

THERMOCHEMICAL ENERGY TRANSPORT
(CHEMICAL HEAT PIPE)

RELATION BETWEEN WORK BY ENERGY CONVERSION GROUP, ANU,
AND WORK ELSEWHERE IN USA, WEST GERMANY AND FRANCE

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JULY, 1979

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DEPARTMENT OF ENGINEERING PHYSICS
THE AUSTRALIAN NATIONAL UNIVERSITY

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1. INTRODUCTION

A requirement of the present contract with NERDDC is to identify the relationship between the work at ANU and similar work being carried out in the USA. This report is a response to that requirement and, in addition, covers work in West Germany and France as well as in the USA. The two European countries have been included on our own initiative in order to make the report more comprehensive since these two countries are prominent in the field of solar energy, and West Germany is particularly advanced in the development of a specific form of chemical heat pipe.

This report is based largely on the observations of the authors: Dr P O Carden who recently visited the three countries named above and Dr O M Williams who visited several additional locations in the USA. Lists of the institutions visited by both, and of the more important contacts made, are appended hereto. Perhaps the most important institutions visited in the USA were the Department of Energy (DOE) programme managers dealing with thermochemical projects viz Sandia Livermore and JPL Pasadena. In addition, the new primary funding agency SERI (Solar Energy Research Institute) at Golden Colorado was visited and also the offshoot of SERI responsible for solar thermal test facilities (STTF) viz the STTF Users Association located at Albuquerque, New Mexico.

One of us (Carden) was engaged by the Users Association to review recent thermochemical proposals submitted to the STTFUA and also attended an STTF two-day workshop at Albuquerque. Both of us attended the ISES conference at Atlanta, Georgia.

Preparation for the visits in West Germany and France began at the respective embassies in Canberra in an endeavour to include all the centres of thermochemical research in the itinerary.

Thus it will be seen that itineraries and activities were sufficiently extensive to ensure that no major thermochemical work which was both current and relevant to solar energy would go unnoticed. Nevertheless, it was clear to us that there exists a considerable body of recently prepared literature which we were not able to explore in the time available.

2. STANDING OF THERMOCHEMICAL-SOLAR WORK

Each of the three countries visited had an official government funded programme on thermochemical energy transport and storage.

In France, an official publication⁽¹⁾ put the 1978 thermochemical budget at 1.4 MF (million francs) out of a total solar energy research budget of 21.1 MF (Australian \$4.5 M). Most of the money for thermochemical work went towards fundamental studies in solar fuels (hydrogen and pyrolysis products from wood), reversible chemical reactions for transport and storage of energy; and the adaptation of chemical processes to solar sources.

In West Germany a major DM 100 Million (Australian \$50 M) demonstration plant is under construction for thermochemical transport of heat energy

from nuclear power stations over distances averaging 70 km. In addition there is a new programme specifically aimed at thermochemical transport of solar energy involving initial studies with NH_3 and SO_3 based systems.

In the USA there is growing realisation of the potential of distributed solar collector systems (as opposed to central receiver systems) with 'chemical' energy transport. For example, a study by Turner⁽²⁾ indicates that, for distributed collector systems, the cost of chemical energy transport is less than any other method (Figure 1) especially for power levels in excess of $1000 \text{ MW}_{\text{th}}$. Moreover, a study by Livingston⁽³⁾ indicates that by year 2000 distributed systems with chemical transport will be less expensive than central receiver systems, and for power levels above about 80 MW_e will be the least expensive systems of all (Figure 2).

It is apparent that there is still considerable ignorance about the potential of thermochemical energy transport and storage, and confusion over their separate requirements with regard to suitable chemical reactions. There is a growing realisation, eg. at the STTFUA workshop, that systems suitable for energy transport are generally unsuitable for energy storage and vice versa. However, if underground gas storage eventuates, reactions based on CH_4 and NH_3 will be superlative for both transport and storage.

3. USA PROJECTS

3.1 Solar-Thermochemical

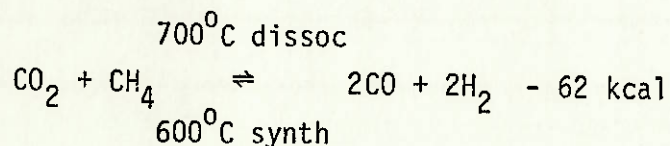
To date the system which has been advanced most is that based on SO_3 proposed by T Chubb⁽⁴⁾ of the Naval Research Establishment, Washington, DC.

Although the concept has been developed theoretically in some detail there does not exist an experimental demonstration of the energy transport loop or any major component. However, in Canada, the Ontario Research

Foundation is collaborating by developing the manufacture of ceramic solar receivers suitable for the high dissociation temperatures and the corrosive environment. Rocket Research Company of Seattle is presently screening catalysts suitable for the dissociation of SO_3 in an experimental programme funded by DOE. The same corporation has also a contract to evaluate several candidate systems including the NH_3 system.

It is becoming apparent that the major drawbacks of the SO_3 system are the materials and catalyst and the fact that the energy transport pipe lines must be kept always at 90°C or greater in order to avoid being plugged with solid SO_3 . A recent paper by Iannucci and Fish⁽⁵⁾ suggests that the system is uneconomical for energy storage largely because of the latent heat required to vaporise the SO_3 out of ambient temperature storage. Nevertheless the SO_3 system has considerable promise for economical energy transport.

Chubb, in collaboration with a group from the University of New Mexico, has very recently submitted a \$30,000 proposal to the STTF Users Association for a small demonstration based on a CH_4 system. The proposal involves a solar heated dissociator and some allied laboratory work. The primary dissociation reaction is:



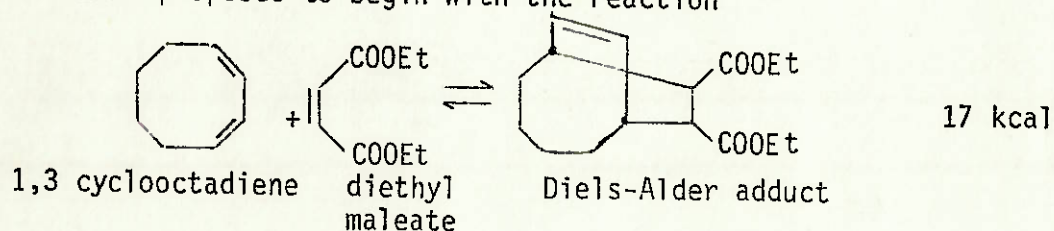
In order to promote reversability within a practical temperature range it is proposed to increase the carbon to hydrogen ratio beyond the stoichiometric value eg. the feedstock for the initial dissociation experiments will be CO_2 , CO , CH_4 in the molar proportions 3:0.2:1.

