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PROCEEDINGS OF THE 24th INTERSOCIETY ENERGY CONVERSION ENGINEERING CONFERENCE

VOLUME 4 Conservation

Environmental Issues

Planning and Operation

Storage

Thermal Management

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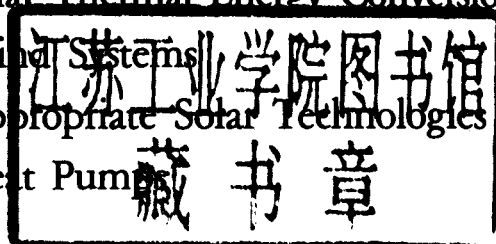
Biomass Energy Conversion Systems

Solar Thermal Energy Conversion

Wind Systems

Appropriate Solar Technologies

Heat Pumps



Editor:
William D. Jackson

Associate Editor:
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PREFACE

The scope of the Intersociety Energy Conversion Engineering Conference (IECEC) series has grown from a straightforward treatment of energy conversion engineering to a broad involvement in all aspects of energy systems, including primary resource processing, utilization and environmental issues in addition to conversion and storage. The other notable trend has been toward increased international participation and, as the papers in this volume attest, IECEC-89 can truly claim to be an international forum on energy engineering with a significant proportion of the papers being presented being from beyond the confines of the United States. It is a particular pleasure to include, for the first time, a sizeable contribution from the USSR.

These developments are indeed appropriate for energy conversion engineering has to be undertaken within the broader context of energy systems and energy issues which are not constrained by national boundaries. The increasing and proper concern for the impact of energy systems not only locally but also globally is particularly reflected in the nearly 500 papers appearing in a Proceedings which has now grown to six volumes. In these, the reader will find a comprehensive coverage of recent work on energy systems and technologies relevant to the expected conditions of the 1990's and beyond. The international character of IECEC-89 shows not just through participation from many countries but the large measure of common ground evident in the contributions made by the many national and international organizations involved in the energy engineering field.

The organization of this large amount of interrelated material poses considerable challenges which have been met in the first instance by dividing the Conference and, by extension, the Proceedings, into a number of major topical areas. From a narrow applications viewpoint, it is tempting to view aerospace and terrestrial energy system issues separately since they generally involve meeting different criteria. This has not been done in the present Conference in large measure because an IECEC objective is to find and emphasize points of commonality. Accordingly, a blend of interests will be found throughout, as for example, in Volume 2 devoted to energy conversion technologies where space and terrestrial photovoltaics are grouped together.

To facilitate use of the volumes, the Table of Contents is repeated in each Volume and an author Index appears in Volume 6. In addition, the now well-established feature of IECEC, the SAE cumulative index for the past four years is also included in Volume 6. This may be used to locate recent related work reported at the IECEC and, in due course, it will be updated to include the current Conference.

The task of preparing these Proceedings has only been possible through the unstinting efforts of the Program Committee, Session Organizers and the IECEC-89 staff whose many contributions are gratefully acknowledged by the Editors. In fairness to other technologies in which the Institute of Electrical and Electronics Engineers is prominently involved, the critical role of computers, FAX machines and other aspects of modern communications technology in permitting the assembly of this body of material also deserves recognition.

William D. Jackson, *Editor*
Dorothy A. Hull, *Associate Editor*

24th Intersociety Energy Conversion Engineering Conference

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IRREVERSIBILITY REDUCTION OF LOW-TEMPERATURE, COMBUSTION HEAT SOURCES

Carl J. Bliem

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ABSTRACT

This paper discusses a concept that will reduce irreversibilities associated with combustion and, thereby, deliver more heat to a process than the heating value of the fuel. The concept involves the use of staged reactions to facilitate the more reversible reactions associated with the combustion process. The system is an open cycle chemical reaction heat pump working between the atmosphere and the desired heat sink. The system is described in the paper. The paper discusses the potential efficiency of this system and the methodology of searching for the required intermediate components.

INTRODUCTION

The use of combustion to supply heat to relatively low temperature heat sources is quite irreversible and, therefore an inefficient use of fuel. The less efficient the combustion is, the greater the amounts of carbon dioxide and other "green-house" gasses for each unit of heat utilized in a given process. A system to make this type of process more reversible and effect abatement of "green-house" gas is discussed in this paper.

