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Nonterrestrial Chemical Synthesis of Food

JULY 1966

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Prepared for BALLISTIC SYSTEMS AND SPACE SYSTEMS DIVISIONS
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Los Angeles, California

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NONTERRESTRIAL CHEMICAL SYNTHESIS OF FOOD

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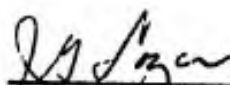
FOREWORD

This report is published by the Aerospace Corporation, El Segundo, California, under Air Force Contract No. AF 04(695)-669.

This report, which documents research carried out from 1 September 1965 through 30 March 1966, was submitted on 1 July 1966 to Captain John T. Allton, SSTRT, for review and approval.

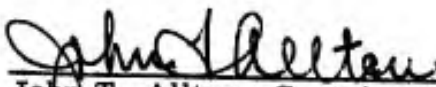
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Research Laboratory

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ABSTRACT

The problem of food logistics in long duration manned space flights is explored. It is concluded that development of a purely chemical method for converting human wastes to useful nutrients is essential, either as a primary system or as a backup for an otherwise primary biological system. The principal requirements for such a chemical system are operational simplicity, efficiency of energy utilization, high yield, and reasonably fast rates of reaction. Various possibilities are considered, based on a thorough analysis of the existing pertinent chemical literature. This is correlated with known metabolic data. Special emphasis is given to the synthesis of structurally simple nutrients rather than the immensely complicated chemical ensembles that we normally know as food. It is recommended that special attention be given to the synthesis of carbohydrates by the base catalyzed polymerization of formaldehyde, since this process fits all the requirements and there is available experimental evidence that the endproducts could be made to be nutritionally acceptable by choice of appropriate reaction conditions.

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I. INTRODUCTION

The conversion of human wastes to foods has recently become a very important technological problem because of space accomplishments that, for the first time in man's history, permit active planning for long duration space flights. This inevitably requires development of a complete ecological system that will continuously regenerate oxygen, useful water, and foodstuffs from metabolic waste products. It is the food aspect, which is perhaps the most difficult to achieve, that is explored here.

Admittedly, most of the emphasis and research in this field has been biological, involving research into the growth of microorganisms. However, as Arnon (Ref. 1) has put it, this has aspects of crop farming in a space ship, with the attendant biological risks of crop failure. Arnon further states that the biological problems of space travel will not be solved unless an adequate supply of oxygen, water, and food can be guaranteed independently of the hazards of such a crop failure. This philosophy accentuates the need for analyzing the possibilities of converting wastes to food by chemical means. By such a process one loses the advantage of having the complex synthesis carried out by living organisms, with their many catalysts that presumably are the results of prolonged evolutionary processes. However, it should be easier for humans to repair errors that develop in a chemical reactor than to cope with mishaps with living cells. It should further be noted that foods need not necessarily be as structurally complex as those to which we are accustomed on earth.

The necessity for conversion of human wastes to useful nutrients arises from the logistics of space travel. Because of the very high propellant-to-payload ratio required for space launchings from the earth, payload weight becomes an overriding factor. For this reason it becomes increasingly difficult to carry along ordinary foods as the duration of the space flight exceeds several days. A partial solution, which is practical for periods of perhaps several months, makes use of dehydrated foods (Ref. 2). However one

reaches a point where even this is impractical, as is easily shown by food logistics. The exact quantity of food required by humans is, of course, not a well defined quantity, varying with factors such as sex, height, build, climate, activity, and, to a large extent, the conditioning of prior eating habits (Refs. 3 - 6). However, for calculational purposes we have selected as representative the value of 500 g of dry food per day per man, based on a 2400 cal diet, of which 10 percent is protein, 35 percent is fat, and 55 percent is carbohydrate (Ref. 7). About 2200 g of water must be added daily to this diet; 1000 g of this is for food rehydration and the rest is for drinking purposes (Ref. 8). A very substantial saving in weight can thus be realized if water is recovered from the human wastes since only a single day's supply of water need be carried along; perhaps several such rations of water would be necessary to correct for gradual leakage of gases from the spacecraft. However, if one multiplies 500 g per day per man of dry food by the number of days planned for the space flight, it is obvious that even dehydrated foods become impractical as the stored food supply equals and then exceeds the weight of the space traveler.

Mention should be made of medical suggestions for drastically reducing the metabolic requirements of space travelers. These include the use of appropriate drugs, either surgical removal or radiation degeneration of the thyroid gland followed by controlled administration of reduced amounts of thyroid extract, and hibernation induced by drastically lowering the body temperature (Ref. 7). The problems inherent in the latter technique are discussed by Hanrahan and Bushnell (Ref. 9). While drastic remedies such as the use of thyroid destruction or prolonged periods of deep freeze cannot be dismissed, they are hardly appealing to a potential space traveler. The deep freeze concept is incompatible with the need for alertness on manned space stations or extraterrestrial explorations of prolonged duration, where presumably observations and data gathering would be of importance, although it might be of use during the flight from earth to the destination.