Presumably this is also necessary to promote the formation of CO_2 rather than H_2O in the synthesis stage but catalysts specific to the required reaction will still be required. It is anticipated that operation of the dissociator below a certain temperature will result in carbon deposition but it is argued that this process should be reversed at subsequent higher temperatures.

The NH_3 system was studied in the USA for the first time by Lenz⁽⁶⁾ under a \$24,000 contract with the STTFUA. Lenz has now before SERI a \$273,000 proposal⁽⁷⁾ to continue this work to the demonstration stage. The proposal includes a solar heated dissociator operating at 10 kW.

In a separate submission Lenz has requested \$37,000 for preliminary work with a reaction which is representative of a promising group known as Diels-Alder reactions⁽⁸⁾. These reactions generally involve complex organic substances and some of them occur between liquid phases. Thus both transportability and storageability may be practicable with the same system. However suitable Diels-Alder reactions appear to be limited to the temperature range 100°C - 500°C .

Lenz proposes to begin with the reaction



We are not aware of any other US work in thermochemical energy transport except that there is evidently collaboration between GE and the organisations in West Germany working on the $\text{CH}_4 + \text{H}_2\text{O}$ system.

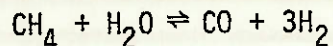
3.2 Other Related Programmes in the USA

Related programmes are in solar fuels and thermochemical storage and transmission for power generating utilities. We make the distinction between thermochemical systems for the utilities and those for solar energy purposes. This is because the utilities are concerned essentially with energy transmission outwards from a centralised source to large numbers of consumers or energy converters whereas solar systems are concerned with the reverse problem: energy transmission inwards to a centralised energy converter from a large number of sources. The former may be called radiational transport systems and the latter corradiational systems⁽⁹⁾. Thermochemical transport systems for which the energy input end is more complicated than the output end are clearly more suitable for radiational systems. On the other hand systems where the opposite is true are more appropriate for energy corradiation.

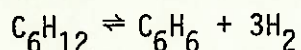
A second distinction between the needs of the utilities and the needs of solar systems is that for the former, energy storage is presently of most importance whereas for the latter energy transmission is the more pressing need (because the most economical solar energy systems possess little energy storage and are hybrids with conventional power generating systems).

A basis for the US work on thermochemical energy storage and transmission for the utilities is provided by two studies, one by Rocket Research Company⁽¹⁰⁾ and one by GE Company⁽¹¹⁾. The first of these emphasises storage and consequently gives prominence to solid-liquid reactions which have high energy density. Liquid-gas reactions are considered to be least suitable.

The second report throws more emphasis on radiational energy transport systems and comes out strongly in favour of the reaction pioneered in West Germany:



It also recommends research into the practicability of a lower temperature reaction:



cyclohexane benzene

The GE report also contains a brief reference to ammonia dissociation which is here quoted in full:

'This reaction is a good example to illustrate the shortcoming of a purely thermodynamic screening of reactions. The system looks attractive because the operating range of 400 to 600 K is ideally suited to light-water reactors, the enthalpy of reaction is reasonable, and the constituent materials are inexpensive and abundant. In addition, catalysts and industrial technology are available.

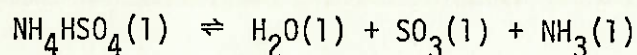
Unfortunately, an analysis of a typical ammonia synthesis process shows that in order to overcome poor kinetics, pressures in the range of 200 to 1000 bars and temperatures in excess of 775 K are required to carry out the exothermic step. To make matters worse, this energy producing step is in reality an energy loser - instead of producing ~ 46 kJ/mole typical plants consume 20 times as much energy for steam and compressor power. One could hope for a major breakthrough in ammonia synthesis catalysts, but this has not happened in the last 20 years and does not appear highly likely. If it should happen, the first impact will not be in CHP applications but in the manufacture of fertilizers, explosives, and countless other end uses for which large quantities of ammonia are currently produced.'

The criticism is in fact quite unfounded and could only arise from ignorance of the commercial synthesis processes and the energy flows therein. As illustration we show in Figure 3 the energy flows of a

modern plant based on figures provided by Vancini⁽¹²⁾. The corresponding energy flow relevant to the converter and to thermochemical energy transport is shown in Figure 4. Clearly the GE claim that there cannot be a net energy output is incorrect and in fact the parasitic energy flows are relatively minor.

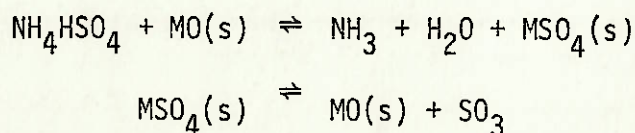
The GE report also regards the SO_3 reaction as useful for energy storage, a contention that has been shown to be incorrect by Fish⁽⁵⁾.

A promising liquid-liquid reaction is being studied by W E Wentworth and colleagues at the Department of Chemistry, University of Houston. The reaction is basically the dissociation and resynthesis of ammonium hydrogen sulphate (AHS)



and is being investigated primarily for energy storage. The reaction is non catalysed and has the distinction that all reactants may be stored as liquids. However, the AHS must be stored at typically 100°C in order to avoid solidification; dissociation of the SO_3 into SO_2 and O_2 is therefore difficult to avoid.

The work is centered on the difficult task of separating the forward reaction products at high temperature. Metal oxide separation processes involving intermediate chemical reactions such as:



are thought to offer a solution despite their disadvantage that the introduction of solids into the reaction scheme makes the systems unsuitable for energy transfer.

Work at the University of Houston is concentrated on the basic chemistry of the separation process. It is recognised that in comparison the ANU work with ammonia is at a more advanced stage of development (W E Wentworth - private communication).

Thus the utility thermochemical programme is in a fairly embryonic stage and this is true also of the solar fuel programme. This is evidenced by the very broad scope of a recent (1979) DOE contract to the University of New Hampshire, valued at \$58 K⁽¹³⁾. The tasks include:

1. Preliminary assessment of potential resources of solar insolation and the following feed stocks and their regional distributions.
(a) wood, (b) biomass, (c) shale, (d) lignite, (e) peat,
(f) anthracite.
2. Initial identification of processes for making fuels from the above.

3.3 ANU Stature in the USA

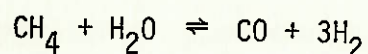
At the STTF Users Association workshop at Albuquerque which was attended by one of us (Dr Carden) the spokesman for the thermochemical panel Dr Chubb reported back to the meeting that it was apparent the ANU Energy Conversion Group was 'furtherest along on thermochemical research'.

