The energy flow in photosynergistically formed, nonlinear systems, Si-Mo Jeewanu, ~~are~~ is more efficient under the conditions they are auto~~synth~~organised.

By

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Key words: Synergetics, ~~Autoorganisation~~, oxygenic anoxygenic, Si-Mo Jeewanu, Sunlight, Memory

Abstract: Under suitable conditions photosynergetic, light assisted autoorganisation helps to form Si-Mo Jeewanu. These particles split water in sunlight producing hydrogen and oxygen, fix molecular nitrogen and under certain conditions indicate loss of nitrogen. It has been observed that if these particles are produced under oxygenic condition, these particles show more energy flow, as indicated by splitting of water in sunlight and fixation of nitrogen, under oxygenic conditions than under anoxygenic conditions. But when these particles are ~~autosynthesised~~ autoorganised under anoxygenic conditions they show more energy flow under anoxygenic conditions than under oxygenic conditions indicating some ~~short~~ sort of memory in these abiogenic particles to recognise the condition of their synergesis ~~synergesis~~ ~~auto organisation~~. ~~It~~ and it becomes still more important because these particles ~~most~~ duplicate by budding.

Introduction:

It has been observed that photosynergetic, light assisted self-organisation, produces such nonlinear systems which (i) show growth and development, (ii) need energy input from outside and (iii) have such properties which are not found in any of its constituents. Such systems have been synthesised[1] and named as Jeewanu, a Sanskrit word for 'Particles of Life' and the work has been repeatedly confirmed[2,3]. Study of such systems or particles is of great interest in the investigation of origin of life and formation of earliest system which had properties of living.

One of such systems which has been extensively studied is molybdenum Jeewanu or self-sustaining coacervate with about 39 % of molybdenum[4]. These particles have ferridoxin-like[5] and nitrogenase-like[6] materials in them, multiply by budding and split water in sunlight and fix molecular nitrogen[7,8,9,10]. This system has been modified by incorporating soluble sodium orthosilicate and sodium chloride[11,12].

So far these particles were produced under oxygenic condition only. These particles produce hydrogen and oxygen on splitting water in sunlight and in Warburg's apparatus these gases cause increase in the pressure in the Warburg's flask (w.f.). If this Warburg's study is done under anoxygenic condition less pressure is produced in w.f. because alkaline pyrogalol kept in the side lobe absorbs the oxygen which is simultaneously produced on splitting of water by these particles in sunlight. However if one wants to have a good amount of hydrogen by splitting of water in sunlight one must have a high ~~yield~~ increase in the pressure inside the ~~W~~ W.f. under anoxygenic condition also even after the simultaneously liberated oxygen is absorbed by ~~the~~ alkaline pyrogallol of the side lobe of the W.f.

A ~~p~~ typical biological system acts better in the environment in which it is produced. We prepared Si-Mo Jeewanu both under oxygenic and anoxygenic conditions and investigated their gas transformations both under oxygenic and anoxygenic conditions in phosphate buffer of pH 5.7, 6.5 and 7.5 in Warburg's apparatus and observed that at all pH the particles produced under oxygenic conditions ~~show the~~ were more efficient and showed more energy flow i.e. splitting of water in sunlight and fixation of molecular nitrogen under oxygenic conditions and the particles produced under anoxygenic condition were more efficient and showed greater energy flow under anoxygenic conditions. This observation shows some parallelism with the living systems which act better in the condition in which they are produced and also this suggests a new method of preparing Si-Mo Jeewanu which show significant increase in the pressure in the W.f. under anoxygenic condition caused by the ~~presence~~ presence of considerable amount of hydrogen.

Experimental: w/v

4% (w/v) ammonium molybdate, 3% (w/v) ~~di~~

diammonium hydrogen phosphate (DAHP), 5% (w/v) soluble sodium orthosilicate and 3% (w/v) sodium chloride solutions were ~~made~~ prepared by dissolving the requisite amount of the salt in 100 ml. of distilled water. ~~The~~ A mineral solution was prepared by dissolving 20 mg. of each of NaCl, K_2SO_4, $MgSO_4$, KH_2PO_4, $MnSO_4$ and $Ca(CH_3COO)_2$ one by one in about 80 ml. of distilled water. 50 mg of ferrous sulphate were dissolved in about 10 ~~ml~~ ml of distilled water. 0·1 ml. of $6N/H_2SO_4$ was added to it to stop the hydrolysis and this was mixed with the above mineral solution. The total volume of this mineral solution was made to 100 ml. 36% formaldehyde was used in the experiment.

70 ml. of the ammonium molybdate solution, 140 ml. of the DAHP solution, 70 ml. of the mineral solution, 70 ml. of the sodium chloride solution and 70 ml. of the soluble sodium ortho silicate solution were taken in 1l. Conical flask and 70 ml. of distilled water was added to it. Total volume of the solution was ~~made up to~~ 500 ml. 250 ml of this solution were taken in two 500 ml. corning conical flasks. The flasks were cotton plugged and sterilised in an autoclave at 15 lb. pressure for 30 min. On ~~cooli~~ Cooling 40 ml. of 36% formaldehyde

were added in each flask aseptically.

In one flask one sterilised test tube reaching up to the neck of the conical flask and two-third filled with alkaline pyrogallol solution was lowered. This flask was tightly closed with a sterilised rubber cork. Alkaline pyrogallol solution was prepared by mixing 10 ml. of 20% pyrogallol solution with 10 ml. of 30% sodium hydroxide solution.

Both the flasks, one having only cotton plug in its mouth and allowing the passage of air through it and thus the over head space of its mixture had air with nitrogen and oxygen and the other flask with rubber stopper in its mouth, which had all its oxygen absorbed in the alkaline pyrogallol solution contained in the test-tube kept in it and having only anoxygenic atmosphere in the over head space of its mixture, were exposed to sunlight for 24 hr. giving 8 hr. exposure each day for 3 days.

The mixtures contained in the flask were gently shaken once a day by giving whirling motion to the flasks taking care that the mixture does not touch the cotton plug or the pyrogallol solution kept in the test-tube lowered in it. This dispersed the sediment of Si-Mo. Jeewanu, getting formed at the bottom of the mixture during exposure, in the whole mixture.

After the completion of the exposure, the mixtures were opened. The alkaline pyrogallol test-tube was removed from the rubber stoppered flask. The mixtures were shaken and the Jeewanu formed in these mixtures were separated by filtration through dried and weighed filter papers (Wattman No-1) and the particles dried in a desiccator and weighed.

Weight of the particles, ~~to~~ Si-Mo Jeewanu prepared under oxygenic condition was 1·0900 gm.

Weight of the Si-Mo Jeewanu prepared under anoxygenic condition was 1·7614 gm.

Thus under anoxygenic condition about 61·4% more of Si-Mo Jeewanu were prepared as compared to the Jeewanu prepared under oxygenic conditions.

The splitting of water in sunlight, fixation of nitrogen and under certain condition loss of fixed nitrogen by ~~the~~ both the types of Si-Mo Jeewanu, one prepared under oxygenic condition (PUOC) and the other prepared under anoxygenic condition (PUAC) were studied using Warburg's apparatus.

When water is split into hydrogen and oxygen by exposing an aqueous mixture of the Jeewanu to sunlight it causes increase in the pressure in the ~~two~~ Warburg's flask (W.f.). These particles have nitrogenase-like material[6] so as soon as hydrogen is produced it starts combining with the nitrogen of the overhead space of the W.f. and the pressure in W.f. decreases. It is ~~to~~ more clearly observed ~~when~~ after the exposure to sunlight is cut off and the increase in the pressure caused by the formation of hydrogen and ~~nitrogen~~ oxygen ~~by~~ by splitting of water stops and the remaining hydrogen of the over head space continues combining with the nitrogen showing decrease in the pressure in w.f. Under certain conditions when the fixation of nitrogen is very fast, the liberated hydrogen immediately combines with the nitrogen in the overhead space of the w.f. and decreases the pressure in the ~~Warburg's~~ w.f. and decrease in the pressure in w.f. is observed as soon as the exposure to sunlight begins. Under certain conditions ~~when~~ although the fixation of nitrogen is very fast; the fixed nitrogen is lost and N_2 is produced causing increase in the pressure in the w.f. The fixation of nitrogen causing decrease in the pressure of the w.f. is due to the formation of diimide, NH=NH and loss of nitrogen is due to decomposition of diimide and ~~of~~ its ~~dispro~~ disproportionation.

It was observed that when the w.f. of the Warburg's apparatus is kept dipped in the water bath of the Warburg's apparatus the sunlight has to pass through the water of the water bath ~~of the Warburg's apparatus~~ to reach the w.f. containing aqueous mixture of the Jeevanu and this sunlight becomes too weak for action. So the water bath of the Warburg's apparatus was removed and a control ~~Warburg's flask~~ w.f. containing same volume of water was kept near the exposed w.f. and the reading of pressure in this w.f. was always recorded together with the readings of pressure in the experimental w.f. The pressure in the control w.f. was due to the temperature difference during the exposure of the w.f. and also when these stood in shade. This reading of pressure in the control w.f. was subtracted from the reading in the controlled flask to get the net change in the pressure of the experimental ~~flask~~ w.f. due to the gas transformations taking place in them. It was assumed that in the atmosphere the temperature between a small space of 30 cm × 20 cm × 20 cm, in which all the ~~flask~~ w.f. and the control flask ~~lied~~ were kept, was the same.

It was further observed that when the liquid in the manometers of the Warburg's apparatus was water the pressure difference during the exposure to sunlight and when the apparatus stood in shade was so much that levels of the liquid went beyond the range of the manometers. So the monometers were filled with mercury and then the pressure difference could be read ~~reading limits of the monometer~~ within the limits of the monometer.

20 mg of the particles were taken in the W.f. with 4 ml. of distilled water and 1 ml. of the phosphate buffer of pH 5.7 in first, pH 6.5 in the second and pH 7.5 in the third W.f. The total volume of each of the conical ~~flask~~ W.f. was 14.5 ml., the bore of the manometer was 1 mm. in diameter and bottom area of each W.f. was 9.5 cm^2. One similar W.f. ~~flask~~ was kept as control which had only 5 ml. of distilled water. For working under oxygenic condition 0.3 ml. of distilled water was kept in the side lobe of each the W.f. For working under anoxygenic condition 0.3 ml. of freshly prepared alkaline pyrogallol was kept in ~~each of~~ the side lobe of ~~a~~ the W.f. which was prepared by mixing 0.15 ml. of 20% pyragallol solution and 0.15 ml. of 30% sodium hydroxide solutions.