Finally, there are nutritional concepts that reduce the magnitude of the logistics problem without requiring a complete ecological system. One such alternative is research into the minimum caloric intakes possible after prolonged conditioning. Mitchell (Ref. 5) describes the remarkable adaptability of individuals to new and seemingly drastic diets; Adams (Ref. 7) refers to a study in which no adverse effects were found to result from a diet providing only 10 to 20 g of protein per day, whereas approximately 70 g are generally considered to be a minimum requirement for average males (Ref. 10). Also of interest is the question of energy-dense synthetic foods, or chemicals that may possibly be nutritive although they are not ordinarily considered so. If such materials can be found in which the useful combustion energy per unit weight exceeds that of normal foods, considerable weight savings in space can be obtained. Several researches in this area have been reported (Refs. 11 - 13). However it must be recognized that these solutions are only of degree and not of kind. At best they would only extend the practicality of carrying along foods by durations of perhaps several months (Ref. 14).

One cannot escape the conclusion that complete ecological systems should be developed which conserve the atoms that are used in eating and then liberated by metabolism. There is a strong likelihood that extended space travel will not be possible without such a system. The problem of complete conservation of these atoms is most acute on spaceships or space stations where all raw materials must be boosted from the earth. On distant planets it is conceivable that part of the raw materials needed for food synthesis could be found locally.

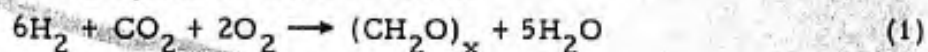
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II. BIOLOGICAL SYSTEMS

The efforts to develop biological models of ecological systems, or scaled-down replicas of the earth's system, are described briefly. It has been suggested that higher plants offer promise in nonterrestrial situations, and there has been considerable research in this area (Refs. 15 and 16). However, a recent review of this endeavor (Ref. 16) states that low production rates and high cost of light and space have conspired against these projects, which are now largely abandoned. Emphasis has shifted to microorganisms, the photosynthetic algae. Much of this experimentation has been with artificial light sources rather than solar energy because of the risk inherent in complete dependence on the constant availability of solar energy. The incident light produced by solar energy decreases markedly with distance from the sun [45 percent of intensity at Earth exists at Mars, and only 3 percent at Jupiter (Ref. 1)] and can vary with attitude of the spacecraft. Chemical changes that might occur in window material or solar trapping devices could also reduce the available solar energy; however, this could possibly be corrected by the space traveler. In any event, planners thus far have not been content to accept complete dependence on solar energy, even though the use of artificial light requires added weight.

Research with photosynthetic algae has also been disappointing (Ref. 16) because of the difficulty in designing a lightweight system in which sufficient light can be transmitted past dense upper layers of algae to reach lower layers without exposing the upper layers to over-radiation. The volume limited situations in spacecraft are evidently incompatible with this requirement, and agitation is expensive in terms of power and complexity of equipment (Ref. 17).

At present the favored biological microorganism is the nonphotosynthetic, hydrogen-fixing *Hydrogenomonas* bacterium, which grows in the dark. It produces carbohydrates by the overall reaction



Although photochemical energy is not needed by this microorganism, energy must be provided to the overall system because hydrogen, which is not a significant human metabolic product, must be prepared from water by electrolysis.

Although biological systems have been under intensive investigation as sources of nutrition in long-duration, manned space ventures, it is still not known whether a practical biological system can be developed on the scale required to feed space travelers. There is also the risk, mentioned above, inherent in depending on the invariability of a living system. Possible dangers are attack upon the essential microorganisms by other microorganisms inadvertently carried aboard the spacecraft and undesirable mutations of the microorganisms caused by innumerable reproductive cycles or by high radiation doses. Clearly, an alternate system is needed, at least as a back-up. Development of a purely chemical method of converting human wastes to foods thus would be insurance against our possible failure to produce a practical biological system. A chemical method also could serve as an alternate system to be used in the event of the unforeseen failure, in space, of a primary biological system.

III. GENERAL CONSIDERATIONS FOR CHEMICAL SYSTEMS

A. ENERGY AND MATERIAL LIMITATIONS

Because of the extreme logistical importance of payload weights, the weights of energy sources are important considerations in evaluating possible chemical methods of food synthesis. Therefore, reactions that are highly efficient in the utilization of energy are necessary, particularly if solar energy is not available. Approximately 2 kw-hr of energy per man per day would be required to convert waste products to nutrients if the reaction used all the available energy in a completely efficient process. If the overall efficiency of utilization of this energy were only 1 percent, a power source of 8.33 kw would have to be provided continuously for each man for food production alone. From weight/power relationships given in Ref. 5, we can estimate a power source weight of 800 lb per man, which would be impractical.

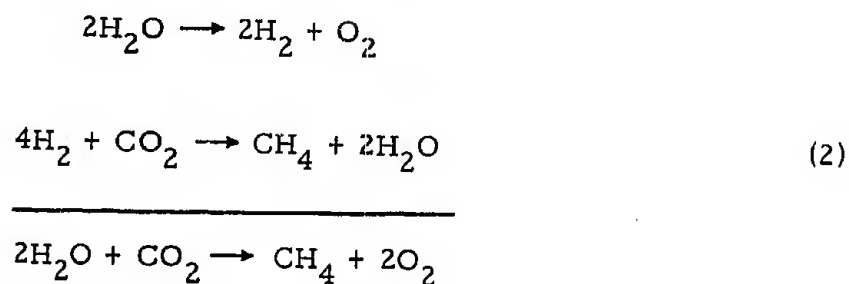
Reaction rates are also very important. Obviously, the food synthesis reaction must be at least as fast as the metabolic process if the ecological system is to be effective.

