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Evaluation of Concepts for a NET Plasma Exhaust Clean-up System

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Summary

The process steps for the off-gas clean-up and direct recovery of the unburned fuel gases deuterium and tritium are, together with the isotope separation and the fuel preparation, the major subsystems within the fuel cycle of a fusion reactor. In this study a comparison between process concepts largely based on experimental work at KfK and other process alternatives discussed in the literature is carried out and the various options evaluated on the basis of the process requirements for NET I. The recovery of most of the unburned hydrogen with a palladium/silver permeator is selected as a first step, common to all seven concepts. The remaining impurity stream is processed either catalytically, with the help of getters, or by oxidation followed by reduction of the produced water. The physicochemical basis of each process alternative is discussed and the corresponding chemical flow sheets (flow diagrams and material flow tables) are presented. The comparison of the various processes shows that concepts employing getters are unattractive because they produce intolerably high amounts of solid waste. Main drawbacks of process options involving an oxidation step are the non-discriminative oxidation of hydrogen and impurities as well as the non-trivial reduction of the produced highly tritiated water at the required elevated throughput. Advantages of the catalytic process are the production of little solid waste, the low steady state inventory and the comparatively easy scale-up. The catalytic process is therefore considered the most promising option for the development of a fuel clean-up process.

Bewertung von Plasma-Abgas-Reinigungskonzepten für NET

Zusammenfassung

Die Prozeßschritte zur Reinigung des Abgases eines Fusionsreaktors und die unmittelbare Zurückführung der unverbrannten Brenngase Deuterium und Tritium sind neben der Trennung der Wasserstoffisotope und Mischung der Brenngase ein wesentliches Untersystem im Brennstoffkreislauf eines Fusionsreaktors. Hier auf der Basis experimenteller Arbeiten entwickelte katalytische Konzepte zur Abgasreinigung werden zusammen mit Prozeßalternativen aus der Literatur verglichen und auf der Grundlage der Anforderungen von NET I bewertet. Allen sieben Konzepten gemeinsam ist die Abtrennung des Hauptanteils der unverbrannten Wasserstoffisotope mit einem Palladium/Silber - Permeator. Nachfolgend werden die Gase unmittelbar katalytisch aufgearbeitet, mit Hilfe von Gettern prozessiert oder oxidiert und anschließend das entstandene Wasser wieder reduziert. Für jede einzelne Prozeßalternative wird die physikalisch chemische Basis diskutiert und daraus chemische Fließbilder (Fließdiagramm und Tabelle der Stoffströme) abgeleitet. Der Vergleich der einzelnen Prozesse untereinander zeigt, daß Konzepte unter Verwendung von Gettern untolerierbar hohe Mengen an festem Abfall erzeugen und daher nicht geeignet sind. Nachteile der oxidativ arbeitenden Prozesse sind die nicht diskriminierende Oxidation von molekularem Wasserstoff und Verunreinigungen und die keineswegs triviale Reduktion von hochtritiertem Wasser bei dem geforderten Durchsatz. Demgegenüber hat das direkt katalytisch arbeitende Verfahren den Vorteil, daß es nur wenig festen Abfall erzeugt, nur geringe Tritiuminventare aufweist und in der notwendigen Skalierung realisiert werden kann. Diese Prozeßalternative wird daher als die aussichtsreichste im Rahmen der Entwicklung eines Abgasreinigungsverfahrens angesehen.

The work given in this report was carried out during 1988 jointly by the Kernforschungszentrum Karlsruhe (KfK) and Kraftanlagen Heidelberg (KAH), as the industrial subcontractor, within the frame of a contract with the Commission of the European Communities and in close contact with staff members of the NET Team.

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1. Executive Summary

Scope of this work was an assessment of seven process alternatives (A - G) for the processing of the primary vacuum exhaust of the NET I fusion reactor during burn & dwell as well as for the recovery of tritium from waste gases produced during bake-out, glow discharge cleaning (He or D₂), carbonization, or pump down (air or He). The various options were compared on the basis of design requirements as specified by the NET Team as well as on engineering, safety and economic criteria.

A palladium / silver alloy hydrogen permeator, which separates practically all the unburned hydrogen isotopes (deuterium and tritium) from the fuel ash helium and the hydrogenated as well as non-hydrogenated impurities, was selected as a first common step to all alternatives. This step was chosen because permeators

- deliver a highly pure hydrogen stream which can be sent directly into an isotope separation system (ISS),
- operate with a low tritium inventory,
- are fairly insensitive to poisoning by impurities such as helium, nitrogen, carbon monoxide, carbon dioxide, methane and water at temperatures above 300°C (experiments at CEA, JAERI, and KfK),
- produce no process solid waste,
- are used on an industrial scale for hydrogen purification, and
- promote cracking of ammonia into the elements (experiments at JAERI and KfK).

In a next step a nearly quantitative recovery of tritium from the bleed gas of the first permeator (He containing Q₂, CQ₄, Q₂O, CO, CO₂, O₂, N₂ etc. with Q = H, D, T) is required not only from a fuel economy point of view, but also to obtain a waste gas with a detritiation level acceptable to a conventional tritium waste treatment system. For this purpose concepts involving

- the direct gettering of carbon, nitrogen, and oxygen on a hot metal bed followed by a separation of hydrogen and helium via a second permeator or another metal bed,

- the oxidation of all impurities and residual hydrogen into water and carbon dioxide employing either an oxygen donor solid bed or molecular oxygen together with a noble metal catalyst followed by a combination cryotrap/electrolysis cell or a combination water gas shift reaction/permeator, or
- the use of selective catalytic reactions to liberate deuterium/tritium from water (water gas shift reaction) and hydrocarbons (nickel catalyst) in combination with a permeator

were compared.

While it is possible to liberate the hydrogen isotopes from the impurities on a hot metal getter bed in a single reaction step (process concepts B, C and D), problems are expected from the required high temperatures, the large amounts of getter material needed to maintain a high conversion, the potential for getter ignition, the complex reactions occurring between reactive gases and products on the getter surface as well as the frequent renewal and disposal of spent tritium contaminated getter.

Very little solid waste is anticipated from the oxidation/electrolysis cell option (alternative E). Disadvantages of this process concept are the non-specific oxidation of impurities, the cryogenic separation of water and carbon dioxide and the high steady state tritium inventory (cryotrap and electrolysis cell). In addition, technical units still need to be demonstrated and long-term tests with tritium are required.

The electrolysis of water proposed in concept E can be replaced by the water gas shift reaction (alternative F). In the latter case, water is volatilized with an appropriate carrier gas to which carbon monoxide is added in a concentration of $\text{CO}/\text{H}_2\text{O} \approx 1.5$. The large amount of argon sweep gas needed to volatilize the water is considered to be a major drawback of this alternative.

A catalytic process concept (alternative A) based on extensive laboratory work, including investigations with tritium, has been proposed at KfK. Main design guidelines were: i) one process for all exhaust gases, ii) few process steps, iii) low

hold up, iv) low solid waste, v) no high temperatures*), vi) minimum conversion of hydrogen isotopes and/or impurities into water, and vii) few replacement operations.

According to the catalytic concept, essentially all molecular hydrogen isotopes from the exhausts of the reactor operations burn and dwell, bake-out, glow discharge and carbonization are recovered in a highly pure form (99.9999 % purity) using a palladium/silver permeator. Tritiated and non-tritiated impurities are processed in two catalytic reaction steps: i) a nickel catalyst to decompose hydrocarbons and ammonia and ii) a zinc stabilized copper chromite catalyst to convert water into hydrogen via the water gas shift reaction. Hydrogen isotopes liberated by the catalytic reactions are recovered with the help of a second palladium / silver permeator. The detritiated gas is sent to waste.

From complementary R & D work carried out during the course of the study work the following results were obtained:

i) Permeation of hydrogen isotopes through palladium silver membranes

The influence of fusion fuel cycle relevant impurities on the rate of hydrogen permeation through Pd/Ag membranes was investigated over a wide range of temperatures and pressures employing commercial permeators with up to 840 cm² permeation surface area. The permeation coefficient of pure hydrogen through these permeators was measured at temperatures between 100 and 450°C and found to be in good agreement with values determined by other investigators, who used only small Pd/Ag fingers. The influence of N₂, H₂O, CH₄, CO, CO₂, C₂H₆, NH₃, and of mixtures of these gases on the rate of hydrogen permeation through Pd/Ag was studied within the same temperature range given above. The maximum partial pressures of CO, water, and ammonia (the three most reactive species) were 400, 30, and 20 mbar, respectively. The results indicate that none of these impurities will reduce significantly the hydrogen permeation rate at temperatures above 250°C. At temperatures below 250°C only CO will cause a reduction in permeation rate. The data show that poisoning by CO

*) While the temperature required for the operation of the nickel catalyst is relatively high, i. e. ~ 500 °C, it is still within the working range of standard structural materials e. g. SS 316 L.

is reversible. Ammonia is decomposed on the Pd/Ag surface into its elements with a rate constant of 3×10^{15} molec/cm²·s when the liberated hydrogen is continuously removed by permeation.

ii) Water gas shift reaction

Water is converted catalytically into hydrogen via the water gas shift reaction. Several catalysts were compared with respect to their catalytic activity at various temperatures. From the obtained data it was concluded that a zinc oxide stabilized copper chromite catalyst is the most favourable. With this catalyst a conversion of at least 98 % could be achieved at temperatures of approx. 200°C and a CO/H₂O ratio higher than 1.3. The catalyst was found to be insensitive to high partial pressures of water, ammonia and CO (1 bar). The amount of catalyst required was very small.

iii) Decomposition of hydrocarbons and / or ammonia on a nickel catalyst

Numerous experiments were carried out to study the decomposition of methane and ethane on an alumina supported nickel catalyst. Both gases are decomposed into molecular hydrogen and carbon. Rate constants and the corresponding activation energy for the decomposition reaction of methane were determined within the temperature range 450 - 580°C. The rate of the methane decomposition was found to be first order in methane and the reaction reversible.

Recent work has concentrated on studies with gaseous mixtures. In particular, the decomposition of methane on a nickel catalyst was investigated in the presence of carbon oxides (typically at the same initial partial pressure as methane) and water. It was observed that high partial pressures of carbon oxides have a favourable effect on the thermodynamic equilibrium of the methane cracking reaction and, within the experimental error, do not influence the rate of the methane decomposition. The catalytic recovery of bonded hydrogen isotopes from hydrocarbons requires no preadjustment of the chemical composition of the gas to be processed.

In experiments with deuterium it was shown that the carbon deposit contains no more than 0.2 atom % of hydrogen. From these experiments the tritium inventory of the catalyst is estimated to be of the order of 0.7 g tritium per kg catalyst.

Typical plasma contaminants like polytritiated methane and polytritiated ammonia were synthesized on a $(1.11 - 1.85) \cdot 10^{11}$ Bq scale; the selfradiolysis constant of both species was determined and found to be $t_{1/2} = 250$ and 550 h respectively. The catalytic cracking reactions of tritiated methane and tritiated ammonia followed the behaviour anticipated from cold experiments.

The amount of catalyst necessary for the normal burn and dwell operation of NET (370 d), obtained from runs with a NET relevant throughput, was estimated to be < 10 kg.

Another concept that was examined uses a regenerable oxygen donating solid bed for the complete oxidation of all impurities (alternative G). While under these conditions the supply of molecular oxygen is avoided, the concept does not appear adequate for process gases containing high partial pressures of carbon monoxide or hydrogen. More experiments are needed for an evaluation of this option.

From the comparison of processes it is concluded that all concepts using selective catalytic reactions for the recovery of tritium from the impurities in the bleed gas of the main permeator (options A and G) are to be favoured because they i) involve little (or no) consumption of solid material, ii) are in line with the process requirement of minimization of solid tritium containing waste, and iii) oxidation / reduction cycles (formation / decomposition of water) are mostly avoided by reaction routes proceeding via the direct liberation of molecular hydrogen from the impurities. Tritiated methane and other hydrocarbons are cracked into carbon and molecular hydrogen at about 500°C on a supported nickel catalyst in combination with a palladium / silver permeator. The amount of catalyst required is moderate, it is further reduced by the carbon volatilization occurring by reaction with carbon dioxide.

The water gas shift reaction has great potential for application in the fuel clean-up process. It has been investigated extensively in laboratories of JAERI, CFFTP, and, to a lesser extent, at KfK. Significant industrial experience with this reaction is also available. The reaction takes place at low temperatures (negligible permeation losses), requires very little catalyst (minimal solid waste), has a low tritium inventory, is insensitive to the presence of methane (experiments at KfK) and proceeds at a sufficiently high conversion and rate.

2. Introductory remarks

The Next European Torus (NET) is conceived as an experimental fusion reactor to follow JET after completion of its mission within the European Fusion Programme. The main objective of NET will be to demonstrate the feasibility of fusion from the point of view of physics and technology. In this context, NET will be concerned with the production of extended plasma burn pulses, the problem of tritium production in the blanket, all aspects concerning the reactor refueling and the development of a plasma exhaust treatment. A schematic diagram of the NET reactor reference concept is shown in Fig. 1. It is anticipated that the design work for the basic NET machine will be started in 1989 and that the construction and assembly phase will be completed within the period 1994 - 2000 /1/.

The fuel to be used during the NET operation phase III consists of a nearly equal molar mixture of deuterium and tritium. Phase III has a duration of 7 years and involves a total of 300 days of operation. The partial burning of the fuel in the fusion reaction chamber could lead to a helium concentration of up to 5 mol % and to impurity concentrations - produced mainly by plasma / wall interactions - totaling approx. 2.5 mol %. The nature and amount of the impurities are still controversial. They will largely be influenced by final design decisions concerning arrangement and materials of the reactor wall, divertor, limiter, etc. . Based on present knowledge, the impurities include compounds like hydrocarbons, oxides of carbon, nitrogen, oxygen, ammonia, water, and of course the fusion 'ash' helium. Protium is also an unwanted species in the fuel. All impurities other than helium and protium must be removed to levels below a total of 1 ppm before the fuel can be processed by the isotope separation system. Higher impurity concentrations can lead to freeze-out in the distillation columns.

Moreover, because impurities in the reacting plasma cause plasma cooling by radiation losses, their build-up is of major concern to the operation of fusion reactors. Fast build-up of impurities will limit fusion reactors to short pulse mode of operation and reduce the efficiency of power production. Impurities must be removed by passing the gas through an external fuel clean-up system. After cleaning the gas, it is isotopically readjusted and recycled into the toroidal reactor chamber.

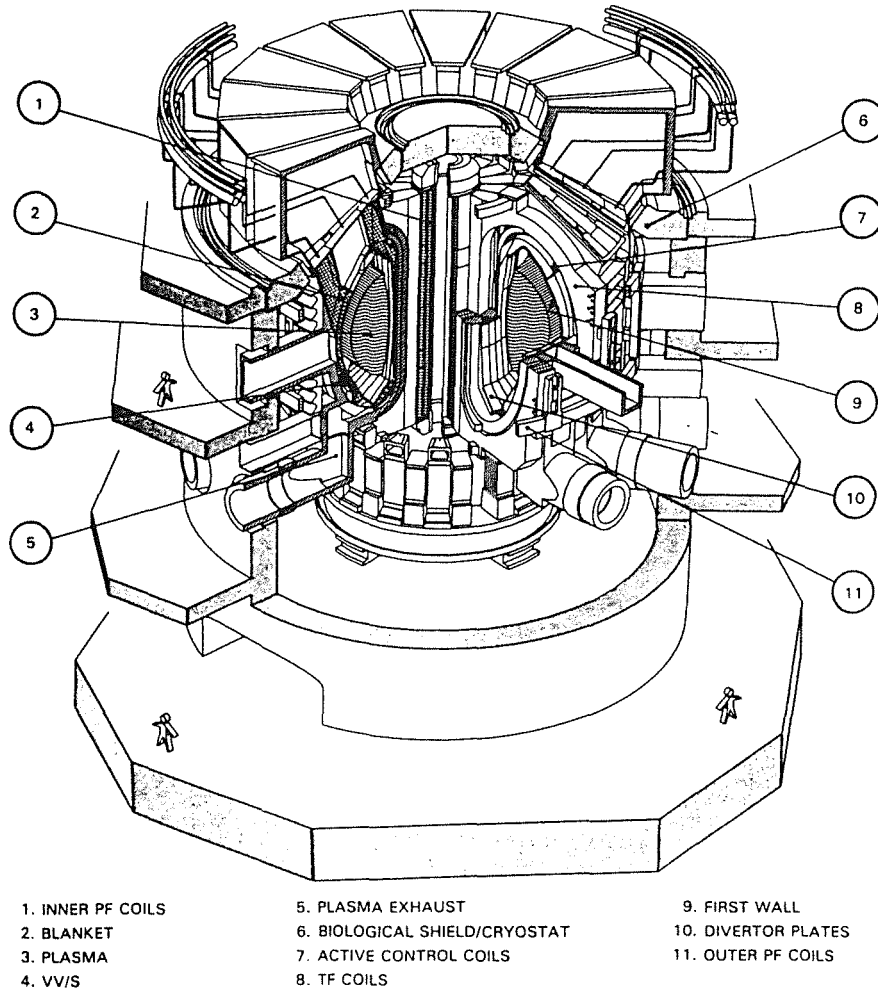


Fig. 1 Schematic diagram of the NET reactor /1/

The fuel processing loop comprises the plasma chamber evacuation, the plasma exhaust transfer, a surge tank, the fuel impurity removal, the impurity processing, the isotope separation system, the fuel supply and the fuel storage. The nominal flow rate in the fuel processing loop is about 500 mol / day, the total tritium inventory is of the order of 200 g.