The system proposed here consists of two (or more) reactors whose net effect is to oxidize the fuel. One of the reactors, an endothermic reactor, takes place at a temperature below ambient. The energy for this reaction is supplied by heat transfer from the atmosphere. The second reactor has an exothermic reaction taking place at a temperature above the desired temperature sink allowing the heat generated in the reaction to transfer to the sink. The reactants, fuel and oxygen are introduced into separate reactors to allow a partial reaction in each reactor. The net effect is an open-cycle, chemical-reaction heat pump that delivers more heat to the sink than would be produced by the conventional reaction of the fuel by the amount of heat transferred from the atmosphere.

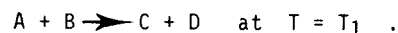
This work is supported by the U. S. Department of Energy (DOE), Energy Conversion and

Utilization Technology Division, under Contract No. DE-AC07-76ID01570. Mr. Marvin E. Gunn is the program manager at DOE Headquarters.

CONCEPT

May and Rudd [1] developed a methodology to synthesize reaction sets using the concepts involved in the Solvey process to produce given reaction products. Lauerhass and Rudd [2] extended this procedure to apply to a series of closed cycle chemical reactions to serve as a heat pump with endothermic reactors receiving heat from an intermediate temperature source and exothermic reactors rejecting heat to higher and lower temperature sinks. This type of chemical heat pump was named a Solvey type chemical heat pump. This methodology gives some insight into the combustion process. In this section, the methodology is reviewed and then applied to the combustion of a hydrocarbon fuel.

The method mentioned in the previous paragraph explores the limits of thermodynamic feasibility of reaction systems through the use of Ellingham diagrams involving "half reactions" with common differences. Considering the chemical reaction, we have



The thermodynamic feasibility of this reaction may be expressed in terms of the change in Gibbs function (free energy) for the reaction, ΔG , where

$$\Delta G = G_C + G_D - G_A - G_B,$$

and is less than zero.

A methodology to determine these conditions [1,2] is:

1. Decompose the reaction into two "half reactions" made up of one reactant paired with one product. For the example given above, these might be:



and



The overall reaction would be



(Note that because of stoichiometry, the "common difference", chemical species left over in each combination will be the same.)

- Plot the Gibbs function of formation for each half reaction as a function of temperature for each half reaction. This Gibbs function-temperature diagram is called an Ellingham diagram.
- The allowed direction of the reaction is that going from higher to lower curve.
- It has been shown [3] that the enthalpy of reaction at a given temperature is the distance between the tangents of the two reaction curves extrapolated to absolute zero. This allows the determination of which reactions are endothermic and which are exothermic as well as the heat of reaction (if reactants and products are at the reaction temperature).

Considering the chemical reaction problem expressed in these terms, Figure 1 shows an Ellingham diagram for the combustion of methane in oxygen. The common difference in each half reaction is atomic oxygen, 0. The values used to generate this plot are taken from Barin and Knacke [4]. In this reference, the enthalpies of formation are taken to be zero for elements at 273 K and absolute entropies are used to determine the Gibbs function. Note that the functional relation between the change in Gibbs function and temperature is approximately linear. The change in slope in the lower curve indicates the in phase of the water in the products from vapor to liquid.

Figure 1 indicates that it is feasible to oxidize methane at any temperature, i.e., go from the oxygen curve to the products minus fuel curve

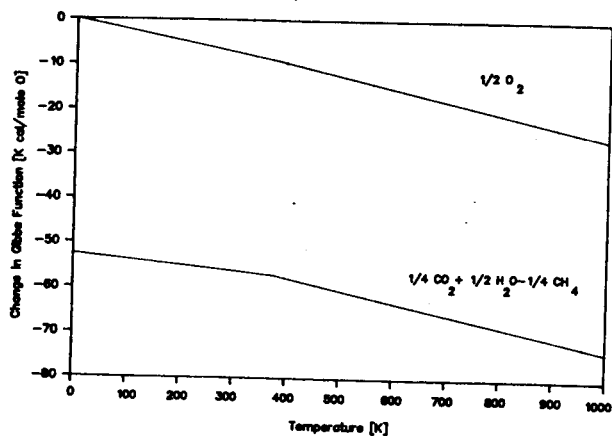


Figure 1. Ellingham Diagram for Combustion of Methane in Oxygen.

at a lower Gibbs function. Extrapolation of the curves to absolute zero indicates that the "heat of reaction" is approximately constant except for the change from water vapor to liquid water in the products.