The principal metabolic wastes in terms of quantity are carbon dioxide and water. In the 500 g per day diet of nonaqueous foods mentioned in Section I, the food is oxidized by 715 g of oxygen to 840 g of carbon dioxide and 350 g of water, assuming about 95 percent digestibility and nearly complete oxidation (Ref. 7). The waste water, which is formed by the metabolic process and is thus, in a sense, extracted from the nonaqueous food, added to the water stored for drinking purposes and food rehydration, would lead to a cumulative storage problem if not converted to food.

After the oxygen required for respiration is extracted from waste carbon dioxide and water, the resultant material has the overall empirical formula $\text{CO}_{0.6}\text{H}_{1.7}$, neglecting only the solid contents of the urinary wastes and the extremely small quantity of useful chemicals available from sources such

as the feces and the flatus. About 50 percent of the urinary solids is urea, the main source of nitrogen for food synthesis. The diet under consideration is only 10 percent protein, thus the nitrogen content relative to $\text{CO}_{0.6}\text{H}_{1.7}$ would not be very large (only about 8 g out of 500 g of food). Much greater amounts of nitrogen for food synthesis could result from diets higher in protein or other sources of nitrogen.

The principal raw materials for nonterrestrial food synthesis from human wastes are $\text{CO}_{0.6}\text{H}_{1.7}$ plus urea or its derivative, ammonia. They are in a far more reduced state than the wastes originally produced because the oxygen necessary for respiration has already been removed by the catalytic thermal reduction of carbon dioxide with hydrogen, which is obtained by electrolysis of water (Ref. 16). The reaction sequence is shown in Eq. (2).



It is therefore realistic to consider that the carbon for food synthesis is mainly available not as CO_2 , but as methane, which is beneficial.

Food production from human wastes has to be an endothermic process, because the reverse reaction of food combustion in the body is exothermic. Accordingly, considerable energy must be expended to convert the wastes to useful nutrients. The heats of overall reactions leading to the simple nutrients glucose and glycine from both methane and carbon dioxide are given in Table 1 to illustrate the magnitudes of energy involved.

Table 1. Energetics of Simple Food Formation Reactions

	$\Delta H,$ Kcal/mole	$\Delta H,$ Kcal/g nutrient
$6\text{CO}_2(\text{g}) + 6\text{H}_2\text{O}(\text{g}) = \text{C}_6\text{H}_{12}\text{O}_6(\text{s}) + 6\text{O}_2(\text{g})$	610.0	3.39
$2\text{CO}_2(\text{g}) + \text{H}_2\text{O}(\text{g}) + \text{NH}_3(\text{g}) = \text{NH}_2\text{CH}_2\text{COOH}(\text{s}) + 3/2\text{H}_2\text{O}(\text{g})$	132.5	1.77
$6\text{CH}_4(\text{g}) + 6\text{H}_2\text{O}(\text{g}) = \text{C}_6\text{H}_{12}\text{O}_6(\text{s}) + 12\text{H}_2(\text{g})$	153.1	0.85
$2\text{CH}_4(\text{g}) + 2\text{H}_2\text{O}(\text{g}) + \text{NH}_3(\text{g}) = \text{NH}_2\text{CH}_2\text{COOH}(\text{s}) + 5\text{H}_2(\text{g})$	38.0	0.51

It can be seen from these data that the reduction of carbon dioxide with water is far more difficult energetically than the oxidation of methane with water. In fact the formation of formaldehyde from $\text{CO}_2 + \text{H}_2\text{O}$ is very difficult to accomplish and to proceed appreciably requires the use of very high energy sources (Ref. 18). The prior reduction of CO_2 by H_2 , the better reducing agent, is thus a reaction which not only releases needed oxygen but facilitates subsequent food syntheses. Since the oxidation by oxygen of methane to formaldehyde is exothermic, we can consider that formaldehyde can be made readily available nonterrestrially from human wastes. This is a key point because of reactions of formaldehyde which are quite relevant to food synthesis, as will be described below.

B. NUTRITIONAL REQUIREMENTS

The foods normally ingested by man have very complex chemical structures. Development of a simple process for synthesizing such complex structures from human wastes, within the limitations imposed by spacecraft logistics, is a formidable task. Admittedly, as complex a system of catalysts as those used by living organisms would not be required, because there would be fewer constraints on permissible reaction conditions such as temperature, pressure, and range and dose of energy sources. Yet when one considers the limitations of resources, both human and material, for

projected space travels it becomes evident that a substantial simplification in the nature of food structures would be very much in order.

Fortunately, some of our essential foodstuffs are required in very small quantities. The required daily amounts of vitamins and minerals are negligible in terms of weight (Refs. 10 and 19), as are those of the essential unsaturated fatty acids (Ref. 20). These materials thus could be carried in space without creating a significant logistics problem, even for space travels of many years.

Proteins are required for synthesis of body enzymes and replacement of constantly degrading body proteins, but, fortunately, only eight of the amino acids found in human protein cannot be synthesized by the body from simpler sources. In adult males the total daily requirement for these essential amino acids is only about 8.5 g (Ref. 21); a small fraction of the 70 g of protein normally considered necessary for good nutrition (about one g per kg of body weight). Although larger than the required amounts of vitamins and minerals, this amount of essential amino acids is sufficiently small that it could be taken along in space. The nonterrestrial food synthesis problem thus involves only carbohydrates, fats, and non-essential amino acids, or their equivalents in function.

