as a result of hydrogen accumulation and the hydride eventually cracked under the action of the stress at the tip of the flaw (Figure 10). The flaw is thus extended and the process is repeated until the crack grows completely through the wall of the tube.

The pressure tube leaks to date have all occurred at temperatures below the operating conditions. This is a result of the hydrogen being taken into solution at the higher temperatures. However, the pressure tubes continually "pick-up" hydrogen from the primary coolant (particularly in the rolled joint area) and eventually the hydrogen levels will be such that hydrides will be present under operating conditions. Provided these hydrides remain in the circumferential-axial orientation, they will not present problems to future reactor operation.

5.5 Calandria Tubes

Calandria tubes are produced from Zircaloy-2 by a process outlined in Figure 11. The seam welding process eliminates Zr-2.5 wt% Nb as calandria tube material because the weld would produce a brittle structure in the tube. Calandria tubes are subjected to a temperature of 80°C and a stress of 0.16 MPa and their main function is to separate the moderator from the pressure tubes. To date there have been no operational difficulties associated with the calandria tubes. There are some studies underway to establish the in-service dimensional stability of the calandria tubes. Extensive elongation could result in high stresses in the end-shields. Also once the pressure tube has settled onto the tube spacers the calandria tube supports the total weight of the fuel bundles and pressure tube which results in calandria tube sag by a creep process. However, in-service measurements and analysis of the projected sag throughout the reactor life do not indicate that significant difficulties of fuel bundle movement will occur.

5.6 Fuel Bundles

The residence time of a fuel bundle in the reactor core is about 18 months. Consequently, long-term effects associated with irradiation creep and growth are not a major concern. Corrosion resistance and ductility are important properties for the fuel sheaths as it is important that fission products do not enter the primary heat transport system. Zircaloy-4 is used for the fuel elements because it has better corrosion properties than Zircaloy-2/19/.

Zircaloy is used for fuel cladding in all water-cooled reactors and extensive investigation has been carried out to determine what factors can lead to cracking of the fuel elements. For CANDU fuel elements the most widely investigated failure mechanism has been stress corrosion cracking (SCC) as a result of the fission product iodine/20,21/. Under normal operating conditions, stress corrosion cracking is not a problem. However, if a fuel bundle is moved from a low power to a high power position then SCC failures become a problem. When the bundle is moved to a high power position the rate of fission gas release is increased which puts extra stress on the cladding; also the fuel pellet can expand and interact with the Zircaloy element. The previous exposure to the neutron irradiation has reduced the ductility of the fuel element (Figure 12), and consequently it cannot readily deform to accommodate the increased internal pressure from the fission gas and/or expanded fuel. This results in high stresses in the cladding and the combination of these stresses and the iodine can result in fuel cladding failures/22/. Such failure occurred with the on-power fuelling of CANDU reactors when only eight bundles of the total twelve in a Pickering type channel were removed. Such fuel shifts resulted in two bundles per channel being moved from low to high power positions (Figure 13).

5.7 Steam Generator Materials

Steam generator tubes must have good thermal conductivity and have good corrosion resistance to high temperature water. For a Pickering 'A' type steam generator there are 2600 tubes arranged in a vertical U-shaped pattern. For each Unit at Pickering 'A' there are twelve steam generators.

The steam generator tubing materials used in the Ontario Hydro nuclear generating stations are listed in Table III and the composition of these alloys is presented in Table IV. Corrosion resistance to the primary side is important as corrosion products will be carried through the core and so become radioactive. Radioactive material in the primary systems increases the background radiation fields and results in high man-rem costs for maintenance programs. From both the primary and secondary sides it is important to minimize the corrosion rates in order to prevent penetration of the tubes. The corrosion rates are largely controlled by the water chemistry as discussed previously. A review of steam generator tubing performance in CANDU reactors is presented in reference /23/.

The world experience with steam generators in nuclear plants has not been very satisfactory as can be noted in Table V. However, Ontario Hydro has only experienced one failure of a Pickering 'A' tube which was a result of a manufacturing defect and in Bruce GS a total of five steam generator tube leaks have occurred. Table VI compares the CANDU performance of steam generator tubing with those in other tubes of reactors.

The most usual failure mechanism of steam generator tubing is stress corrosion cracking/24,25/. In many cases the stress can be a result of tube manufacture which produces residual stresses in the tube. One area where such stresses occur is at the U-bend (Figure 14) and it is necessary to use careful stress relieving treatments of such areas if leaks are to be avoided in operation. Thermal stresses and possibly residual stresses in the U-bend region may have been the reason for the tube leaks in Bruce GS. Stress corrosion cracking can also occur if aggressive elements are concentrated in stagnant flow areas of the steam generator. To date this has not proved to be a problem area for the Ontario Hydro nuclear stations.

5.8 References

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TABLE I

Element	Zircaloy-2	Zircaloy-4	Zr-2.5 wt% Nb
Tin	1.2 - 1.7 wt%	1.2 - 1.7 wt%	-
Iron	0.07 - 0.2 wt%	0.18 - 0.24 wt%	-
Chromium	0.05 - 0.15 wt%	0.07 - 0.13 wt%	-
Nickel	0.03 - 0.08 wt%	-	-
Niobium	-	-	2.4 - 2.8 wt%
Oxygen	1400 ppm (max)	1400 ppm (max)	900 - 1300 ppm
Balance	Zirconium plus impurities.		

The chemical composition of various zirconium alloys used in CANDU Reactors

TABLE II

Alloy	Components
Zircaloy-2	Pressure tubes Calandria tubes Guide tubes
Zircaloy-4	Fuel bundle elements
Zr-2.5 wt% Nb	Pressure tubes (post PGSA-2)

The Zirconium Alloys used for specific components in the core of CANDU Reactors

TABLE III

Steam Generator Materials

Station	Material	Reason for Selection
Pickering 'A'	Monel	Cheaper than Inconel and resistant to SCC from chlorides
Bruce 'A'	Inconel 600	Higher oxygen levels in PHT and Inconel has better corrosion resistance than Monel in such conditions.
Pickering 'B'	Monel	
Bruce 'B'	Inconel	
CANDU-600s) Darlington)	Incoloy 800	Improved corrosion resistance than Inconel and lower Ni content; therefore less activation products.