The large change in Gibbs function indicates a large irreversibility in the combustion process. A possible alternative is to carry out the reaction in two steps. Figure 2 shows the Ellingham diagram for such a reaction system and Figure 3 shows a schematic diagram of this system. For this example, consider a sink for the heat of combustion, T_S , at 600 K and ambient temperature, T_A , at 300 K. There are two classes of this type of system. Consider a Class I system. Here an exothermic reaction takes place at 600 K oxidizing the intermediate product, M_1 . In its simplest form M_1 might be a metal or metal oxide that is oxidized with one additional oxygen atom. (M_1 might be a complex system of molecules, but with the effect of being oxidized with one further oxygen

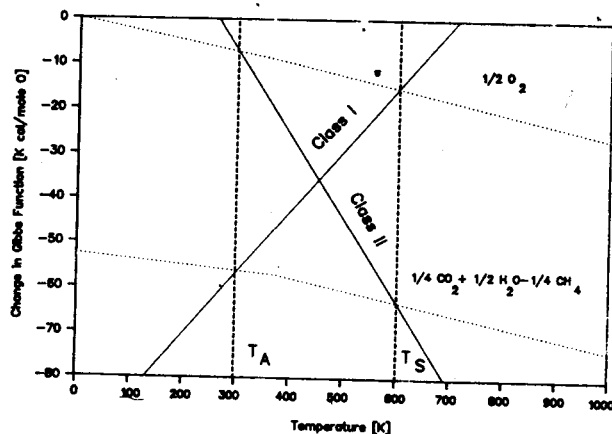


Figure 2. Ideal Two-Reactor Systems

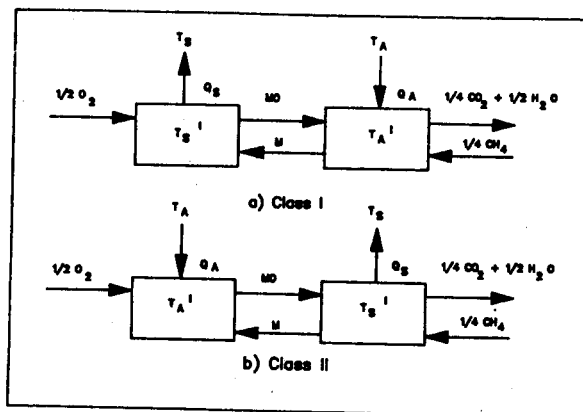


Figure 3. Two Reactor Systems

atom.) The M_1O is then carried to a second reactor where it is reduced by the fuel to form M_1 and the normal combustion produces, carbon dioxide and water. The M_1 is, thereby, regenerated and recycled to the original reactor. The second reactor will be endothermic, receiving its heat from the atmosphere.

Note that the intersection of the oxygen and intermediate, $M_1O - M_1$, half reactions with the absolute zero temperature line indicates a much larger heat of reaction than that of the original combustion because it is carried out in a more reversible manner. This result is not surprising in that the thermodynamic potential of the chemical reactants to produce work is nearly that of the heating value of the fuel. That is, if a perfect fuel cell were developed, its output in work would be nearly the heating value of the fuel. If the same process was carried out in a reversible conversion of chemical energy to heat to work, a substantially larger amount of heat would be generated to create the reversible work, if the heat were produced at a relatively low temperature. This should mean that if reversible combustion is achieved, a substantially larger amount of heat than the fuel heating value can be delivered at a low temperature.