This acknowledgement was underscored by the engagement of one of us (Dr Carden) to review the thermochemical research proposals submitted to the STTF Users Association.

4. WEST GERMANY

4.1 Eva-Adam System

Without any doubt the largest research and development programme of all in thermochemical energy transport is being undertaken by the KFA (Kernforschungsanlage-Atomic Energy Commission) Julick in West Germany. The system, commonly called the Eva-Adam system⁽¹⁴⁾, is based on the reaction



and the motivation is the promise that high grade heat may be extracted from nuclear power stations and transported distances averaging 70 km to a variety of industrial consumers. There is therefore a tremendous potential to save oil and coal and to substitute nuclear power for these fuels which are both becoming painfully expensive. (The coal is extracted from deep mines.) Moreover, the high grade heat delivered to the consumer may be divided among a variety of end uses in proportions to suit the individual consumer. For example, some high grade heat might be used for on-the-spot power generation with an above-normal sink temperature in order to meet a low grade heat demand as well.

The programme has now reached the point where a 10 MW demonstration is under construction and nearing completion. This demonstration will cost DM 100M (A\$50M), the capital items amounting to DM 40M or the equivalent of $2\$/W_{th}$. Despite this high initial cost, there is obviously a strong commitment to this technology and confidence that it will become economical in the near future.

One way in which costs will be reduced is through the further development of catalysts that will enable a reduction in reformer temperature and an increase in methylation temperature. Considerable work also needs to be done on the materials problem at high temperature.

A schematic diagram and description of the Eva-Adam system is shown in Figure 5.

4.2 NH₃-SO₃ for Solar Applications

Quite recently the interest at KFA in thermochemical energy transport has shifted to include systems suitable for solar energy applications. This is a natural outcome of the combined effects of KFA's new general interests in solar energy and their past experience in thermochemical energy transport. KFA's interest in solar energy includes the development of mirror concentrators and the testing of other mirror concentrators produced in West Germany and elsewhere.

The move to high temperature solar energy is based almost entirely on prospects of developing a new and lucrative export market for it is generally acknowledged that this will neither be economical or able to make a significant contribution in West Germany for some time yet.

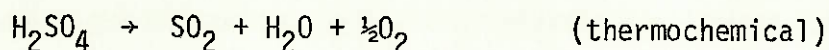
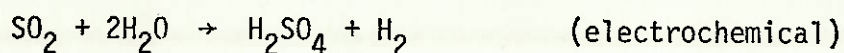
The solar thermochemical programme is presently in the formative stage. It is realised that the Eva-Adam system may not necessarily be the best for solar applications and that other systems such as those based on SO₃ and NH₃ should also be examined. With this in mind the West German researchers approached us early this year with a view to establishing a collaborative agreement.

5. FRANCE

The man in charge of thermochemical work in France is André Vialaron Director of the Laboratoire d'Energetique Solaire du CNRS. The programme described by Prof Vialaron contains several original and imaginative ideas but none of them appear to have been developed to any great extent yet. Some initial work in transport based on SO₃ has begun. This system was chosen primarily because of previous experience with SO₃ reactions.

Another system was described as a hybrid thermochemical-thermoelectric system for splitting water⁽¹⁴⁾. The idea was to split water by electrolysis but to lower the voltage by introducing compounds that could be formed through thermochemical processes.

The following example was given:



The probable advantage of this scheme over what we believe is the thermodynamic equivalent: raising the temperature of electrolysis; is that much higher temperatures may be practically employed in a separate thermochemical process and this would enable a greater proportion of heat to be absorbed by the dissociating water molecules.

Other related projects include a battery rechargeable by heat and new cements of the Portland variety which may be fired at low, solar produced temperatures.

6. COLLABORATIVE POSSIBILITIES

As a result of our visits in the USA and Europe five possibilities for collaborative programmes have been identified: four in the USA and one in Germany. Three of the USA possibilities would result in a division of work where systems studies and demonstrations would be concentrated in Australia while catalyst development and special materials development would be done in the USA. The latter activities could involve one or more of the following:

1. Rocket Research Company: already working on a contract for screening catalysts for the SO_3 system. Experience in materials and catalysts for hydrazine which is similar in many respects to ammonia. Actively pursuing possibility of collaboration.
2. Professor Manasse, University of New Hampshire: excited over the possibility of producing ammonia from sun, water and air. Fits into solar fuels study for which he is presently funded. Expertise in catalysts and materials available.
3. Dr Lenz, Colorado State University: foremost US worker in the ammonia system but very much behind us. Expertise in catalysts.

In addition to the above, it is also possible that an arrangement may be made through the STTF Users Association. This association is already international (we are members, for example, and the French facilities at Odeillo are included in the Association's facilities). There was some discussion about our solar-thermochemical demonstration also becoming a facility of the Association. In some quarters it was thought that we should submit a proposal to the Users Association for funds, in order to test precedence as much as for other reasons.

At KFA Julich, the Director, Professor Hecker proposed an information exchange: we would provide results of our work on ammonia in exchange for unclassified information on the Eva-Adam system. This could include information on costs which would enable us to use the Eva-Adam system applied to solar energy, as a standard against which other systems could be compared.

Exciting as these prospects for collaboration may appear, it was quite evident in the USA that many people, including several top decision makers, believed that competition rather than collaboration was the way to go. Competition is deliberately built into the American system for funding research, and collaboration, even on an international scale, is philosophically abhorrent to some.

The same questions should perhaps be raised here: what are the advantages and disadvantages of collaboration? We pursue it because it would add authenticity to our work and that seems to be important in the context of obtaining Australian support. We also pursue it because we like the prospect of interacting with new and interesting people from whom we may learn something. Collaboration also saves money. But these are the only advantages. One has the feeling that once equality is achieved through collaboration it would then be exceedingly difficult for us to maintain a position of leadership.

7. REFERENCES

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5. J J Iannucci* and J D Fish*, 'Analysis of solar energy conversion processes: systems economic impact of thermochemical storage efficiencis', Proceedings of ISES, Atlanta 1979, *Sandia Laboratories, Livermore, California, 94550.
6. T G Lenz, 'Conversion of solar energy to chemical energy through ammonia dissociation', Proc Solar High-temperature Industrial Process Workshop, (Solar Thermal Test Facilities Users Association), Atlanta Georgia, September 1978 (available from National Technical Information Service).
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*Colorado State University, Fort Collins, Colorado.
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9. P O Carden, Solar Energy 19, 1977, pp 365-378.
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11. General Electric Company (H B Vakil and J L Flock), 'Closed loop chemical systems for energy storage and transmission (chemical heat pipe)', prepared for DOE. ERDA contract EY-76-C-02-2676 February 1978.
12. C A Vancini, 'Synthesis of ammonia'(English translation by L Pirt), MacMillan, London 1971.
13. +F K Manasse*, V K Mathur*, 'A study of alternate fuels manufactured from high temperature solar thermal equipment', Proposal to DOE 1979, *University of New Hampshire.