This paper deals with the impurity removal and processing system only. Seven process concepts were selected for a comparative study. Within the scope of this study all requirements for a detailed engineering layout of the selected fuel clean-up system will be defined. An analysis of various concept alternatives will be carried out from the point of view of the Design Requirements for NET I.

3. Main Design Requirements for NET I /2/

The design requirements as well as the functional requirements for the NET I fuel processing loop are given in the NET document NE/86/S-033 from Nov. 6th 1987. A summary of the performance requirements (PR) relevant to the fuel clean-up system is given below.

PR 1 Process primary vacuum exhaust during burn and dwell periods. Typically (600 MW Fusion Power):

- 20 moles/h of DT
- 1 moles/h of He
- 0.5 moles/h of impurities (CO, CO₂, N₂, CQ₄, NQ₃, N₂, O₂, Q₂O, where Q = H, D or T)

The (He + impurity)/DT ratio depends primarily on the divertor efficiency. The impurity concentration may be up to 10 x higher in outgassing from 1st wall protection tiles during prolonged "down time".

PR2 Process tritium in waste gas from Torus

(a) under normal pumpdown from atmosphere

- 40 kmoles of air or He in 0.3 h (volume)
- 5 moles of water vapour, air 3.0 h (surface)
- frequency 2/mo.

(b) during bake-out

For POCO AXF-5Q graphite baked to 750 °C after 5 days exposure to air approximately 25 moles of impurity may be recovered over 100 hours with the following approximate composition /3/:

H ₂ (DT)	9.6 %
H ₂ O (DTO)	48.5
CO	17.6
CO ₂	10.9
hydrocarbons	13.3

(c) to maintain negative pressure during loss of vacuum condition (100 cm² break)

- multiple breaks possible
- 1 kmole of air or He in 1.0 h per window break
- design frequency 1/a.

(d) during glow discharge cleaning (steel)

- 65 moles/h of D₂
- 0.5 moles/h of impurities
- GDC duration \leq 150 hours max. following systems situation (1/mo)
- frequency 1 d > 2 h.

(e) during carbonization (if used)

- 55 moles/h of D₂
- 10 moles/h of CD₄
- carbonization duration \leq 8 hours
- frequency 1/mo

PR3 NBI and pellet injector cryopanel loads.

PR4 Process NBI neutralizer flow.

PR5 Process dilute T in D and H Streams from blanket or coolant reprocessing.
Typically:

- 10 g T/day average (peak 40 g T/day)
- T: (D + H) = 1 : 100 - 1000
- He: T = 1 : 100 (decay He)
- impurities \leq 10 ppm.

PR 6 Provide pure D₂, T₂ and DT for pellet injection NBI and gas puffing fuelling equipment

- appm impurity content in pellet injector feed
- < 100 appm impurity content in gas puffing and NBI feed
- < 1000 ppm T in D₂ to NBI
- Deliver 21 g moles/h of DT + D₂ + T₂.
- Proportions are undefined, but D : T = 1 : 1 in plasma.

PR 7 Reject a minimum of tritium to the tritium waste treatment system as solid, liquid or gaseous wastes. The volume of solid wastes produced during operation and maintenance is to be minimized. Tritium extraction from components for repair or disposal is an important design consideration. Total non-recoverable tritium in wastes is to be ≤ 0.01 g/d (maximum) ≤ 70 μ mol/h.

PR 8 The product of failure rate leading to a fuel loop outage greater than 8 hours, and the mean down-time for the fuel-processing loop should not exceed 10⁻³. Repair time, or time to remove and replace major redundant components, should be minimized. Individual system elements may have lesser availabilities if the above overall target can be met without excessive impact on process inventories. During a single pulse (≤ 1000 s) it must be possible to sustain fuel flow to and from the plasma to prevent disruptions.

PR9 Tritium inventory of any component, or group of components, which could be released by a single process failure must not exceed 150 g.

PR 10 Gas is to be received from the plasma system and delivered to the fuel introduction systems at 300 K, and < 1000 mbar. Maximum working pressure in the loop should be ≤ 2 bar.

PR 11 All components should be maintainable in a glove box (caisson) or other remote handling environment. In general, double confinement is to be provided for tritium handling processes. Secondary confinement must be maintainable at all times during maintenance.

PR 12 The need to ensure an operation compatible with tritium requires the avoidance of organics in contact with the process streams containing tritium. Any lubricants used must be tritium compatible to minimize maintenance.

The overall external leak rate into the fuel processing loop shall not exceed 10^{-6} mbar · l/s at 1 bar He differential pressure.

PR 13 It will be necessary to use materials which minimize retention and generation of impurity gases and protium.

No outleakage from the fuel processing loop is permitted under normal operation.

PR 14 The fuel processing loop must have a design lifetime of 15 years.

An inventory of 500 g in interim storage (fresh fuel) is to be initially assumed.

PR 15 The design should accommodate changes of ± 50 % in the nominal process conditions specified in the requirements.

PR 16 A bake-out temperature of 150 °C and helium purge capability are required for components where practical.

PR 17 Loose contamination (dust) shall be prevented from migrating through the process system.

4. Description of the selected process concepts

Altogether seven process concepts (A-G) were selected for comparison and evaluation (see Table 1). Process concepts B /4/, C /4/, D /5/, E /6/, and F /7/ were taken from the literature, process concepts A and G were proposed by KfK /8/.

The latter two concepts are based, to a large extent, on laboratory work carried out at KfK.

In this section a detailed description of each of the processes is given. Common to all seven processes is a palladium/silver permeator (K1), which serves to recover highly pure hydrogen isotopes from the impurities. Also common to all processes are the buffer vessels at the facility inlet (B1) and the facility outlet (B3) as well as the pumping system (P1) on the permeate side. Pumps and other components indicated in the chemical flowsheets are, in size and quantity, of symbolic nature to facilitate the comparison of the processes.

The design of the vacuum and transfer system will be carried out during phase II of this study, once type and configuration of the permeators and reactors are defined. For permeator K1 a two (or three) stage metal bellows compressor roughing pump combined with a NORMETEX dry pump, which can provide vacua down to 10^{-3} mbar at sufficient pumping speed, appear to be suitable. In fact, tests with a 15 Nm³/h NORMETEX dry pump combined with a diaphragm compressor of 7 l/min gave ultimate vacua of 5×10^{-4} and 10^{-3} mbar with nitrogen and helium respectively, at a constant outlet pressure of 1000 mbar. In case a second permeator is used (K4), an independent pump combination may be necessary.

Hydrogen isotopes removed from the plasma exhaust gas via a palladium/silver diffuser permeate in a highly pure form and may therefore be sent directly into the isotope separation system.*) The mass flow through the diffuser remains constant as long as the pressure difference between the high pressure side and the low pressure side is not changed. The required total membrane surface is of the order of ≤ 1 m². Operation of a palladium/silver diffuser has been demonstrated at TSTA, Los Alamos for a period of 450 days with tritium /9/. Arrhenius equations for the permeation rate of hydrogen through palladium/silver membranes of various surface areas are given in Table 2.

* According to the Pionics Co., Japan a hydrogen purity of 99.9999 % can be achieved.

Table 1: Evaluated Processes

Process	Definition
A	Catalytic conversion process (Cu chromite and Ni/Al ₂ O ₃) combined with Pd/Ag permeation
B	Gettering process combined with Pd/Ag permeation
C	Process entirely based on gettering
D	Oxidation process (CuO), combined with molecular sieve sorption and water reduction with a getter
E	Catalytic oxidation process (Pd/Pt) combined with cryo-trapping and electrolysis
F	Catalytic conversion process (Pd/Pt and Cu chromite) combined with Pd/Ag permeation
G	Catalytic conversion process (CuO and Cu chromite) combined with Pd/Ag permeation

Table 2:

**Arrhenius equations for the hydrogen permeation rate
through a palladium/silver diffuser**

Pd/Ag membrane		Q_H cm . mol / cm ² . min . kPa ^{1/2}	Author
surface area cm ²	thickness cm		
0.24	0.013	$1.28 \cdot 10^{-6} \exp(-1572/RT)$	Ackermann et al. 1972
4.37	0.016	$7.31 \cdot 10^{-7} \exp(-1370/RT)$	Yoshida et al. 1983
9.4	0.025	$9.40 \cdot 10^{-7} \exp(-1400/RT)$	Chabot 1986
289 - 578	0.020	$1.00 \cdot 10^{-6} \exp(-1300/RT)$	this work

Recent experiments at KfK on the effect of the carrier gases He, N₂ and CH₄ as well as of the impurities CO, CO₂, and H₂O on the permeation rate coefficient Q_H of hydrogen through a Pd/Ag diffuser are given at several temperatures in Tables 3 - 5. These short-term experiments show that fairly high levels of impurities are tolerated by a permeator without loss of permeation efficiency. Contamination by oil is not expected because only oil-free pumps will be employed in the fuel processing system. In case of an oil contamination, however, the permeator can be regenerated by a treatment with oxygen or air at 300 - 450 °C.

Ammonia, if present, is cracked into the elements on the surface of the palladium/silver permeator according to the reaction



Due to the high catalytic activity of palladium for the reaction



oxygen is anticipated to be only a minor impurity in the off-gas of permeator K1 /10/.

The selected follow-up processes can be divided into two groups:

- processes avoiding the conversion of hydrogen isotopes into water, e.g. processes A, B, and C, and
- processes involving the conversion of free and bonded hydrogen isotopes into water, e.g. processes D, E, F, and G.

They can further be subdivided into processes based entirely on catalytic reactions like process A or entirely on gettering reactions like process C or combinations of these.

Table 3: Effect of carbon monoxide on the permeation coefficient of hydrogen at an initial pressure of 20 kPa diluted in He or N₂ through a commercial Pd/Ag membrane diffuser operated at 450 °C

carrier gas	P _{carr. gas} kPa	impurity	P _{imp} kPa	Q _H · 10 ⁷ mol · cm/cm ² · min · kPa ^{1/2}	Nr. of runs
N ₂	82.6	--	--	3.60 ± 0.29	2
He	82.6	--	--	3.17 ± 0.27	3
He	80.6	CO	2.07	3.20 ± 0.13	2
He	72.3	CO	10.03	2.71 ± 0.07	2
He	62.0	CO	20.7	3.27 ± 0.31	5

Table 4:

Permeation coefficient of hydrogen diluted in various carrier gases through a commercial Pd/Ag membrane diffuser operated at 400 °C

Pd/Ag membrane area	P_{H_2}	carrier gas	$P_{carr. gas}$	impurity	P_{imp}	$Q_H \cdot 10^7$
cm ²	kPa		kPa		kPa	mol · cm/cm ² · min kPa ^{1/2}
289	36.2	He	87.8	--	--	2.45
578	38.2	He	86.8	--	--	2.13
289	20.7	He	82.6	CO	2.07	2.36
289	20.7	He	80.6	CO	20.7	2.19
289	20.7	He	70.0	CO ₂	20.7	2.14
578	20.7	CH ₄	82.6	--	--	2.36

Table 5: Influence of water on the permeation rate of hydrogen ($P_{H_2} = 25$ kPa) diluted in helium ($P_{He} = 75$ kPa) through a palladium/silver membrane (surface area = 289 cm²) at various temperatures.

Temp. °C	$P_{H_2O}^*$ kPa	$Q_H \cdot 10^7$ mol . cm/cm ² . min . kPa ^{1/2}
400	--	2.96
400	--	2.66
400	0.49	2.90
400	1.23	2.78
400	2.46	2.60
300	--	2.46
300	--	2.52
300	0.49	2.46
300	1.23	2.48
300	2.46	2.36
200	--	1.57
200	--	2.44
200	0.49	1.97
200	1.23	1.98
200	2.46	2.08

* Volume of closed loop = 5.6 l

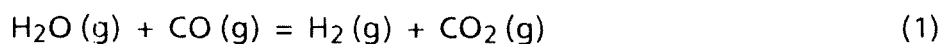
In the following a description of each process is given together with a simplified chemical flow sheet and an equipment list. A quantitative evaluation of each process, mainly based on thermodynamic considerations, is presented in the form of tables for the process conditions

- normal burn and dwell
- glow discharge (Appendix)
- bake-out (Appendix)
- carbonization (Appendix).

4.1 Description of Process A

In this process hydrogen isotopes are recovered directly from the impurities by catalytic reactions without intermediate conversion into water.

In a first step water vapor is reduced by carbon monoxide over a ZnO-stabilized copper chromite catalyst according to the reaction



which is well known as the water gas shift reaction. In industry this chemical conversion is used for the large scale production of hydrogen from water /11/. The application of this reaction to the recovery of tritium in the fusion fuel clean-up system has been reported in previous studies /7, 12/.

Even though the rate of reaction (1) increases with temperature, low catalyst temperatures are preferred because of thermodynamical considerations and the fact that permeation losses are minimized under these conditions. In experiments carried out at KfK with a commercial zinc oxide stabilized copper chromite catalyst (Niki Co, Japan) it was shown that the optimal temperature range of operation of the catalyst is 150 - 250 °C. The addition of methane to a helium carrier gas containing the reactants CO and H₂O was found to have no effect on the chemical equilibrium.

Reaction (1) involves no change in volume and therefore yields do not depend upon pressure. For a high conversion a ratio CO/H₂ \leq 1.5 is required /12c/.

Screening tests were carried out at KfK with catalysts of the following composition:

- (A) CuO - ZnO - Al₂O₃
- (B) Cr₂O₃ - CuO
- (C) Fe₂O₃ - Cr₂O₃
- (D) CoO - MoO₃ - Al₂O₃
- (E) CuO - ZnO - Cr₂O₃ - Al₂O₃

In all experiments a gaseous mixture of 20 mbar H₂O, 60 mbar CO and 920 mbar He was employed. An almost complete conversion of water into hydrogen was only attained with catalyst B at 160 °C and catalyst E at 150 °C. Of these two catalysts type E is favoured out of kinetic considerations. Only very small amounts of catalysts are required for the operation time of NET I (i.e. of the order of a few 10 g).

The equilibrium conversion EC

$$EC = (H_2)_{\text{prod}} / (H_2O)_{\text{init}}$$

which is very high at 150 - 200 °C, can be increased to practically 100 % by recycling once under continuous removal of hydrogen by selective permeation with an integrated Pd/Ag-diffuser.

Only small tritium permeation losses are expected from a water gas shift reactor, because of the comparatively low reaction temperature ($T \leq 250^\circ\text{C}$) and the small size of the catalyst reactor. The tritium inventory in the catalytic reactor, estimated from the solubility of hydrogen in the catalyst and the reactor wall, is considered to be low /12b,c/

Methane is subsequently cracked into the elements on a nickel catalyst (Ni on γ -Al₂O₃):



at a temperature of about 500 °C. The reaction has been thoroughly studied by a number of investigators employing a variety of catalysts /13-16/. The conversion increases with decreasing methane partial pressure /16/.

The steady state inventory of tritium in the nickel catalyst reactor is determined by the amount of tritium in the reactor wall, the amount dissolved in nickel, the amount present in the gas phase, and the amount trapped in the carbon deposit produced during methane cracking.

Whereas the inventory in structural materials, that in the gas phase, and that dissolved in nickel/ Al_2O_3 (of the order of $1.4 \cdot 10^7$ Bq tritium/g of Ni) can easily be estimated from literature data, the hydrogen trapped in the carbon deposit must be determined experimentally. To this effect two different tests were carried out at KfK with 10 g Ni catalyst containing the total carbon deposit from the cracking of 1.08 mol of CH_4 :

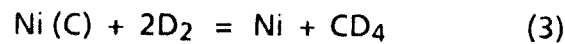
- 1) Elemental analysis of carbon and hydrogen
- 2) Regeneration of the catalyst by treatment with D_2 and analysis of the products (HD , CD_3H and CD_4) by gas chromatography, mass spectrometry and fourier transform infrared spectroscopy.