If the search for a possible intermediate reaction with a single reaction is too restrictive, perhaps two intermediate half reactions will be necessary. Figures 4 and 5 illustrate such systems. Here three reactors will be needed and two recyclable chemical pairs. Two classes of systems, Class III and Class IV, are shown. As with the single intermediate reaction systems, the differences are in the temperatures at which the individual reactor operate. The Class III system has two endothermic (low temperature) reactors and one exothermic (high temperature) reactor, while the Class IV system has a single endothermic (low

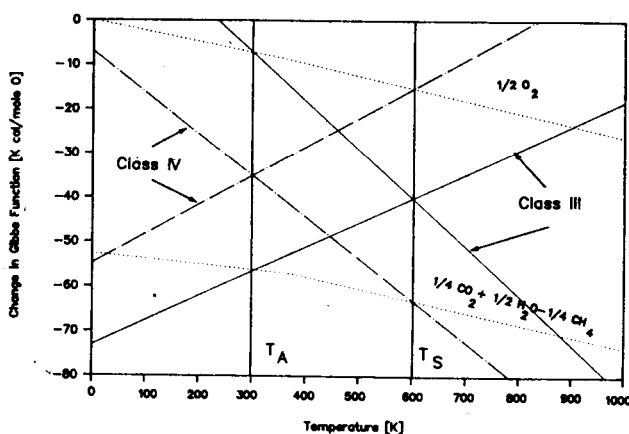


Figure 4. Ideal Three-Reactor Systems

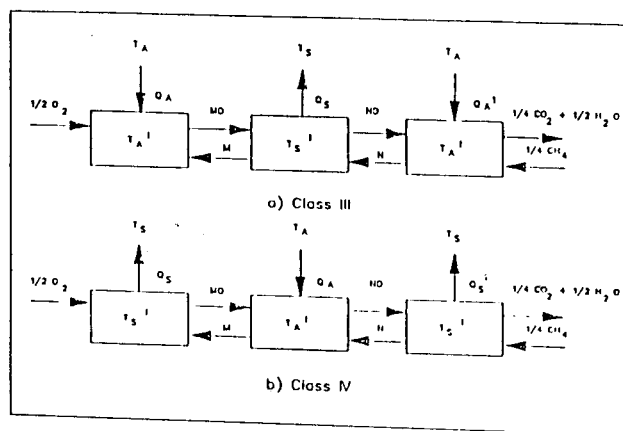


Figure 5. Three Reactor Systems

temperature) reactor and two exothermic (high temperature) reactors. Figure 4 shows the Ellingham diagram for the ideal temperature situation.

RESULTING PERFORMANCE

The systems depicted in the previous section represent idealized systems. Assumptions were made that the reaction would take place at realistic rates up to the point at which the change in Gibbs function between reactants and products is zero. Experience dictates that this should be a negative number in the order of 5 k cal/mole O. Similarly, in order to transfer heat to a sink at T_S , the reactor must be at a higher temperature; to receive heat from the environment, the reactor temperature must be below the ambient temperature, T_A .

Figure 6 shows how these constraints effect the desired half-reaction line on the Ellingham diagram for a Class I system. The line labeled ideal is the same line shown on Figure 2. The exothermic reactor is moved to a temperature T above T_S to allow heat transfer to T_S , and the endothermic reactor operates below T_A by an amount T to allow heat transfer from the environment. At these operating points, a Gibbs function decrease of G for each reaction is also forced to occur. Note that this decreases the heat generated in the exothermic reactor (the distance between the intercepts of the zero absolute temperature line by the half reaction lines).

These constraints were used to estimate the performance of these systems as a function of the sink temperature. Figure 7 shows the performance as a ratio of the actual heat delivered to the