14. R Harth, K Kugeler, H F Niessen, V Boltendahl and K A Theis, Nuclear Technology 38, April 1978, pp 252-7.
15. A C Vialaron, 'Hybrid chemical concepts for storage of solar thermal energy', Internal Report CNRS/PIRDES Toulouse, 31 France.

+ These submissions are not yet generally available and their status at present is confidential.

8. FIGURE CAPTIONS

Figure 1: Results of a study of Turner [2] of the cost of various forms of energy transport for distributed solar collector systems

(1) hot water	315°C output,	200°C return	100 Ats
(2) steam/water	510°C output,	40°C return	100 Ats
(3) liquid metal	650°C output,	170°C return	7 Ats
(4) hot helium	650°C output,	370°C return	100 Ats
(5) $\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$ *	40°C output,	40°C return	100 Ats
(6) Therminol 66	315°C output,	200°C return	7 Ats

* three pipe lines (H_2O separate)

Figure 2: Results of a study by Livingston [3] of the expected cost of various solar power generating systems by year 2000. The 'dish-chemical' costs are based on paraboloidal mirror collectors and thermochemical energy transport using $\text{CH}_4 + \text{H}_2\text{O}$.

Figure 3: Energy and steam flows of a modern ammonia synthesis plant. Based on Vancini [12].

Figure 4: Energy and steam flows of the section of Figure 3 relevant to the converter and hence relevant to thermochemical energy transport.

Figure 5: Schematic of the Eva-Adam system developed in West Germany for thermochemical energy transport from nuclear power stations.

9. INSTITUTIONS VISITED

1. Bureau de Recherches Géologiques et Minières, Orleans, Cedex, France.
2. Colorado State University, Fort Collins, Colorado.
3. Department d'héliophysique, Marseille, France, Université de Provence.
4. Institut du Genie Chimique, Toulouse, France.
5. JPL, Pasadena, California.
6. Kernforschungsanlage (KFA) Julich, Federal Republic of Germany, (Atomic Energy Research Establishment).
7. McDonnell Douglas Astronautics, Huntington Beach, California.
8. Naval Research Laboratory, Washington, DC.
9. Martin Marietta Aerospace, Denver, Colorado.
10. Pullman Kellogg, Houston, Texas.
11. Rocket Research Company, Seattle, Washington.
12. Sandia Laboratories, Albuquerque, New Mexico.
13. Sandia Laboratories, Livermore, California.
14. Société Française d'Études Thermiques et d'Energie Solaire (SOFRETES), Montargis, France.
15. Solar Energy Research Institute, Golden, Colorado.
16. Solar Thermal Test Facilities Users Association, Albuquerque, New Mexico.
17. University of Houston, Houston, Texas.

10. OVERSEAS CONTACTS

1. Dr L C Axelrod, Senior Vice President, Pullman Kellogg, Houston, Texas.
2. Dr M G Bowman, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.
3. Dr E C M Chen, Department of Chemistry, University of Houston, Houston, Texas.
4. Dr T A Chubb, Head, Solchem Project, Naval Research Laboratory, Washington, DC.

5. Dr M G Clemont, Manager,
SOFRETES, Montargis, France.
6. Dr T Cole, Senior Engineer,
Ford Motor Company, Dearborn, Michigan.
7. Dr R J Copeland, Senior Systems Engineer,
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8. Dr J E Dwan II, Manager, International Marketing,
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9. Dr J D Fish,
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13. Dr M U Gutstein,
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17. Dr R J Holl, Senior Systems Engineer,
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18. Dr J J Iannucci,
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19. Dr H P Leftin, Manager of Research,
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20. Dr L P Leibowitz, Task Manager,
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21. Professor T G Lenz,
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22. Dr F K Manasse, Professor Electrical and Computer Engineering,
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23. Dr A T Marriott, Technical Manager,
Jet Propulsion Laboratory, Pasadena, California.
24. Mr J J Nemecek, Program Manager Solchem Project,
Naval Research Laboratory, Washington, DC.
25. Dr R L Parker, Manager Requirements,
Martin Marietta Aerospace, Denver, Colorado.
26. Professor G Peri, Head, Department d'Heliophysique,
Université de Provence, Marseille, France.
27. Dr E W Schmidt, Senior Staff Scientist,
Rocket Research Company, Seattle, Washington.
28. Dr F B Smith, Manager,
STTF Users Association, Albuquerque, New Mexico.
29. Dr R J Smith, Principal Investigator,
Rocket Research Company, Seattle, Washington.
30. Dr C J Swet, Consultant to DOE,
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31. Dr J Vanet, Head, Department of Geothermie,
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32. Dr L Vant-Hull, Energy Laboratory,
University of Houston, Houston, Texas.
33. Professor A Vielaron, Director,
Laboratoire d'Energetique Solaire du, CNRS, Toulouse, France.
34. Professor W E Wentworth,
Department of Chemistry, University of Houston, Houston, Texas.
35. Dr C E Wyman, Senior Engineer,
Solar Energy Research Institute, Golden, Colorado.