The results from these experiments conclusively indicate that the steady state tritium inventory in the Ni catalyst reactor is similar to that observed in other solid catalyst beds and that therefore the process requirement that the total tritium inventory in a single component should not to exceed 150 g can be met (PR9).

The catalyst consumption without loss of catalytic activity was found to be

$$< 4 \text{ g catalyst / mol CH}_4$$

This "consumption" of catalyst *) can be considerably reduced if the catalyst is regenerated with deuterium according to the reaction



Several authors have investigated the gasification of carbon catalyzed by nickel metal as well as by nickel catalysts /17-20/. As shown by the experimental results in Table 6, methane cracking followed by carbon gasification can be carried out quantitatively without loss of catalyst effectiveness a number of times. For catalyst regeneration in a fusion fuel clean-up system deuterium should be employed.

To reduce the formation of CTD_3 produced from the interaction of deuterium with a tritium contaminated catalyst, swamping at temperatures below that of carbon gasification (i.e. $\leq 400^\circ\text{C}$) is proposed.

Even without regeneration the total catalyst consumption is low, i.e. for the total normal burn and dwell operation of NET I (370 d) an amount of only ≤ 11 kg Ni catalyst is estimated to be necessary.

By recycling the process gas over the Ni catalyst and simultaneously removing the hydrogen produced by cracking of methane with a palladium/silver permeator it is possible to reduce the methane concentration to practically zero. This high degree of conversion can be achieved with low number of cycles, because the calculated conversion per passage over the catalyst rapidly increases as the total pressure (methane + hydrogen) decreases (see Table 7). The equilibrium constant K_p for reaction (2) is given by the relation

$$K_p = \frac{P_{\text{H}_2}^2}{P_{\text{CH}_4}} \quad (4)$$

* The catalyst is actually not consumed, only its activity is reduced by the carbon deposit.

Table 6: Repeated cracking/regeneration cycles on 10 g Ni catalyst

CH ₄ mbar	He mbar	Temp. °C	% cracking +	% regeneration *
329	705	460	91.1	98.5
341	711	470	98.8	91.7
339	811	470	99.4	n.d.
336	775	460	99.9	100
331	752	490	93.7	100
333	773	490	99.4	100
321	776	490	95.8	100
330	786	490	97.0	100
210	800	490	96.9	100
205	801	490	99.1	100
107	900	490	98.1	100
312	701	490	99.3	100
257	751	500	98.1	100
153	859	490	96.5	100
302	698	490	97.6	100
302	706	500	99.0	100
416	586	490	97.1	100
417	594	490	95.4	

n.d. = not determined

+ = no attempt was made to crack methane quantitatively in these experiments

* = the % regeneration was estimated from the amount of methane obtained by carbon gasification

Table 7: Methane cracking efficiency at 500 °C
(all pressures are given in bar)

cycle No.	pH ₂ init.	pCH ₄ init.	pH ₂ final	pCH ₄ final	actual conversion	total conversion
1	0.0300	0.3000	0.2672	0.1814	39.53 %	39.53 %
2	0	0.1814	0.1863	0.08824	51.36 %	70.59 %
3	0	0.08824	0.1123	0.03208	63.64 %	89.31 %
4	0	0.03208	0.05096	0.006601	79.42 %	98.00 %
5	0	0.006601	0.01242	0.0003920	94.06 %	99.87 %
6	0	0.0003920	0.0007809	0.000001550	99.60 %	99.9995 %

p_{CH_4} and p_{H_2} being the partial pressures of methane and hydrogen respectively.

The value obtained for $\log (1/Kp)$ [bar⁻¹] at 500 °C is 2.542.

The partial pressure of hydrogen p_{H_2} can be calculated from the initial hydrogen pressure and that produced from the cracking of methane employing the expression

$$p_{\text{H}_2} = p_{\text{H}_2}^i + 2 (p_{\text{CH}_4}^i - p_{\text{CH}_4}) \quad (5)$$

where $p_{\text{CH}_4}^i$ is the initial partial pressure of methane.

Combining equations (4) and (5) one obtains

$$p_{\text{H}_2}^i + 2 (p_{\text{CH}_4}^i)^2 / p_{\text{CH}_4} - \frac{1}{kp} = 0 \quad (6)$$

From this equation the equilibrium partial pressure of methane given in Table 7 as $p_{\text{CH}_4}^{\text{final}}$, can be easily obtained by Newton iteration, if $p_{\text{H}_2}^i$ and $p_{\text{CH}_4}^i$ are known.

The presence of CO or CO₂ in the helium/methane gas mixture is not anticipated to be a problem. During methane cracking CO will partially disproportionate according to

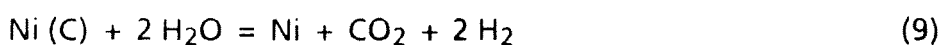


or be converted into methane and water according to



/21/. The products from reaction (7) and (8) are identical with the main components of the process gas.

Another reaction that has been observed is /17/



Therefore, residual water will be converted into hydrogen by reaction with the carbon deposit on the nickel catalyst.

Recovery and recycling of molecular hydrogen initially present in the off-gas from permeator K1 together with the hydrogen liberated from water and hydrocarbons in reactions (1) and (2) occurs with the help of a second permeator K4. Permeated gas is recycled into the main purified fuel stream.

Experimentally it was shown at KfK that at 400 - 450 °C impurity concentrations of up to

40 mol % CO
40 mol % CO₂
10 mol % H₂O

have no significant influence on the permeation coefficient of hydrogen. The hydrogen permeation coefficient through a palladium/silver membrane is essentially not dependent upon the type of carrier gas employed, i.e. He or N₂ (see Tables 2 - 5).

Although methane can be decomposed quantitatively by recycling the gas over a nickel catalyst combined with a palladium/silver permeator in a closed loop operation, a once-through process is also possible. Such a process may either consist of a direct combination catalyst/permeator or of a cascade of catalyst beds with permeators in between.

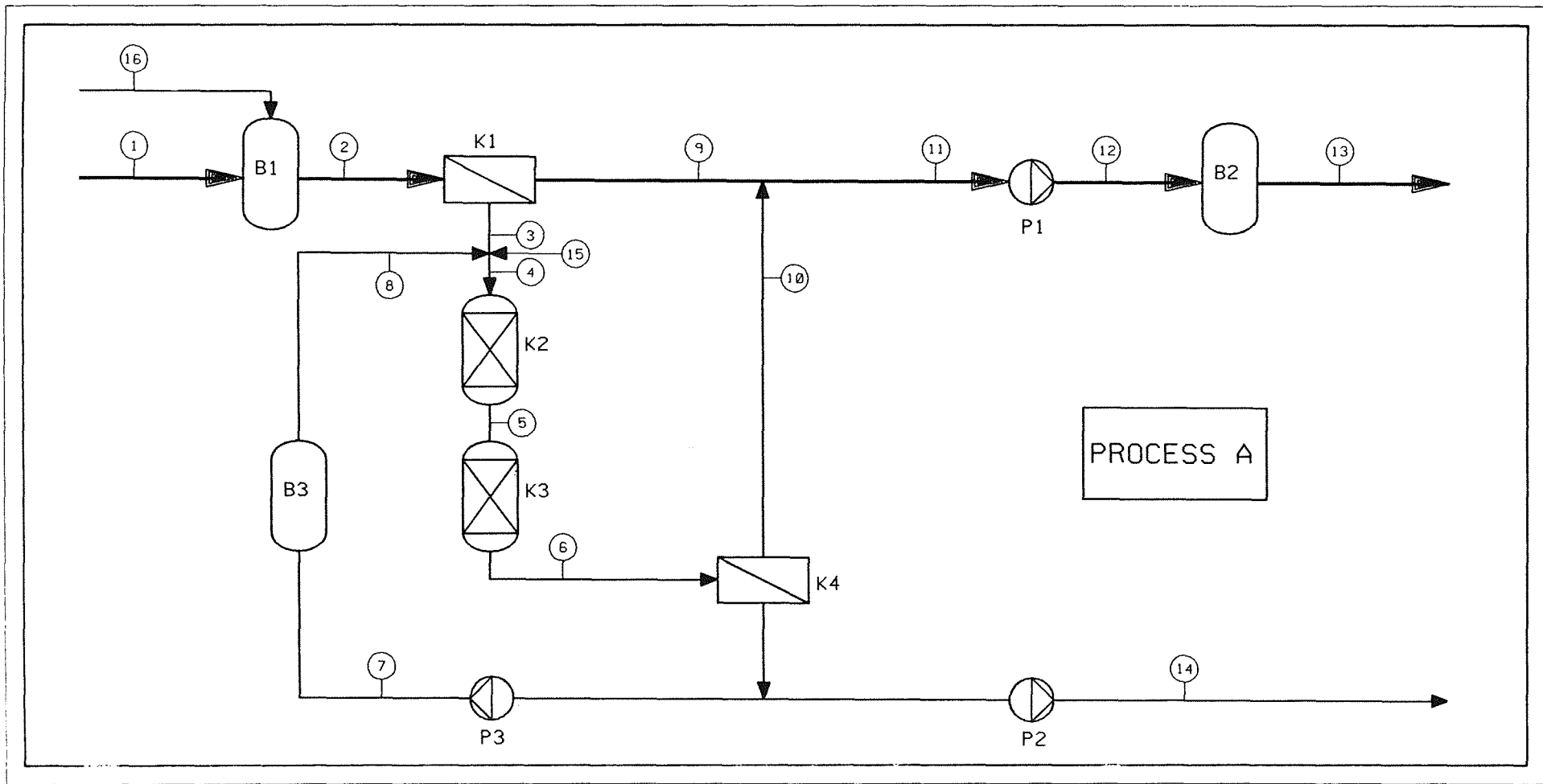


Fig. 2: Chemical flowsheet for process A

EQUIPMENT LIST OF PROCESS A

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25			
K1	Mainstream Pd/Ag permeator	min.350	100		
P1	Main vacuum pump				
B2	Purified fuel collection tank	25	< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25			
K2	CuO/Cr ₂ O ₃ /ZnO catalyst	170	< 100		
K3	Ni-catalyst	500	< 100		
K4	Pd/Ag-permeator	min. 350	100		

Table 8: Streams / flows for process A (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	20.00	20.00	0.03	0.03	0.077	0.681			20.045	0.681	20.726			0.00					
He M/h g/h g/	1.00	1.00	1.00	1.00	1.00	1.00			0.00	0.00	0.00			1.00					
CO ₄ M/h mg/h —	0.30	0.30	0.30	0.30	0.30	0.00			0.00	0.00	0.00			0.00					
CO M/h mg/h —	0.05	0.05	0.05	0.05	0.003	0.00			0.00	0.00	0.00			0.00	0.00				
CO ₂ M/h mg/h —	0.05	0.05	0.05	0.05	0.097	0.10			0.00	0.00	0.00			0.10					
O ₂ O M/h mg/h —	0.05	0.05	0.051	0.051	0.003	0.00			0.00	0.00	0.00			0.00					
O ₂ M/h mg/h —	0.001	0.001	0.00	0.00	0.00	0.00			0.00	0.00	0.00			0.00					
N ₂ M/h mg/h —	0.001	0.001	0.026	0.026	0.026	0.026			0.00	0.00	0.00			0.026					
NO ₃ M/h mg/h —	0.05	0.05	0.00	0.00	0.00	0.00			0.00	0.00	0.00			0.00					

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS A

STREAM/

FLOW No.

1 NET normal burn and dwell composition

-	DT (50:50)	20	mol/h
-	He	1	mol/h
-	impurities total	0.5	mol/h

Assumptions as to impurities:

-	CQ ₄ is the dominating constituent	0.3	mol/h
-	O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible		
-	CO	0.05	mol/h
-	CO ₂	0.05	mol/h
-	Q ₂ O	0.05	mol/h
-	NQ ₃	0.05	mol/h
	(Q = DT)		

2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration

3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K1

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS A

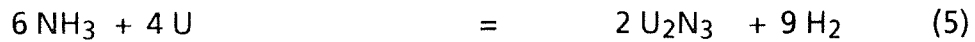
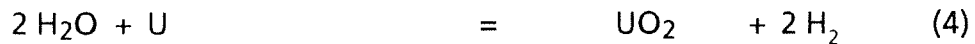
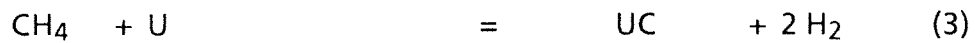
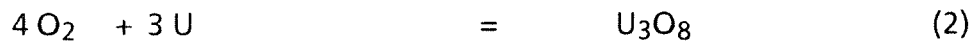
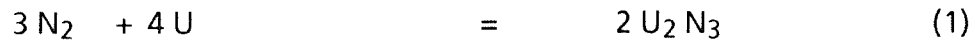
STREAM/
FLOW No.

- 9 includes hydrogen from ammonia cracking and separation in K1
- 10,11 100 % recovery of hydrogen isotopes (molecular hydrogen, and hydrogen from impurities) is assumed and considered possible depending upon recycling effort and/or permeator development
- 14 comprises all hydrogen-free converted and non-converted impurities
- 5 The residual water content is determined by the thermodynamic equilibrium constant at 150 °C and the initial impurity concentrations. The residual water content can be reduced to practically zero by recycling once after removal of all molecular hydrogen, or with a large carbon monoxide excess.
- 6 Methane cracking progresses to 99.9995 % after recycling (see Table 7) provided that all molecular hydrogen is removed prior to each recycling step.
- The catalyst consumption has been determined experimentally to be about 4 g per mole of methane without loss of catalytic efficiency. This leads to a catalyst consumption of 10.7 kg for the life time of NET.

4.2 Description of Process B

According to this process concept, hydrogen isotopes and helium are separated from the impurities by gettering C, N and O on an uranium bed. The intermediate conversion of hydrogen isotopes to water is thus avoided.

Impurities react in the uranium bed according to the following reactions:



While many investigations have dealt with the reaction of single gases like N_2 , NH_3 , D_2 , H_2 , CH_4 , CO , and CO_2 /22-32/ with uranium in various forms of aggregation, only few studies have focused attention on the interaction of mixtures of these gases with the metal /33-34/. In general, depending upon the reaction conditions, a large number of reaction products have been observed (reactions 1-7 and others), which can undergo further reaction with species present in the gas phase. Because of the complicated nature of the reaction kinetics inside of a U bed the operation temperature needs to be carefully selected and maintained in order to ensure complete C, N and O removal and minimize reverse reactions.

High temperatures in the range of 600-900 °C are needed not only for kinetic reasons but also to promote C, N, and O diffusion into the bulk of the getter material to avoid surface saturation. Excessive heating, on the other hand, may cause sintering which is accompanied by a substantial reduction in the reaction rate.