A Description of the Steam Generator Tube Material used in Different CANDU Reactors & the reason for selection of these Materials

TABLE IV

Alloy	Composition
Monel	70% Ni, 30% Cu
Inconel 600	75.5% Ni, 15.5% Cr, 1.0% Cu, 1.0% Fe
Incoloy 800	32% Ni, 20% Cr, 47% Fe

Composition of Steam Generator Tubing Material Used in CANDU Reactors

TABLE V

Reactor	Type	Tube Material	No Tubes/ Reactor	Reported Failures to Dec '79
N-Reactor	LWGR	Stainless Steel	19,160	
Tarapur-1	BWR	Stainless Steel	3,200	4
Tarapur-2	BWR	Stainless Steel	3,200	209
Shippingport-1	PWR	Stainless Steel	6,034	426
Indian Point-1	PWR	Stainless Steel	3,224	Shut down?
Dresden-1	BWR	Stainless Steel	7,204	180
Yankee-Rowe	PWR	Stainless Steel	6,480	95
KWL Lingen	BWR	Stainless Steel	10,000	112
Ardennes	PWR	Stainless Steel	6,648	
MZFR Karlsruke	PHWR	Stainless Steel	4,226	0
KRB Gundremmign	BWR	Stainless Steel	5,787	364
Trino Vercelles	PWR	Stainless Steel	6,648	6
Garigliano	BWR	Monel-400	3,570	332+
Douglas Point	PHWR	Monel-400	15,600	2
Pickering 1	PHWR	Monel-400	31,200	0
Pickering 2	PHWR	Monel-400	31,200	1
Pickering 3	PHWR	Monel-400	31,200	0
KANUPP	PHWR	Monel-400	8,130	0
RAPP-1	PHWR	Monel-400	15,600	0
Beznau-1	PWR	Inconel-600	5,208	1007
Mihami-1	PWR	Inconel-600	8,852	2208
Point-Beach-1	PWR	Inconel-600	6,520	678
KWO(Obrigheim)	PWR	Inconel-600	5,214	273
Shippingport-2	PWR	Inconel-600	6,034	
HB Robinson-2	PWR	Inconel-600	9,780	157
San Onofre-1	PWR	Inconel-600	11,382	265
Haddam Neck	PWR	Inconel-600	15,176	32
NPD	PHWR	Inconel-600	2,069	47
Mihami-2	PWR	Inconel-600	6,520	297
Surrey-1	PWR	Inconel-600	10,164	1798
Point Beach-2	PWR	Inconel-600	6,520	25
R.E. Ginna-1	PWR	Inconel-600	6,520	205

TABLE V (Contd)

Reactor	Type	Tube Material	No Tubes/ Reactor	Reported Failures to Dec '79
Palisades	PWR	Inconel-600	17,038	3696
Jose Cabrera	PWR	Inconel-600	2,604	7
Maine Yankee	PWR	Inconel-600	17,109	15
Beznau-2	PWR	Inconel-600	5,208	275
Turkey Point-3	PWR	Inconel-600	9,780	1655
Bruce 1	PHWR	Inconel-600	33,600	0
Bruce 2	PHWR	Inconel-600	33,600	11*
Bruce 3	PHWR	Inconel-600	33,600	0
Bruce 4	PHWR	Inconel-600	33,600	0

*Tubes actually plugged, only 4 actually failed.

A list of reported Steam Generator Tube Failures to December 31, 1979 in different reactors throughout the world. A more detailed list can be obtained from O.S. Tatone and R.S.Pathania, AECL 7251, March 1981.

TABLE VI

Type of Reactor	No of Reactors	No of S/G Tubes	No of Tube Defects	% of Tubes with Defects
CANDU	12	300 599	63	0.02
BWR	85	1 061 688	22 351	2.1

Steam Generator Tube Performance to December 31, 1980

47 of the CANDU failures have occurred in recent years at NPD. Only eight of the remaining 16 tubes have actually leaked, 2 at Douglas Point, 1 at Pickering 'A' and 5 at Bruce 'A'. These plants have all had good quality condenser cooling water, from Lakes Huron or Ontario and exceptionally good condenser and condenser tube performance.

(The above table is from Tatone & Pathania of AECL)

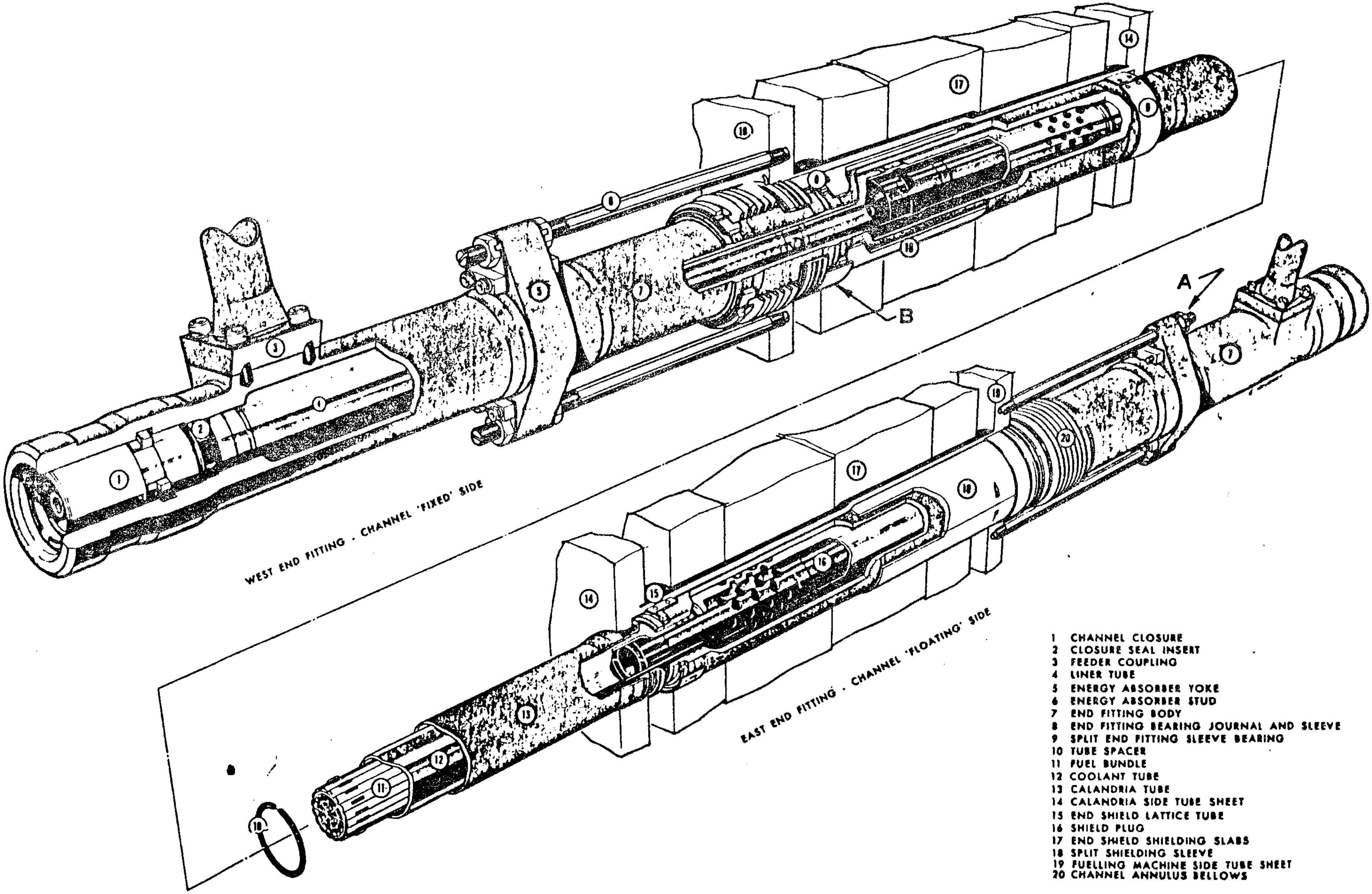


FIGURE 1

SCHEMATIC DIAGRAM OF A PICKERING GENERATING STATION "A" FUEL CHANNEL
(FROM CANDU 500 PICKERING GENERATION STATION 1969)