The carbohydrates and fats, unlike the proteins, are used principally for energy release; proteins are used for this purpose only if they are present in excess. With the exception of the small required amounts of essential fatty acids, which are similar to vitamins in function, the body can synthesize sugars from fats or vice versa. Thus there is no fixed ratio for these two ingredients in the human diet; in fact, this ratio varies widely all over the world. Although fats are usually eaten for their culinary effects, they do have higher energy densities than carbohydrates, and, because of their slower absorption and metabolism, they tend to appease hunger pangs for longer periods. However, space logistics problems suggest the simplification of making either fats or carbohydrates, rather than both; we must therefore determine which is easier to synthesize from available ingredients.

There seems to be no physiological reason to preclude a diet in which carbohydrates completely replace fats, provided the minimal amounts of essential fatty acids and vitamins are carried along from earth. In fact, one major reason for consumption of fats is to ensure ingestion of minimal amounts of the fat-soluble vitamins that normally accompany fats in nature. A diet in which fats replace carbohydrates seems less satisfactory because of possible ketogenic effects. Many authors maintain that a diet too high in saturated fats leads to a high buildup of acetone bodies in the blood (acetone, acetoacetic acid, and hydroxybutyric acid), which results in a marked lowering of blood pH. Anderson (Ref. 22) even cites a method for computing the minimal ratio of sugar to fat in the diet necessary to prevent this ketosis. However, it has been pointed out that ketosis has only been studied in diabetic or starving subjects, and diets that were very high in saturated fats and excluded carbohydrates produced no harmful effects when fed to animals (Ref. 23). This has led to the formulation of a calorie deficient hypothesis of ketosis (Ref. 23). It is well known that Eskimos exist on a diet with a very high ratio of unsaturated fats to carbohydrates without apparent harmful effects. Sinclair points out that on a purely chemical basis such diets should actually be antiketogenic (Ref. 20). Unless research should establish the safeness of a diet in which saturated fats can completely replace carbohydrates, it would seem prudent to consider diets in which unsaturated fats are the replacement for much of the carbohydrates. Diets that included unsaturated fats have also been considered in theories concerning atherosclerosis, although the facts are still obscure.

We might also consider a diet containing only protein or amino acids, supplemented by the minimal quantities of essentials described above. It is well known that the body can use excess protein as a source of energy. After deamination of amino acids the residues can be metabolized by either the sugar mechanism or the fat mechanism, depending on the chain structure of the amino acid. Studies in which the human diet consisted almost totally of proteins have been carried out for periods up to one year without observable harmful effects (Ref. 21). Data are lacking for longer periods but it has been

pointed out that the only conceivable problem in such extended diets might be possible damage to overworked kidneys (Ref. 21). This damage is conjectural, and it should be noted that humans have exhibited extraordinary abilities in adapting to drastic dietary changes (Ref. 5).

Synthesis of protein diets could be simplified considerably if the amino acids did not have to be polymerized to protein. There do not appear to be harmful physiological effects associated with amino acid diets, except that amino acids seem somewhat less efficient than protein in providing energy release (Ref. 24). This may be caused by the use of d-form amino acids in addition to the l-forms which occur in proteins of higher species (Ref. 25). It is known that the d-forms of some amino acids are utilized much less effectively than the l-forms, due presumably to the lower inversion rates of some d-amino acids and their slower absorption rates in the intestine (Ref. 26). Additionally, some d-amino acids markedly inhibit growth even when the l-form is present in sufficient amounts (Ref. 27).

Because of the inhibitory effect of some d-amino acids, optical resolution of stereoisomers must be accomplished for some, if not all, of the synthesized amino acids. In a batch process this would probably entail complete resolution of the entire batch of amino acids. Thus the acids other than glycine, which is optically inactive, would not only have to undergo further reaction to isolate the l-isomers, but the d-forms would represent wasted energy input and would have to be reprocessed for conversion to further racemic mixtures of amino acids. For this reason, and because of the difficulty of devising a simple procedure for synthesizing all of the nonessential amino acids, a further simplification, involving substitution of a simple nitrogen source for the nonessential amino acids, should be investigated.

It has been shown that the ammonium salts of citric and acetic acids, when fed as supplements to diets deficient only in nonessential amino acids, are excellent sources of nitrogen for *in vivo* synthesis of the nonessential amino acids by rats (Refs. 28 and 29). Rats fed on such diets grew much more rapidly than those that were not fed these nitrogen sources, and this also led to a marked improvement in the metabolic nitrogen balance.

Further, the rats fed ammonium citrate grew as rapidly as rats fed the relevant nonessential amino acids (Ref. 29). L-glutamic acid was equally effective, while glycine was less effective and urea still less effective (Ref. 28). Yet both glycine and urea did serve as effective nitrogen sources, in that growth was faster than with diets lacking in nitrogen sources other than minimal amounts of the essential amino acids. Rose and coworkers (Ref. 28) concluded that the effectiveness of a nitrogen source depends upon the facility with which the cells can either convert the source to ammonia or undergo a transamination reaction.

Several of the amino acids are known to undergo antagonistic reactions when present in excess (Ref. 26), a problem which is eliminated by using a single nitrogen source. Use of a single nitrogen source also ensures that the body will synthesize the nonessential amino acids simultaneously in amounts necessary for effective conversion to other amino acids; this may not be true in unbalanced diets of nonessential amino acids.