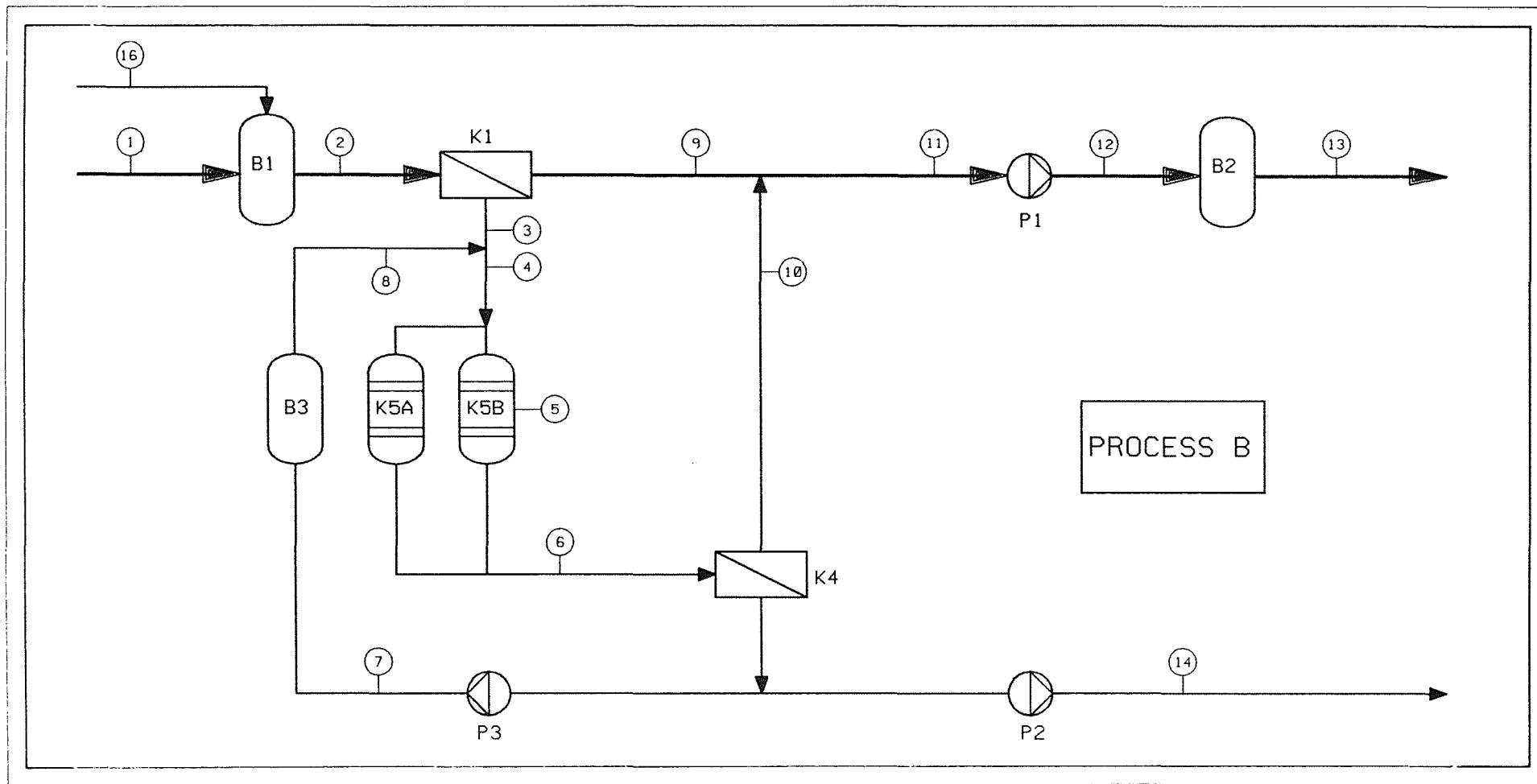
The amount of uranium required for the stoichiometric removal of the C, O, and N impurities in the plasma exhaust during normal burn and dwell is calculated from reactions (1) - (7) to be 1025 kg. A much larger amount of uranium will have to be used for the quantitative recovery of pure tritium. If non-tritiated impurities predominate in the process gas the getter consumption is very inefficient.

A process improvement can be achieved by the use of several U beds operated at different temperatures. In this way the more reactive gases (O_2 , H_2O) can be processed at lower temperatures than the others.

To avoid high tritium permeation losses a large design effort must be invested into the construction of the U beds. Difficult material problems (metal/ceramic seals) need to be solved. Appropriate safety measures are necessary to cope with the pyrophoricity of uranium powder.

All liberated hydrogen isotopes are finally separated from the remaining impurities with a palladium/silver permeator and recycled to the purified fuel stream.

Fig. 3: Chemical flow sheet for process B



EQUIPMENT LIST OF PROCESS B

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25	< 100		
K1	Mainstream Pd/Ag permeator	min.350	100		
P1	Main vacuum pump				
B2	Purified fuel collection tank	25	< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25	< 100		
K5A/B	Uranium getter beds	min. 700	< 100		
K4	Pd/Ag-permeator	min. 350	100		

Table 9: Streams / flows for process B (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T	M/h	20.00	20.00	0.03	0.03				0.681						0.00				
	g/h																		
	g/																		
He	M/h	1.00	1.00	1.00	1.00				1.00						1.00				
	g/h																		
	g/																		
CO ₄	M/h	0.30	0.30	0.30	0.30				0.00						0.00				
	mg/h																		
	gU/h					71.4													
CO	M/h	0.05	0.05	0.05	0.05				0.00						0.00	0.00			
	mg/h																		
	gU/h					17.8													
CO ₂	M/h	0.05	0.05	0.05	0.05				0.00						0.0				
	mg/h																		
	gU/h					11.9													
O ₂ O	M/h	0.05	0.05	0.051	0.051				0.00						0.00				
	mg/h																		
	gU/h					6.07													
O ₂	M/h	0.001	0.001	0.00	0.00				0.00						0.00				
	mg/h																		
	gU/h					0.00													
N ₂	M/h	0.001	0.001	0.026	0.026				0.00						0.026				
	mg/h																		
	gU/h					8.23													
NO ₃	M/h	0.05	0.05	0.00	0.00				0.00						0.00				
	mg/h																		
	gU/h					0.00													

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS B

STREAM/
FLOW No.

1	NET normal burn and dwell composition			
	- DT (50:50)	20		mol/h
	- He	1		mol/h
	- impurities total	0.5		mol/h

Assumptions as to impurities:

	- CQ ₄ is the dominating constituent	0.3		mol/h
	- O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible			
	- CO	0.05		mol/h
	- CO ₂	0.05		mol/h
	- Q ₂ O	0.05		mol/h
	- NQ ₃	0.05		mol/h

(Q = DT)

- 2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration
- 3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K11

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS B

STREAM/
FLOW No.

- 9 includes hydrogen from ammonia cracking and separation in K1
- 10,11 100 % recovery of hydrogen isotopes (molecular, and from impurities) is assumed and considered possible depending upon recycling effort and/or permeator design development
- 14 comprises all hydrogen-free converted and non-converted impurities
- 5 The total consumption of uranium, expressed in g/h, was calculated assuming stoichiometry of the involved chemical reactions. The actual amount of uranium required may be up to 10 times higher than 1025 kg/370 days.
- 6 100 % hydrogen recovery and complete impurity retention was assumed.

4.3 Description of Process C /4/

Hydrogen isotopes and helium are separated from the impurities by gettering C, N, and O on one (or more) hot uranium beds.

Some of the main reactions taking place inside the uranium bed are the same as described for process B. This process alternative avoids the intermediate conversion of hydrogen isotopes into water.

For the separation of the hydrogen isotopes from helium a uranium bed held at room temperature is employed. The p-c isotherms for the hydrogen/uranium system as well as the kinetics of the reaction of hydrogen isotopes with uranium are well known /35, 36/. Numerous traps have been designed to purify tritium from helium /37/. Since the efficiency of an uranium trap in scavenging hydrogen (tritium) from a noble gas carrier was found to vary inversely both with the flow rate and with the hydrogen/uranium ratio, it is necessary to monitor continuously the trap off-gas for tritium /38/.

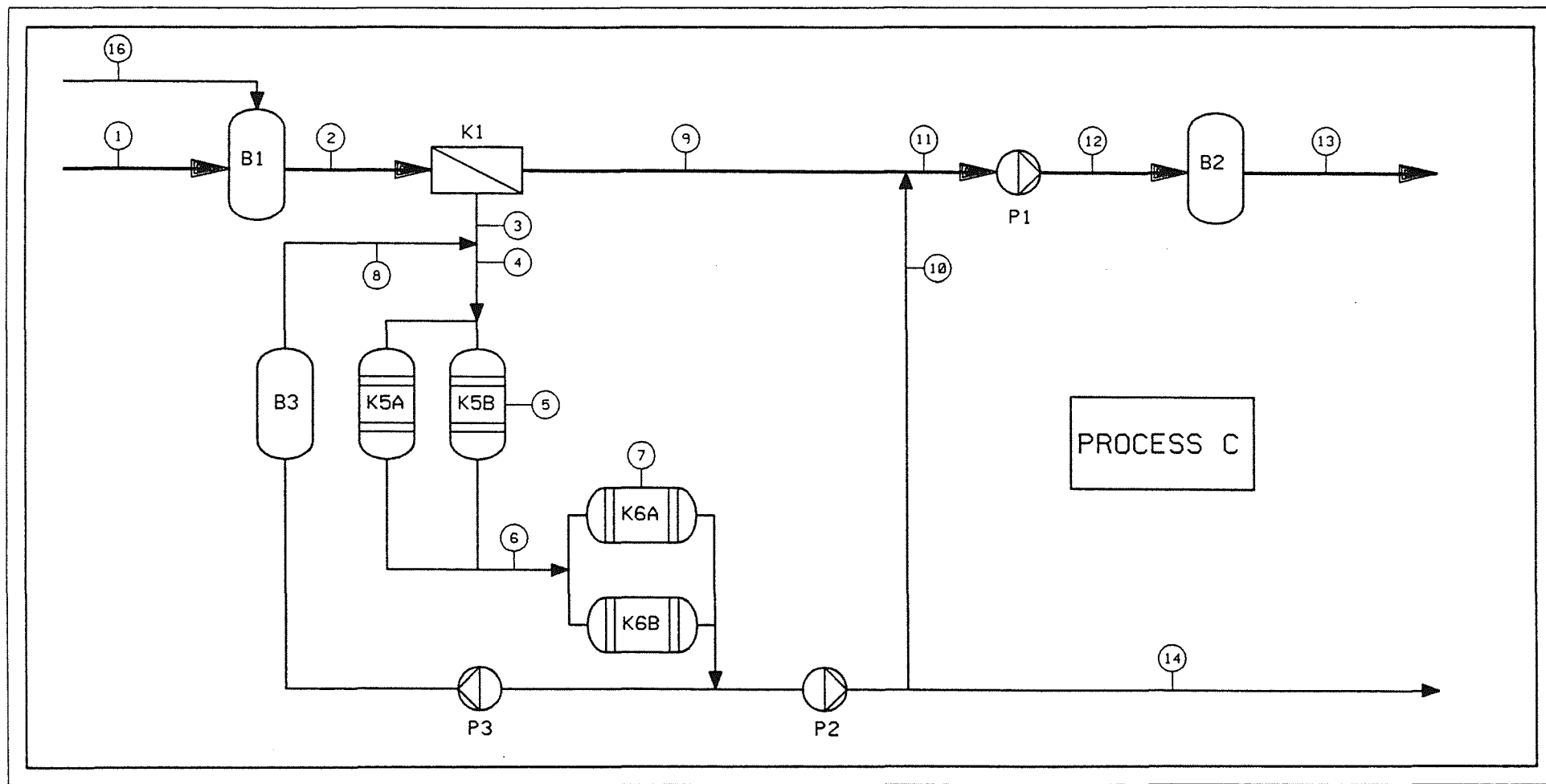


Fig. 4: Chemical flow sheet for process C

EQUIPMENT LIST OF PROCESS C

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25	< 100		
K1	Mainstream Pd/Ag permeator	min. 350	< 100		
P1	Main vacuum pump				
B2	Purified fuel collection tank	25	< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25	< 100		
K5A/B	Uranium getter beds	min. 700	< 100		
K6A/B	Uranium getter beds	approx. 400	< 100		

Table 10 : Streams /flows for process C (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	20.00	20.00	0.03	0.03		0.681			20.045	0.681	20.726			0.00					
He M/h g/h g/	1.00	1.00	1.00	1.00		1.00			0.00	0.00	0.00			1.00					
CO ₄ M/h mg/h gU/h	0.30	0.30	0.30	0.30		0.00			0.00	0.00	0.00			0.00					
					71.4														
CO M/h mg/h gU/h	0.05	0.05	0.05	0.05		0.00			0.00	0.00	0.00			0.00					
					17.8														
CO ₂ M/h mg/h gU/h	0.05	0.05	0.05	0.05		0.00			0.00	0.00	0.00			0.00					
					11.9														
O ₂ O M/h mg/h gU/h	0.05	0.05	0.051	0.051		0.00			0.00	0.00	0.00			0.00					
					6.07														
O ₂ M/h mg/h gU/h	0.001	0.001	0.00	0.00		0.00			0.00	0.00	0.00			0.00					
					0.00														
N ₂ M/h mg/h gU/h	0.001	0.001	0.026	0.026		0.00			0.00	0.00	0.00			0.00					
					8.23														
NO ₃ M/h mg/h gU/h	0.05	0.05	0.00	0.00		0.00			0.00	0.00	0.00			0.00					
					0.00														

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS C

STREAM/
FLOW No.

1 NET normal burn and dwell composition

-	DT (50:50)	20	mol/h
-	He	1	mol/h
-	impurities total	0.5	mol/h

Assumptions as to impurities:

-	CQ ₄ is the dominating constituent	0.3	mol/h
-	O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible		
-	CO	0.05	mol/h
-	CO ₂	0.05	mol/h
-	Q ₂ O	0.05	mol/h
-	NQ ₃	0.05	mol/h

(Q = DT)

2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration

3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K1

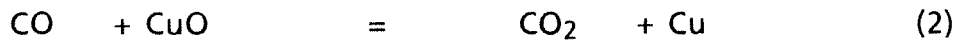
NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS C

STREAM/
FLOW No.

- 9 includes hydrogen from ammonia cracking and separation in K1
- 10,11 100 % recovery of hydrogen isotopes (molecular hydrogen, and hydrogen from impurities) is assumed and considered possible depending upon recycling effort and/or permeator design development
- 14 comprises all hydrogen-free converted and non-converted impurities
- 5 The total consumption of uranium, expressed in g/h, was calculated assuming stoichiometry of the involved chemical reactions. The actual amount of uranium required may be up to 10 times higher than 1025 kg/370 days. Additional uranium consumption arises from the uranium beds K6 A and K6 B.
- 6 100 % hydrogen recovery and impurity retention was assumed.

4.4 Description of Process D

Methane, carbon monoxide, and molecular hydrogen isotopes are oxidized to water and carbon dioxide on a hot copper oxide bed according to the following reactions /39/:



The oxygen donator bed is operated at 520 °C. This high temperature is mainly required for the oxidation of methane. Hydrogen and carbon monoxide will reduce CuO at temperatures below 250 °C /40/. At high temperatures copper oxide tends to sinter, which is associated with a loss of reactivity and an incomplete reactivation.

In a next step water and carbon dioxide are absorbed on a molecular sieve bed. Recovery of water and carbon dioxide from the molecular sieve bed is rather ineffective and time consuming. In addition, the residual water trapped in the sieve after regeneration at < 250 °C is of the order of 1 wt %, which represents a considerable inventory.

Hydrogen isotopes are recovered from water by oxygen gettinger on a hot uranium bed, according to the reaction



The reaction of carbon dioxide with uranium is described by



A high temperature uranium bed is employed (700 °C) in order to minimize the tritium inventory of the bed.

A considerable improvement of this process concept could be achieved by replacing the molecular sieve bed with a cryo trap. At a trap temperature of 160 K water is retained quantitatively and separated from most of the carbon dioxide, which at that temperature has a vapor pressure of approx. 50 mbar. As a result, a large reduction in uranium getter consumption is possible.