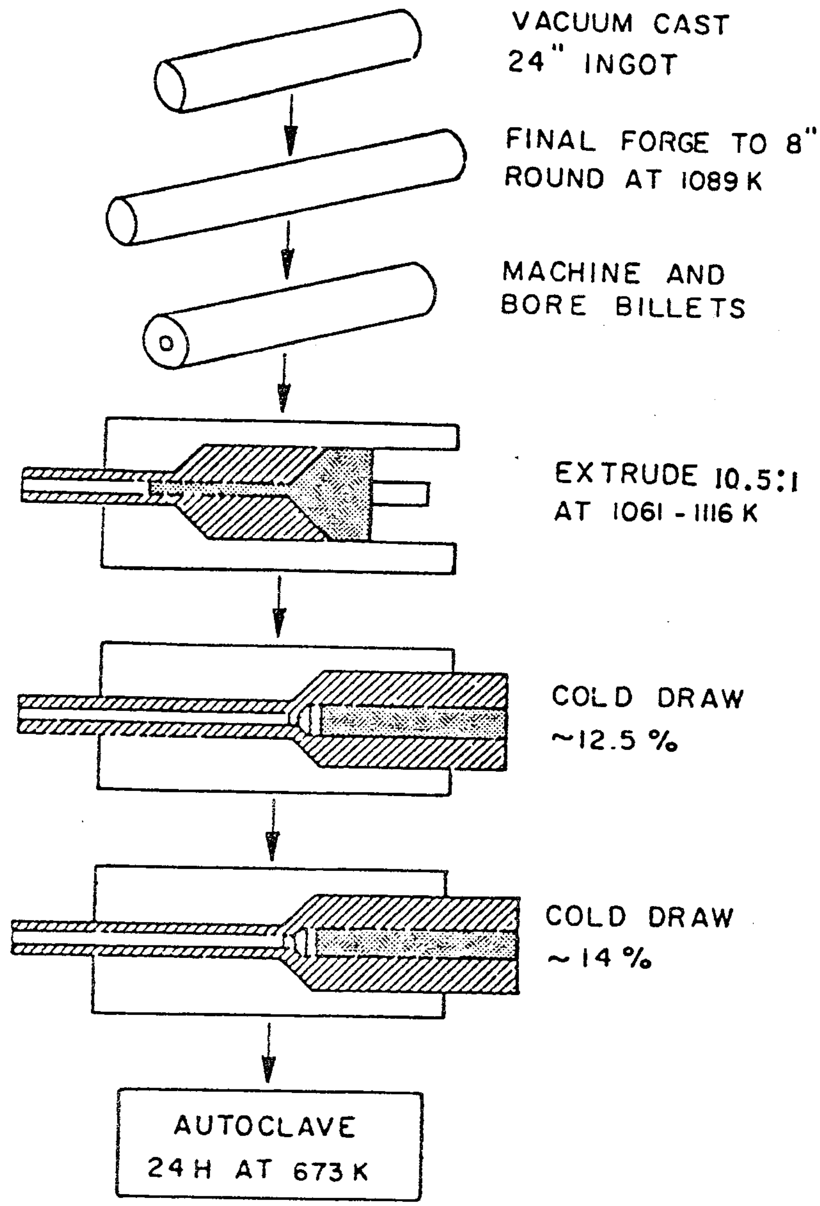


FIGURE 2

FABRICATION ROUTE USED TO PRODUCE PRESSURE TUBES FOR PICKERING GENERATING STATION 'A'

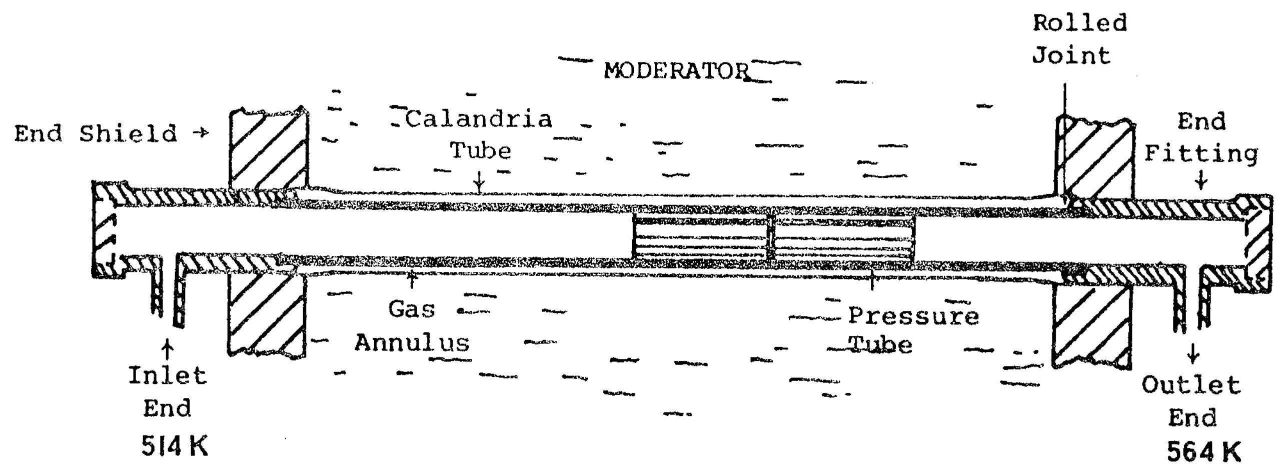


FIGURE 3

SCHEMATIC DRAWING OF A FUEL CHANNEL

PRESSURE TUBE: LENGTH 6.29m (20 ft 8 in)

INTERNAL DIAMETER: 10.34 cm (4.07 in)