One is encouraged to consider simple nitrogen sources, probably ammonium salts or simple amino acids, as synthetic foods that might be prepared rather simply, in space, from human wastes. We should also consider the carbohydrates or high fatty acids that are most readily prepared from such wastes by simple processes, rather than those carbohydrates and fats to which we normally are accustomed. In this connection it is interesting to note dietary experiments with dimethylheptanoic acid and butanediol, which have been shown to be nutritive within limits (Refs. 11-13).

It should be emphasized that the energy contents per unit weights of foods are not primary factors in our present context as they are in shorter duration space ventures in which all food is prepared terrestrially. Since the metabolic products are to be continuously converted to food, the initial ration can be considerably larger than would otherwise be practical. Thus substitution of carbohydrates, or the relatively low energy glycine, for fats represents no problem despite the much higher energy density available in the fats due to their relatively higher reduced states. The relevant factors are the kinetics, energy requirements, and efficiencies of the proposed chemical

reactions. Provided that the materials are not in themselves toxic or irritant, and do not lead to undesirably high concentrations of certain metabolic byproducts such as acetone bodies, any materials that can be readily prepared from human wastes, in a form that is efficiently metabolized with the resultant release of a high fraction of the theoretical energy, should be considered. One might consider the effect on the human digestive system of synthetic foods of low molecular weight and high solubility, since the digestive system would be essentially bypassed. It might be argued that atrophy of the digestive organs might occur if the diet were continued for a number of years, but this is purely conjectural at this point. Questions might be raised as to the psychological acceptability of synthetic foods, but we can assume that space travelers are highly motivated in this regard.

IV. CHEMICAL SYNTHESIS

A. PRIMARY REACTANTS

The primary reactants available from human wastes are water, ammonia, and methane or formaldehyde. We must develop an operationally simple chemical process for producing foodstuffs from these compounds. The possibility of developing such a process can be evaluated from literature on chemical evolution from hypothesized primordial constituents and on the mechanism of plant photosynthesis.

1. PHOTOCHEMICAL REACTIONS IN THE VACUUM ULTRAVIOLET RANGE

We might start this discussion with photochemical reactions involving these substances. In the absence of suitable catalysts, photolyses involving methane, water, and ammonia must be carried out in the vacuum ultraviolet since it is only in this region that appreciable absorption occurs (Ref. 30). Of these gases ammonia has an absorption maximum at the highest wavelength, 1870 Å. This is an important factor since the light intensities available for photolytic reactions tend to fall with decreasing wavelength in the vacuum ultraviolet region of the spectrum, for reasons concerned with the ease of constructing complex reactor shapes out of materials that are transparent at successively lower wavelengths in this spectral region. Accordingly it would be desirable to initiate reactions by radiation near the ammonia maximum, using the radicals generated by the ammonia photolysis to attack methane. This has been studied by Groth and Weysenhoff (Ref 31) who photolyzed mixtures of CH_4 , H_2O , and NH_3 at 1849 Å. However no detectable reaction on the part of the methane was noted. Since ethane has a lower dissociation energy for the C-H bond than does methane by some 5 kcal/mole (Ref. 32), greater reactions might be expected by substitution of ethane for methane in these mixtures. This was confirmed by Groth and Weysenhoff, who found that small amounts of glycine and alanine were formed, along with traces of α -amino butyric acid. Quantitative data for all products and reactants were not provided and no attempt was made to ascertain products other

than the amino acids and unsubstituted carboxylic acids, which were formed in amounts one order of magnitude higher than the amino acids. However, from the qualitative description it is obvious that the amino acid fraction was a very small fraction of the total product mixture. As might be expected, methane reacted at 1295 Å but the yield of amino acids fell approximately proportionately with the lower light intensity at 1295 Å. Not only were yields very low, even at 1849 Å, but approximately one day was required to obtain significant amounts of amino acids.

We can conclude that this type of reaction offers very little promise for converting methane, water, and ammonia to amino acids at either the rate or the efficiency required, even if methane were to be initially converted to higher saturated hydrocarbons, as seems necessary.

2. PHOTOCHEMICAL REACTIONS IN THE NONVACUUM ULTRAVIOLET RANGE

One might next consider methods of shifting the absorbing region to longer wavelengths. Terenin (Ref. 33) shows that the wavelengths at which water and ammonia absorb appreciably can be shifted well into the nonvacuum ultraviolet by adsorbing these gases upon appropriate catalysts. Evidently it is the new bond, created by the adsorption process, that absorbs such wavelengths. Ellenbogen (Ref. 34) reports a variation of this procedure, irradiating aqueous ammonium-containing solutions through which natural gas was bubbled slowly in the presence of catalysts. A number of amino acids were formed in these experiments, but the overall production of amino acids was small. On the basis of the only yield reported quantitatively, only 0.2 percent of the hydrocarbon had been converted to amino acids after six days of continuous radiation. Thus, this procedure for forming amino acids seems to be no more practical than that of Groth and Weysenhoff. Yet it is interesting that, whereas Groth and Weysenhoff formed only the simplest amino acids (essentially only amino acetic and propionic acids), Ellenbogen obtained far more complex amino acids, such as phenylalanine, valine, leucine, and methionine after only 8 hr of irradiation. This could be attributed to the longer contact times in Ellenbogen's study, since he slowly passed

hydrocarbon through the aqueous solution while Groth and Weysenhoff had a circulating system with presumably short contact times. An intriguing possibility is that the higher amino acids formed in Ellenbogen's studies resulted from reactions of higher hydrocarbons, probably present as small components of the natural gas. Since the solubility of hydrocarbons in water increases with chain length, the mole fractions of higher hydrocarbons would tend to be disproportionately high in the aqueous solutions. Also, the bond strengths of secondary C-H bonds in hydrocarbons tend to weaken with increasing chain length, so that higher hydrocarbons would be more susceptible to attack by primary photolytic radicals, a factor leading mainly to secondary hydrocarbons in photolytic studies (Ref. 30).