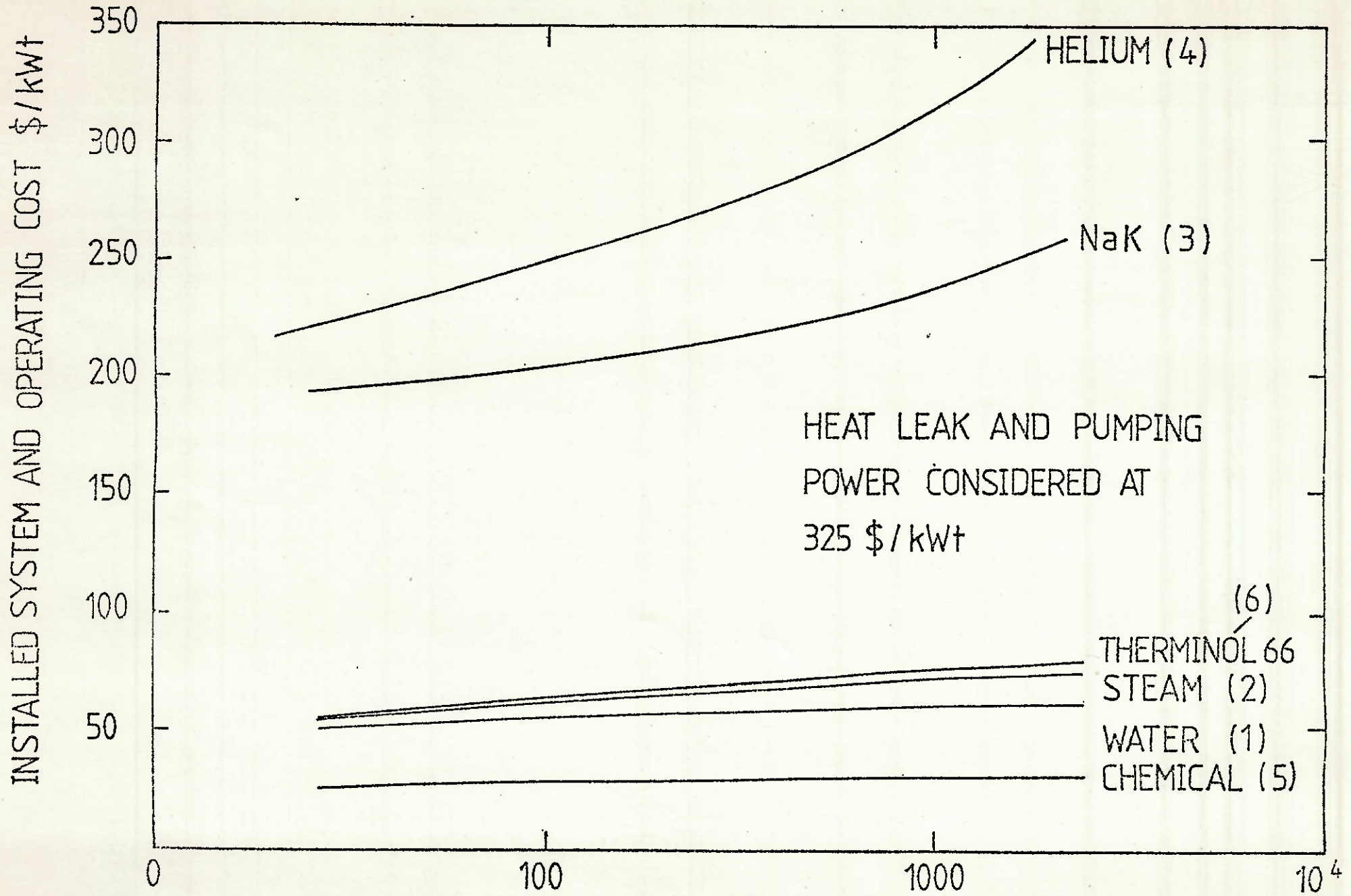


Fig. 1

DELIVERED THERMAL POWER (MWt)