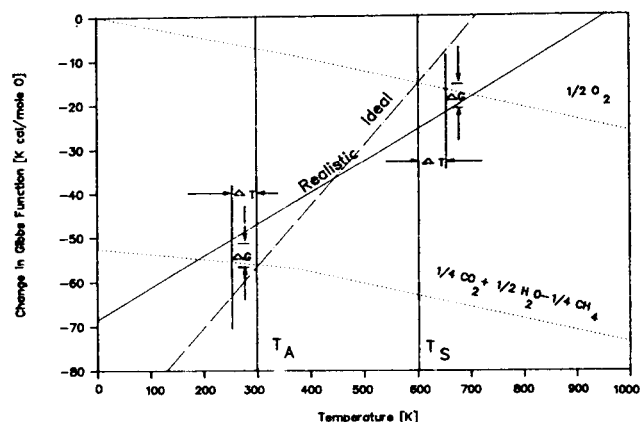


Figure 6. Realistic Class I System

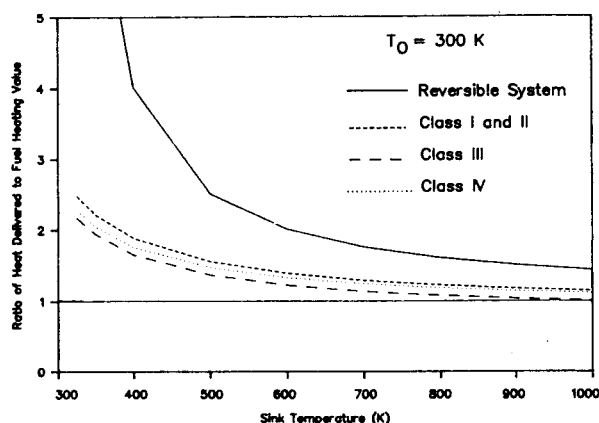


Figure 7. Performance of Systems with a 300 K Ambient Temperature

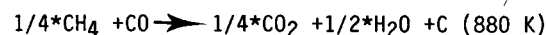
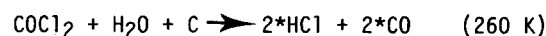
sink to the fuel heating value for different sink temperatures. (This parameter is essentially the heat pump coefficient of performance for a prime energy driven device.) For this comparison, the ambient temperature was taken to be 300 K. The highest curve represents the reversible (ideal) situation in which the reactors are at the temperature of the respective sources and sinks of heat, and that the change in Gibbs function between half reactions is zero. For this case, the results are the same for each class of system. For the more realistic situation it was assumed that the difference in temperature between the reactors and the source/sink was 50 K, and that a 5 k cal/mole O decrease in Gibbs function was used for each reaction to proceed. Note that the results are the same for the two reactor systems. For the three reactor systems there is a difference, however. For a sink at 600 K, a totally reversible system would deliver approximately twice the fuel heating value. A

realistic two reactor system would deliver 1.39 times the fuel heating value and a three reactor system, 1.33 or 1.22.

The greatest benefit of this type of device would be at low sink temperatures, below 500 K. Here, the benefit would be substantial. An additional use of this type of system would be for space conditioning. Figure 8 shows similar information to the previous figure for an ambient temperature of 255 K (0 °F). Here, for normal inside temperatures, the "effective coefficient of performance would be two, i.e., twice the heating value of the fuel would be delivered to the heated space at the inside temperature

Figure 9 shows the Ellingham diagram for a system with actual reactions for a three reactor system (Reactors operating at 880, 760, and 260 K). The high temperatures associated with this reaction set is higher than the optimum for energy saving (see Figure 7). This system would allow the production of steam and superheating it to around 800 K (1000°F). Figure 10 is a schematic diagram showing the reactors for this Class IV system.

The reaction system under consideration consists of three reactions given as



If no regenerative heat transfer is used, the system will produce only as much heat in the high temperature reactors as the heating value of the fuel. If two pairs of streams are recuperated (two gas/gas heat exchangers), this increases to 1.17. This is around 90% of the maximum possible increase which that require at least twice that number of heat exchangers.

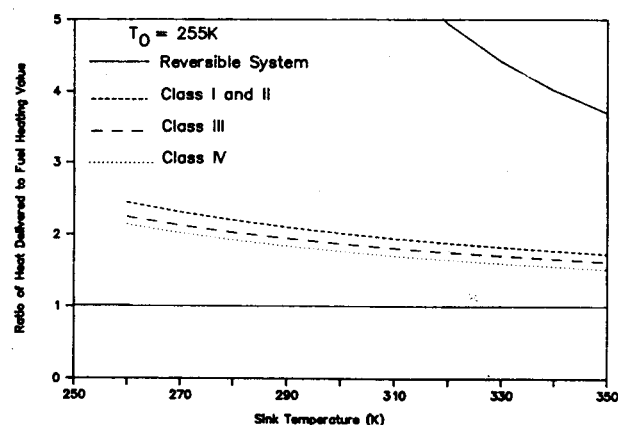


Figure 8. Space Heating Performance

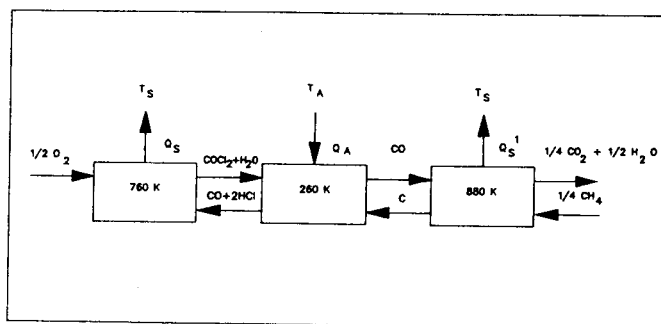


Figure 9. Schematic Diagram of Proposed System.