Photolyses of aqueous formaldehyde solutions with ammonium salts for long irradiation times, leading to very small amounts of various amino acids, have also been reported (Ref. 35). Variations of this process are irradiation of paraformaldehyde solutions with nitrate ion (Ref. 36) and irradiation of formaldehyde solutions with nitrate or nitrites (Ref. 37), both of which also lead to the formation of extremely small amounts of various amino acids. These were clearly static experiments with long contact times. Baly, et al. (Ref. 37) demonstrated that formaldehyde reacts with the nitrate or nitrite to form formhydroxamic acid, $\text{HC}(\text{NOH})\text{OH}$, which can react further with an additional formaldehyde molecule to form glycine, or with more formaldehyde molecules to form higher amino acids. However, other paths leading to the formation of a number of organic nitrogenous bases and their alkaloid condensation products, were also possible. Free ammonia was detected in these experiments, and it has been shown (Ref. 38) that nitrates can be readily reduced to ammonia by ultraviolet irradiation in the presence of oxidizable organic matter, leading to amino acid formation.

In summary, it has been shown that either glycine or more complex amino acids can be prepared in very low yields by simple photochemical reactions of the primary reactants available for nonterrestrial food synthesis.

Despite the attractive simplicity of such processes, it is apparent from the data that such techniques have little chance of producing amino acids as major products. This evidently is because a multitude of other products is formed from such simple reactants, given C, H, O, and N.

3. PHOTOCHEMICAL FORMATION OF SUGARS

Since formaldehyde is related so closely to sugars, photochemical polymerization of formaldehyde alone should result in higher yields of carbohydrates than would reactions of methane, water, and ammonia. Baly (Ref. 39) discusses the problems in the photochemical formation of sugars from aqueous formaldehyde solutions. He describes a yield of about 20 percent sugars, calculated as glucose, after 14 days of continuous irradiation, and implies that this was the practical limit because of a number of disadvantageous factors, mainly the photochemical destruction of the sugars themselves. In these reactions additives that are generally classified as catalysts are used. Baly disputes that catalysis is involved, claiming that the function of the photochemically absorbing additives is to protect the formaldehyde from decomposition by absorption of shorter wavelengths. However, aqueous formaldehyde solutions absorb appreciably only at wavelengths below about 2300 Å (Ref. 40), and the ultraviolet absorption band of formaldehyde at longer wavelengths disappears when formaldehyde is in contact with polar substances (Ref. 41). It seems that catalysis or photosensitization is required for this reaction to proceed at wavelengths much above 2300 Å.

It is significant that only hexoses were detected among the sugars formed. It appears that the long irradiation times might be reduced by further research into this procedure, but the photochemical method does not appear to be as attractive as the thermal polymerization of formaldehyde.

4. THERMAL POLYMERIZATIONS

Thermal reactions are particularly attractive as nonterrestrial sources of food because they may use energy more efficiently than either complex photochemical reactions in the high energy regions of the ultraviolet or electrical discharge reactions. We next consider, therefore, the possibility of preparing

foods from the primary reactants by purely thermal means. Harada and Fox (Ref. 42) report that amino acids can be formed by the passage of gaseous mixtures of methane, water, and ammonia over hot solids at temperatures of 950° to 1050°C. In these experiments almost all the amino acids common to protein were formed in varying amounts, although the simplest ones, glycine and alanine, were the major products. It is significant that the mole fraction of glycine was much higher at 950° than at 1050°, and that there was substantial conversion of this simple amino acid to higher amino acids at the higher temperature; the amino acids that increased the most at the higher temperature were the dicarboxylic amino acids--aspartic and glutamic acids--and serine. Although not stated in Ref. 42, the contact times for these reactions were very short. In a personal communication to the author, Harada and Fox indicated that these contact times were probably much less than 1 sec, although they were not specifically measured. They further indicated that the total yield of amino acids after long reaction times (many passes of reactants) were of the order of a few milligrams. This type of simple synthesis, called a "pan" synthesis, has been advocated as a possible solution to the space nutrition problem (Ref. 43). However, the yields under the conditions described are very low, not really an improvement over those given by photochemical reactions. While improvement could undoubtedly occur with further research, it appears that the chances of developing such a process of synthesis from methane, water, and ammonia that satisfies the requirements of yield and reaction rate are not very good.

The base-catalyzed thermal polymerization of formaldehyde is a more promising approach, although this method would yield carbohydrates, rather than amino acids or mixtures of amino acids with other food ingredients. This reaction is relatively fast, especially when small amounts of sugars are present as additional catalysts (Ref. 44). The polymerization of formaldehyde solutions in the presence of calcium hydroxide and traces of fructose is completed in less than one-half hour at 60°C, at least with regard to disappearance of formaldehyde. At this stage lower carbohydrates are the main products, but further condensations to higher sugars occur if the reaction time is extended.