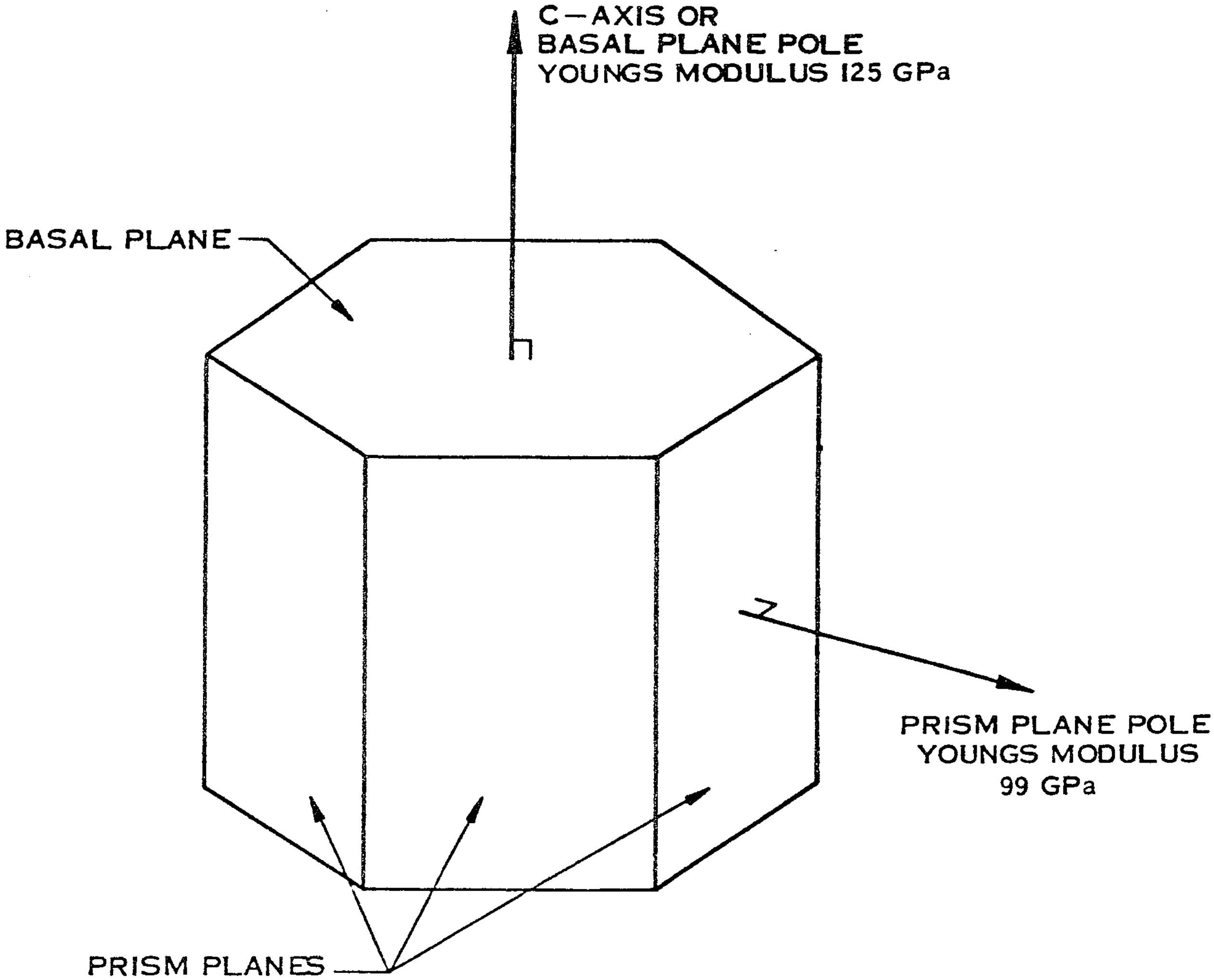


FIGURE 4

HEXAGONAL CRYSTAL STRUCTURE OF ZIRCONIUM.
TEXTURE MEASUREMENTS ON PRESSURE TUBES HAVE BEEN MAINLY CONCERNED WITH THE DISTRIBUTION OF THE BASAL PLANE POLES IN THE THREE PRINCIPAL DIRECTIONS OF THE PRESSURE TUBE

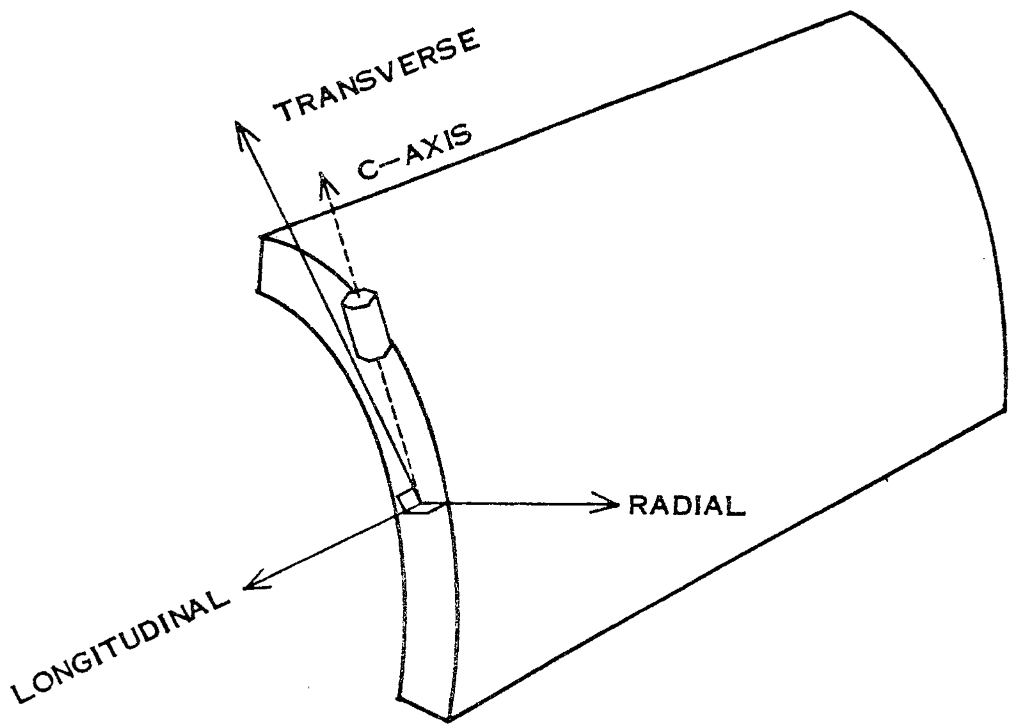


FIGURE 5

SCHEMATIC REPRESENTATION OF THE PREFERRED ORIENTATION OF C—AXIS (BASAL POLES) OF ZIRCONIUM CRYSTALS (GRAINS) IN PRESENT CANDU PRESSURE TUBES. THE FRACTION OF THE BASAL POLES IN THE THREE PRINCIPAL DIRECTIONS ARE DETERMINED

F_T — FRACTION OF BASAL POLES IN TRANSVERSE DIRECTION -0.56
F_R — FRACTION OF BASAL POLES IN RADIAL DIRECTION -0.36
F_L — FRACTION OF BASAL POLES IN LONGITUDINAL DIRECTION -0.09