The lobes of the W.f. were covered with ~~2.5~~ same size (2.5 cm^2) of a black cloth to avoid any decomposition of alkaline pyrogallol during the exposure to sunlight. To keep the results of working under oxygenic condition and anoxygenic condition comparable, the ~~black cloth~~ ~~was~~ side lobe of the ~~to~~ W.f. was covered with this black cloth even while working under oxygenic condition when the side ~~tube~~ had only distilled water.

Discussion:

When ~~the~~ Si-Mo Jeewanu ~~which~~ were prepared under oxygenic condition, these particles were most active under oxygenic condition at pH 5.7 and the pressure increase in the W.f. goes upto about 2.8 cm of mercury but ~~but the under particles~~ under ~~oxy~~ anoxygenic condition, the fixation of nitrogen is so much ~~activated~~ at this pH that there is decrease in pressure in the W.f. even during the exposure of the Si-Mo Jeewanu agnes mixture to sunlight. ~~There is~~ However this fixed nitrogen is soon lost as is indicated by the increase in pressure in the W.f. on standing in shade after all the ~~liberated~~ hydrogen liberated by splitting of water is consumed in nitrogen fixation and decrease in pressure due to this stops.

At pH 6.5 and 7.5 the trend of water splitting in sunlight and fixation of nitrogen remains same both under oxygenic and anoxygenic conditions. ~~It is ab~~ There is about 2.5 cm. increase in pressure in sunlight and about the same decrease in pressure in shade due to fixation of nitrogen. Under oxygenic condition this increase in the pressure during exposure is less than 0.5 cm of mercury during exposure and about 1 cm. decrease in ~~p~~ the pressure in the W.f. in shade. The decrease in value of the pressure in W.f. during exposure under anoxygenic condition is due to ~~the fact~~ the oxygen ~~being~~ simultaneously produced

~~by~~ during water splitting being ~~is~~ absorbed in the alkaline pyrogallol of the side lobe.

When Si-Mo Jeewanu are produced under anoxygenic condition and their gas transformation is examined under oxygenic condition there is increase of only about 1·8 cm. of mercury pressure in the c.f. at pH 5·7 but under anoxygenic condition there is an increase of about 4·4 cm. of mercury in the c.f. during exposure ~~and~~ at this pH. This increase in pressure under ~~oxy~~ anoxygenic condition where alkaline pyrogallol is kept in the side lobe to absorb all the oxygen which is ~~produ~~ simultaneously produced during the splitting of water by sunlight together with hydrogen is of great interest as ~~all the oxygen~~ it is free of oxygen and ~~most of it~~ is ~~the~~ due to the presence of hydrogen in the c.f. The results obtained at the other two pH values studied are similar.

The experiments described here show ~~synergetic~~ synergistic particles produced by the exposure of sterilised aqueous mixture of ammonium molybdate, DAHP, biological minerals, formaldehyde, sodium chloride and soluble sodium orthosilicate to sunlight under oxygenic condition are better in splitting water in sunlight ~~and~~ under oxygenic condition and the same particles prepared by ~~exposure~~ exposing the same mixture under anoxygenic condition are better in water splitting under anoxygenic condition. Similar results were obtained in the case of Ti, Zn, Co, Cd and La doped Si-Mo Jeewanu and will be published elsewhere (11).

It can be safely concluded that the synergistic particles act more efficiently under the conditions in which they are autoorganised.

The observation becomes of considerable interest because like any living system these abiogenic photosynergistic particles also act better under the condition in which they are auto-organised. What is more interesting is that this shows that these synergistic particles have memory of their own and recoganise the condition of their formation and act better in that particular condition. Thus there appears to be inbetween the typical nucleic acid memory apparatus of the present-day living system, the crystal structure memory apparatus suggested by Cairns-Smith (13) and has a memory apparatus which is non nucleic acid and non crystal structure, because X-ray diffraction shows that Jeewanu have non crystal structure, and is inbetween the both of them.

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Table No. 1.

Pressure variation in cm. of mercury in the Warburg's flasks under oxygenic condition, having phosphate buffer of different pH values and aqueous mixture of Si-Mo Jeewanu ~~was~~ prepared under oxygenic condition.

Time in min.	Pressure variation in W.f. in cm. of mercury at different pH		
	5·7	6·5	7·5
Sunlight			
5	0·5	1·1	0·9
10	1·8 ~~2·3~~	1·8	1·2
15	2·3	1·9	1·9
20	2·4	2·3	2·1
25	2·8	2·6	1·9
30	2·5	2·3	2·1
Shade			
5	1·7	1·7	1·0
10	1·0	0·9	0·2
15	0·5	0·7	0·0
20	0·3	0·5	-0·2
25	0·1	0·5	-0·1
30	-0·1	0·3	-0·1
Sunlight			
5	1·0	0·8	0·6
10	1·9	2·1	1·3
15	2·8	2·9	1·6
20	2·9	2·9	1·6
25	3·1	3·2	1·5
30	3·6	3·7	1·5
Shade			
5	2·1	1·9	0·8
10	1·1	0·9	0·1
15	0·6	0·7	-0·2
20	0·4	0·5	-0·3
25	0·2	0·4	-0·3
30	0·1	0·4	-0·4

Table No. 2.

Pressure variation in cm. of mercury in the Warburg flasks under anoxygenic condition, having phosphate buffers of different pH values and aqueous mixture of Si-Mo-geewanu prepared under oxygenic condition.

Time in min.	Pressure variation in wf in cm. of mercury at different pH		
	5.7	6.5	7.5
	Sunlight		
5	−2.0	0.5	0.2
10	−1.9	0.5	0.0
15	−1.9	−0.2	0.4
20	−1.3	−0.2	0.4
25	−1.6	−0.5	0.1
30	−1.0	−0.2	0.3
	Shade		
5	0.2	−0.8	−1.0
10	0.5	−0.7	−1.0
15	0.4	−0.7	−0.8
20	0.2	−0.6	−1.0
25	0.2	−0.6	−0.8
30	0.2	−0.6	−0.8
	Sunlight		
5	−1.1	−0.8	−0.1
10	−1.7	−0.2	−0.1
15	−0.9	0.0	0.6
20	−0.9	−0.1	0.4
25	−0.6	0.0	0.4
30	−0.4	0.0	0.2
	Shade		
5	0.5	−0.4	−0.7
10	0.9	−0.6	−0.8
15	0.6	−0.7	−0.7
20	0.5	−0.5	−0.8
25	0.2	−0.5	−0.9
30	0.0	−0.5	−1.0

Table No. 3

Pressure variation in cm. of mercury in the Warburg's flasks under oxygenic condition having phosphate buffer of different pH values and aqueous mixture of Si-Mo Jeewanu prepared under anoxygenic condition.

Time in min	Pressure variation in w/. in cm. of mercury at diff. pH		
	5.7	6.5	7.5
Sunlight			
5	1.2	1.2	1.5
10	1.4	1.2	1.9
15	1.5	1.0	1.9
20	1.5	1.5	1.8
25	1.6	1.1	1.6
30	1.6	1.2	1.7
Shade			
5	1.4	0.7	1.6
10	1.2	1.1	0.7
15	1.1	0.8	0.5
20	1.0	0.4	0.4
25	0.8	0.3	0.2
30	0.7	0.3	0.1
Sunlight			
5	1.3	1.0	1.4
10	1.5	1.1	1.8
15	1.5	1.0	1.7
20	1.4	1.0	1.7
25	1.5	1.1	1.7
30	-0.2	1.2	1.5
Shade			
5	0.6	1.2	0.4
10	-0.4	1.3	0.8
15	-0.8	0.0	0.0
20	-0.8	-0.2	-0.3
25	-0.8	-0.1	-0.3
30	-0.9	0.0	-0.2

Table No. 4.

Pressure variation in cm. of mercury in the Warburg's flasks under anoxygenic condition, having phosphate buffer of different pH values and aqueous mixture of Si-Mo Jeevanu prepared under anoxygenic conditions.

Time in min	Pressure variation in w.f. in cm. of mercury at different pH		
	5·7	6·5	7·5
Sunlight			
5	-0·5	-0·6	1·1
10	0·9	-0·7	1·1
15	2·2	-0·5	0·6
20	1·9	-0·9	1·2
25	2·1	-0·6	1·3
30	3·6	-0·7	0·6
Shade			
5	4·4	-1·3	-0·6
10	2·5	-1·7	-1·5
15	1·8	-1·5	-1·4
20	1·6	-1·9	-1·4
25	0·8	-2·6	-1·4
30	0·2	-2·1	-1·9
Sunlight			
5	-0·4	-1·4	-1·0
10	0·1	-0·8	0·3
15	2·8	-0·5	0·7
20	3·7	-0·5	0·9
25	3·6	-0·3	1·3
30	3·7	-0·2	1·3
Shade			
5	3·8	-0·2	-1·1
10	2·7	-1·5	-1·7
15	2·7	-1·7	-1·9
20	2·0	-2·0	-2·0
25	1·6	-2·2	1·9
30	0·3	-1·7	1·5

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THE ENERGY FLOW IN PHOTOSYNERGISTICALLY FORMED, NONLINEAR SYSTEMS, SI-MO JEEWANU IS MORE EFFICIENT UNDER THE CONDITIONS THEY ARE AUTO ORGANISED

By

Bahadur, K., Ranganayakai, S. and Mridula Bahadur
(Chemistry Department, University of Allahabad, India)

Key Words: Synergetics, Oxygenic, Anoxygenic, Si-Mo Jeewanu, Sunlight, Memory.

Abstract:

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"ABIOSYMBIOSIS OR MOLECULAR SYMBIOSIS, SYNERGISTIC COLLABORATON PROCESSES AN ESSENTIAL EARLIER STAGE IN ORIGIN OF LIFE"

(Key Words): Molecular Symbiosis, Semes, Molybdenum Jeewanu, Light, Water splitting, Nitrogen fixation, Synergetics.

(By :Bahadur, K., S. Ranganayaki, Mridula Bahadur* and Vandana Mathur, Chemistry Department, Allahabad University, INDIA).