- YEAR 2000 PLANT STARTUP
- ANNUAL CAPACITY FACTOR: 0.55

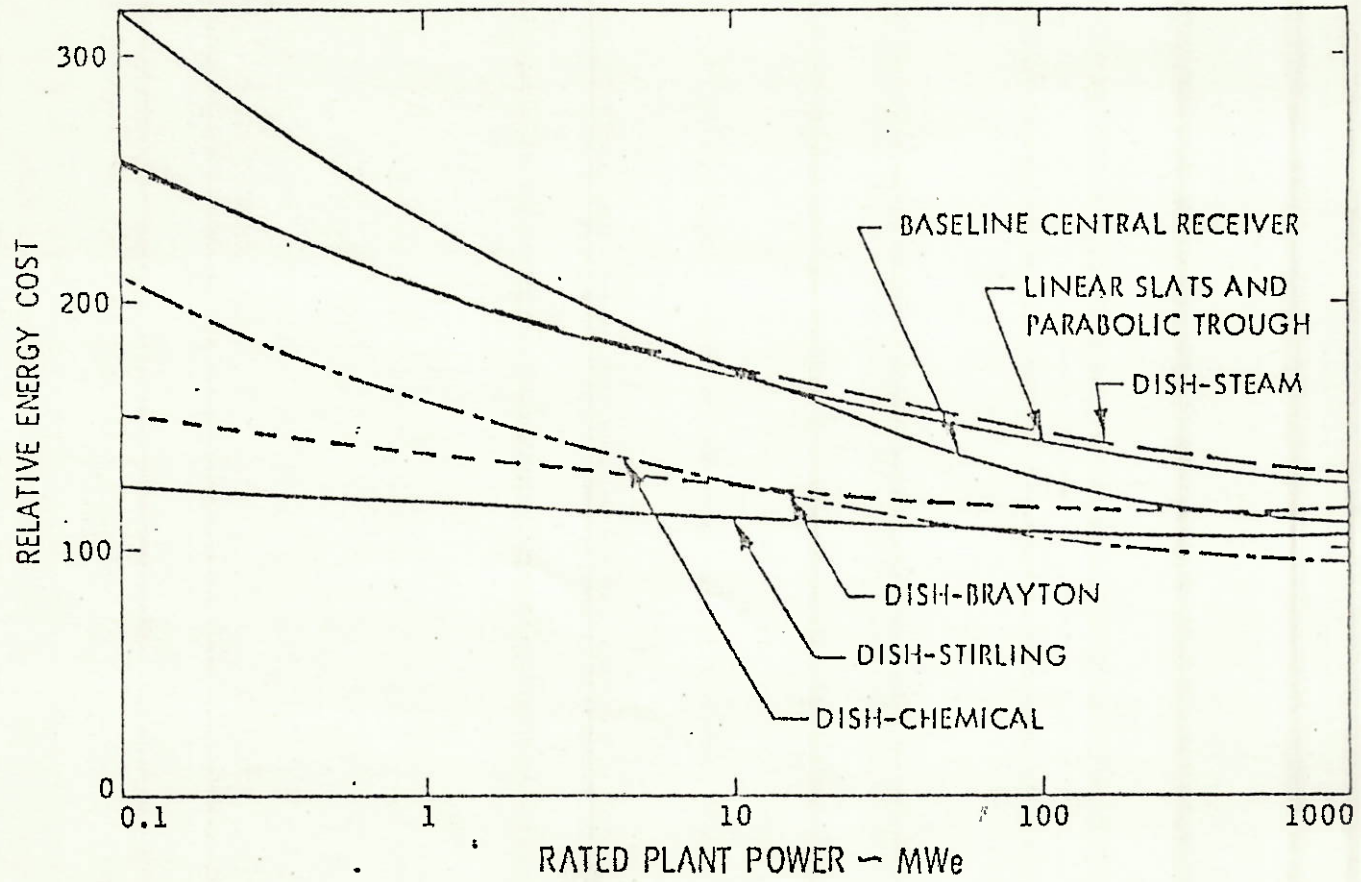


Fig. 2

AMMONIA PRODUCTION 1000 t/d

N^o 3

STEAM PRODUCTION 45.2 t/h