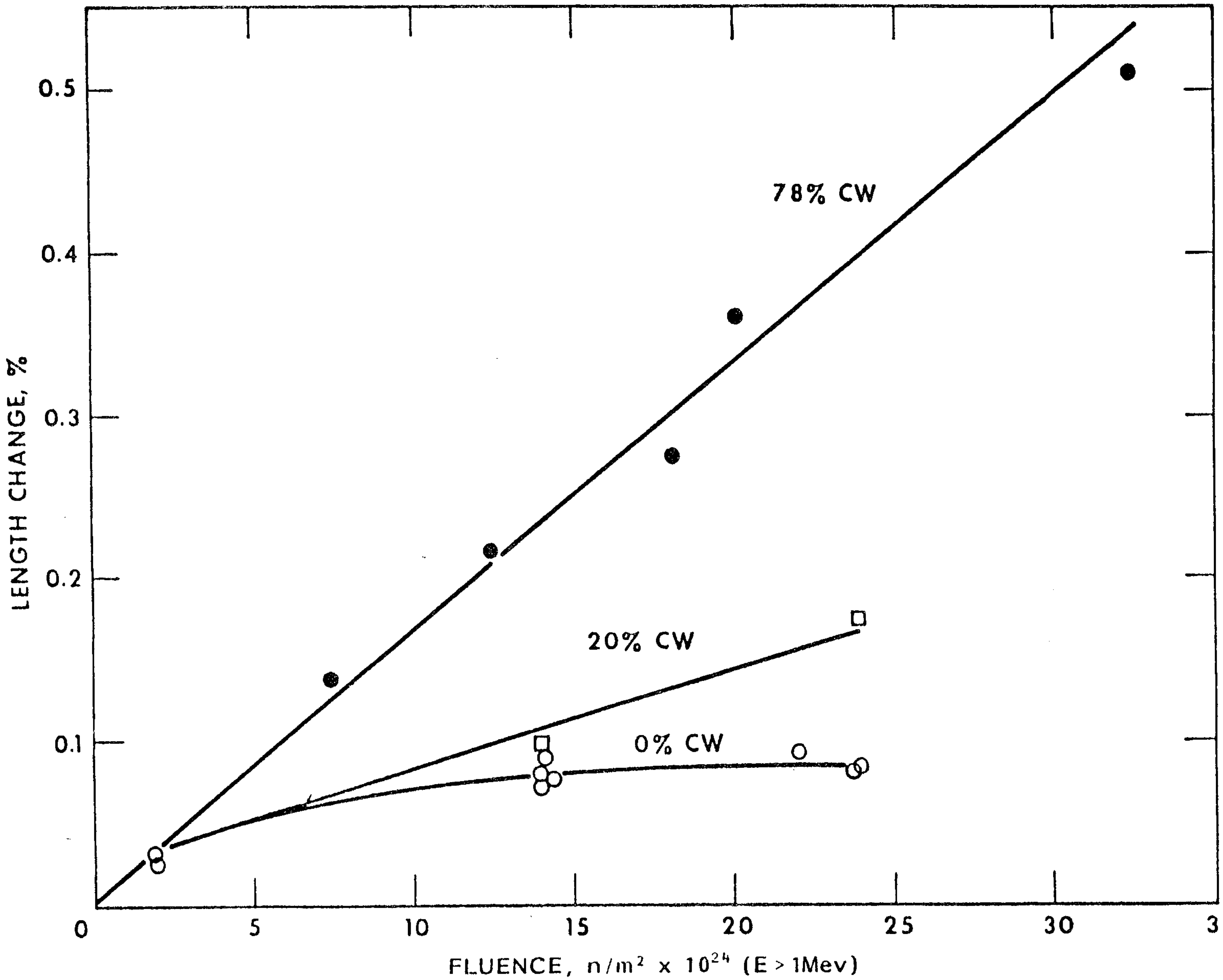
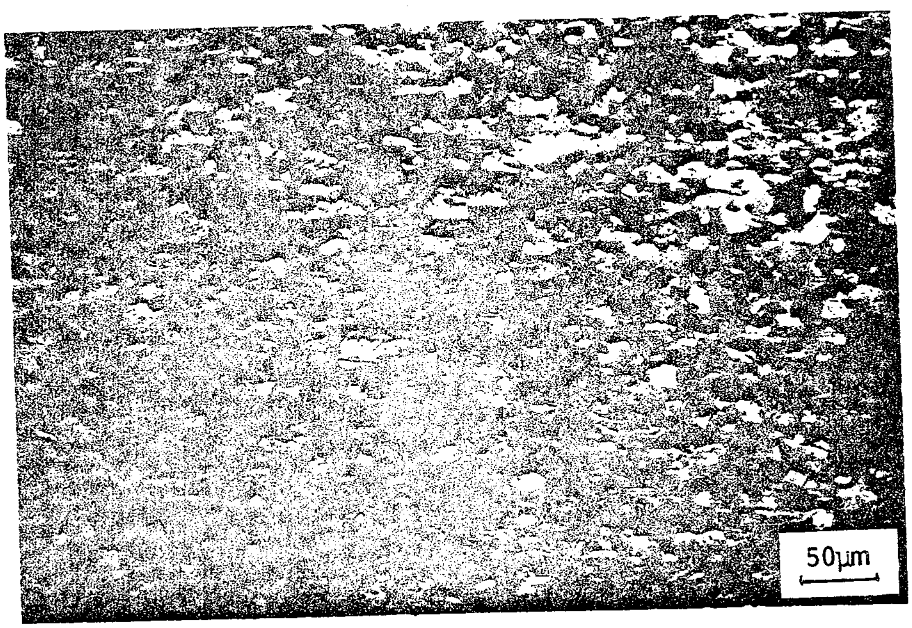


FIGURE 6

THE EXTENT OF RADIATION GROWTH (LENGTH CHANGE) AS A FUNCTION OF FAST NEUTRON FLUENCE IN ZIRCALOY SAMPLES WITH DIFFERENT LEVELS OF COLD WORK (AFTER ADAMSON