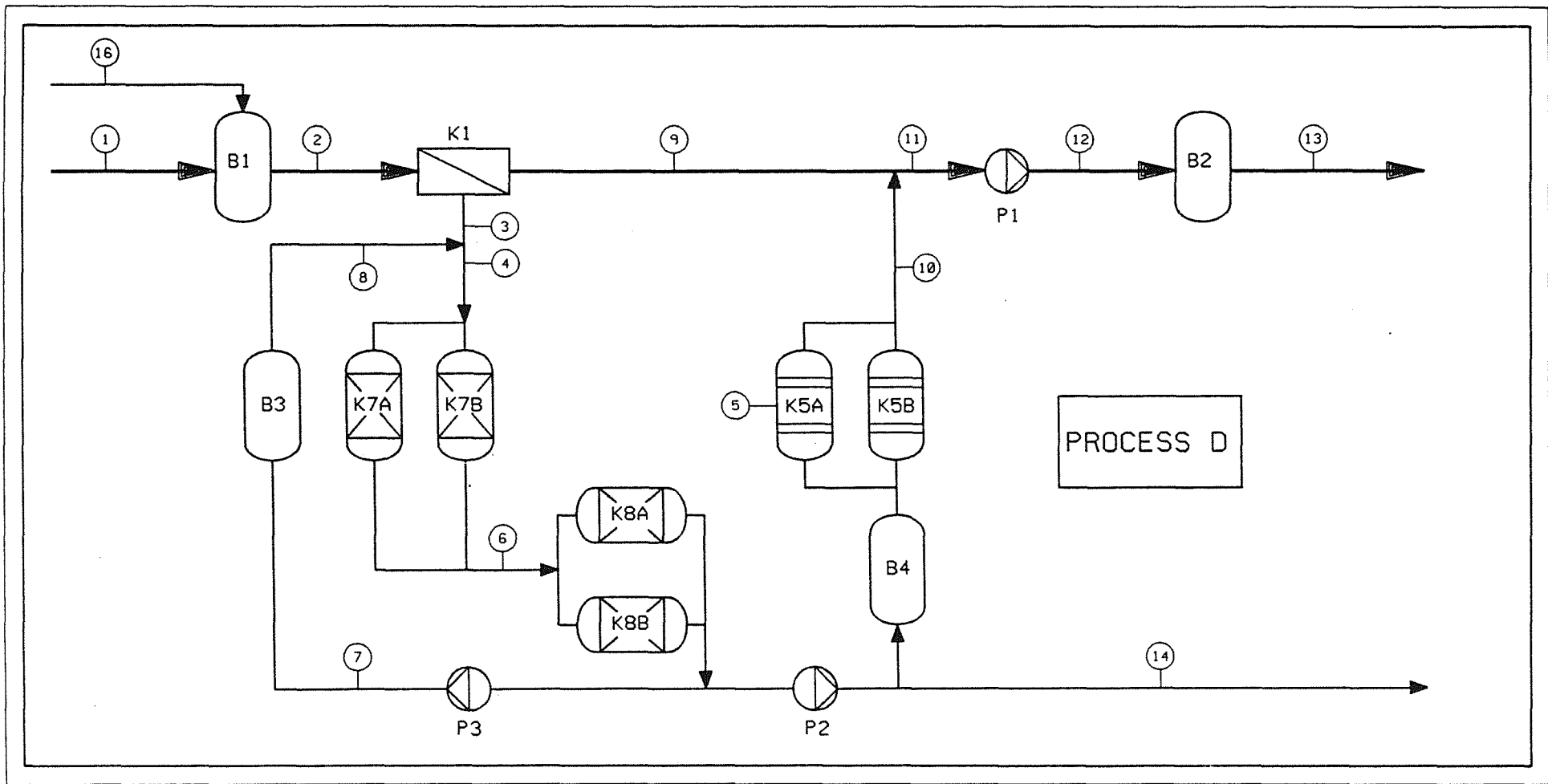


Fig. 5 : Chemical flow sheet for process D

EQUIPMENT LIST OF PROCESS D

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25	< 100		
K1	Mainstream Pd/Ag permeator	min.350	< 100		
P1	Main vacuum pump				
B2	Purified fuel collection tank		< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25	< 100		
K5A/B	Uranium getter beds	700	< 100		
K7A/B	Copper oxide beds	520	< 100		
K8A/B	Molecular sieve beds	20/150	< 100		
B4	Condensate collection tank	25	< 100		

Table 11 : Streams / flows for process D (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T	M/h	20.00	20.00	0.03	0.03					20.045	0.681	20.726							
	g/h														0.00				
	g/																		
He	M/h	1.00	1.00	1.00	1.00					0.00	0.00	0.00							
	g/h														1.00				
	g/																		
CO ₄	M/h	0.30	0.30	0.30	0.30					0.00	0.00	0.00							
	mg/h														0.00				
	gU/h																		
CO	M/h	0.05	0.05	0.05	0.05					0.00	0.00	0.00							
	mg/h														0.00				
	gU/h																		
CO ₂	M/h	0.05	0.05	0.05	0.05					0.00	0.00	0.00							
	mg/h														0.00				
	gU/h					95.2													
O ₂ D	M/h	0.05	0.05	0.051	0.051					0.681									
	mg/h														0.00				
	gU/h					81.0													
O ₂	M/h	0.001	0.001	0.00	0.00					0.00	0.00	0.00							
	mg/h														0.00				
	gU/h																		
N ₂	M/h	0.001	0.001	0.026	0.026					0.00	0.00	0.00							
	mg/h														0.00				
	gU/h																		
NO ₃	M/h	0.05	0.05	0.00	0.00					0.00	0.00	0.00							
	mg/h														0.00				
	gU/h																		

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS D

STREAM/
FLOW No.

1 NET normal burn and dwell composition

-	DT (50:50)	20	mol/h
-	He	1	mol/h
-	impurities total	0.5	mol/h

Assumptions as to impurities:

-	CQ ₄ is the dominating constituent	0.3	mol/h
-	O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible		
-	CO	0.05	mol/h
-	CO ₂	0.05	mol/h
-	Q ₂ O	0.05	mol/h
-	NQ ₃	0.05	mol/h

(Q = DT)

2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration

3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

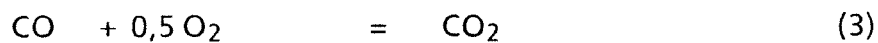
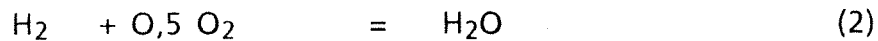
- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K1

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS D

STREAM/ FLOW No.	
9	includes hydrogen from ammonia cracking and separation in K1
10,11	100 % recovery of hydrogen isotopes (molecular hydrogen, and hydrogen from impurities) is assumed and considered possible depending upon recycling effort and/or permeator design development
14	comprises all hydrogen-free converted and non-converted impurities
6	It is assumed that hydrogen, methane and carbon monoxide are quantitatively oxidized into water and carbon dioxide. From the stoichiometry of the involved chemical reactions a minimum copper oxide consumption of 927 kg/370 days is calculated. Kinetic aspects have so far been ignored, hence a larger consumption of the oxygen-donator is anticipated.
5	The total consumption of uranium was calculated from the stoichiometry of the reactions taking place. The minimum amount required is calculated to be 176.2 g/h. Since no consideration has been given to kinetics aspects, this amount may be up to 10 times higher.

4.5 Description of Process E /6/

Impurities are oxidized by molecular oxygen in the presence of a palladium/platinum catalyst according to the following reactions:



The oxidation of methane, hydrogen, and carbon monoxide by oxygen employing a noble metal catalyst requires a minimum operating temperature of 510 °C /41, 42/. To ensure complete oxidation an excess of oxygen is needed, which depends upon the composition of the gaseous mixture. All impurities are converted into water and carbon dioxide. Small amounts of commercially available catalyst appear to be sufficient for the operation time of NET I.

Moisture present in the effluent gas of the catalyst bed is retained quantitatively in a cold trap operated at 160 K. At this temperature solid water has a vapor pressure of $\approx 10^{-6}$ mbar. Oxygen is not condensed at 160 K and will appear in the bleed stream. Carbon dioxide, on the other hand, with a vapor pressure of approx. 50 mbar at 160 K and a certain chemical affinity for water, can be partially captured in the cold trap.

Water accumulated in the trap needs to be periodically vaporized and transferred into a storage tank either by gravity, cryogenic procedures, or with a sweep gas. In either case the operation is time consuming.

Tritium from water collected in the storage tank can be recovered by electrolysis. Electrolytic oxygen is used to complete that required for the catalytic oxidation of the impurities. Three electrolysis concepts are presently under development /9, 43-45/:

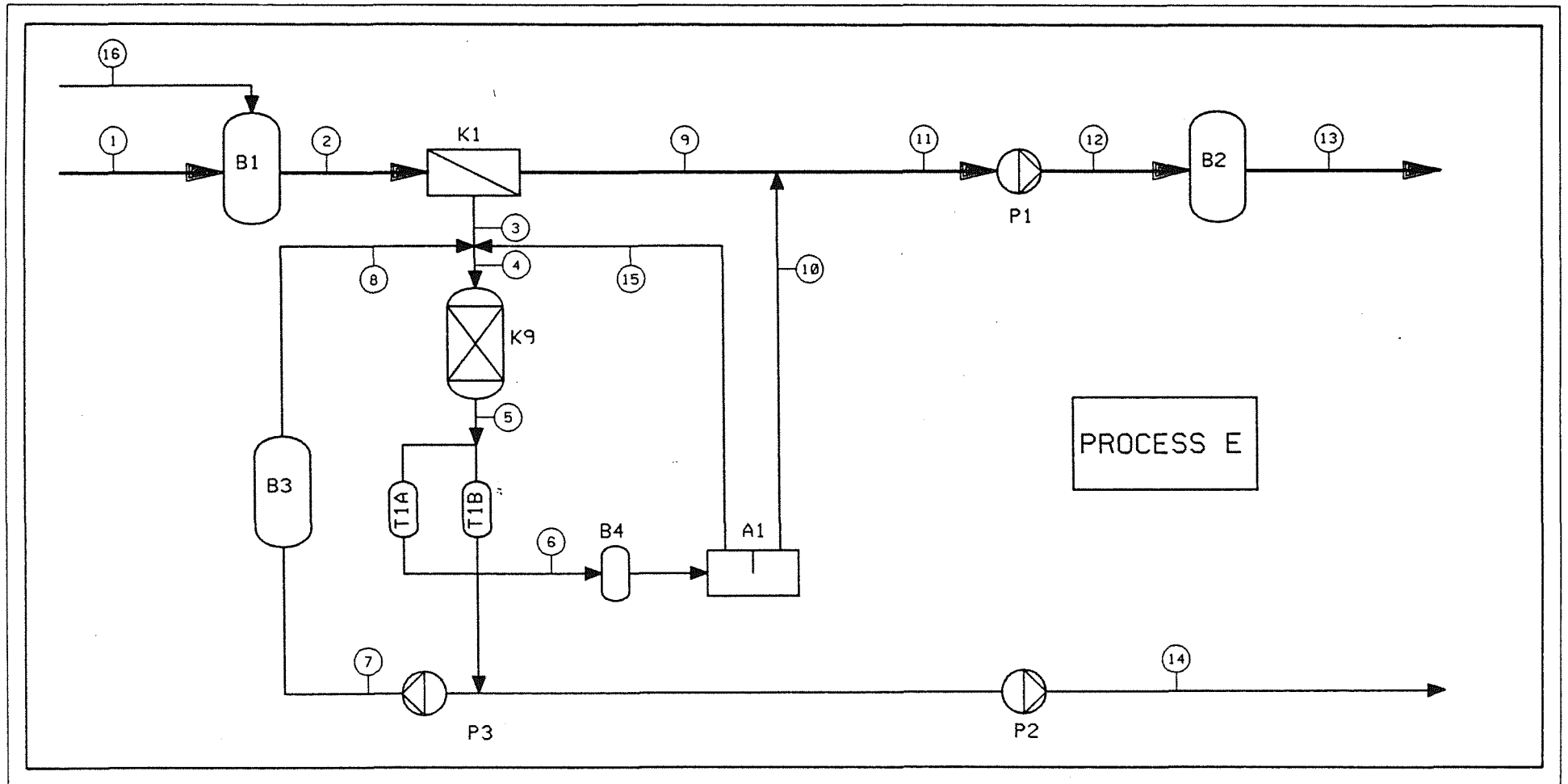
Japanese concept: Water vapor in a carrier gas (97 % He + 3 % T₂) is decomposed in a solid oxide electrolysis cell. The stationary tritium inventory is comparatively small ($\approx 1.5 \cdot 10^{12}$ Bq), but a high working temperature (≤ 630 °C) is required (problems with structural materials). The electrolytical

hydrogen needs to be separated from the helium carrier gas and decontaminated from residual water. The fate of carbon dioxide in the cell is not known. At TSTA a ceramic electrolysis cell has been operated with tritium ($1.5 \cdot 10^{12}$ Bq in He) for a period of one year at a processing rate of 7.4×10^{14} Bq/day. For NET I a scale up factor of 25 is required /42/.

Belgian concept: Tritiated water is electrolysed employing a 6 M H_2SO_4 solution. The inventory of each cell is estimated to be 2.7 g T_2 . The low operation temperature of only 8 °C has two positive effects: negligible permeation losses and low contamination of the electrolytical hydrogen with tritiated water. The concept design requires little scale up. Sulfur oxides contaminating the electrolytical oxygen must be removed from the feed gas of the noble metal catalyst. Electrolytical hydrogen needs to be decontaminated from tritiated water.

French concept: Tritiated water is electrolysed from a NaOH 1 molar solution using a hollow palladium cathode. The working temperature is comparatively low (80 °C). The through-put of 11 mol DTO/day requires scale up. Electrolytical hydrogen is of high purity and can be directly recycled. Electrolytical oxygen needs to be decontaminated from tritiated water and hydrogen. The fate of carbon dioxide cotrapped with water in the cold traps is not known.

Fig..6 : Chemical flow sheet for process E



EQUIPMENT LIST OF PROCESS E

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25	< 100		
K1	Mainstream Pd/Ag permeator	min.350	< 100		
P1	Main vacuum pump				
B2	Purified fuel collection tank	25	< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25	< 100		
K9	Pd/Pt-catalyst	520			
T1A/B	Cold traps	-113			
B4	Condensate collection tank	25	< 100		
A1	Electrolytical cell				

Table 12 : Streams / flows for process E (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	20.00	20.00	0.03	0.03	0.00	0.00			20.045	0.681	20.726			0.00					
He M/h g/h g/	1.00	1.00	1.00	1.00	1.00	0.00			0.00	0.00	0.00			1.00					
CO ₄ M/h mg/h —	0.30	0.30	0.30	0.30	0.00	0.00			0.00	0.00	0.00			0.00					
CO M/h mg/h —	0.05	0.05	0.05	0.05	0.00	0.00			0.00	0.00	0.00			0.00					
CO ₂ M/h mg/h —	0.05	0.05	0.05	0.05	0.40	?			0.00	?	?			0.40					
O ₂ O M/h mg/h —	0.05	0.05	0.051	0.051	0.681	0.681			0.00	0.00	0.00			0.00					
O ₂ M/h mg/h —	0.001	0.001	0.00	1.171	0.531	0.00			0.00	0.00	0.00			0.531	1.171				
N ₂ M/h mg/h —	0.001	0.001	0.026	0.026	0.026	0.00			0.00	0.00	0.00			0.026					
NO ₃ M/h mg/h —	0.05	0.05	0.00	0.00	0.00	0.00			0.00	0.00	0.00			0.00					

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS E

STREAM/
FLOW No.

1 NET normal burn and dwell composition

-	DT (50:50)	20	mol/h
-	He	1	mol/h
-	impurities total	0.5	mol/h

Assumptions as to impurities:

-	CQ ₄ is the dominating constituent	0.3	mol/h
-	O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible		
-	CO	0.05	mol/h
-	CO ₂	0.05	mol/h
-	Q ₂ O	0.05	mol/h
-	NQ ₃	0.05	mol/h

(Q = DT)

2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration

3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K1

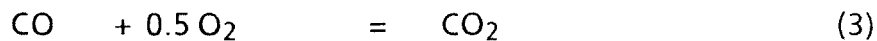
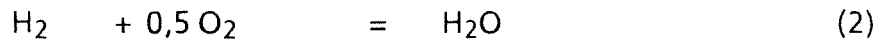
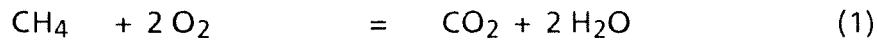
NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS E

STREAM/
FLOW No.

- 9 includes hydrogen from ammonia cracking and separation in K1
- 10,11 100 % recovery of hydrogen isotopes (molecular hydrogen, and hydrogen from impurities) is assumed and considered possible depending upon recycling effort and/or permeator design development
- 14 comprises all hydrogen-free converted and non-converted impurities
- 4 Twice the stoichiometric amount of oxygen as required for the oxidation of the impurities is considered necessary. Therefore, the electrolytically produced oxygen is not sufficient.
- 5 Quantitative oxidation of the impurities is assumed.
- 6 At 160 K 100 % of the water, containing an unknown amount of carbon dioxide, will be retained in the cold traps.

4.6 Description of Process F /7/

Impurities are oxidized by molecular oxygen in the presence of a palladium/platinum catalyst according to the following reactions:

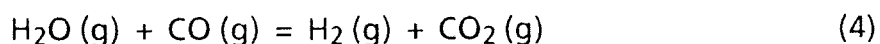


The oxidation of methane, hydrogen, and carbon monoxide by oxygen employing a noble metal catalyst requires a minimum operating temperature of 510 °C /41, 42/. To ensure complete oxidation an excess of oxygen is needed, which depends upon the composition of the gaseous mixture. All impurities are converted into water and carbon dioxide. Small amounts of commercially available catalyst appear to be sufficient for the operation time of NET I.

Moisture present in the effluent gas of the catalyst bed is retained quantitatively in a cold trap operated at 160 K. At this temperature solid water has a vapor pressure of $\leq 10^{-6}$ mbar. Oxygen is not condensed at 160 K and will appear in the bleed stream of the cold trap. Oxygen needs to be removed to protect the zinc stabilized copper chromite catalyst from oxidation. Carbon dioxide with a vapor pressure of approx. 50 mbar at 160 K and a certain chemical affinity for water, can be partially captured in the cold trap.

The gases condensed in the cold trap are periodically volatilized employing an adequate sweep gas (argon) and then send over a zinc stabilized copper chromite catalyst (K2) operated at $T \leq 250$ °C. For the volatilization of the daily water production approx. 1m³ STP of carrier gas is needed.

Carbon monoxide must be added to the sweep gas up to a concentration of CO/H₂O $\cong 1.5$. Under these conditions water is converted into hydrogen according to the water gas shift reaction



Liberated hydrogen isotopes are continuously removed by a palladium/silver permeator operated at 300 °C. Permeated hydrogen isotopes are of high purity and can be sent directly to the purified fuel stream.