Abstract

Abiosymbiosis or Molecular Symbiosis the synergistic collaboration processes are earlier steps which helped in the auto organisation of photoautotrophs like molybdenum Jeewanu, the protocell in which an alkaline material with strong properties of splitting water in sunlight to hydrogen and oxygen and poor nitrogen fixing ability forms a symbiotic association with an acidic material which has poor ability of splitting water in sunlight but strong property of fixation of molecular nitrogen, resulting in a more efficient system, the molybdenum Jeewanu with good property of water splitting in sunlight and fixation of molecular nitrogen. The efficient flow of energy through these systems, stabilised the resultant nonlinear systems and these develop boundary wall and internal structures, grow from within, multiply by budding and show metabolic activity.

142/Prod.

Organisation of Biological Matrix

By

Mridula Bahadur and Krishna Bahadur

Why and how the molecules constituting the first living system organised ~~into~~ in a specific steric position, the biological matrix, which started showing the properties of biological order, is a very significant aspect of the study of the process of origin of life. Under suitable conditions matter has ~~inherent~~ inherent property of duplication and a system of matter in equilibrium has inherent property of adaptability. So whenever conditions were ~~favourable~~ suitable for a set of molecules they arranged themselves in a pattern which could show these properties of ~~l~~ matter. Such systems had a ~~boundary~~ boundary wall and internal structures and if appropriate physico-chemical conditions were present in the environment this system could grow from within, multiply by budding and had metabolic activity.

Such particles were photosynthesised and named as Jeewanu in 1963 and the worked by M.H. Briggs in 1964 and 1965. Jeewanu can be synthesised ~~by~~ with a variety of organic and inorganic matter and Copper Jeewanu, Cerous Jeewanu and Nickel and Cobalt Jeewanu were prepared. The most interesting amongs these are Molybdenum Jeewanu and Silica-molybdenum Jeewanu.

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Molybdenum Jeewanu can be fixed with biological fixatives and stained with biological dyes as Sudan Black, Eosin, Haematoxylin, Pyronin-Y and Gentian Violet and are sensitive to the presence of antibiotics and sulphur drugs in their parental environmental medium.

Molybdenum Jeewanu have ferredoxin - like material and nitrogenase - like material. These Jeewanu in D_2O form CHD=CHD from CH = CH of the over head space and organic material with ^{14}C from $NaH^{14}CO_3$ present in the environmental medium on exposure to light from a clinial U.V. lamp.

There is a close relationship between Jeewanu and crystals and in all probability life originated as a highly deformed crystal. In a particular mixture of molybdenum Jeewanu after the mixture have been exposed for a certain number of hours to sunlight in the evening after a full one day's exposure the mixture is full of blue rod shaped structures like crystals. When this mixture is allowed to stand in dark over night and is examined microscopically next morning it is full of well formed Jeewanu and rod shaped structure disappear. Again on next day after exposure, in the evening, the mixture is full of rod shaped crystals and next morning again it is full of Jeewanu. And this process can be repeatedly be seen for a few days.

MICROFOSSILS, BIOGENIC OR ABIOGENIC ?

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It has been suggested by us that under suitable conditions, matter organises itself in cell-like structure which have boundary wall and internal structures, (Bahadur et al. 1986) and where necessary substrate molecules were present in its surroundings and the energy needed for the formation of the body material of these autoorganised cell-like structures was available, these structures started showing the properties of growth multiplication and metabolic activity (Bahadur, K., 1964, 1967). These particles could adapt to mild changing conditions and thus could evolve. Such structures were from 0.2 to 3.0 μ (Bahadur et al. 1964), as against cell-like autoorganised structures without the ability to grow from within, to multiply and to exhibit metabolic activity and the latter may be called merely as cell-models or models of the Protocell. Jeewanu are not cell models but still are far from the present day cells and these could have been of a variety of materials not present in today's cells.

Kirpotin et al. (1986) have demonstrated that some autoorganised cell-models on artifical fossilisation resemble natural microfossils. Several cell-models have been described viz. microspheres (Fox and Dose, 1977), marigranules (Yanagawa and Egami, 1977) and others. These look like cells under high power optical microscope and some of these under specific conditions in presence of some specific a

Energy Flow in Photosynergistic
Nonliner Systems.
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Micrographs of multiplying molybdenum Jewanu showing boundary wall and internal structures (X1500, by Varma, P.K.).

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Typist:

1. For Warburg's flask w.f. is written, both w & f are small letters.

2. Pyrogallol is correct spelling.

3. Weight by volume is written as (w/v) both small letters.

4. Do not underline the title.

5. Type in electric typewriter in ~~single~~ double spacing.

6. For graph type at the bottom as I have written in single spacing.

P.T.O.

THE ENERGY FLOW IN PHOTOSYNERGISTICALLY FORMED, NONLINEAR SYSTEMS, SI-MO JEEWANU IS MORE EFFICIENT UNDER THE CONDITIONS THEY ARE AUTOORGANISED

By

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Key words: Synergetics, Oxygenic, Anoxygenic, Si-Mo Jeewanu, Sunlight, Memory.

Abstract:

Under suitable conditions photosynergetic, light assisted autoorganisation helps to form Si-Mo Jeewanu. These particles spilt water in sunlight producing hydrogen and oxygen, fix molecular nitrogen and under certain conditions indicate loss of nitrogen. It has been observed that if these particles are produced under oxygenic condition, these particles show more energy flow, as indicated by splitting of water in sunlight and fixation of nitrogen, under oxygenic conditions than under anoxygenic conditions. But when these particles are autoorganised under anoxygenic conditions they show more energy flow under anoxygenic conditions than under oxygenic conditions indicating some sort of memory in these abiogenic particles to recognise the conditions of their synergesis and it becomes still more important because these particles duplicate by budding.

Introduction:

It has been observed that photosynergetic, light assisted self-organisation, produces such nonlinear systems which (i) show growth and development, (ii) need energy input from outside and (iii) have such properties which are not found in any of its constituents. Such systems have been synthesised[1] and named as Jeewanu, a Sanskrit word for 'Particles of Life' and the work has been repeatedly confirmed[2,3]. Study of such systems of particles is of great interest in the investigation of origin of life and formation of the earliest system which had the properties of living.

One of such systems which has been extensively studied is molybdenum Jeewanu or self sustaining coacervate with about 39% of molybdenum[4]. These particles have ferridoxin - like[5] and nitrogenase - like[6] materials in them, multiply by budding and split water in sunlight and fix molecular nitrogen[7,8,9,10]. This system has been modified by incorporating soluble sodium orthosilicate and sodium chloride[11,12].

So far these particles were produced under oxygenic condition only. These particles produce hydrogen and oxygen on splitting water in sunlight and in Warburg's apparatus these gases cause increase in the pressure in the Warburg's flask (wf.). If this Warburg's study is done under anoxygenic condition less pressure is produced in wf. because alkaline pyrogallol kept in the side lobe absorbs the oxygen which is simultaneously produced on splitting of water by these particles in sunlight. However, if one wants to have a good amount of hydrogen by splitting of water in sunlight one must have a high increase in the pressure inside the wf. Under anoxygenic condition also even after the simultaneously liberated oxygen is absorbed by alkaline pyrogallol of the side lobe of the wf.

A typical biological system acts better in the environment in which it is produced. We prepared Si-Mo Jeewanu both under oxygenic and anoxygenic conditions and investigated their gas transformations both under oxygenic and anoxygenic conditions in phosphate buffer of pH 5.7, 6.5 and 7.5 in Warburg's apparatus and observed that at all pH the particles produced under oxygenic conditions were more efficient and showed more energy flow i.e., splitting of water in sunlight and fixation of molecular nitrogen under oxygenic conditions and the particles produced under anoxygenic condition were more efficient and showed greater energy flow under anoxygenic condition. This observation shows some parallelism with the living systems which act better in the condition in which they are produced and also this suggests a new method of preparing Si-Mo Jeewanu which show significant increase in pressure in the wf. under anoxygenic condition, caused by the presence of considerable amount of hydrogen.

Experimental

4% (w/v) ammonium molybdate, 3% (w/v) diammonium hydrogen phosphate (DAHP), 5%(w/v) soluble sodium orthosilicate and 3% (w/v) sodium chloride solutions were prepared by dissolving the requisite amount of the salt in 100 ml. of distilled water. A mineral solution was prepared by dissolving 20 mg. each of NaCl, K_2SO_4, $MgSO_4$, KH_2PO_4, $MnSO_4$ and

$Ca(CH_3COO)_2$ one by one in about 80 ml. of distilled water. 50 mg of ferrous sulphate were dissolved in about 10 ml. of distilled water. 0.1 ml. of $6N/H_2SO_4$ was added to it to stop the hydrolysis and this was mixed with the above mineral solution. The total volume of this mineral soultuion was made to 100 ml. 36% formaldehyde was used in the experiment.

70 ml. of the ammonium molybdate solution, 140 ml. of the DAHP solution, 70 ml. of the sodium chloride solution and 70 ml. of the soluble sodium orthosilicate solution were taken in a one l., conical flask and 70 ml. of distilled water was added to it. Total volume of the solution was 500 ml. 250 ml. of this solution were taken in two 500 ml. corning conical flasks. The flasks were cotton plugged and sterilised in an autoclave at 15 lb pressure for 30 min. On cooling 40 ml. of 35% formaldehyde were added in each flask aseptically.

In one flask one sterilised test tube reaching up to the neck of the conical flask and two - third filled with alkaline pyrogallol solution was lowered. This flask was tightly closed with a sterilised rubber cork. Alkaline pyrogallol solution was prepared by mixing 10 ml. of 20% pyrogallol solution with 10 ml. of 30% sodium hydroxide solution.

Both the flasks, one plugged with only cotton and allowing the passage of air through it and thus having in the over head space of its mixture air with nitrogen and oxygen and the other flask with rubber stopper, which had all its oxygen absorbed in the alkaline pyrogallol solution contained in the testtube kept in it and having only anoxygenic atmosphere in the over head space of its mixture, were exposed to sunlight for 24 hr. giving 8 hr. exposure each day for 3 days.

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(7)

The mixtures contained in the flask were gently shaken once a day by giving whirling motion to the flasks, taking care that the mixture does not touch the cotton plug or the pyrogallol solution kept in the test-tube lowered in it. This dispersed the sediment of Si-Mo. Jeewanu, getting formed at the bottom of the mixture during exposure, in the whole mixture.