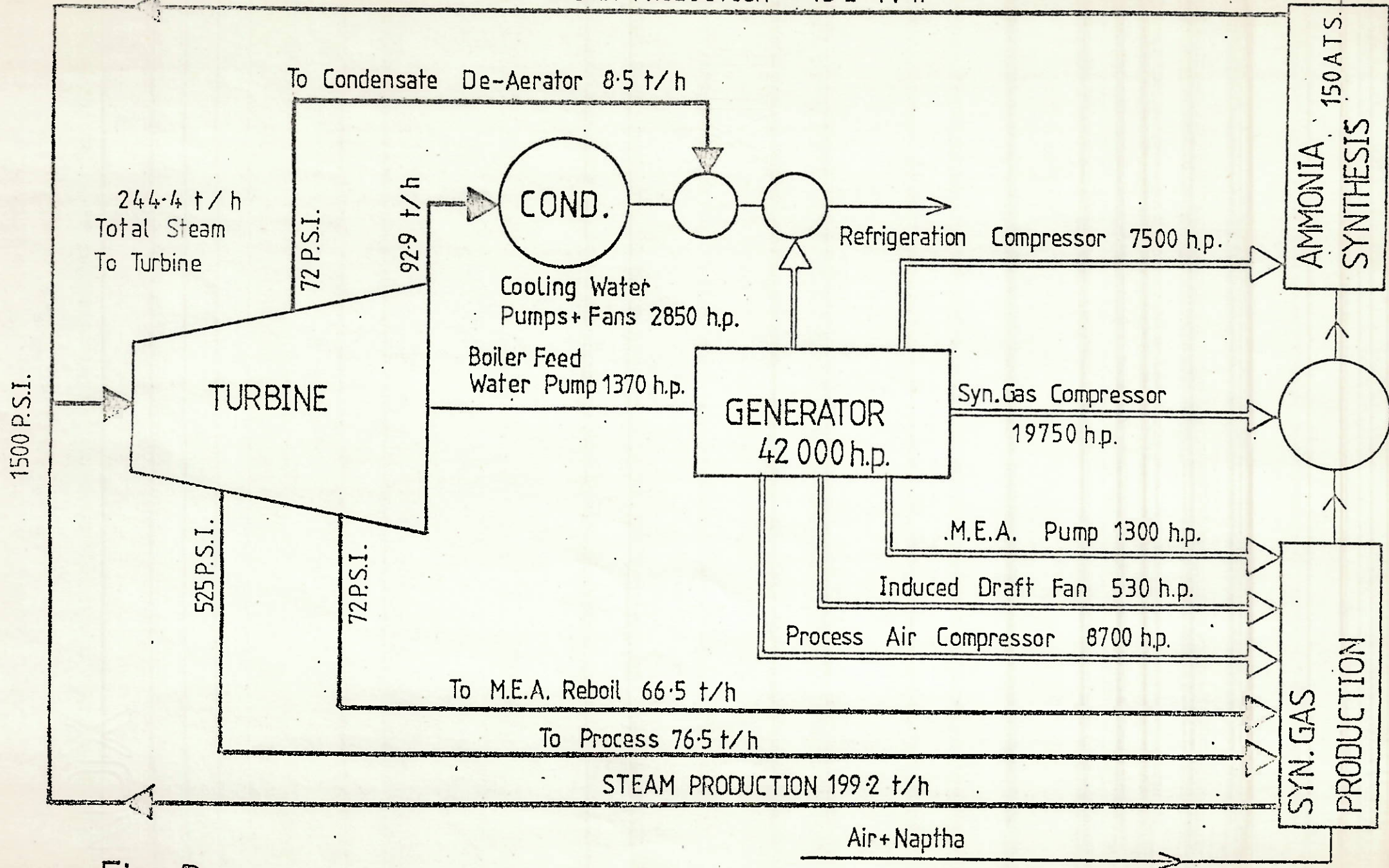


Fig. 3

AMMONIA PRODUCTION 5000 t/d

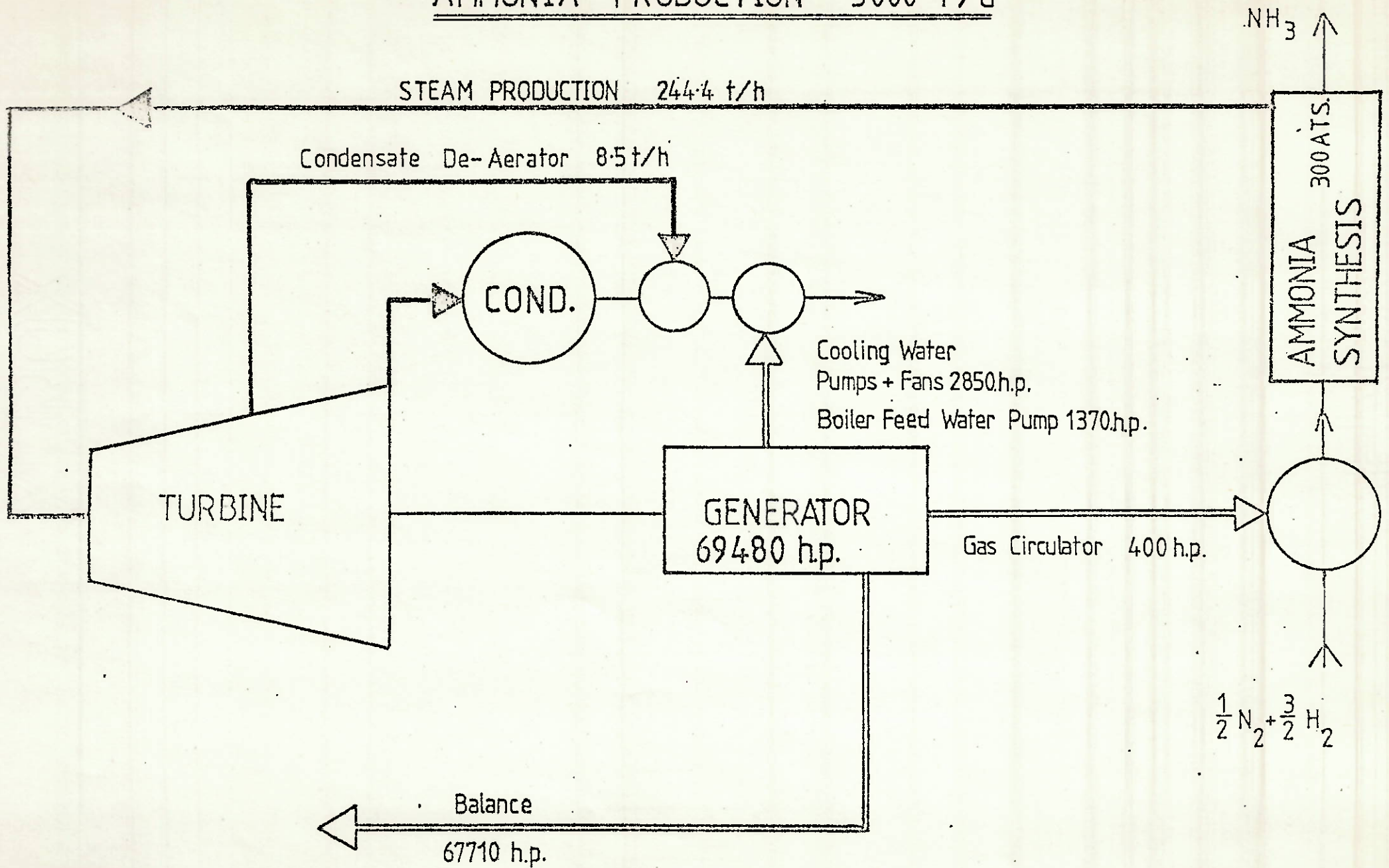


Fig. 4

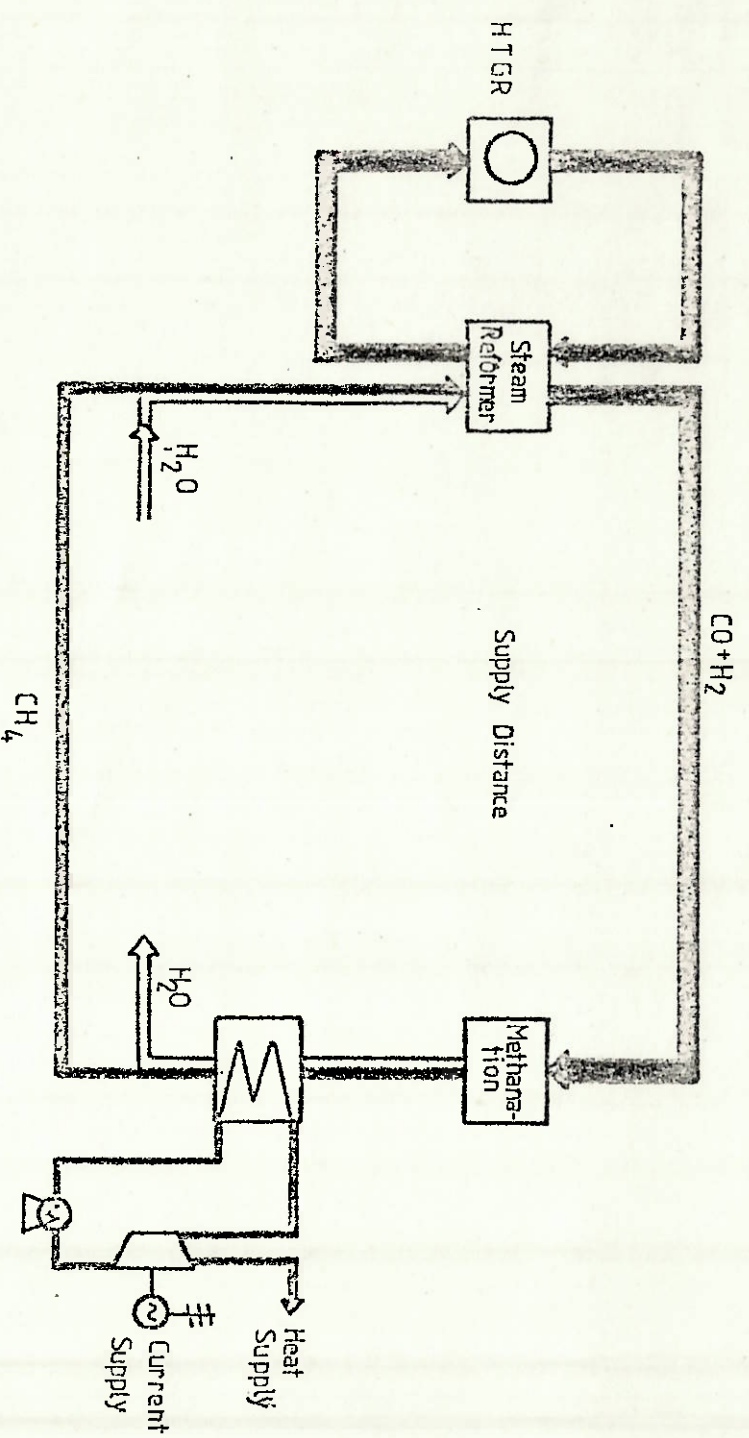


Fig. 5a.