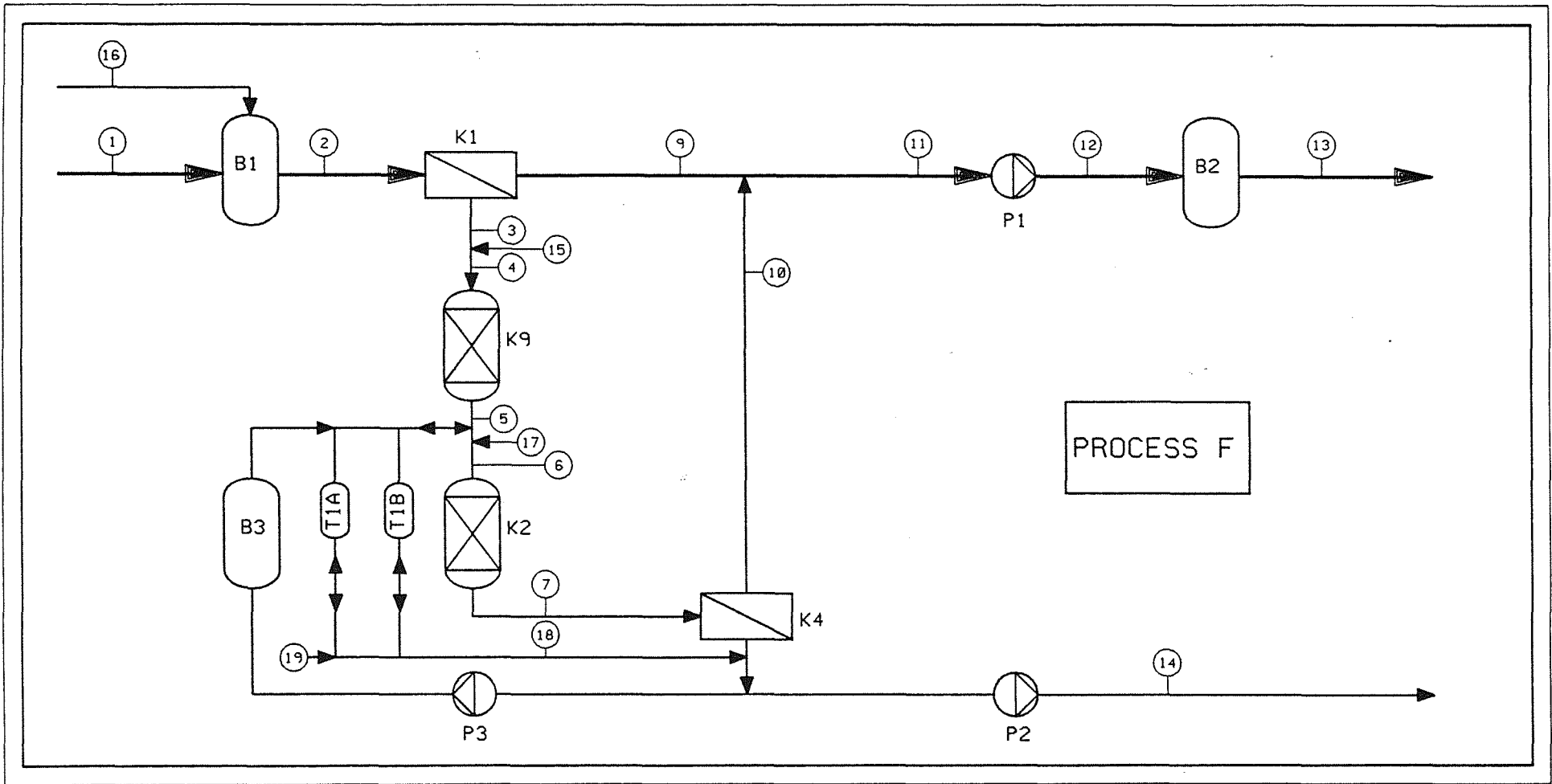


Fig. 7 : Chemical flow sheet for process F

EQUIPMENT LIST OF PROCESS F

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25	< 100		
K1	Mainstream Pd/Ag permeator	min.350	< 100		
P1	Main vacuum pump				
B2	Purified fuel collection tank	25	< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25	< 100		
K9	Pd/Pt-catalyst	520			
T1A/B	Cold traps	-113			
K2	CuO/Cr ₂ O ₃ /ZnO catalyst	170	< 100		
K4	Pd/Ag-permeator	min. 350	< 100		

Table 13 : Streams / flows for process F (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	20.00	20.00	0.03	0.03	0.00	0.00	0.681		20.045	0.681	20.726			0.00				0.00	
He M/h g/h g/	1.00	1.00	1.00	1.00	1.00	1.00	0.00		0.00	0.00	0.00			1.00				1.00	
CO ₄ M/h mg/h —	0.30	0.30	0.30	0.30	0.00	0.00	0.00		0.00	0.00	0.00			0.00				0.00	
CO M/h mg/h —	0.05	0.05	0.05	0.05	0.00	>1.36	>0.681		0.00	0.00	0.00			>0.681				0.00	>1.36
CO ₂ M/h mg/h —	0.05	0.05	0.05	0.05	0.40	?	0.681		0.00	0.00	0.00			0.681				0.40	
O ₂ O M/h mg/h —	0.05	0.05	0.051	0.051	0.681	0.681	0.00		0.00	0.00	0.00			0.00				0.00	
						14.300													
O ₂ M/h mg/h —	0.001	0.001	0.00	1.171	0.531	0.00	0.00		0.00	0.00	0.00			0.531	1.171			0.531	
N ₂ M/h mg/h —	0.001	0.001	0.026	0.026	0.026	>50	>50		0.00	0.00	0.00			>50				0.026	>50
																			7.2E5
NO ₃ M/h mg/h —	0.05	0.05	0.00	0.00	0.00	0.00	0.00		0.00	0.00	0.00			0.00				0.00	

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS F

STREAM/

FLOW No.

1 NET normal burn and dwell composition

-	DT (50:50)	20	mol/h
-	He	1	mol/h
-	impurities total	0.5	mol/h

Assumptions as to impurities:

-	CQ ₄ is the dominating constituent	0.3	mol/h
-	O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible		
-	CO	0.05	mol/h
-	CO ₂	0.05	mol/h
-	Q ₂ O	0.05	mol/h
-	NQ ₃	0.05	mol/h

(Q = DT)

2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration

3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K1

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS F

STREAM/
FLOW No.

- 9 includes hydrogen from ammonia cracking and separation in K1
- 10,11 100 % recovery of hydrogen isotopes (molecular hydrogen, and hydrogen from impurities) is assumed and considered possible depending upon recycling effort and/or permeator design development
- 14 comprises all hydrogen-free converted and non-converted impurities
- 4 Twice the stoichiometric amount of oxygen as required for the oxidation of the impurities is considered necessary.
- 5 Quantitative oxidation of the impurities is assumed.
- 18 At 160 K 100 % of the water is retained in the cold traps. All oxygen is removed, therefore avoiding oxidation of catalyst in K2.
- 6 Water is vaporized and transported with a carbon monoxide/nitrogen sweep gas into K2.
- 7 The residual water content can be reduced to practically zero by recycling while continuously removing hydrogen.

4.7 Description of Process G

Methane, carbon monoxide and hydrogen are converted into water and carbon dioxide in the presence of copper oxide (or other oxygen donators like NiO) according to the following reactions:



at temperatures of the order of 500 °C [39]. By using an oxygen donator (oxygen is supplied as stoichiometrically required) the external supply of molecular oxygen is avoided. For the normal burn and dwell operation a CuO consumption of 920 kg/370 days is estimated. Sintering of copper oxide at high temperatures cannot be excluded. This may lead to a loss of reactivity and to an incomplete regenerability of the solid bed. To avoid condensation of water a carrier gas may be necessary.

Another potential oxygen donator is NiO [39, 46]. This oxide is thermally much more stable than CuO and could possibly be regenerated periodically. However, experimental work under conditions relevant to fusion fuel clean-up is needed.

In a next step water vapor is converted into hydrogen by reaction with carbon monoxide on a zinc stabilized copper chromite catalyst kept at $T \approx 250$ °C. The following reaction describes the process:



CO must be added up to a concentration of $\text{CO}/\text{H}_2\text{O} \approx 1.5$. To achieve a high degree of conversion the gaseous mixture must be recycled. At the same time hydrogen isotopes are continuously removed by a palladium/silver permeator operated at 300 °C. Highly pure permeated hydrogen isotopes are recycled to the purified fuel stream.

Option G is not adequate to process an impurity stream containing high partial pressures of carbon monoxide or molecular hydrogen.

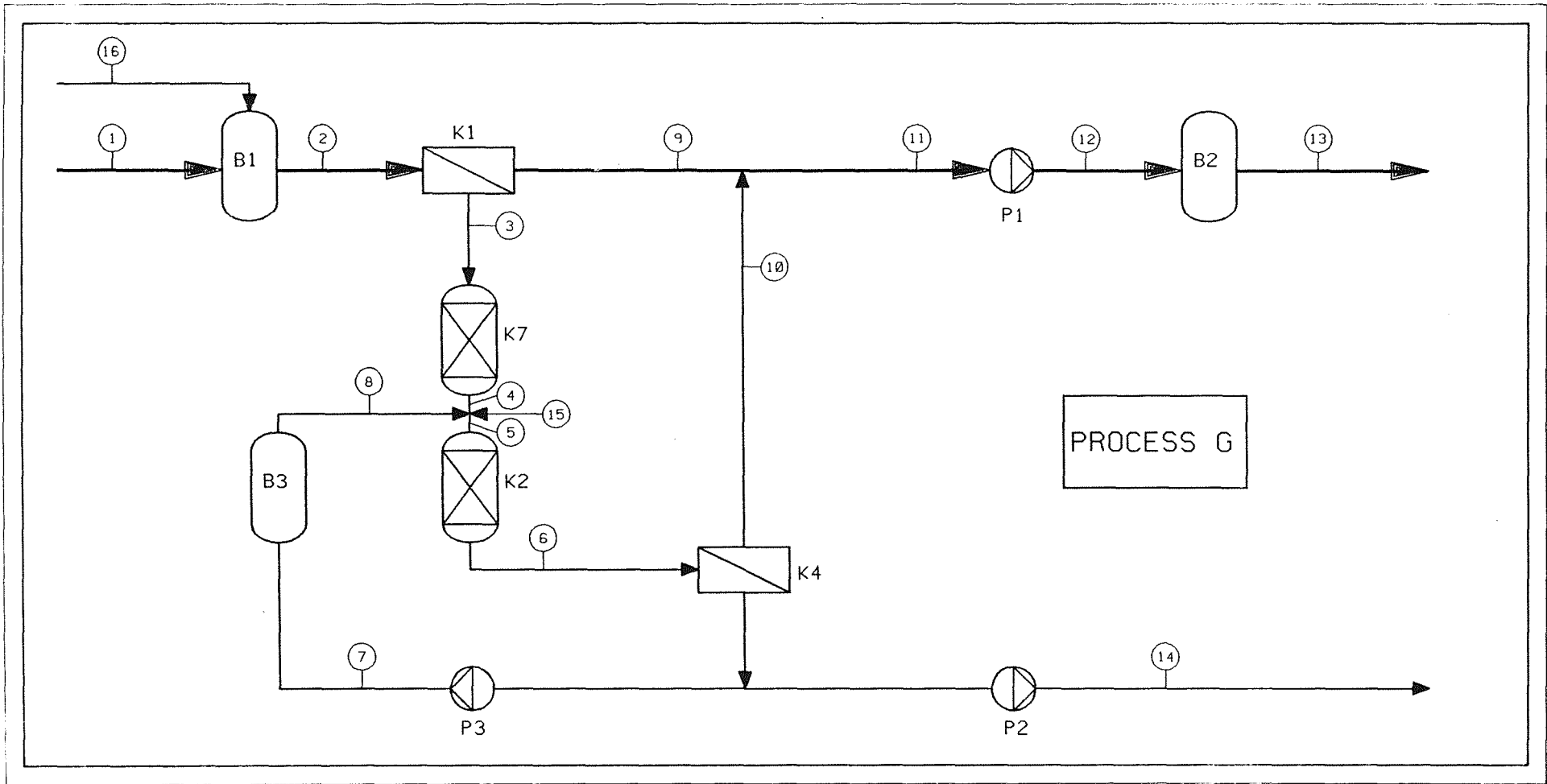


Fig. 8 : Chemical flow sheet for process G

EQUIPMENT LIST OF PROCESS G

DESIGNATION	FUNCTION	OPERATING CONDITIONS		DIMENSIONS (mm)	REMARKS
		TEMP. (°C)	PRESSURE (kPa)		
B1	Equalizing/collection tank	25	< 100		
K1	Mainstream Pd/Ag permeator	min. 350	< 100		
P1	Main vacuum pump				
B2	Purified fuel collection tank	25	< 100		
P2	Waste pump				
P3	Recirculation pump				
B3	Buffer tank	25	< 100		
K7	Copper oxide beds	520	< 100		
K2	Copper chromite zinc stabilized catalyst	170	< 100		
K4	Pd/Ag permeator	min. 350	< 100		

Table 14 : Streams / flows for process G (normal burn and dwell)

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = O T M/h g/h g/	20.00	20.00	0.03	0.00	0.00	0.681			20.045	0.681	20.726			0.00					
He M/h g/h g/	1.00	1.00	1.00	1.00	1.00	1.00			0.00	0.00	0.00			1.00					
CO ₄ M/h mg/h —	0.30	0.30	0.30	0.00	0.00	0.00			0.00	0.00	0.00			0.00					
CO M/h mg/h —	0.05	0.05	0.05	0.00	1.362	0.681			0.00	0.00	0.00			0.681	1.362				
CO ₂ M/h mg/h —	0.05	0.05	0.05	0.40	0.40	1.081			0.00	0.00	0.00			1.081					
O ₂ O M/h mg/h —	0.05	0.05	0.051	0.681	0.681	0.00			0.00	0.00	0.00			0.00					
O ₂ M/h mg/h —	0.001	0.001	0.00	0.00	0.00	0.00			0.00	0.00	0.00			0.00					
N ₂ M/h mg/h —	0.001	0.001	0.026	0.026	0.026	0.026			0.00	0.00	0.00			0.026					
NO ₃ M/h mg/h —	0.05	0.05	0.00	0.00	0.00	0.00			0.00	0.00	0.00			0.00					

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS G

STREAM/

FLOW No.

1 NET normal burn and dwell composition

-	DT (50:50)	20	mol/h
-	He	1	mol/h
-	impurities total	0.5	mol/h

Assumptions as to impurities:

-	CQ ₄ is the dominating constituent	0.3	mol/h
-	O ₂ and N ₂ will be most likely converted to water and ammonia respectively and are considered negligible		
-	CO	0.05	mol/h
-	CO ₂	0.05	mol/h
-	Q ₂ O	0.05	mol/h
-	NQ ₃	0.05	mol/h

(Q = DT)

2 Equal to No.1, possible influence by addition of fresh fuel, No. 16, not yet taken into consideration

3 Permeation efficiency approx. 99.85 % as deduced from CFFTP report G 86020 prepared for NET in Sept. 86

Assumptions as to impurities:

- possible reactions between impurities in K1 are neglected (to be verified experimentally)
- the permeation rate for the impurities is taken as zero (to be verified experimentally)
- the permeation rate of hydrogen isotopes is not affected by the impurities
- ammonia is cracked into its elements completely by K1

NOTES PERTAINING TO CHEMICAL FLOW SHEET OF PROCESS G

STREAM/
FLOW No.

- 9 includes hydrogen from ammonia cracking and separation in K1
- 10,11 100 % recovery of hydrogen isotopes (molecular hydrogen, and hydrogen from impurities) is assumed and considered possible depending upon recycling effort and/or permeator design development
- 14 comprises all hydrogen-free converted and non-converted impurities
- 6 It is assumed that hydrogen, methane and carbon monoxide are quantitatively oxidized into water and carbon dioxide. From the stoichiometry of the involved chemical reactions a minimum copper oxide consumption of 927 kg/370 days is calculated. Kinetic aspects have so far been ignored, hence a larger consumption of the oxygen-donor is anticipated.
- 7 The residual water content can be reduced to practically zero by recycling while continuously removing hydrogen.

5. Process Evaluation

In this chapter the seven process concepts A - G are evaluated with respect to compliance with the Performance Requirements (PR) of the Fuel Processing System as specified in the NET document NE/86/S-033 and already summarized in chapter 2. Because of insufficient experimental data the evaluation is mostly qualitative.