PICKERING 2 - FAST ELONGATING TUBE



PICKERING 2 - SLOW ELONGATING TUBE

FIGURE 7

OPTICAL MICROGRAPHS OF LONGITUDINAL SECTIONS
FROM ZIRCALOY PRESSURE TUBES

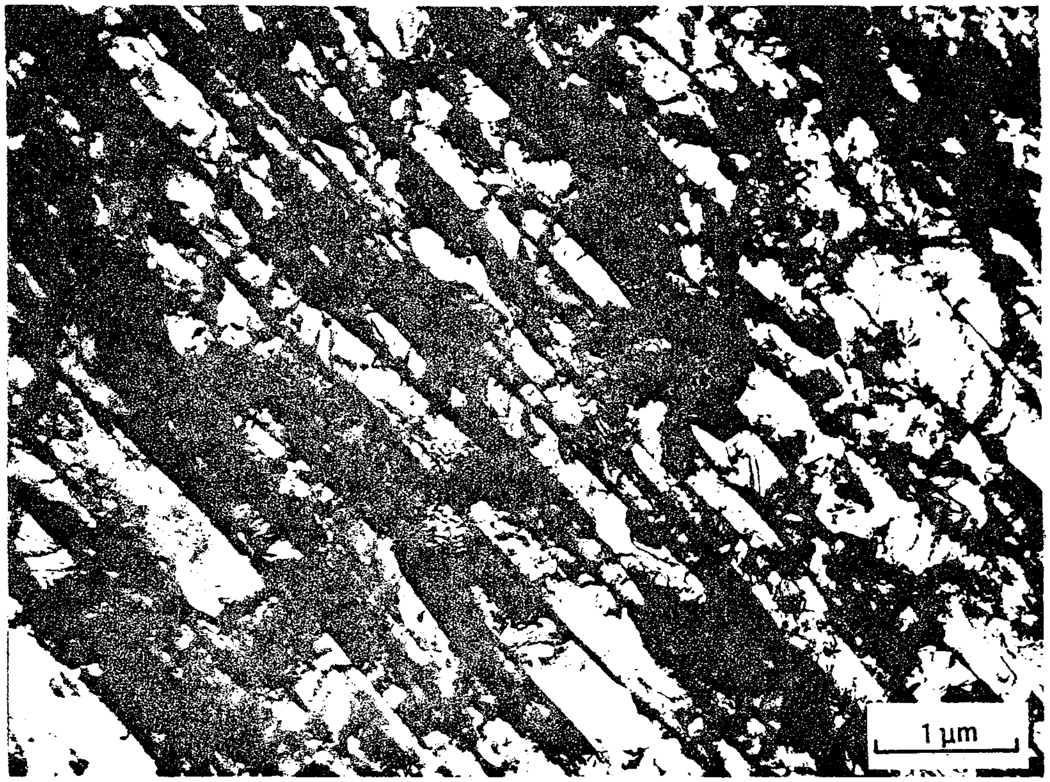


FIGURE 8

TRANSMISSION ELECTRON MICROGRAPH OF A LONGITUDINAL SECTION FROM A Zr-2.5 WT % Nb PRESSURE TUBE

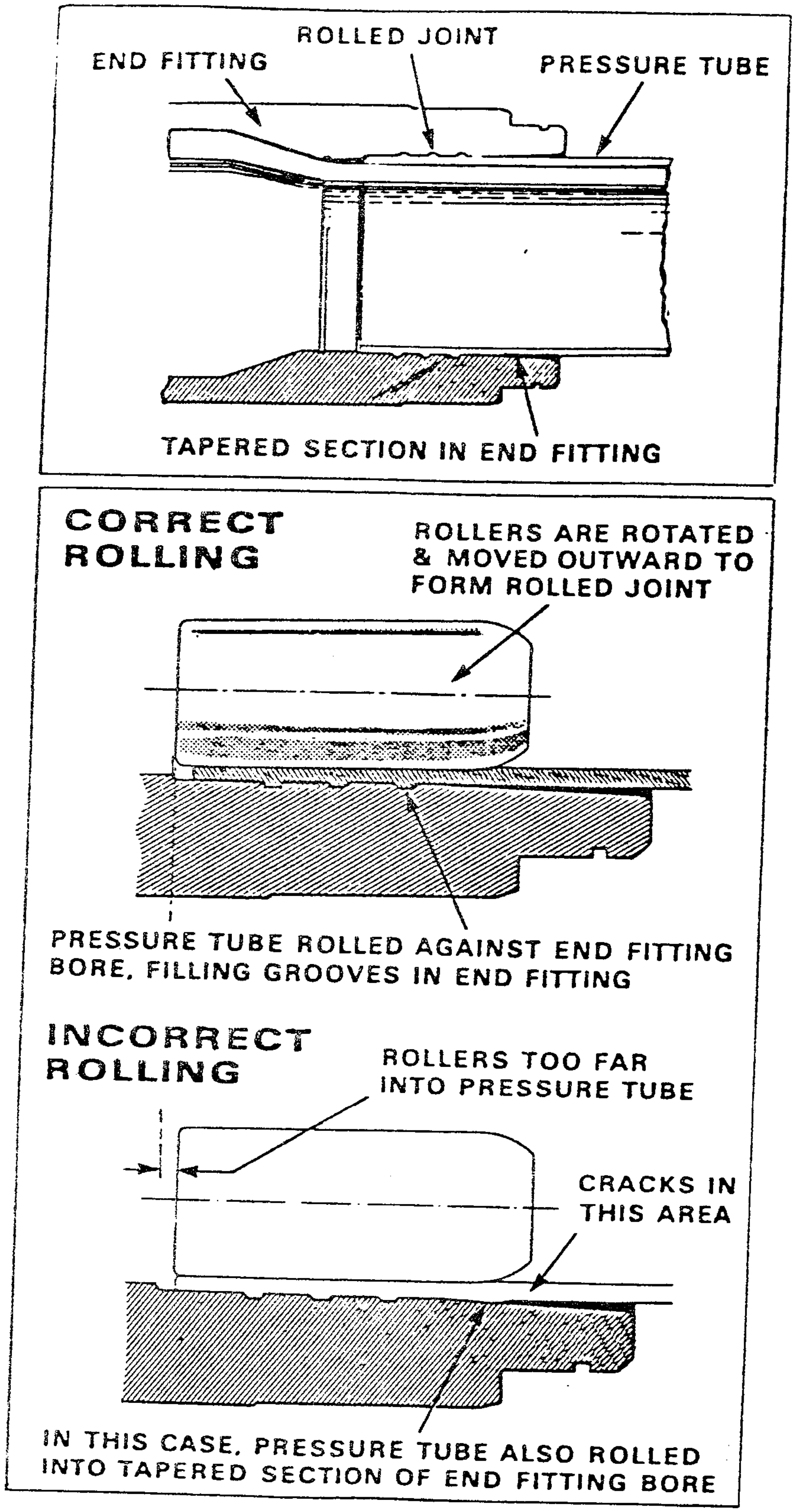
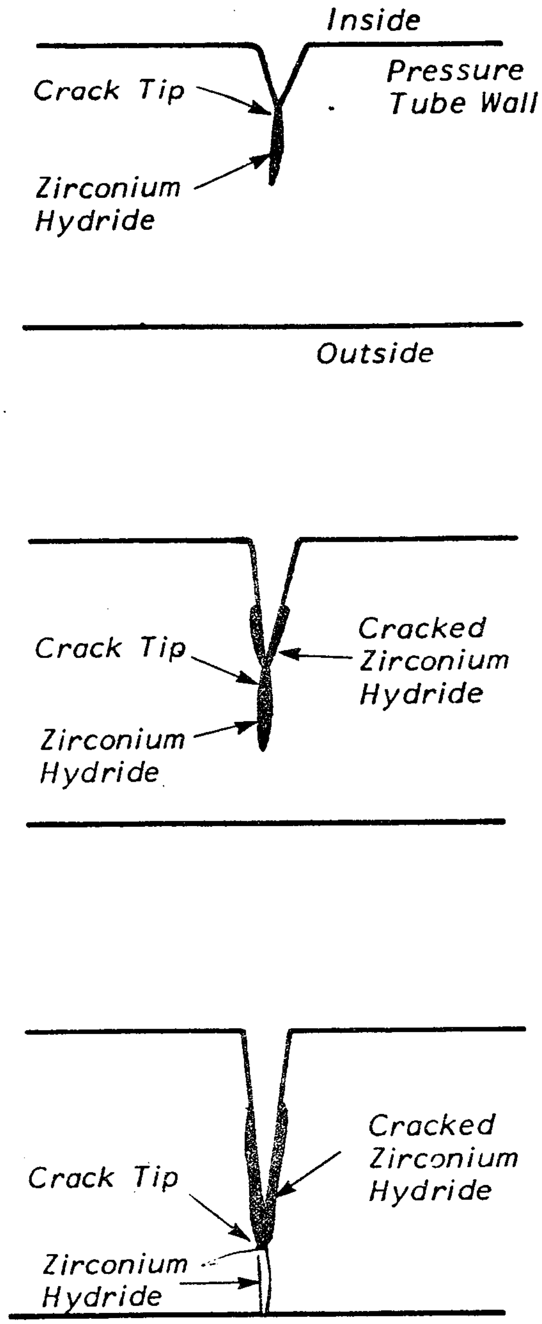
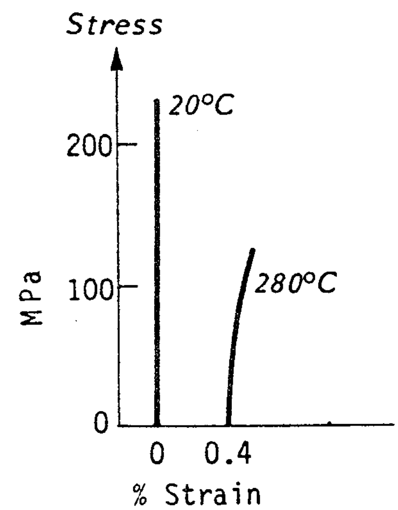