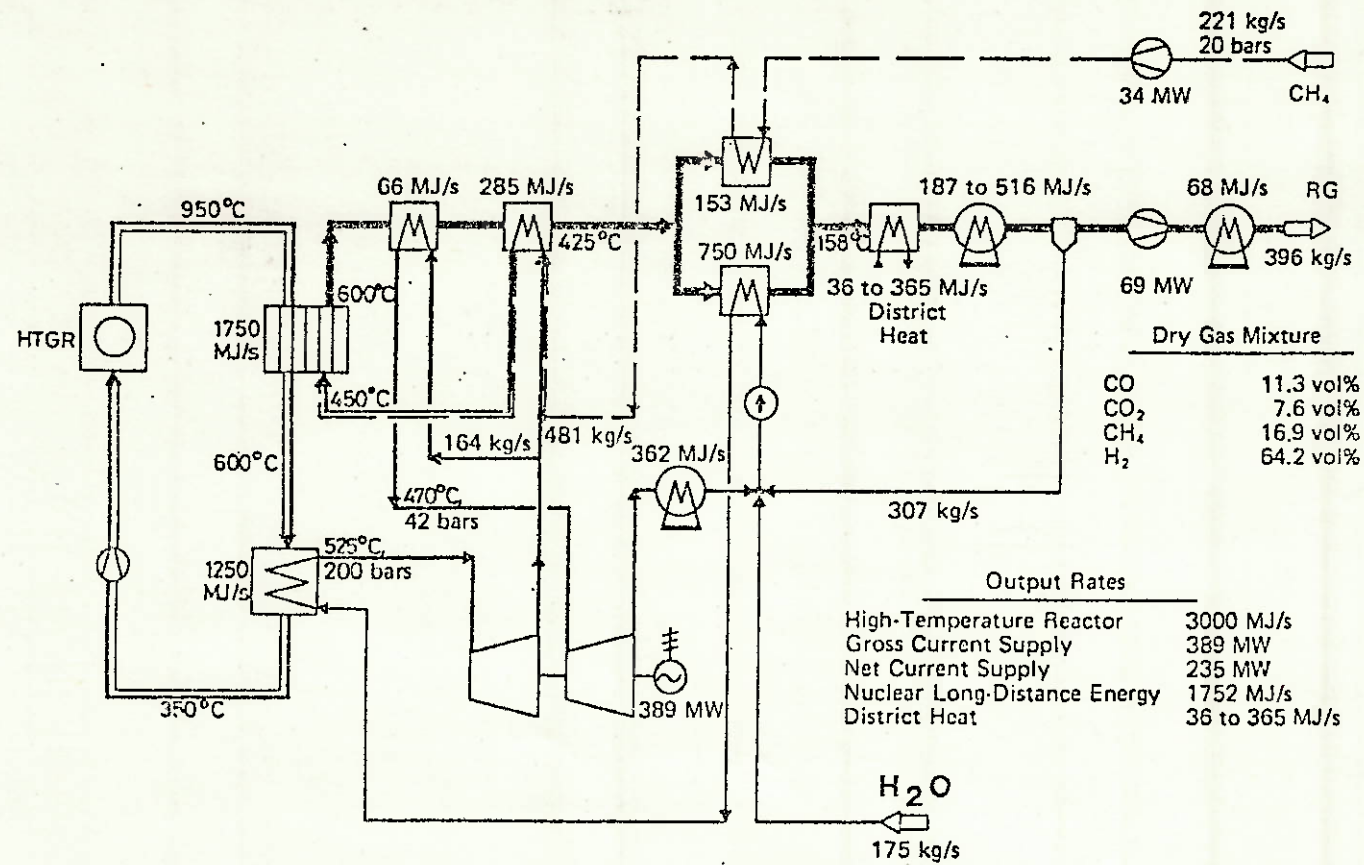


Fig. 5b

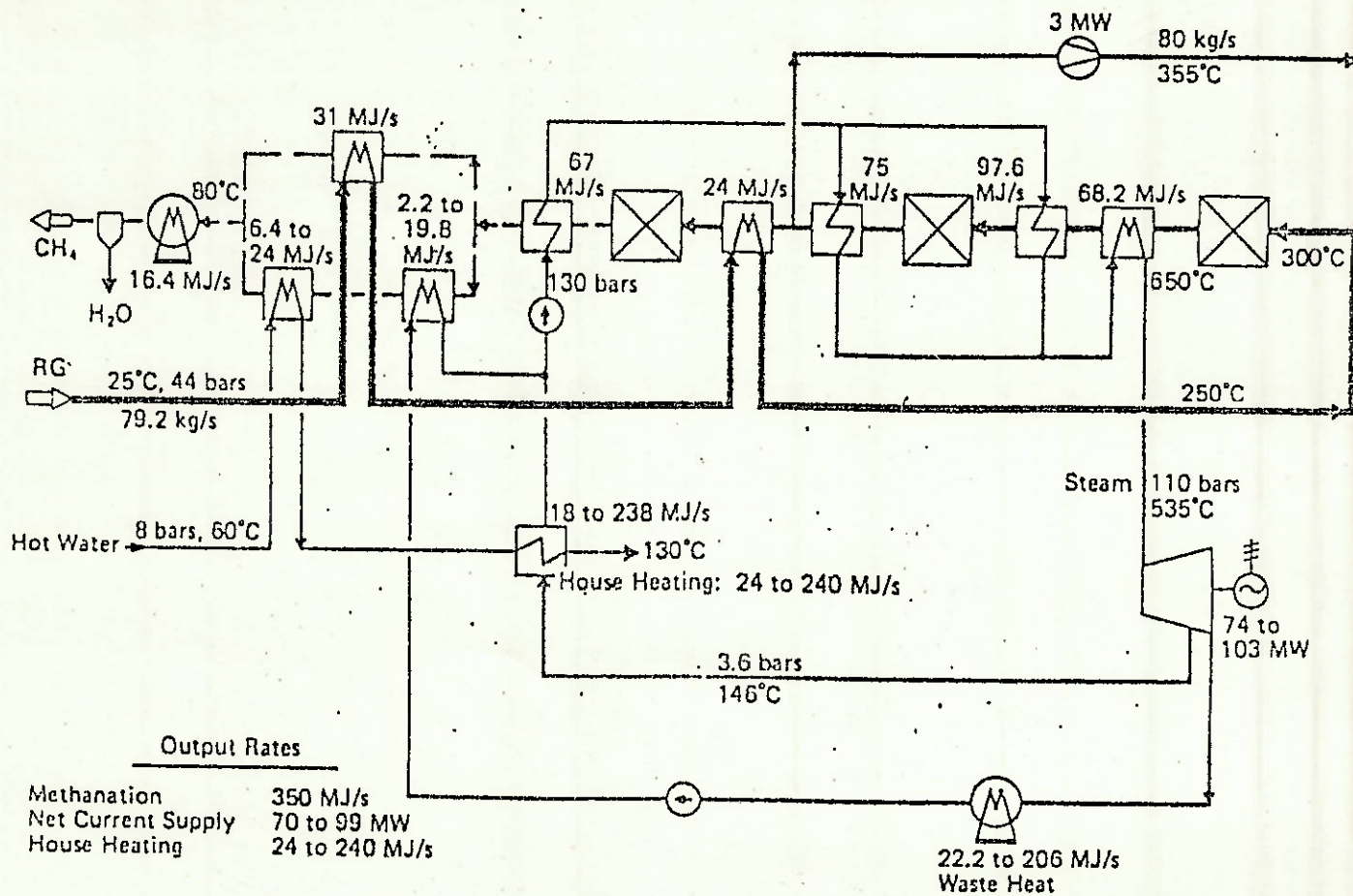


Fig. 5c