PR1 Process primary vacuum exhaust during burn and dwell

All processes under consideration can handle the plasma exhaust gas stream arising during normal burn and dwell periods. A process stream containing the same impurity spectrum in 10 times higher concentrations, such as may occur during prolonged "down time", is associated with an additional consumption of solid reagents (getter, oxygen donator, etc.) and more prolonged recycling or electrolysis times.

Based on current knowledge it appears that the main permeator (K1) can tolerate high impurity levels without loss of permeation efficiency.

13 criteria have been selected for a comparison of the advantages/disadvantages of the various processes (see Table 15). The evaluation represents a "grosso modo" judgment of the processes. It may be used to identify the most promising alternatives suitable for a more detailed investigation.

Process tritium losses or tritium losses to waste (crit. 1) should be minimized. The losses are limited to the extremely low daily release quantity allowed by the NET I-DDR. Time and energy consuming recycling stages and/or additional equipment, e.g. scavenging getter beds with direct influence on the total processing time, the process economy and operability, may become necessary.

Table 15: Process Evaluation Scheme

	Criterion	Process						
		A	B	C	D	E	F	G
1	D-T losses to waste	L	L	L	L	E	L	L
2	D-T inventory	L	L	E	E	E	E	L
3	processing time	S	S	S	L	L	L	E
4	final product quality	G	G	G	G	P	G	G
5	operating temp.	L-E	H	H	E-H	E	L	E-L
6	gas volume to be treated	L	L	L	L	E	H	E
7	media requirements	L	L	L	L	E	H	E
8	operability	NC	NC	NC	NC	NC	C	NC
9	availability	DDR	DDR	DDR	DDR	DDR	DDR	DDR
10	credibility of functional predictions	sat.	sat.	sat.	sat.	sat.	sat.	sat.
11	T-waste total gaseous/solid	L/L	H/L	H/L	H/L	L/L	L/H	E/E
12	risk level	L	E	E	E	E	E	L
13	economical aspects	G	P	P	P	G	F	G

L = low
 E = elevated
 S = short
 G = good
 C = complicated

NC = not complicated
 DDR = draft design requirements met
 sat = satisfactory
 P = poor
 H = high

High process losses can be anticipated for process E and F involving cold traps because non-condensibles (excess oxygen, carbon dioxide) are sent directly to waste.

The inventory (crit. 2) is classified as low whenever small size catalyst beds are involved (see also Table 16, Appendix).

Duration (crit. 3) or processing time is classified as low for nominally once-through processes.

A less good or poor product quality (crit. 4) is assigned to electrolytically separated hydrogen isotopes when recycled without further treatment (exception: electrolytical hydrogen as obtained according to the French electrolysis concept).

Operating temperature (crit. 5) and process duration (crit. 3) may be used for the estimation of possible permeation losses. An estimation of the permeation rate of hydrogen through 2-3 mm SS (AISI 304) at 450 °C and a partial pressure difference of 1 bar give values of about 50 cm³ STP/d.m². This indicates that double containment with tritium recovery will become necessary for several components.

The original gas volume to be treated may have to be increased (crit. 6) by the addition of reaction gases or the necessity of introducing sweep gases (process E and F). This is reflected by crit. 7 (see also Table 5 and the Appendix).

Operability (crit. 8) comprises aspects like ease of operation, possibility of maloperation, extent of instrumentation and process control.

Availability predictions (crit. 9) can be met by assuming redundancy or standby for all heated components or components subject to corrosive attack.

The credibility of functional predictions (crit. 10) is considered satisfactory when experimental results on the process are available and the scale-up to a NET relevant size becomes possible.

The amount of gaseous waste (crit. 11) is considered to be high, whenever additional reaction and/or sweeping gases are required. High quantities of solid waste will result from getter and catalyst consuming processes (see also Table 10 and 16 in Appendix).

The risk level (crit. 12) is considered elevated when pyrophoric getter materials heated up to > 700 °C are involved or explosive gas mixtures can be formed, particularly when gas mixtures are handled at elevated temperatures. The radiotoxic hazards associated with the handling of tritium are well known. The containment barriers are dictated by the hazard potential of the tritium species involved.

Safety precautions in systems containing hydrogen are well developed. It is known that gaseous hydrogen has flammability limits in air of 4 - 75 %, the limits for methane instead are rather narrow, e.g. 5-15 %. In fuel clean-up processes, where oxygen is deliberately admitted, i.e. added for the combustion of hydrogen or hydrogenated gases, process control reliability is of utmost importance. Wherever molecular oxygen is needed provisions have to be made to exclude its unintentional admission.

It is known that tritium will be absorbed on a U-bed initially with high rate and substantial heat evolution. The absorption of tritium is therefore self-limiting, because of the exothermic nature of the reaction. An emergency cooling is not necessary from a tritium absorption point of view.

Excess oxygen needs to be avoided to prevent overheating and ignition of uranium.

High catalyst and/or getter material consumption require frequent renewal of solid reagents, which is associated with additional complicated handling operations. The effect on the economical aspects (crit. 13) will be negative.

PR 2 Process tritium in waste gas from Torus

(a) under normal pumpdown from atmosphere

A flow rate of 0.83 m³/s of air or helium cannot be processed by any of the Fuel Impurity Removal Systems (processes A-G). The main air or helium waste gas should therefore be sent to the Detritiation System. The residual gas containing the species outgassing from the Torus wall (i.e. up to 1.5 mol water/h) can be processed by the alternatives A-G.

(b) during bake-out

All processes under consideration can process the bakeout exhaust gas. This process gas should not be sent through permeator K 1 because water in high concentration may reduce the efficiency of the Pd/Ag membranes. Therefore, DTO should first be converted into DT via the water gas shift reaction (dilution with He/CO required) or reaction with uranium.

Alternatively, DT could be oxidized and then all DTO trapped cryogenically for subsequent electrolytical decomposition. For the transport of 0.12 moles DTO/h at a concentration of about 50 % additional carrier gas and/or heated transport tubes are necessary. More information on the various process streams as well as on the consumption of solid reagents are given in tables 17 and 20 to 26 (see Appendix).

(c) to maintain negative pressure during loss of vacuum condition

The amount of air or He required to maintain a negative pressure in the Torus after a loss of vacuum cannot be processed by any of the processing loops A-G. It is anticipated, however, that most of the tritiated gaseous inventory present in the Torus before the break will be retained by the cryopumps during the period immediately after the break. Once the cryopumps are saturated, the air/He stream needed to maintain negative pressure could be sent directly into the air detritiation system.

The gaseous species retained by the cryopumps could be processed separately (up to 150 g of tritium).

In case of a rupture of a water cooling pipe (not specified in the process requirements) the partial pressure of water in the fuel clean-up system should be maintained below the dew point. The preferred options A and G do not appear to be particularly sensitive to water and air in-breaks. If the water dew point is exceeded problems with the operation of pumps are possible.

Water vapors will not affect the hydrogen permeation within the Pd/Ag diffusers (expm. at KfK). Air in-breaks must be viewed from hydrogen safe handling considerations.

(d) during glow discharge cleaning

To cope with this PR permeator K 1 must be capable of processing > 65 mol/h of hydrogen. The total amount of impurities is the same as that in PR 1. The composition of the impurity stream was estimated after Dylla et al /47/. Estimated process streams are given in Tables 27 to 33 (see Appendix). The total glow discharge cleaning time was taken to be 150 hours.

(e) during carbonization (if used)

It appears possible to process 10 mol CD_4 /h with any of the alternative processes A - G under discussion. Solid reagent consumptions and flow streams are given in Table 19 and 34 to 40 (see Appendix).

General remark: A buffer storage tank capable of handling untreated plasma exhaust with 21.5 mol/h for at least 8 h requires to have a volume of more than 3.85 m³. This would lead to a tritium inventory of 515 g.

PR3 NBI and Pellet Injector Cryopanel Loads

After /2/ the NBI and the Pellet Injector Cryopanel Loads do not interface with the Impurity Removal System. Therefore they need not be considered.

PR 4 Process NBI Neutralizer Flow

Same as PR 3.

PR 5 Process dilute T in D and H Streams for Blanket or Coolant Reprocessing

140 mol Q2 (Q = D, H, T)/h can be processed by an appropriately designed palladium/silver permeator.

PR6 Provide pure D2, T2 and DT for pellet injection NBI and gas puffing fuelling equipment.

Not within the scope of the Fuel Processing Loop.

PR7 Reject a minimum of tritium to the tritium waste treatment system as solid, liquid, or gaseous wastes.

This requirement is particularly severe for processes based on metallic getters (process B and C). Tritium permeated through structural materials should be recovered to reduce gaseous wastes. To improve the overall decontamination factor (removal of trace amounts) of the Fuel Processing Loops a hydrogen getter bed (ZrCo or U) operated at room temperature and placed in the effluent gas stream can be considered.

PR8 The product of failure rate leading to a Fuel Loop outage greater than 8 hours, and mean down-time for the fuel processing loop should not exceed 10^{-3} .

Redundancy or spare supply of critical components prone to failure or to wear and tear, such as pumps, circulation pumps, electrical heating, etc. is required.

PR9 Tritium inventory of any components which could be released by a single process failure must not exceed 150 g.

All components can be designed to comply with the requirement of a tritium inventory smaller than 150 g.

PR10 Gas is to be received from the plasma system and delivered to the fuel introduction systems at 300 K, and < 1 bar. Maximum working pressure in the loop should be ≤ 2 bar.

All processes can be designed to comply with these requirements.

PR 11 All components should be maintainable in a glove box (caisson) or other remote handling environment.

Same as PR 10.

PR 12 The need to ensure operation compatible with tritium requires the avoidance of organics in contact with the process streams containing tritium. The overall external leak rate into the fuel processing loop shall not exceed 10^{-6} torr l/s at 1 bar He differential pressure.

Organics and lubricants can be avoided. In one case, i.e. valves with a Vespel seat, tritium compatibility of organics has been demonstrated (TSTA).

A total leak rate lower than 10^{-6} torr · l/s for an all metal subsystem can be achieved.

PR 13 No outleakage from the fuel processing loop is permitted under normal operation.

This PR will be accomplished by design measures.

PR 14 The fuel processing loop must have a design lifetime of 15 years.

A lifetime of 15 years can only be achieved for static components such as vessels, tubing, sensors, etc. All other components which are subject to wear and tear will have a shorter lifetime (see also PR8).

PR 15 The design should accommodate changes of $\pm 50\%$ in the nominal process conditions specified in the requirements.

50 % changes in the nominal process conditions can be achieved by design measures.

PR 16 A bake-out temperature of 150 °C and helium purge capability are required for components where practical.

Since all components employed are of stainless steel or other temperature resistant metals bake-out at temperatures up to 150 °C is possible. This also holds for valves with Vespel seat.

PR 17 Loose contamination (dust) shall be prevented from migrating through the process system.

Appropriate mechanical filters will be installed at the inlet and outlet of all components containing dust producing solids, i.e. metal getters, oxygen donating solids, catalyst, etc.

6. Conclusion and recommendation

All processes which include one (or more) impurity gettering step by metals like uranium will require large amounts of solid reagent and will consequently yield substantial amounts of tritium-contaminated solid waste. Uranium reactions require very high temperatures and a large reagent excess to achieve high decontamination factors. To overcome the permeation problems associated with high temperatures and to provide adequate containment of the getter material considerable engineering efforts are needed.

Oxidation of all impurities into carbon dioxide and water, either by catalytic oxidation with molecular oxygen or by reaction with an oxygen donor, can be carried out at temperatures below those required for gettering. In addition the amount of tritium-contaminated solid waste is lower than that from metal getter reactions. The main drawback of processes including an oxidation step is seen in the non-discriminative oxidation of all impurities (i.e. CO, molecular hydrogen) and in the difficulty of separating quantitatively carbon dioxide from water, for instance by cryogenic methods. The latter problem can be circumvented by using the water gas shift reaction for the conversion of water into hydrogen. Also of concern in processes that include the formation of water are problems like incidental condensation, the necessity of an inert sweep gas, etc.

Electrolytic methods are attractive because they produce very little (or no) process waste. Presently considered alternatives involve either low temperatures and high tritium inventory or high temperatures and very low tritium inventory. Purification of one or both of the electrolytical gases may become necessary.

The main advantages of selective catalytic reactions are seen in the low solid waste production, acceptable tritium inventories, comparatively moderate reaction temperatures (low permeation losses) and the fact that recovery of tritium is carried out directly (water gas shift reaction, hydrocarbon cracking) and not via intermediate oxidation / reduction steps. All components of an impurity processing system based on selective catalytic reactions are relatively small.

The impurity stream is not carried through temperature cycles and may be processed in a once-through or in a recycling modus. Recovered deuterium / tritium are of high purity and may be sent directly into the Isotope Separation System.

From the above it is concluded that Process A, backed up by process G can be considered the most promising concepts for the development of NET fuel clean-up.

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10 Appendix

Comparison of processes A - G with respect to processing tritium in waste gas from Torus during normal burn and dwell, carbonization, glow discharge and bake-out operation.

Table 16

Process comparison: Normal Burn and Dwell

Process	A	B	C	D	E	F	G	H
getter material consumption kg/370 d	none	uranium >> 1025	uranium >> 1025	uranium >> 1566	none	none	none	
catalyst consumption kg/370 d	Ni-cat.* none 10.7	none	none	CuO* >> 925	none	none	CuO* >> 925	
additional reaction gas	CO	none	none	N ₂ /He for vol.	O ₂ , N ₂ /He for vol.	O ₂ , N ₂ , CO	CO	
gaseous waste - type - Nm ³ /370 d - DT content g/370 d	He, CO ₂ , N ₂ 224	He 199	He 199	He/N ₂ 204	He, CO ₂ , O ₂ N ₂ 389	He, CO, CO ₂ O ₂ , N ₂ 575 + vol.	N ₂ He, CO, CO ₂ 554	
solid waste - type - Nm ³ /370 d - DT content g/370 d	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	

* can be regenerated

vol. = volatilization

Table 17

Process comparison: Bake-out

Process	A	B	C	D	E	F	G	H
getter material consumption kg	none	uranium >> 4.4	uranium >> 4.4	uranium >> 3.8	none	none	none	
catalyst consumption kg	Ni-cat.* 0.13 *	none	none	CuO* >> 15.9 *	none	none	CuO* >> 15.9 *	
additional reaction gas	CO	none	none	N ₂ /He for vol.	O ₂ , N ₂ /He for vol.	O ₂ , N ₂ , CO	CO	
gaseous waste - type - Nm ³ - DT content g	He, CO ₂ , N ₂ 0.6	none	none	none	CO ₂ , CO 0.4	CO, CO ₂ N ₂ 1.3 + vol.	CO, CO ₂ 1.2	
solid waste - type - Nm ³ - DT content g	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	

* can be regenerated

vol. = volatilization

Table 18

Process comparison: Glow Discharge

Process	A	B	C	D	E	F	G	H
getter material consumption kg	none	uranium >> 19.6	uranium >> 19.6	uranium >> 60.8	none	none	none	
catalyst consumption kg	Ni-cat.* 2.4	none	none	CuO* 21.5	none	none	CuO* >21.5	
additional reaction gas	none **	none	none	He, N ₂ for vol.	O ₂ , N ₂ /He for vol.	O ₂ , CO, N ₂ for vol.	CO	
gaseous waste - type - Nm ³ - DT content g	CO 0.34	none	none	He, N ₂	O ₂ , CO ₂ , He N ₂ for vol. > 0.32	O ₂ , CO, N ₂ for vol. > 0.45	CO ₂ , CO 0.77	
solid waste - type - Nm ³ - DT content g	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	

* can be regenerated

vol. = volatilization

** K₂ not used

Table 19:

Process comparison: Carbonization

Process	A	B	C	D	E	F	G	H
getter material consumption kg	none	uranium >> 19	uranium >> 19	uranium >> 57.2	none	none	none	
catalyst consumption kg	Ni-cat.* 3.2	none	none	CuO* 25.4	none	none	CuO* >25.4	
additional reaction gas	none	none	none	none	O ₂ , N ₂ /He for vol.	O ₂ , N ₂ /He for vol.	CO	
gaseous waste - type - Nm ³ - DT content g	none	none	none	none	O ₂ , CO ₂ N ₂ /He 3.6 + vol.	O ₂ , CO ₂ , CO N ₂ 10.8 + vol.	CO ₂ , CO 9.0	
solid waste - type - Nm ³ - DT content g	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	to be det.	