Mayer and Jäscke (Ref. 45) have studied the mechanism of this base-catalyzed thermal polymerization of formaldehyde. They conclude that initially formaldehyde dimerizes to glycolaldehyde. Further reaction depends upon the strength of the basic catalyst. With weak bases, glycolaldehyde reacts with an additional formaldehyde molecule to form dihydroxyacetone, which reacts with glycolaldehyde to form ribulose, a ketopentose. Eventually the ribulose is converted into the aldopentoses ribose, xylose, and arabinose. In strong alkali, glycolaldehyde dimerizes to tetroses, which are unstable under these conditions and react further to form pentoses and trioses. Hexoses are also formed in strongly alkaline reactions, but not in weakly alkaline solutions. Pfeil and Ruckert (Ref. 46) report that not only glycolaldehyde and dihydroxyacetone, but also glyceraldehyde, are initial products of the strongly alkaline reaction. The formation of a complex mixture of tetroses, pentoses, and hexoses is described by interactions of these initially formed compounds among themselves and with formaldehyde.

The yields, reaction rates, and inherent simplicity of these base-catalyzed thermal polymerizations of formaldehyde make this type of reaction particularly attractive for nonterrestrial chemical food synthesis. In question are the metabolic utility of the types of products formed by this type of reaction and the possible toxicity of some of these materials, factors that can be resolved by research.

5. ELECTRICAL DISCHARGE TECHNIQUES

Although electrical discharge reactions are notoriously inefficient in the conversion of input energy into chemical energy, we shall briefly consider this technique because of its operational simplicity. It has long been known (Ref. 47) that mixtures of carbon monoxide and hydrogen, or carbon dioxide and hydrogen, can be partially converted to polymers of formaldehyde by the silent electrical discharge technique, using long contact times. Mixtures of carbon monoxide, water, and hydrogen have also been converted by this technique to product mixtures containing large amounts of formaldehyde and glycolaldehyde (Ref. 48), from which most sugars can be prepared by base-catalyzed reactions. However, the product mixtures from such techniques

appear to be more complex than those produced by the thermal reactions described above, and it does not seem likely that the electrical discharge technique offers any advantage over the more efficient thermal procedure. Further, it has been reported that, in reactions of lower hydrocarbons with air in electrical discharges, the yields of formaldehyde are lower than for comparable catalytic thermal processes (Ref. 49).

There have been a number of studies in which very small yields of amino acids were prepared from simple reactants by electrical discharge techniques (Refs. 35 and 50 - 53). Generally, the amino acids formed in these experiments were of short chain lengths if methane, CO_2 , or CO served as sources of carbon. Higher amino acids have been reported by this procedure if higher hydrocarbons were substituted for the C_1 compounds (Ref. 54). A mechanism has been advanced for glycine formation from methane, water, and ammonia that requires the initial formation of HCN and formaldehyde (Ref. 53). Reaction between HCN and formaldehyde yields an hydroxy nitrile which can be aminated and then hydrolyzed to glycine by the well known Strecker synthesis. The extremely low yields of amino acids by the electrical discharge technique suggests that this procedure is no more practical for the synthesis of amino acids in nonterrestrial situations than are the comparable photochemical or thermal reactions.

B. COMPLEX REACTIONS

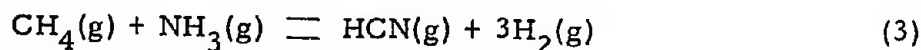
1. AMINO ACIDS

It was shown in Section IV-A that more than the mere application of various forms of energy to mixtures of primary reactants is required to produce amino acids in acceptable yields and rates for space nutrition. In contrast to such nondirected reactions, a significant amount of direction must be applied. Cultrera and Ferrari (Refs. 55 and 56) found that very small yields of amino acids were formed by ultraviolet irradiations of aqueous solutions that were similar to the irradiation techniques described above, except that higher organic molecules were substituted for the C_1 compounds. These included glycerol, alcohols, mono- and dicarboxylic acids, lactic and citric

acids, and sugars. Very small yields of amino acids were also found when aqueous solutions containing ammonia and HCN were heated for long contact times (Refs. 57 and 58). What is evidently needed is restriction of reactants to those that form only amino acids, or at least mainly amino acids.

A number of reaction types are known for general amino acid formation (Ref. 59). These require either quite complex reactants or a number of reaction steps in the amino acid formation sequence. In some cases, higher amino acids can be produced in large quantity, from glycine itself and from other reactants that are primary products available from human wastes, by operationally simple processes (Refs. 60 and 61). However, this requires prior formation of glycine. Since glycine is the only optically inactive α -amino acid, and because of the nutritional arguments made in a previous section, we might consider the problem of synthesizing only this simplest of amino acids.

Oro, et al., report an operationally simple procedure for the synthesis of glycine from formaldehyde and hydroxylamine by a thermal reaction (Ref. 62). A 0.20 molar yield of glycine was obtained after 90 hours of reaction time. The order of magnitude of the rate discourages further consideration of such a process. However, the mechanism of this process is probably the initial reaction of formaldehyde and hydroxylamine to form CH_2NOH , which decomposes to HCN and water, followed by a Strecker-type synthesis of glycine. Clearly the process would be faster if HCN could be generated at a faster rate, say directly from methane and ammonia. The standard free energy change for the reaction of Eq. (3) goes from positive to negative at about 1060°K , indicating that this transformation could



proceed readily at high temperatures. The electrical discharge method, less efficient from an energy conservation viewpoint, is known to produce HCN in good yields from simple reactants such as CO, CH_4 , N_2 , H_2 , and NH_3 (Refs. 63 - 66). Another possibility is the nearly quantitative formation of glycine by

either acid or base hydrolysis of a low molecular weight HCN polymer. This reaction was reported nearly a century ago by Wipperman (Ref. 67), who prepared what was called $(\text{HCN})_3$ by base catalysis of an aqueous HCN solution. It has been observed that a tetramer of HCN, diaminomaleonitrile, is the probable precursor of glycine (Ref. 58).