After the completion of the exposure, the mixtures were opened. The alkaline pyrogallol testtube was removed from the rubber stoppered flask. The mixtures were shaken and the Jeewanu formed in these mixtures were separated by filtration through dried and weighed filter papers (Whatman No. 1) and the particles dried in a desiccator and weighed.

Weight of the, Si-Mo Jeewanu prepared under oxygenic condition was 1.0900 gm.

Weight of the Si-Mo Jeewanu prepared under anoxygenic condition was 1.7614 gm.

Thus under anoxygenic condition about 61.4% more of Si-Mo Jeewanu were prepared as compared to the Jeewanu prepared under oxygenic conditions.

The splitting of water in sunlight, fixation of nitrogen and under certain condition loss of fixed nitrogen by both the types of Si-Mo Jeewanu, one prepared under oxygenic condition (PUOC) and the other prepared under anoxygenic condition (PUAC) were studied using Warburg's apparatus.

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When water is split into hydrogen and oxygen by exposing an aqueous mixture of the Jeewanu to sunlight it causes increase in the pressure in the wf. These particles have nitrogenase- like material[6], so as soon as hydrogen is produced, it starts combining with the nitrogen of the overhead space of the wf. and the pressure in wf. decreases. It is more clearly observed after the exposure to sunlight is cut off and the increase in the pressure caused by the formation of hydrogen and oxygen by splitting of water stops and the remaining hydrogen of the over head space continues combining with the nitrogen showing decrease in the pressure in wf. Under certain conditions when the fixation of nitrogen is very fast, the liberated hydrogen immediately combines with the nitrogen in the overhead space of the wf. and decreases the pressure in the wf. and decrease in the pressure in wf. is observed as soon as the exposure to sunlight begins. Under certain conditions although the fixation of nitrogen is very fast, the fixed nitrogen is lost and N_2 is produced causing increase in the pressure in the wf. The fixation of nitrogen causing decrease in the pressure of the wf. is due to the formation of diimide. NH = NH and loss of nitrogen is due to decomposition of diimide and its disproportionation.

(2) It was observed that when the wf. of the Warburg's apparatus is kept dipped in the water bath of the Warburg's apparatus the sunlight has to pass through the water of the water bath to reach the wf. containing aqueous mixture of the Jeewanu and this sunlight becomes too weak for action. So the water bath of the Warburg's was removed and a control

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wf. containing same volume of water was kept near the exposed wf. and the reading of pressure in this wf. was always recorded together with the readings of pressure in the experimental wf. The pressure in the control wf. was due to the temperature difference during the exposure of the wf. and also when these stood in shade. This reading of pressure in the control wf. was subtracted from the reading in the control flask to get the net change in the pressure of the experimental wf. due to the gas transformations taking place in them. It was assumed that in the atmosphere the temperature ~~between~~ within a small space of 30 cm x 20 cm x 20 cm, in which all the wf. and the control flask were kept, was the same.

It was further observed that when the liquid in the manometers of the Warburg's apparatus was water, the pressure difference during the exposure to sunlight and when the apparatus stood in shade was so much that levels of the liquid went beyond the range of the manometers. So the manometers were filled with mercury and then the pressure difference could be read within the limits of the manometer.

(3) 20 mg of the particles were taken in the wf. with 4 ml. of distilled water and 1 ml. of the phosphate buffer of pH 5.7 in the first, pH 6.5 in the second and pH 7.5 in the third wf. The total volume of each of the conical wf. was 14.5 ml. the bore of the manometer was 1 mm. in diameter and bottom area of each wf. was 9.5 cm^2. One similar wf. was kept as control which had only 5 ml. of distilled water. For working under oxygenic condition 0.3 ml. of distilled water was kept in the side lobe of each of the wf. For working under anoxygenic condition 0.3 ml. of freshly prepared alkaline pyrogallol was kept in the side lobe of the wf. This was prepared by mixing 0.15 ml. of 20% pyrogallol solution and 0.15

ml. of 30% sodium hydroxide solution. The lobes of the wf. were covered with same size (2.5 cm^2) of a black cloth to avoid any decomposition of alkaline pyrogallol during the exposure to sunlight. To keep the results of working under oxygenic conditions and anoxygenic condition comparable, the side lobe of the wf. was covered with this black cloth even while working under oxygenic condition when the side lobe had only distilled water.

Discussion:

When Si-Mo Jeewanu were prepared under oxygenic condition, these particles were most active under oxygenic condition at pH 5.7 and the pressure increase in the wf. goes upto about 2.70 cm of mercury but under anoxygenic condition, the fixation of nitrogen is so much at this pH that there is decrease in pressure in the wf. even during the exposure of the Si-Mo Jeewanu aqueous mixture to sunlight. However, this fixed nitrogen is soon lost as is indicated by the increase in pressure in the wf. on standing in shade, after all the hydrogen liberated by splitting of water is consumed in nitrogen fixation and decrease in pressure due to this stops.

At pH 6.5 and 7.5 the trend of water splitting in sunlight and fixation of nitrogen remains same both under oxygenic and anoxygenic conditions. There is about 2.5 cm increase in pressure in sunlight and about the same decrease in pressure in shade due to fixation of nitrogen. Under oxygenic conditions this increase in the pressure during exposure is less than 0.5 cm of mercury during exposure and there is about 1 cm. decrease in the pressure in the wf. in shade. The decrease in value of the pressure

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in wf. during exposure under anoxygenic condition is due to the oxygen simultaneously produced during water splitting being absorbed in the alkaline pyrogallol of the side lobe.

(6) When Si-Mo Jeewanu are produced under anoxygenic condition and their gas transformation is examined under oxygenic condition there is increase of only about 1.8 cm of mercury pressure in the wf. at pH. 5.7 but under anoxygenic condition there is an increase of about 4.4 cm. of mercury in the wf. during exposure at this pH. This increase in pressure under anoxygenic condition where alkaline pyrogallol is kept in the side lobe to absorb all the oxygen which is simultaneously produced during the splitting of water by sunlight together with hydrogen is of great interest as it is free of oxygen and is due to the presence of hydrogen in the wf. The results obtained at the other two pH values studied are similar.

The experiments described here show that the synergistic particles produced by the exposure of sterilised aqueous mixture of ammonium molydate, DAHP, biological minerals, formaldehyde, sodium chloride and soluble sodium Orthosilicate to sunlight under oxygenic condition are better in splitting water in sunlight under oxygenic condition and the same particles prepared by exposing the same mixture under anoxygenic condition are better in water splitting under anoxygenic condition. Similar results were obtained in the case of Ti, Zn, Co, Cd and Ba doped Si-Mo Jeewanu (11) and will be published else where.

It can be safely concluded that the synergistic particles act more efficiently under the conditions in which they are autoorganised.

The observation becomes of considerable interest because like any living system these abiogenic photosynergistic particles also act better under the condition in which they are autoorganised. What is more interesting is that this shows that these synergistic particles have memory of their own and recognise the condition of their formation and act better in that particular condition. Thus these appear to be in between the typical nucleic acid memory apparatus of the present day living system, the crystal structure memory apparatus suggested by Cairns-Smith (13) and has a memory apparatus which is of non nucleic acid and non crystal structure, because X-Ray diffraction shows that Jeewanu have non crystal structure, ~~and are in between both of them~~.

Acknowledgement:

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CHARACTERISTIC NON BROWNIAN MOTION IN THE JEEWANU, THE MODEL OF PROTOCELLS.

Bahadur, K., Ranganayaki, S., Bahadur, M., Mathur, V. and Gupta, V.K.

Chemistry Department, University of Allahabad, India.

In all probability phosphorylation on protocells made of phospholipid or phospholipid-like materials would have induced photobiological motility in the earlier stages of evolution (Matsuno, 1986). The origin of photobiological and biological motility is an evolutionary event. Protocells made of phospholipids or phospholipid-like material would have served as material units carrying free equilibration. The JEEWANU (Bahadur et al., 1964, Bahadur and Ranganayaki, 1981) with boundary wall made of phospholipid and phospholipid-like materials (Singh, 1973) would have served this purpose.

The molybdenum Jeewanu have phospholipid membrane (Verma, 1981). These particles also have phosphotase-like (Briggs, 1968), ATP-ase-like (Bahadur and Ranganayaki, 1970) and acid phosphatase-like (Bahadur and Gupta, 1984) materials. These particles also have ferridoxin-like (Rao et al., 1978) and nitrogenase-like (Smith et al., 1981) materials in them. These Jeewanu have boundary wall and internal structures (Bahadur and Rangnayaki, 1981). These particles can be fixed with biological fixatives and stained with biologial dyes (Bahadur and Gupta, 1972). In sunlight these can split water to hydrogen and oxygen and also are capable of fixing molecular nitrogen.

Silicon - molybdenum Jeewanu are prepared by incorporating silicon in molybdenum Jeewanu. These are prepared by exposing an aqueous mixture of 1 volume of 4% ammomium molybdate, 2 volumes of 3% dihydrogen ammonium phosphate, 5 volumes of the mineral solution, 1 vol. of 36% formaldehyde 1 vol. of 3% sodium chloride and 1 vol. of 5% soluble sodium orthosilicate solution to sunlight in a cotton plugged, conical flask giving two hours exposure every day for 24 days. The mineral solution was prepared by mixing 20 mg of each of NaCl, K_2SO_4, $CaAc_2$, $MgSO_4$, KH_2PO_4, $MnSO_4$ and 50 mg of $FeSO_4$ in 100 ml. of distilled water. After 2 hr. of exposure to sunlight the mixture was allowed to stand in shade for the whole day.

The mixture on exposure produces spherical particles with boundary wall and internal structure and graduallly turns blue. These silicon molybdenum Jeewanu show a charecteristic shivering and oscillating motion after 10 days of exposure and this continues for 24 days. With further exposure this motion gradually stops.

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Introduction:

It has been observed that photosynergetic, light assisted self-organisation, produce such nonlinear systems which (i) show growth and development, (ii) need energy input from outside and (iii) have such properties which are not found in any of its constituents. Such systems have been synthesised [1] and named as Jeewanu, a Sanskrit word for 'Particles of Life' and the work has been repeatedly confirmed [2,3]. Study of such systems of particles is of great interest in the investigation of origin of life and formation of the earliest system which had the properties of living.