* can be regenerated

vol. = volatilization

Table 20

GLOW DISCHARGE
PROCESS A

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = O T M/h g/h g/	65.00	65.00	0.10	0.10	0.10	0.90			64.90	0.90	65.80	65.80	65.80	0.00					
He M/h g/h g/	0.00																		
CO ₄ M/h mg/h gh/h	0.40	0.40	0.40	0.40	0.40	0.00			0.00	0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h gh/h	0.10	0.10	0.10	0.10	0.10	0.10			0.00	0.00	0.00	0.00	0.00	0.10					
CO ₂ M/h mg/h —																			
O ₂ O M/h mg/h —																			
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

ZYS-NR. JA-YC-100031-001-01.16-DEC-87

Table 21
GLOW DISCHARGE
PROCESS B

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	65.00	65.00	0.10	0.10		0.90			64.90	0.90	65.80	65.80	65.80	0.00					
He M/h g/h g/	0.00																		
CO ₄ M/h mg/h gh/h	0.40	0.40	0.40	0.40		0.00			0.00	0.00	0.00	0.00	0.00	0.00					
					95.20														
CO M/h mg/h gh/h	0.10	0.10	0.10	0.10		0.00			0.00	0.00	0.00	0.00	0.00	0.00					
					35.70														
CO ₂ M/h mg/h —																			
O ₂ O M/h mg/h —																			
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

ZYS-NR. JA-YC-100031-001-01 16-DEC-87

Table 22

GLOW DISCHARGE
PROCESS C

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
D ₂ = D T M/h g/h g/	65.00	65.00	0.10	0.10		0.90			64.90	0.90	65.80	65.80	65.80	0.00					
He M/h g/h g/	0.00																		
CO ₄ M/h ng/h gh/h	0.40	0.40	0.40	0.40		0.00			0.00	0.00	0.00	0.00	0.00	0.00					
					95.20														
CO M/h ng/h gh/h	0.10	0.10	0.10	0.10		0.00			0.00	0.00	0.00	0.00	0.00	0.00					
					35.70														
CO ₂ M/h ng/h —																			
D ₂ O M/h ng/h —																			
O ₂ M/h ng/h —																			
N ₂ M/h ng/h —																			
NO ₃ M/h ng/h —																			

Table 23
 GLOW DISCHARGE
 PROCESS D

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	65.00	65.00	0.10	0.10		0.00			64.90	0.90	65.80	65.80	65.80	0.00					
He M/h g/h g/	0.00																		
CO ₄ M/h mg/h —	0.40	0.40	0.40	0.40		0.00			0.00	0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h —	0.10	0.10	0.10	0.10		0.00			0.00	0.00	0.00	0.00	0.00	0.00					
CO ₂ M/h mg/h gh/h	0.00	0.00	0.00	0.00	298.00	0.60			0.00	0.00	0.00	0.00	0.00	0.00					
O ₂ D M/h mg/h gh/h	0.00	0.00	0.00	0.00		0.90			0.00	0.00	0.00	0.00	0.00	0.00					
						107.10													
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

ZVS-NR, JA-YC-100031-001-01 10-DEC-87

Table 24
GLOW DISCHARGE
PROCESS E

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g'	65.00	65.00	0.10	0.10	0.00	0.00			64.90	0.90	65.80	65.80	65.80	0.00					
He M/h g/h g'	0.00																		
CO ₄ M/h mg/h —	0.40	0.40	0.40	0.40	0.00	0.00			0.00	0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h —	0.10	0.10	0.10	0.10	0.00	0.00			0.00	0.00	0.00	0.00	0.00	0.00					
CO ₂ M/h mg/h —	0.00	0.00	0.00	0.00	0.50				0.00	0.00	0.00	0.00	0.00	0.50					
O ₂ D M/h mg/h —	0.00	0.00	0.00	0.00	0.90	0.90			0.00	0.00	0.00	0.00	0.00	0.00					
O ₂ M/h mg/h —	0.00	0.00	0.00	0.90	0.45	0.00			0.00	0.00	0.00	0.00	0.00	0.45	0.45				
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

ZVS-NR. JA-YC-00031-001-01.16-DEC-87

Table 25
GLOW DISCHARGE
PROCESS F

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	
O ₂ = O T M/h g/h g/	65.00	65.00	0.10	0.10	0.00	0.00	0.90		64.90	0.90	65.80	65.80	65.80	0.00				0.00		
He M/h g/h g/	0.00																			
CO ₄ M/h mg/h —	0.40	0.40	0.40	0.40	0.00	0.00	0.00		0.00	0.00	0.00	0.00	0.00	0.00				0.00		
CO M/h mg/h —	0.10	0.10	0.10	0.10	0.00	1.80	0.90		0.00	0.00	0.00	0.00	0.00	0.00	1.80			0.00		
CO ₂ M/h mg/h —	0.00	0.00	0.00	0.00	0.50		0.90		0.00	0.00	0.00	0.00	0.00	0.90						
O ₂ O M/h mg/h —	0.00	0.00	0.00	0.00	0.90	0.90	0.00		0.00	0.00	0.00	0.00	0.00	0.00				0.00		
O ₂ M/h mg/h —	0.00	0.00	0.00	0.90	0.45	0.00	0.00		0.00	0.00	0.00	0.00	0.00	0.45				0.45		
N ₂ M/h mg/h —																				
NO ₃ M/h mg/h —																				

ZVSS-NR. JA-YC-10000-CI-YC-100-DEC-91-87

Table 26
 GLOW DISCHARGE
 PROCESS G

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T	M/h	65.00	65.00	0.10	0.00	0.00	0.90			64.90	0.90	65.80	65.80	65.80	0.00				
	g/h																		
	g/																		
He	M/h	0.00																	
	g/h																		
	g/																		
CO ₄	M/h	0.40	0.40	0.40	0.00	0.00	0.00			0.00	0.00	0.00	0.00	0.00	0.00				
	mg/h																		
	—																		
CO	M/h	0.10	0.10	0.10	0.00	1.80	0.90			0.00	0.00	0.00	0.00	0.00	0.90	1.80			
	mg/h																		
	—																		
CO ₂	M/h	0.00	0.00	0.00	0.50	0.50	1.40			0.00	0.00	0.00	0.00	0.00	1.40				
	mg/h																		
	—																		
B ₂ O	M/h	0.00	0.00	0.00	0.90	0.90	0.00			0.00	0.00	0.00	0.00	0.00	0.00				
	mg/h																		
	—																		
O ₂	M/h																		
	mg/h																		
	—																		
N ₂	M/h																		
	mg/h																		
	—																		
NO ₃	M/h																		
	mg/h																		
	—																		

87-DEC-98-10-10-10-10000-C-V-AP-NR-SZ

Table 27
BAKEOUT
PROCESS A

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.024	0.145	0.211				0.211	0.211	0.211	0.211	0.00		0.00			
He M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₄ M/h mg/h gU/h	0.033	0.033	0.033	0.033	0.033	0.00				0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h gU/h	0.044	0.044	0.044	0.242	0.121	0.121				0.00	0.00	0.00	0.00	0.121	0.198				
CO ₂ M/h mg/h gU/h	0.027	0.027	0.027	0.027	0.148	0.148				0.00	0.00	0.00	0.00	0.148					
H ₂ O M/h mg/h gU/h	0.121	0.121	0.121	0.121	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
O ₂ M/h mg/h gU/h																			
N ₂ M/h mg/h gU/h																			
NO ₃ M/h mg/h gU/h																			

78-DEC-3-10--10010000-CY-AF-UR-SVZ

Table 28
BAKEOUT
PROCESS B

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.024	0.00	0.211				0.211	0.211	0.211	0.211	0.00		0.00			
H ₂ M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₄ M/h mg/h gJ/h	0.033	0.033	0.033	0.033		0.00				0.00	0.00	0.00	0.00	0.00					
					7.95														
CO M/h mg/h gJ/h	0.044	0.044	0.044	0.044		0.00				0.00	0.00	0.00	0.00	0.00					
					15.71														
CO ₂ M/h mg/h gJ/h	0.027	0.027	0.027	0.027		0.00				0.00	0.00	0.00	0.00	0.00					
					6.43														
O ₂ O M/h mg/h gJ/h	0.121	0.121	0.121	0.121		0.00				0.00	0.00	0.00		0.00					
					14.39														
O ₂ M/h mg/h gJ/h																			
N ₂ M/h mg/h gJ/h																			
NO ₃ M/h mg/h gJ/h																			

Table 29
BAKEOUT
PROCESS C

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.024	0.00	0.211				0.211	0.211	0.211	0.211	0.00		0.00			
He M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₄ M/h mg/h gJ/h	0.033	0.033	0.033	0.033		0.00				0.00	0.00	0.00	0.00	0.00					
					7.85														
CO M/h mg/h gJ/h	0.044	0.044	0.044	0.044		0.00				0.00	0.00	0.00	0.00	0.00					
					15.71														
CO ₂ M/h mg/h gJ/h	0.027	0.027	0.027	0.027		0.00				0.00	0.00	0.00	0.00	0.00					
					6.43														
O ₂ D M/h mg/h gJ/h	0.121	0.121	0.121	0.121		0.00				0.00	0.00	0.00		0.00					
					14.39														
O ₂ M/h mg/h gJ/h																			
N ₂ M/h mg/h gJ/h																			
NO ₃ M/h mg/h gJ/h																			

LB-330-6-10-10013000-C-A-V-R-S-SZ

Table 30
BAKEOUT
PROCESS D

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.024		0.00				0.211	0.211	0.211	0.211	0.00		0.00			
He M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₄ M/h mg/h gU/h	0.033	0.033	0.033	0.033		0.00				0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h gU/h	0.044	0.044	0.044	0.044		0.00				0.00	0.00	0.00	0.00	0.00					
CO ₂ M/h mg/h gU/h	0.027	0.027	0.027	0.027		0.104				0.00	0.00	0.00	0.00	0.00					
					12.85														
O ₂ M/h mg/h gU/h	0.121	0.121	0.121	0.121		0.211				0.00	0.00	0.00	0.00	0.00					
					25.11														
O ₂ M/h mg/h gU/h																			
N ₂ M/h mg/h gU/h																			
NO ₃ M/h mg/h gU/h																			

Table 31
BAKEOUT
PROCESS E

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.024	0.00	0.00				0.211	0.211	0.211	0.211	0.00		0.00			
He M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₄ M/h mg/h —	0.033	0.033	0.033	0.033	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h —	0.044	0.044	0.044	0.044	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₂ M/h mg/h —	0.027	0.027	0.027	0.027	0.104					0.00	0.00	0.00	0.00	0.104					
D ₂ O M/h mg/h —	0.121	0.121	0.121	0.121	0.211	0.211				0.00	0.00	0.00	0.00	0.00					
O ₂ M/h mg/h —				0.134	0.067	0.00								0.067	0.134		0.067		
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

Table 32
BAKEOUT
PROCESS F

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.024	0.00	0.00	0.211			0.211	0.211	0.211	0.211	0.00				0.00		
H ₂ M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00	0.00			0.00	0.00	0.00	0.00	0.00				0.00		
CO ₄ M/h mg/h —	0.033	0.033	0.033	0.033	0.00	0.00	0.00			0.00	0.00	0.00	0.00	0.00				0.00		
CO M/h mg/h —	0.044	0.044	0.044	0.00	0.00	>0.422	>0.211			0.00	0.00	0.00	0.00	>0.211				0.00		
CO ₂ M/h mg/h —	0.027	0.027	0.027	0.027	0.104		0.315			0.00	0.00	0.00	0.00	0.315						
O ₂ O M/h mg/h —	0.121	0.121	0.121	0.121	0.211	0.211	0.00			0.00	0.00	0.00	0.00	0.00						
O ₂ M/h mg/h —				0.134	0.067	0.00	0.00							0.00	0.134			0.067		
N ₂ M/h mg/h —						>100	>100							>100					>100	
NO ₃ M/h mg/h —																				

ZWS-NR-JA-YC-00001-01-3-DEC-07

Table 33
 BAKEOUT
 PROCESS G

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	0.024	0.024	0.024	0.00	0.00	0.211				0.211	0.211	0.211	0.211	0.00					
He M/h g/h g/	0.00	0.00	0.00	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO ₄ M/h mg/h —	0.033	0.033	0.033	0.00	0.00	0.00				0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h —	0.044	0.044	0.044	0.00	0.422	0.211				0.00	0.00	0.00	0.00	0.211	0.422				
CO ₂ M/h mg/h —	0.027	0.027	0.027	0.104	0.104	0.315				0.00	0.00	0.00	0.00	0.315					
O ₂ O M/h mg/h —	0.121	0.121	0.121	0.211	0.211	0.00				0.00	0.00	0.00	0.00	0.00					
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

ZYS-NR. JA-YC-10031-001-01 3-DEC-87

Table 34
 CARBONIZATION
 PROCESS A

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	55.00	55.00	0.00	0.00	0.00	20.00			54.92	20.00	75.00	75.00	75.00	0.00					
He M/h g/h g/																			
CO ₄ M/h mg/h gU/h	10.00	10.00	10.00	10.00	10.00	0.00								0.00					
CO M/h mg/h —															0.00				
CO ₂ M/h mg/h —																			
D ₂ O M/h mg/h —																			
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

ZYS-NR. JA-YC-10010000-CY-10010000-DE-C-87

Table 35

CARBONIZATION
PROCESS B

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19		
O ₂ = D T M/h g/h g/	55.00	55.00	0.00	0.00		20.00				20.00	75.00	75.00	75.00	0.00							
He M/h g/h g/																					
CO ₄ M/h mg/h gU/h	10.00	10.00	10.00	10.00		0.00				0.00	0.00	0.00	0.00	0.00							
						2300															
CO M/h mg/h —																					
CO ₂ M/h mg/h —																					
O ₂ O M/h mg/h —																					
O ₂ M/h mg/h —																					
N ₂ M/h mg/h —																					
NO ₃ M/h mg/h —																					

18-DEC-9 10-100-100001-01-VF JIN SYS

Table 36
 CARBONIZATION
 PROCESS C

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	55.00	55.00	0.00	0.00		20.00				20.00	75.00	75.00	75.00	0.00					
He M/h g/h g/																			
CO ₄ M/h mg/h gU/h	10.00	10.00	10.00	10.00		0.00				0.00	0.00	0.00	0.00	0.00					
					2300														
CO M/h mg/h —																			
CO ₂ M/h mg/h —																			
D ₂ O M/h mg/h —																			
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

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Table 37
 CARBONIZATION
 PROCESS D

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	
O ₂ = D T M/h	55.00	55.00	0.00	0.00		0.00			54.92	20.00	75.00	75.00	75.00	0.00						
He M/h																				
CO ₄ M/h	10.00	10.00	10.00	10.00		0.00			0.00	0.00	0.00	0.00	0.00	0.00						
					2380															
CO M/h																				
CO ₂ M/h						10.00				0.00	0.00	0.00	0.00	0.00						
					4760															
O ₂ D M/h						20.00				0.00	0.00	0.00	0.00	0.00						
					2389															
O ₂ M/h																				
N ₂ M/h																				
NO ₃ M/h																				

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Table 39
 CARBONIZATION
 PROCESS F

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h	55.00	55.00	0.00	0.00	0.00	0.00	20.00		54.92	20.00	75.00	75.00	75.00	0.00					
g/h																			
g/																			
He M/h																			
g/h																			
g/																			
CO ₄ M/h	10.00	10.00	10.00	10.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00	0.00	0.00					
mg/h																			
—																			
CO M/h						40.16	20.00							20.00				40.16	
mg/h																			
—																			
CO ₂ M/h					10.00	?	?							30.00					
mg/h																			
—																			
O ₂ O M/h					20.00	20.00	0.00			0.00	0.00	0.00	0.00	0.00					
mg/h																			
—																			
O ₂ M/h				20.00	10.04									10.04	20.00				10.04
mg/h																			
—																			
N ₂ M/h																			
mg/h																			
—																			
NO ₃ M/h																			
mg/h																			
—																			

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Table 40
 CARBONIZATION
 PROCESS G

Stream/flow	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
O ₂ = D T M/h g/h g/	55.00	55.00	0.00	0.00	0.00	20.00			54.92	20.00	75.00	75.00	75.00	0.00					
He M/h g/h g/																			
CO ₄ M/h mg/h —	10.00	10.00	10.00	10.00	0.00	0.00			0.00	0.00	0.00	0.00	0.00	0.00					
CO M/h mg/h —					40.16	20.00				0.00	0.00	0.00	0.00	20.00	40.16				
CO ₂ M/h mg/h —				10.00	10.00	30.00				0.00	0.00	0.00	0.00	30.00					
O ₂ O M/h mg/h —				20.00	20.00	0.00				0.00	0.00	0.00	0.00	0.00					
O ₂ M/h mg/h —																			
N ₂ M/h mg/h —																			
NO ₃ M/h mg/h —																			

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