2. CARBOHYDRATES

It is difficult to conceive of an operationally simple synthesis of long chain unsaturated fatty acids from human wastes. This increases the interest in carbohydrate synthesis by the operationally simple polymerization of formaldehyde in basic solutions. There is considerable disagreement between the results of the two most complete studies of this reaction (Refs. 45 and 46). However, it does appear that the ketopentose ribulose is the principal carbohydrate product formed under conditions of very weak alkalinity and moderate reaction times (Ref. 45). We might, then, examine the probable metabolic effects of ingesting ribulose. Research with the aldopentoses ribose, xylose, lyxose, and arabinose has demonstrated that humans effectively metabolize D-ribose but not the others (Refs. 68 - 70), or at least that the nonuseful aldopentoses are metabolized far more slowly. Since D-ribose is effectively metabolized, it appears that the corresponding ketopentose, ribulose, should be even more effectively utilized.

Under different experimental conditions, tetroses and hexoses can also result from the base-catalyzed polymerization of formaldehyde. There are no comparable human metabolic data available on tetroses, but research with rats indicates that D-erythrose and L-erythrulose are rapidly metabolized (Ref. 71). In this study a considerable amount of material was not accounted for. However, more recent studies¹ indicate that the metabolism of D-erythrose is not only rapid but quantitative, the erythrose being converted

¹A. L. Fluharty, University of Southern California, Personal Communication.

to fructose by rat liver homogenates. Nutritional experiments are generally lacking for the nonnatural hexoses. However, it is encouraging that the reported hexoses products are fructose, glucose, and galactose, all known to be rapidly and effectively metabolized.

In view of the above metabolism data on pentoses and tetroses one might be encouraged to further investigate the base-catalyzed polymerization under various experimental conditions. It has been suggested that not only the base strength but the nature of the base cation markedly affect the numbers and ratios of sugars which are formed by this reaction. Because of the formation of racemic mixtures among the tetroses, pentoses, and hexoses, it is also worth considering the possibility of merely synthesizing dihydroxyacetone, a known intermediate and byproduct of the base catalyzed polymerization of formaldehyde since it is optically inactive. Because dihydroxyacetone is important in human intermediary metabolism, it may very well be nutritionally effective.

Finally, it must be pointed out that the available data on metabolism of chemically synthesized sugars were generally obtained on quantities of sugars considerably less than those under consideration here. It is possible that sugars that can be utilized in small amounts may not be useful in larger quantities. For example, 1, 3 butanediol is a very effective food for young rats in amounts as great as 40 percent of the total diet, but it is toxic in greater quantities (Ref. 72). There are no data for tetroses or pentoses in quantities sufficient for our purposes, and such data must be collected if synthesis of these sugars is ever to be used in space.

C. CONCLUSIONS

The problems inherent in the synthesis of the nonessential amino acids from human waste products by operationally simple procedures have been discussed. It was concluded that several simple types of reaction, which lead to very simple amino acids, are possible. These might be used by humans as nitrogen sources for the in vivo synthesis of all the nonessential amino acids. However, because these amino acids may have toxic effects, and because of the problem of the wasteful production of racemic mixtures, we should consider producing nitrogen sources by hydrolysis of urea to simple ammonium salts.

In the author's view, the most promising synthetic foodstuff that can be synthesized simply from human wastes under nonterrestrial conditions is a carbohydrate prepared by base-catalyzed polymerization of formaldehyde solutions at moderately elevated temperatures. It is believed that, under appropriate reaction conditions, nutritionally useful sugars can be prepared by this procedure in rates and yields appropriate to the problem. However, the formation of racemic mixtures, with the present uncertain utility of some of enantiomorphs invites consideration of altering the polymerization to preparing largely dihydroxyacetone, an optically inactive carbohydrate that may be a very good food. The special problems created by space nutrition invite nutritional research into the utility of these synthetic foodstuffs, particularly unnatural hexoses and the unnatural enantiomorphs of the natural sugars, in quantities adequate for good human nutrition.

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13. ABSTRACT The problem of food logistics in long duration manned space flights is explored. It is concluded that development of a purely chemical method for converting human wastes to useful nutrients is essential, either as a primary system or as a backup for an otherwise primary biological system. The principal requirements for such a chemical system are operational simplicity, efficiency of energy utilization, high yield, and reasonably fast rates of reaction. Various possibilities are considered, based on a thorough analysis of the existing pertinent chemical literature. This is correlated with known metabolic data. Special emphasis is given to the synthesis of structurally simple nutrients rather than the immensely complicated chemical ensembles that we normally know as food. It is recommended that special attention be given to the synthesis of carbohydrates by the base catalyzed polymerization of formaldehyde, since this process fits all the requirements and there is available experimental evidence that the endproducts could be made to be nutritionally acceptable by choice of appropriate reaction conditions.		

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