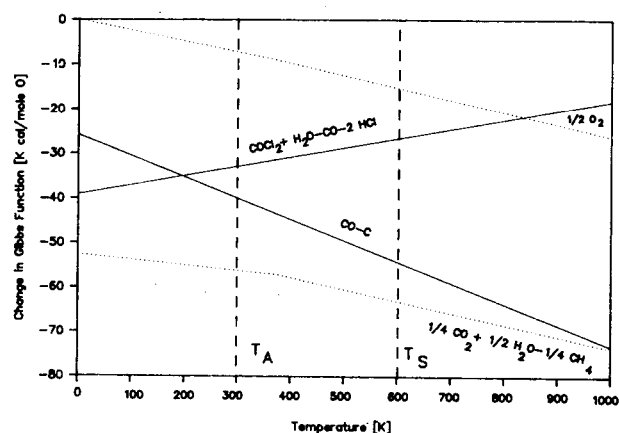


Figure 10 Ellingham Diagram of Proposed System

There is one point that makes this choice of intermediate products unworkable. There will be a tendency of the carbon to form carbon dioxide instead of the desired products. It may be possible to overcome this tendency by exerting appropriate chemical control, but in practice the scheme may fail. It would be better to use intermediate products that will not form carbon dioxide and water. A more appropriate search for intermediate compounds would be for those with no carbon or hydrogen if the goal is to burn a hydrocarbon fuel. Candidate intermediate compounds should then be searched for among inorganic substances. The Ellingham Diagram gives the specifications for Gibbs function temperature relationships for postulated half reactions.

One additional problem remains in the implementation of this type of system. Products leaving the individual reactors must be separated. For example, in the third reactor, carbon must be separated from the carbon dioxide and water vapor that are the products of combustion. This will be a simple separation

because the carbon will be in the solid state and the combustion products gaseous. In the second reactor, the products are equal numbers of moles of carbon monoxide and hydrogen chloride. Half of the carbon monoxide is fed to the third reactor and the remainder of the products is fed back into the first reactor. This separation will be more difficult.

A second problem results if the reactions do not go to completion. In these cases, the reactants must be separated from the products and fed back into the reactor. This will create additional separation problems. One potential solution to this problem is to investigate the degradation in performance if this separation is not made. This strategy will put additional reactants into the given reactors and may adversely affect the reactor performance. If the final products (which are released from the system) contain any of the intermediate chemicals, additional makeup of the intermediate chemicals will be necessary. In addition, environmental problems may result from the release of these chemical into the atmosphere.

The kinetics of the individual reactions must be suitably rapid to allow the system to operate at an appropriate rate. Each of these problems must be handled on an individual basis when a thermodynamically appropriate set of reactions is determined.

CONCLUSIONS AND RECOMMENDATIONS

This paper has presented a scheme by which the combustion process can be made more reversible when supplying heat to sinks at relatively low temperatures. The following conclusions may be drawn from the work to date:

1. Systems with two or three reactors can substantially increase the heat released from the combustion of a hydrocarbon fuel when the ultimate use of the heat is a sink in the 400 to 500 K range.
2. With realistic assumptions about the temperatures and Gibbs function changes in reactions, the heat delivered can be 1.5 to 2 times the fuel heating value.
3. Space conditioning heat pumps with coefficients of performance in the range of 2 can be devised using this type of system when pumping heat from 255 K (0°F) to typical inside conditions.
4. Proper choice of the intermediate reacting compounds is necessary for the implementation of this class of system. The text of the paper discusses the main considerations in choosing these substances.

It is recommended on the basis of the conclusions above that a search be instituted

to find appropriate chemical reactions for the implementation of this type of system. At that time, extensive investigation of the feasibility of this device can be carried out.

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