One of such systems which has been extensively studied is molybdenum Jeewanu or self sustaining coacervate with about 39% of molybdenum [4]. These particles have ferridoxin - like [5] and nitrogenare - like [6] materials in them, multiply by budding and split water in sunlight and fix molecular nitrogen [7,8,9,10]. This system has been modified by incorporating soluble sodium orthosilicate and sodium chloride [11,12].

So far these particles were produced under oxygenic condition only. These particles produce hydrogen and oxygen on splitting water in sunlight and in Warburg's apparatus there gases cause increase in the pressure in the Warburg's flask (wf.) If this Warburg's study is done under anoxygenic

Typist: W is small w.

condition less pressure is produced in Wf. because alkaline pyrogallol kept in the side lobe absorbs the oxygen which is simultaneously produced on splitting of water by these particles in sunlight. However if one wants to have a good amount of hydrogen by splitting of water in sunlight one must have a high increase in the pressure inside the wf. Under anoxygenic condition also even after the simultaneously liberated oxygen is absorbed by alkaline pyrogallol of the side lobe of the wf.

A typical biological system acts better in the environment in which it is produced. We prepared Si-Mo Jeewanu both under oxygenic and anoxygenic conditions and investigated their gas transformations both under oxygenic and anoxygenic conditions in phosphate buffer of pH 5.7, 6.5 and 7.5 in Warburg's apparatus and observed that at all pH the particles produced under oxygenic conditions were more efficient and showed more energy flow i.e., splitting of water in sunlight and fixation of molecular nitrogen under oxygenic conditions and the particles produced under anoxygenic condition were more efficient and showed greater energy flow under anoxygenic condition. This observation shows some parallelism with the living systems which act better in the condition in which they are produced and also this suggests a new method of preparing Si-Mo Jeewanu which show significant increase in the pressure in the Wf. under anoxygenic condition, caused by the presence of considerable amount of hydrogen.

Experimental

4% (W/V) ammonium molybdate, 3 % (W/V) diammonium hydrogen phosphate (DAHP), 5 % (W/V) soluble sodium orthosilicate and 3% (W/V) sodium chloride solutions were prepared by dissolving the requisite amount of the salt in 100 ml. of distilled water. A mineral solution was prepared by dissolving 20 mg. of each of NaCl, K_2SO_4, $MgSO_4$, KH_2PO_4, $MnSO_4$ and Ca $(CH_3COO)_2$ one by one in about 80 ml. of distilled water. 50 mg of ferrous sulphate were dissolved in about 10 ml. of distilled water. 0.1 ml. of $6N/H_2SO_4$ was added to it to stop the hydrolysis and this was mixed with the above mineral solution. The total volume of this mineral solution was made to 100 ml. 36% formaldhydr was used in the experiment.

70 ml. of the ammonium molybdate solution, 140 ml. of the DAHP solution, 70 ml. of the mineral solution, 70 ml. of the sodium chloride solution and 70 ml. of the soluble sodium orthosilicate solution were taken in one conical flask and 70 ml. of distilled water was added to it, Total volume

of the solution was 500 ml. 250 ml. of this solution were taken in two 500 ml. corning conical flasks. The flasks were cotton plugged and sterilised in an autoclave at 15 lb pressure for 30 min. On cooling 40 ml. of 35% formaldehyde were added in each flask aseptically.

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Both the flasks, one ~~having~~ plugged with only cotton ~~plug in its mouth~~ and allowing the passage of air through it and thus having in the over head space of its mixture ~~had~~ air with nitrogen and oxygen and the other flask with rubber stopper ~~in its mouth~~, which had all its oxygen absorbed in the alkaline pyrogallol solution contained in the test tube kept in it and having only anoxygenic atmosphere in the over head space of its mixture, were exposed to sunlight for 24 hr. giving 8 hr. exposure each day for 3 days.

The mixtures contained in the flask were gently shaken once a day by giving whirling motion to the flasks, taking care that the mixture does not touch the cotton plug or the pyrogallol solution kept in the test-tube lowered in it. This dispersed the sediment of Si-Mo. Jeewanu, getting formed at the bottom of the mixture during exposure, in the whole mixture.

After the completion of the exposure, the mixtures were opened. The alkaline pyrogallol test-tube was removed from the rubber stoppered flask. The mixtures were shaken and the Jeewanu formed in these mixtures were separated by filtration through dried and weighed filter papers (~~Weltman~~ Whatman No.41) and the particles dried in a desiccator and weighed.

Weight of the ~~particles~~, Si-Mo Jeewanu prepared under oxygenic condition was 1.0900 gm.

Weight of the Si-Mo Jeewanu prepared under anoxygenic condition was 1.7614 gm.

Thus under anoxygenic condition about 61.4% more of Si-Mo Jeewanu were prepared as compared to the Jeewanu prepared under oxygenic conditions.

The splitting of water in sunlight, fixation of nitrogen and under certain condition loss of fixed ~~of~~ nitrogen by both the types of Si-Mo Jeewanu, one prepared under oxygenic condition (PUOC) and the other prepared under anoxygenic condition (PUAC) were studied using Warburg's apparatus.

When water is split into hydrogen and oxygen by exposing an aqueous mixture of the Jeewanu to sunlight it causes increase in the pressure in the ~~Warburg's flask (Wf.)~~ Wf. These particles have nitrogenase- like material[6], so as soon as hydrogen is produced, ~~it~~ starts combining with the nitrogen of the overhead space of the ~~Wf.~~ Wf. and the pressure in ~~Wf.~~ Wf. decreases. It is more clearly observed after the exposure to sunlight is cut off and the increase in the pressure caused by the formation of hydrogen and oxygen by splitting of water stops and the remaining hydrogen of the over head space continues combining with the nitrogen showing decrease in the pressure in ~~Wf.~~ Wf. Under certain condition when the fixation of nitrogen is very fast, the liberated hydrogen immediately combines with the nitrogen in the overhead space of the ~~Wf.~~ Wf. and decreases the pressure in the ~~Wf.~~ Wf. and decrease in the pressure in ~~Wf.~~ Wf. is observed as soon as the exposure to sunlight begins. Under certain conditions although the fixation of nitrogen is very fast, the fixed nitrogen is lost and N_2 is produced causing increase in the pressure in the ~~Wf.~~ Wf. The fixation of nitrogen causing decrease in the pressure of the Wf. is due to the formation of diimide. NH = NH and loss of nitrogen is due to decomposition of diimide and its disproportionation.

It was observed that when the Wf. of the Warburg's apparatus is kept dipped in the water bath of the Warburg's apparatus the sunlight has to pass through the water of the water bath to reach the Wf. containing aqueous mixture of the Jeewanu and this sunlight becomes too weak for action. So the water bath of the Warburg's apparatus was removed and a control Wf. containing same volume of water was kept near the exposed Wf. and the reading of pressure in this Wf. was always recorded together with the readings of pressure in the experimental Wf. The pressure in the control Wf. was due to the temperature difference during the exposure of the Wf. and also when these stood in shade. This reading of pressure in the control Wf. was subtracted from the reading in the control flask to get the net change in the pressure of the experimental Wf. due to the gas transformations taking place in them. It was assumed that in the atmosphere the temperature between a small space of 30 cm x 20 cm x20 cm, in which all the Wf. and the control flask were kept, was the same.

It was further observed that when the liquid in the manometers of the Warburg's apparatus was water the pressure difference during the exposure to sunlight and when the apparatus stood in shade was so much that levels of the liquid went beyond the range of the manometers. So the manometers were filled with mercury and then the pressure difference could be read within the limits of the manometers

20 mg of the particles were taken in the Wf. with 4 ml.

of distilled water and 1 ml. of the phosphate buffer of pH 5.7 in the first, pH 6.5 in the second and pH 7.5 in the third Wf. The total volume of each of the conical Wf. was 14.5 ml. the bore of the manometer was 1 mm. in diameter and bottom area of each Wf. was 9.5 cm^2 One similar Wf. was kept as control which had only 5 ml. of distilled water, For working under oxygenic condition 0.3 ml. of distilled water was kept in the side lobe of each of the Wf. For working under anoxygenic condition 0.3 ml. of freshly prepared alkaline pyrogallol was kept in the side lobe of the Wf. This was prepared by mixing 0.15 ml. of 20% pyrogallol solution and 0.15 ml. of 30% sodium hydroxide solution. The lobes of the Wf. were covered with same size (2.5 cm^2) of a black cloth to avoid any decomposition of alkaline pyrogallol during the exposure to sunlight. To keep the results of working under oxygenic condition and anoxygenic condition comparable, the side lobe of the Wf. was covered with this black cloth even while working under oxygenic condition when the side lobe had only distilled water.

Discussion:

When Si-Mo Jeewanu were prepared under oxygenic condition, these particles were most active under oxygenic condition at pH 5.7 and the pressure increase in the Wf. goes upto about 2.70cm of mercury but under anoxygenic condition, the fixation of nitrogen is so much at this pH that there is decrease in pressure in the Wf. even during the exposure of the Si-Mo Jeewanu aqueous mixture to sunlight. However this fixed nitrogen is soon lost as is indicated by the increase in pressure in the Wf. on standing in shade, after all the hydrogen liberated by splitting of water is consumed in nitrogen fixation and decrease in pressure due to this stops.

At pH. 6.5 and 7.5 the trend of water splitting in sunlight and fixation of nitrogen remains same both under oxygenic and anoxygenic conditions. There is about 2.5 cm increase in pressure in sunlight and about the same decrease in pressure in shade due to fixation of nitrogen. Under oxygenic conditions this increase in the pressure during exposure is less than 0.5 cm of mercury during exposure and there is about 1 cm. decrease in the pressure in the Wf. in shade. The decrease in value of the pressure in Wf. during exposure under anoxygenic condition is due to the oxygen simultaneously produced during water splitting being absorbed in the alkaline pyrogallol of the side lobe.

When Si-Mo Jeewanu are produced under anoxygenic condition and their gas transformation is examined under oxygenic condition there is increase of only about 1.8 cm of mercury pressure in the Wf. at pH. 5.7 but under anoxygenic condition there is an increase of about 4.4 cm. of mercury in the Wf. during exposure at this pH. This increase in pressure under anoxygenic condition where alkaline pyrogallol is kept in the side lobe to absorb all the oxygen which is simultaneously produced during the splitting of water by sunlight together with hydrogen is of great interest as it is free of oxygen and is due to the presence of hydrogen in the Wf. The results obtained at the other two pH values studied are similar.