FIGURE 9

SCHEMATIC DRAWING TO ILLUSTRATE CORRECT AND INCORRECT ROLLING OF PRESSURE TUBE INTO THE END FITTING





Stress - Strain Curves for Zirconium Hydride

FIGURE 10

SCHEMATIC ILLUSTRATION OF DELAYED HYDRIDE CRACKING

DOUBLE CONSUMABLE ARC MELT

↓

FORGE AT 1305 K (1030°C)

↓

WATER QUENCH FROM 1305 K (1030°C) AFTER 30 min SOAK

↓

HOT ROLL 1060 $^{+15}_{-30}$ K (785°C) TO 19 mm

↓

HOT ROLL 1060 $^{+15}_{-30}$ K (785°C) TO 3.5 mm

↓

AIR ANNEAL 1060 − 15 K (785°C) 1 h ± 15 min

↓

COLD ROLL TO 2.15 mm

↓

ANNEAL 1005 ± 15 K (730°C) 2 h ± 15 min IN VAC

↓

COLD ROLL TO 1.37 − 1.42 mm

↓

ANNEAL 1005 ± 15 K (730°C) 2 h ± 15 min IN VAC

↓

CALANDRIA STRIP

↓

WESTINGHOUSE CANADA LTD

↓ BRAKEFORM TO ROUND
↓ WELD (TIG)
↓ BEAD ROLL
↓ VACUUM ANNEAL
ZONE ANNEAL
1273 K/3 min (805°C)
UP TO PGSA 5
1013 K (740°C) PGSA5
ONWARDS
↓ SINK 1 − 1.6%
↓ STRAIGHTEN
↓ BELL ENDS

BRISTOL AEROSPACE

↓ BRAKEFORM TO ROUND
↓ WELD (TIG)
↓ BEAD ROLL
↓ VACUUM ANNEAL
(FURNACE)
1005 − 1033 K
(730 − 760°C) − 30 min
↓ SINK ~ 1%
↓ STRAIGHTEN
↓ STRESS RELIEF (725 K − 1 h)
↓ STRAIGHTEN
↓ BELL ENDS

CARPENTER

↓ BRAKEFORM TO ROUND
↓ WELD (TIG)
↓ BEAD ROLL
↓ VACUUM ANNEAL
(FURNACE)
1020 K (745°C)
30 min
↓ SINK ~ 0.5%
↓ STRAIGHTEN
↓ BELL ENDS

FIGURE 11

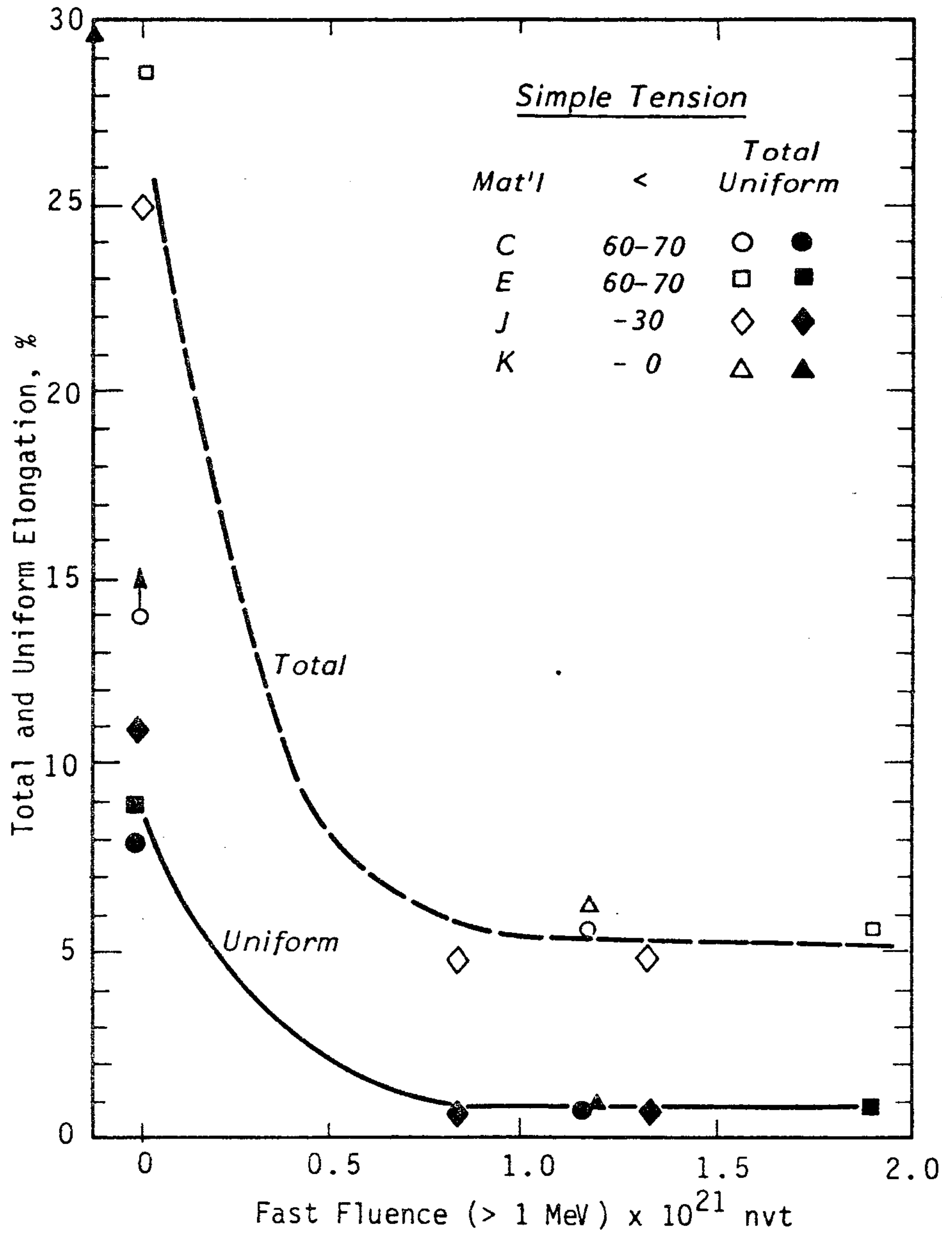
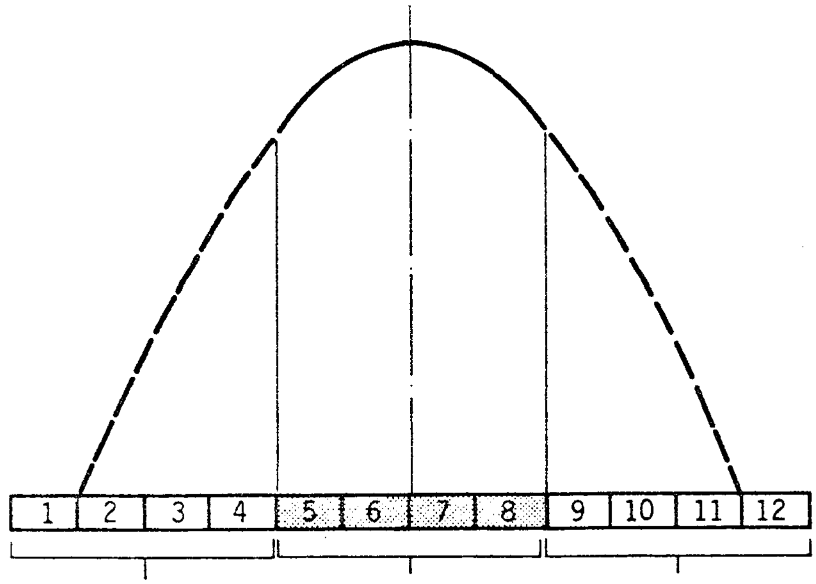