The experiments described here show that the synergistic particles produced by the exposure of sterilised aquueous mixture of ammonium molybdate, DAHP, biological minerals, formaldehyde, sodium chloride and soluble sodium orthosilicate to sunlight under oxygenic condition are better in splitting water in sunlight under oxygenic condition and the same particles prepared by exposing the same mixture under anoxygenic condition are better in water splitting under anoxygenic condition. Similar results were obtained in the case of Ti, Zn, Co, Cd and Ba doped Si-Mo Jeewanu (11) and will be published else where.

It can be safely concluded that the synergistic particles act more efficiently under the conditions in which they are autoorganised.

The observation becomes of considerable interest because like any living system these abiogenic photosynergistic particles also act better under the condition in which they are auto-organised. What is more interesting is that this shows that these synergistic particles have memory of their own and recognise the condition of their formation and act better in that particular condition. Thus these appear to be in between the typical nucleic acid memory apparatus of the present day living system, the crystal structure memory apparatus suggested by Cairns-Smith (13) and has a memory apparatus which is of non nucleic acid and non crystal structure, because X-Ray diffraction shows that Jeewanu have non crystal structure, and are in between both of them.

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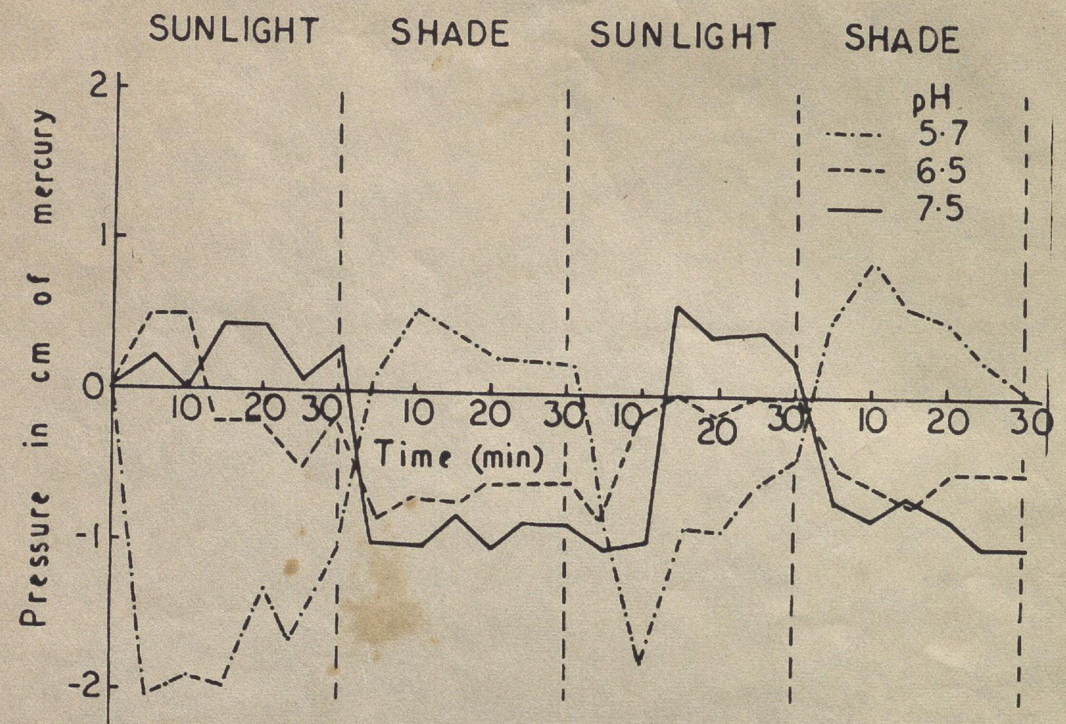

Fig. 1: Energy flow under anoxygenic condition in Si-Mo Jeewanu autoorganised under oxygenic condition, as indicated by pressure increase in cm. of mercury due to splitting of water in sunlight and decrease caused by fixation of nitrogen.

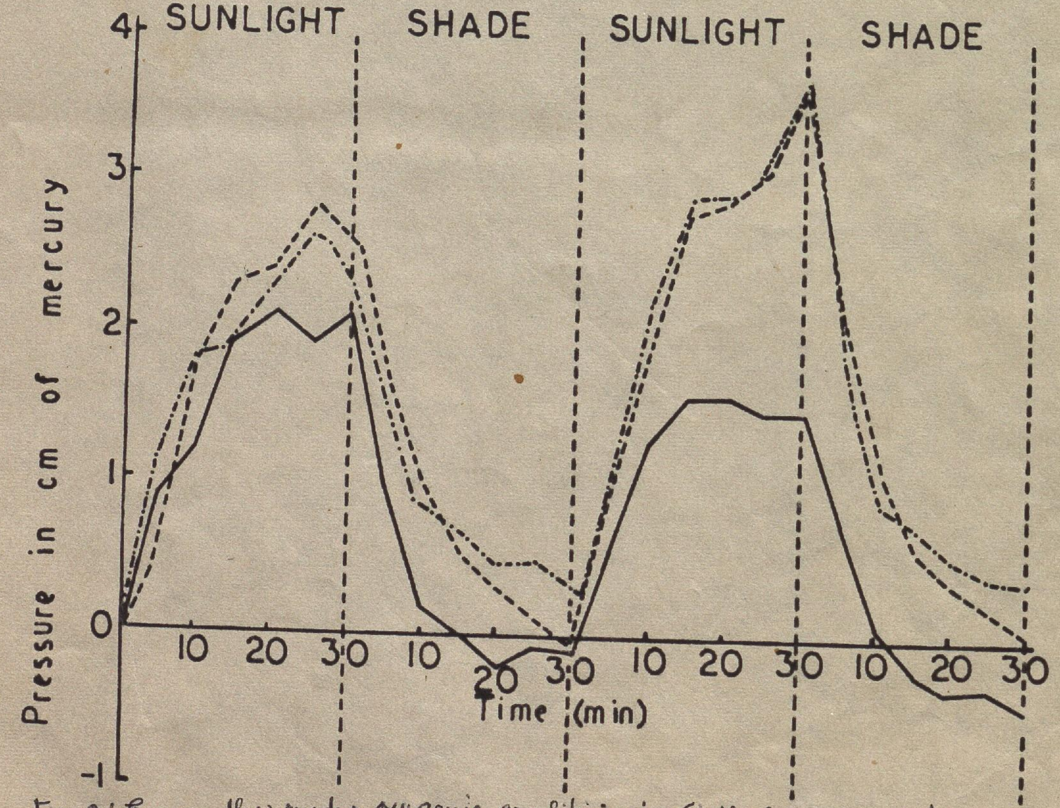

Fig. 2: Energy flow under oxygenic condition in Si-Mo Jeewanu autoorganised under oxygenic condition, as indicated by pressure increase in cm. of mercury due to splitting of water in sunlight and decrease caused by fixation of nitrogen.

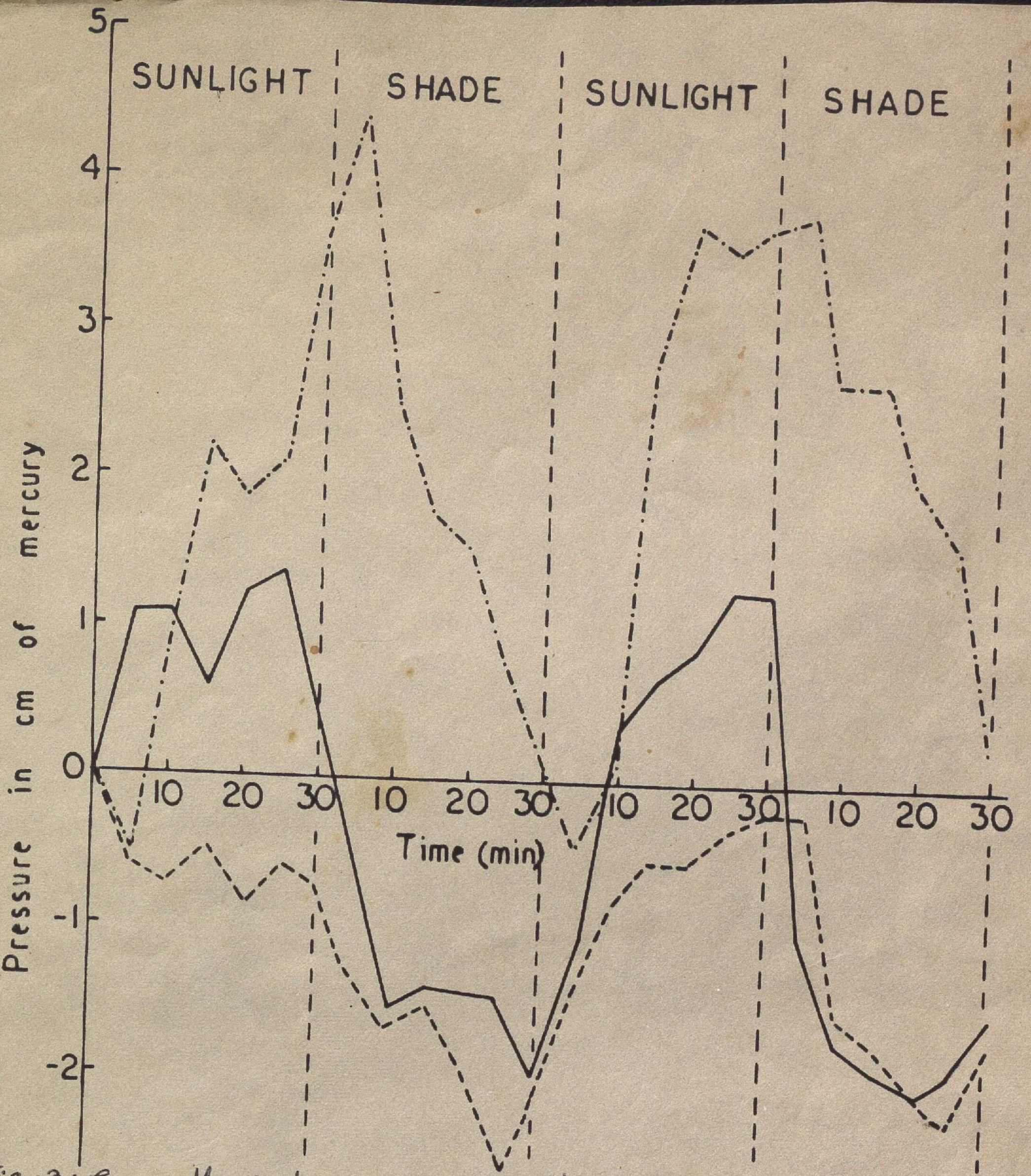

Fig. 3: Energy flow under anoxygenic condition in Si-Mo Jeewanu autoorganised under anoxygenic condition, as indicated by pressure increase in cm. of mercury due to splitting of water in sunlight and decrease caused by fixation of nitrogen.