FIGURE 12

TOTAL AND UNIFORM ELONGATION AS FUNCTION OF FAST FLUENCE OF ZIRCALOY-2 WITH VARIOUS TEXTAURES. ALL TESTS WERE MADE AT 250°C AT THE STRAIN RATE OF 0.05/min

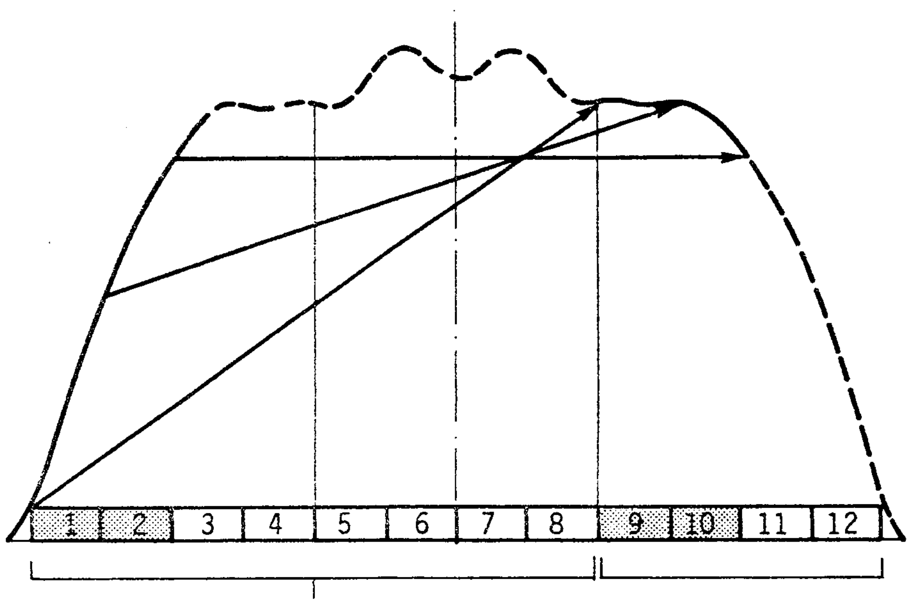


4-Bundle Shift Positions

Flow →

FIGURE

DOUGLAS POINT AXIAL FLUX PROFILE



8-Bundle Shift Positions

Flow →

FIGURE 13

PICKERING AXIAL FLUX PROFILE

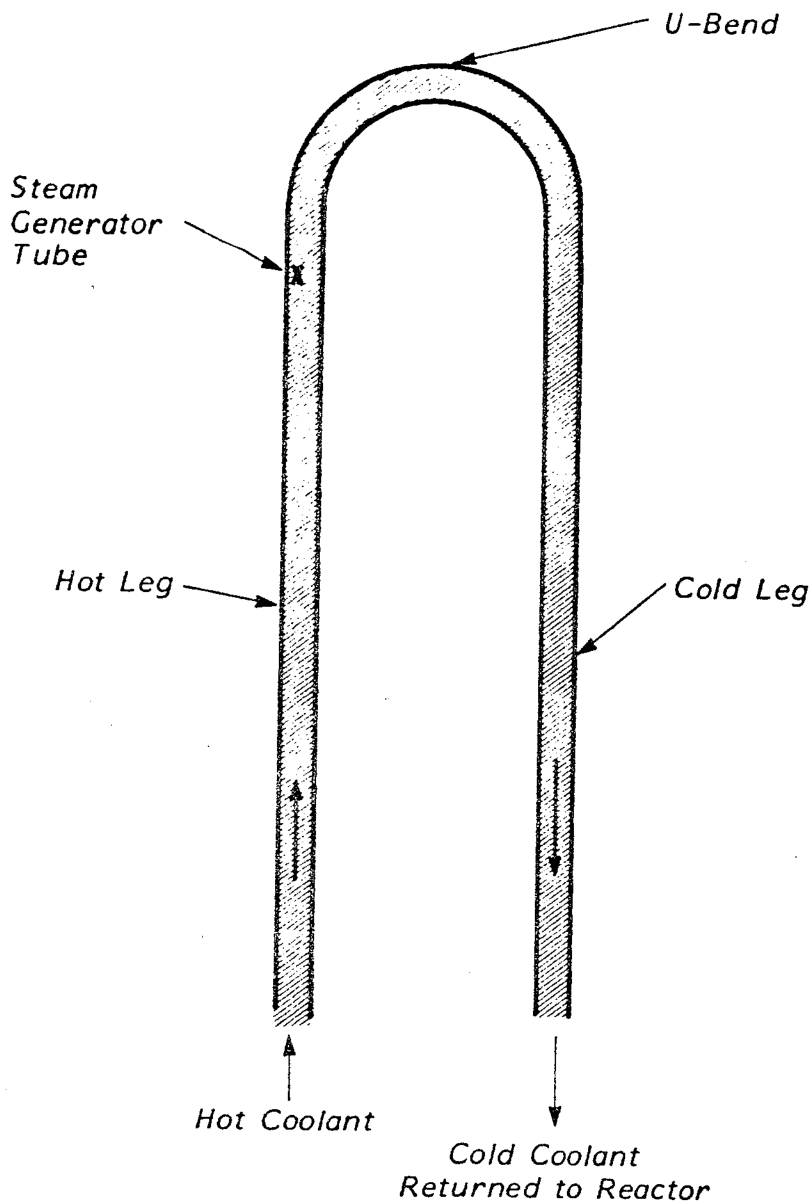


FIGURE 14
SCHEMATIC DRAWING OF A STEAM GENERATOR TUBE