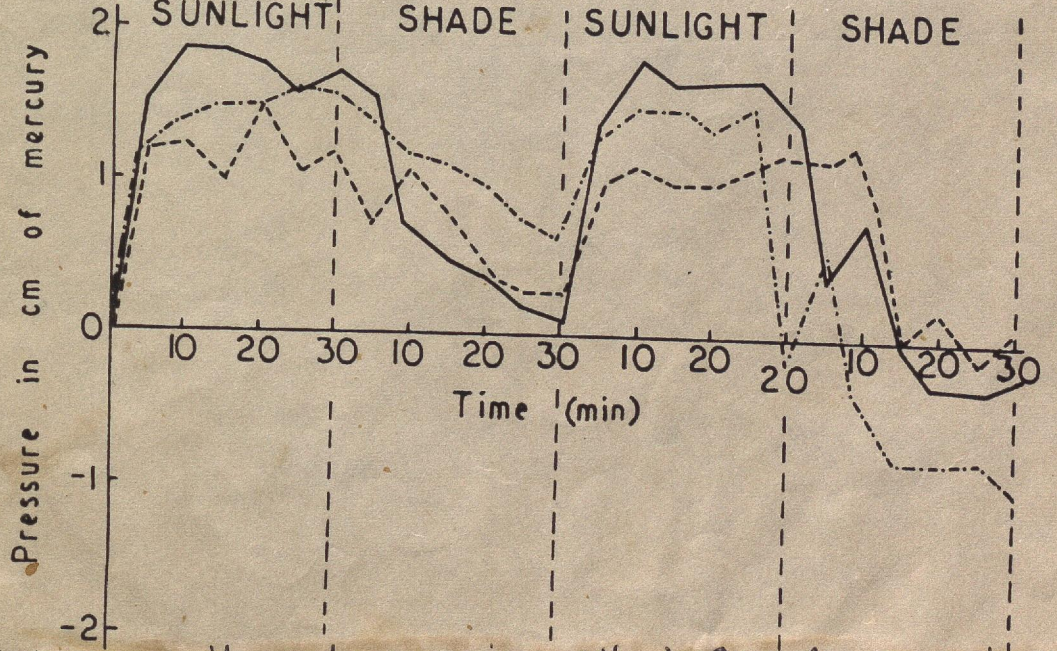

Fig. 4: Energy flow under oxygenic condition in Si-Mo Jeewanu autoorganised under anoxygenic condition, as indicated by pressure increase in cm. of mercury due to splitting of ~~nitrogen~~ water in sunlight and decrease caused by nitrogen fixation.

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Formation of JEEWANU, the molecular association with properties of biological order *

By

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The short history of the modern approach to the problem of Origin of life by a process of molecular evolution has been adventurous. There had been satisfactory progress in abiogenesis of compounds of biological order and also in the study of self-duplicating systems and in the investigations of the molecular associations with the properties of biological order. The first phase of abiogenesis was the search of the natural processes which could form compounds of biological interest as amino acids, peptides, purines and pyrimidines. Many processes effecting the synthesis of these compounds abiogenically were discovered. All the processes which were used to effect these syntheses might have helped in the formation of these substances in prebiological era to some extent. However one needs to differentiate between those factors which were present on the Earth for considerable length of time and those which were only of phenomenal types, if a some what clear picture of the major factors controlling the abiogenesis which led to the formation of the first living systems is to be had and it is of great interest in exobiological studies to have at least an estimate of the cosmogany of life in the universe.

Here the chief difficulty is of the remoteness of the period when probably these reactions were taking place and forming the materials with which the earliest living systems were made. It is still difficult to have a correct guess of the physico-chemical conditions which were present at that time. From the very beginning of the Origin of the Earth itself and its atmosphere to the period when living systems made their first appearance, nothing can be said with certainty for which there is not an alternative suggestion. In a condition like this it is best to follow one of the basic principles of science known as Lyell Principle which he suggested in his investigations of geological findings. The rule says that one should consider the past as the backward extension of the present unless there are positive reasons to believe otherwise.

Taking the case of abiogenesis of amino acids starting from the first observation of Loeb in 1913 who reported the formation of amino acids as glycine and alanine by passing silent electric discharge in a mixture of formaldehyde, ammonia and water(1), amino acids have been synthesised by passing electric discharge in mixture of gases(2) and also by exposing a sterilised aqueous mixtures containing organic substances and inorganic catalysts to light(3). Hasselstrom exposed aqueous solutions of ammonium acetate and observed the formation of glycine and aspartic acid(4) and synthesis of amino acids and other compounds of biological interest have been done using isocyanates(5), energy from ultra violet rays(6) to X-ray radiations(7) and other sources of energies have also been used for this abiogenesis.

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* English translation of the original paper in Hindi under print in Vijnan Parisad Anusandhan Patrika, Nov. (1966)

If one evaluates the probability of the availability of these agencies of energy in the prebiological era, the presence of water excludes the possibility of appreciable abiogenesis by short ultra violet and other short radiations, for the ozone layer of the Earth would have soon cut off their supply on the surface of the Earth. Water appeared early on the Earth and its presence also excludes the high temperature processes. In presence of water these processes might have been probable near volcanoes within a very limited zone. Other remaining sources of energy as electrical discharges and others must have been phenomenal. Organic molecules and inorganic catalysts were present in abundance in the oceans of the primitive Earth and sunlight was available since the very beginning of the Earth. So the photochemical processes apear to be the major contributors of not only the amino acids but also for the formation of peptides and other molecules of biological importance as descussed later.

The next important step in abiogenesis was the formation of peptide linkage. Fox (8) synthesised peptides by heating a mixture of amino acids to 180°C for a few hours. Akabori(9) synthesised peptides by exposing aqueous mixtures of amino acids to ultra-violet light. Terenin(10) suggested the possibility of effecting the reactions needing quanta of larger amount of energy by radiations as shorter quanta of energy if the substrate molecules are adsorbed on solid substances. Bernal(11) reported the formation of peptides by the radiations available on the Earth in the mud.

In 1958 Bahadur and Ranganayaki observed the formation of peptides in aqueous mixture of amino acids containing colloids of iron and molybdenum oxides as catalyst on irradiation with sunlight or artificial light from an electric bulb(12). The photochemical formation of peptides in aqueous mixtures has been investigated in detail(12). Perti and Pathak (13) observed the formation of peptides in aqueus mixtures using visible light and ultra-violet light in presence of inorganic catalysts. Briggs confirmed the observation on photochemical formation of peptides(16). The synthesis of peptides in aqueous mixtures using hydrogen cyanide as the dehydrating agent has been observed by Lowe(14).

The photochemical formation of peptides in aqueous mixtures is interesting and becomes important because put together with the observations of photochemical formation of amino acids, it is quite probable that first amino acids were formed in the oceans of the primitive oceans and then these were subsequently utilized for the formation of peptides.

The abiogenesis of purines, pyrimidine, adenosine and nucleotides have been studied by a number of scientists.

The next important step was the formation of molecular associations with characteristics of living system. Though there appears considerable difference in the difinition of a living system, if a system has properties of growth, multiplication and metabolic activity, where growth is the increase in size of the system from within by the actual synthesis of the material with which the system is made, multiplication is the increase in the number of the system where newer systems come in existence through the parent one and metabolic acitvity is any series of chemical reactions within a boundary the result of which is that at least a part of the environmental molecules entering the system is converted into the material with which the units are made and if such a system shows mutational and adaptive changes, this system can be admitted to the category of living systems. Though these properties do not demark the line of separation between the living and non-living systems and these include properties which are of systems which are more towards living side and living systems with lesser properties than these may be possible, it can be safely said that if a system has all these properties it will be considered as a living system (19,21).

A few earlier efforts of synthesis of the molecular associations with properties of biological order are well known. Oparin has reviewed this work (15). In these experiments an attempt was made to duplicate the morophology of cells by the interaction of simple inorganic and organic substances. Referring to these particles Briggs(16) writes, "While there is no doubt that the products obtained by many of these workers do bear a morphological resemblance to living cells this is the only feature in common, in that the products are dissimilar in chemical composition are metabolically inert, do not grow or reproduce etc. Moreover, most of these artefacts are produced from substances and under conditions that were probably quite absent from the primitive Earth-".

Coacervates appear to have boundary wall and divide by fission. However, they lose their shape on centrifugation and do not have a definite shape. Microspheres are made of solid and brittle substance and break radially on pressing. They retain their shape for a few hours only and then coalesce forming clumps at the bottom of the mixture. Upto 1963 when the work on JEEWANU was first published, it had been observed that microspheres have boundary wall, their contents dissolve on adjusting the hydrogen ion concentration of the environmental medium and they appear to divide by binary fission.

The difficulty about these experiments was that it was not clear as how the nucleic acid which has the properties of duplication and which controls the sequence of amino acid in protein molecules and thus is the essential compound in all biological systems, would have come in function to begin with, because the enzymes necessary for its activity could have been themselves formed by its activity only.

Appearance of propeties of duplication and adaptability in abiogenic molecular associations thus becomes a very important aspect of biopoesis. A few earlier theoretical considerations of this aspect may be of interest. Henderson in his book "Fitness of Environment" writes "the subsequent complex systems show the inherent properties of the simple systems already in existence. Wald (17) writes, that the cell was formed because of the inherent properties of the matter constituting it. In 1963 Bahadur and Ranganayaki (18) suggested that matter has inherent properties of duplication and adaptability and under appropriate conditions any system of matter will show these properties. A system of matter in equilibrium has inherent property of adjusting itself in a way as to nullify the mild constraints caused on the system. According to Bahadur (19) the quantum mechanical resonance special stability force (20) and Le Chatelier's Principles are the expressions of the inherent properties of duplication and adaptability of the matter respectively. Thus on quantum mechanical resonance consideration a system AA is more stable than another system AB where A and A are identical and A and B are similar but not identical. Thus in a system at dynamic equilibrium where there is a possibility of the formation of n molecular structures all nearly at the same thermodynamic level and needing about the same amount of energy of activation, if a molecular structure A, one of the n possible synthesis/able molecular structures, is introduced more of A will be formed in the system than other possible molecular structures. A living system is a system at equilibrium. If we substitute the term " a system in equilibrium" with " a living system" then Le Chatelier's Principle will read, "If a living systme is subjected to a constraint a change occurs in the system if possible of such a type that the constraint is partially annulled" But this is how adaptability in living systems can also be explained.

If matter has inherent properties of duplication and adapatbility it is only the question of creating the suitable conditions when a system of matter will start showing the properties of growth, multiplication and metabolic activity. These systems have been prepared and have been named as JEEWANU (20). In photochemical series the constituent substances were synthesised photochemically and Jeewanu having the properties of growth, multiplication and metabolic activity have been prepared using light as the source of irradiation(21). Thermal peptides have also been used for the preparation of Jeewanu (22). Jeewanu have also been prepared from non-proteinous matter. These contain copper, molybdenum, carbon, hydrogen, oxygen, nitrogen and biological minerals but no proteins. These are basically inorganic in composition with an ash content of about 88 percent (23,19). Photomicrographs of the internal structures of Jeewanu have been taken (23).

The work on photochemical formation of Jeewanu has been confirmed by Briggs (16). The phenomenon of growth, multiplication and metabolic activity has been recently observed by Fox in his particles microspheres prepared from thermal peptides, as reported in the contribution No. 050 of the Institute of Molecular Evolution, Miami University, Miami, U.S.A., in 1965 using different procedures than employed in the preparation of Jeewanu from the thermal peptides. Comparative study of the Jeewanu prepared from thermal peptides in 1963 and the microspheres with the above properties as reported in 1965 is as follows:

Table No. 1. Similarities in the properties of Jeewanu prepared from thermal peptid es - ammonium molybdate complex as observed in 1963 (18,22) and in microspheres prepared from thermal peptides which were discovered from 1963-1965 as reported in 1965 (24).

S.No.	Observations made on Jeewanu prepared from thermal peptides - a mmonium molybdate complex as reported in 1963 (18).	Observations made on microspheres prepared from thermal peptides from 1963 onwards and reported in 1965 (24).
1.	Experiments discribing the growth and multiplication by budding reported. Jeewanu multiply by budding and show growth.	On page number 10 of the cyclostyled copy of the report mentioned above time lapse photomicrographs of separation of daughter fragments is given. On page 12 time-lapse photomicrographs of a bud getting formed and increasing in size has been shown.

S.No.	Observations made on Jeewanu prepared from thermal peptides-ammonium molybdate complex as reported in 1963(18)	Observations made on microspheres prepared from thermal peptides from 1963 onwards and reported in 1965(24).
2.	It has been reported that the Jeewanu have characteristic motility.	On page No. 13 and 14 of the above paper timelapse photomicrographs showing motility produced.
3.	The data showing different amino acid patterns of the peptides of the Jeewanu body material and the peptides in the envirnmental medium of Jeewanu used as nutrition given.	On page 8 of the above paper the difference in the amino acid patterns of the peptides in microspheres and the peptides in the boundary given.
4.	Whirling motion reported in Jeewanu.	It is reported that the particles rotate.
5.	It was suggested that matter has inherent properties of duplication and adaptability and therefore it is possible to prepare particles showing growth, multiplication and metabolic activity.	On page 15 it is reported "such developments are consistent with a general prediction made by George Wold in 1954 that the origin of cell eventually be understood as due to internal properties of the matter of which it was composed".
6.	It has been reported that Jeewanu are similar to "Organised elements" in morphological appearance.	On page 3 resemblance of microspheres with Organised elements in morphology suggested.

However there are basic differences between Jeewanu and microspheres. These differences are as tabulated below.

Table No. 2: Difference between Jeewanu and Microspheres.

S.No.	JEEWANU.	S.N.	Microspheres.
1.	Jeewanu are prepared from a variety of substances of organic and inorganic natures. Jeewanu are also prepared from thermal peptides - ammonium molybdate complex.	1.	Microspheres are prepared from thermal peptides only.
2.	The properties of growth, multiplication and metabolic activity have been observed, all in the same environmental medium, without introducing any chemical to affect any of these specific properties.	2.	Separation is observed on addition of magnesium chloride, motility when ATP is introduced.
3.	Inside meterial remains viscous.	3.	Inside material is brittle and solid.

S.N. JEEWANU.	S.N. Microspheres.
4. Retain their shape indefinitely.	4. Coalesce after 24 to 48 hours.
5. Jeewanu show mutational and adaptability changes and are capable of evolution and thus have all the properties necessary to be admitted to the category of living systems.	5. Microspheres are artefacts and not living.

Thus except for the fact that thermal peptides have been used for the preparation of a type of Jeewanu and thermal peptides are the basic building material of microspheres, there are fundamental differences in these two types of particles.

According to Bahadur Jeewanu which have properties of growth, multiplication and metabolic activity unaided by any additional chemical to effect any of these properties like microorganisms, can be subcultured and show mutational adaptability, were in all probability the first living systems, the precursors of the cellular life. Commenting on microspheres on the page number 15 of the above referred report Fox writes:

"Incidentally, the difficulty of ~~difficulty of~~ defining life is more vividly understood on the basis that various attributes of life might be successively introduced into a simple lifeless system as the model suggested."

Other investigations in our laboratory not presented here, yield phenomenon which can also simulate behaviour otherwise associated with vital processes. These apparently complex units and processes occur in a simple way, as in natural experiments. The products are in a large sense of the word, artefacts, though produced by human hands, the mode of production is imputable to the geological locale."

This appears to be a sound statement about microspheres for the various properties which are observed in a living system can be observed in artefacts by creating suitable physico-chemical conditions. Thus a drop of rain water breaks up into innumerable droplets as it falls down from the sky and is not multiplication in biological sense. A gel particle swells up by hydration and it is not biological growth and many chemical reactions take place within a boundary and these cannot be called as metabolic activity. So far these so called biological properties are observed in single or in twos in a system and that too after some physico-chemical factor is artificially introduced in the environmental medium of the system, these functions have nothing in common with biological functions. And it has been observed that many artefacts have a morphological appearance.

However (i) if a system grows in an environmental medium by the actual synthesis of the material, with which it is made, within the system from the environmental substrate molecules entering the system, (ii) after achieving a specific size if the system multiplies and thereby there is an increase in the number of the system by a process where the new systems ~~come~~ in existence through the parent one by itself without the aid of any additional chemical for this process (iii), if a series of reactions is taking place within this system resulting in the synthesis of the material of the system and providing the energy need, (iv) if these systems can be subcultured in their specific environmental medium like micro-organisms and (v) if these systems show mutational and adaptive changes and are thus capable of evolution, such a system will be admitted to the category of living system whatever may be accepted as the definition of life or may be considered as basic properties of living system. Jeewanu have these properties. After all, as Morrison (25) emphasised, life must have arisen from some kind of artefact and the important question is from which kind of artefact and how was it produced? Synthesis of Jeewanu provides answer to this question. The work on photochemical formation of Jeewanu has been recently confirmed by Briggs(16).

It appears that the phenomena of growth, multiplication and metabolic activity can be observed in a number of particles. Observation of growing and multiplying of elements with metabolic activity has been made in an entirely different environment by Gumpert(26) who writes,

"I am very much interested in your working field firstly pure theoretically for the problem of origin of life is very important for microbiologists too. The first organisms have been surely "bacteria - like". On the other hand we have now studied a problem which seems to be very common. In bacteriological media with sera, as they are used for culturing PPLO and L-forms as well as in serumconserves as they are used in medicine, spherical elements are formed which are not to be distinguished in the phase-contrast-microscope from L-forms and PPLO: These elements seem to propagate by means of budding, seem to have 'metabolism' but are not (terrestrial) living organisms. In this connection your results are very interesting for me."

The molecular mechanism resulting in the biological type of growth, multiplication and metabolic activity has been suggested(19). Recently Gabel (27) has suggested a probable mechanism of growth and multiplication of a polymeric molecular pattern with basic polyphosphate skeleton. Whether such growth, multiplication and metabolic activity can be compared to what is observed in biological system has been mentioned above and is discussed in details by Bahadur(19) In the description of the essential properties which if present together in a system may entitle it to be admitted to the category of living, adaptability has not been mentioned because adaptability has been suggested as the inherent property of all the systems in equilibrium and a living system is a system at dynamic equilibrium. Jeewanu have shown mutational changes, quick changes leading to drastic alteration in appearance and morphology and adaptive changes, viz slow changes caused by mild changes in the environment. Thus Jeewanu have been trained to utilise organic sources as their carbon nutrition different from what they were using in the beginning(23). These can be subcultured in their specific media (21,22). Thus though no effort has been made to draw a line of demarkation between living and non-living systems which appears to be a hopeless job, Jeewanu have been found to show properties of accepted biological systems, i.e., they grow, multiply by budding, have metabolic activity, show mutational and adaptability changes can be subcultured and thus are capable of undergoing evolution.

SUMMARY.

The abiogenic approach of the problem of origin of life has been discussed and the importance of the photochemical reaction taking place in aqueous mixtures, having inorganic

catalysts with large surface areas as in colloidal conditions is stressed. Amino acids and peptides are formed in these mixtures. It has been suggested that matter has inherent properties of duplication and adaptability and under suitable conditions these help in the formatio of molecular associations with properties of growth, multiplication and metabolic activity. Such molecular associations have been synthesised by several processes. These have been named as Jeewanu. Jeewanu grow by utilising the nutrition from the environmental medium, after reaching a specific size they multiply by budding, they show mutational and adaptive changes and are capable of evolution,. Jeewanu can be subcultured in their specific media. Jeewanu satisfy all the essentials for a living system but have been synthesised from non-living materials. Jeewanu can be synthesised under natural conditions and were thus in all probability the immediate precursors of cellular life.

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