A Computational Study of the Prebiotic Photochemical Synthesis of L-Ascorbic Acid, and D-sugars: Glucose, Amylose and Cellulose.

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Abstract: - Ab initio applied computing is used to determine a viable and plausible mechanism for the formation of L-ascorbic acid, and D-sugars: glucose, amylose and cellulose. from planetary or interstellar gases that contain the necessary essential elements. Each compound is formed from carbon monoxide, hydrogen and water. The surface catalyst for the formation of the monosaccharides and polysaccharides is taken to be the photochemically activated surface catalyst of the metalloporphyrin, ferrous porphin, itself formed from diacetylene cyanide. The bonding of the carbon monoxide to form a hexadentate Fe. porphin, may produce each of the monosaccahrides, whereas the formation of a glucose furanoside susceptiple to polymerization enables the formation of both α(1-4) amylose and β(1-4) cellulose. The intermediates for the formation of these residues are characterized by their reaction enthalpies and activation energies. The reactions have been shown to be feasible from the overall enthalpy changes in the ZKE approximation at the HF and MP2 /6-31G* level, and with acceptable activation.

Key-Words: - Prebiotic photochemical synthesis, L-ascorbic acid, D-glucose, D-amylose,, D-cellulose, Fe.porphin

Received: March 9, 2025. Revised: May 11, 2025. Accepted: June 3, 2025. Published: July 9, 2025.

1 Introduction

The early chemistry of the Earth provides both the geochemistry, [1] and the prebiotic formation of the biological molecules. D-glucose is the most abundant hexose, the monosaccharide from which the oligosaccharides and polysaccharides are derived. It is the major fuel for most organisms and the basic building block of starch and cellulose, [2]. Starch is the main form of fuel storage in most plants. It occurs in two forms, α-amylose and amylopectin. Amylose consists of long unbranched chains in which all the D-glucose units are bound in α(1-4) linkages, Fig.4. Amylopectin has a backbone of glycosidic linkages α([1-4), [3] but branched with some α(1-6) linkages. Cellulose is the most abundant cell-wall and structural polysaccharide in the plant world, a linear polymer of D-glucose in β(1-4) linkage, Fig.5. It is a major component of wood.

L-ascorbic acid, or Vitamin C, is the γ-lactone of a hexonic acid having an enediol structure at carbon atoms 2 and 3, [4], [5], Fig.6. It is unstable and readily undergoes oxidation to dehydroascorbic acid.

From a prebiotic perspective, [6], [7] it is desirable if these molecules formed spontaneously from a supposed prebiotic atmosphere to be plausibly present as micellular walls of rudimentary cells. It has been often held that the atmosphere of the Earth was originally mildly reducing, [2], [6] implying the presence of concentrations of carbon monoxide and formaldehyde, [7], the partially reduced substrates of modern photosynthesis. It has also been demonstrated that porphin, [8] may act as a catalyst for the formation of sugars, [9] and polyenes, [10].

This paper proposes a model for the catalytic formation of D-sugars using the diamagnetic response of the catalyst with adducts to photochemical excitation where the catalyst, designated Fe.porphin has six carbon monoxide adducts as charge transfer complexes. The bonding in such a complex is highly versatile and may account for the prebiotic formation of D-glucose and the ubiquitous polysaccharides amylose and cellulose. The reactions described have been deduced as kinetically and thermodynamically viable, but photochemical excitation is required to form the saccharide backbone.

2 Problem Formulation

The computations tabulated in this paper used the Wavefunction, [11] and GAUSSIAN98 [12] commercial packages. The standard calculations at

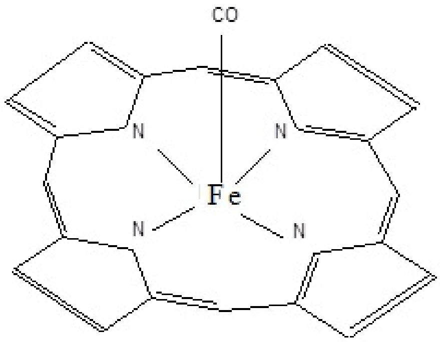
the HF and MP2 levels using the same basis set, 6-31G*. include zero-point energy corrections that are scaled by a factor of 0.9, [11], [13]. The charge transfer complexes are less stable when calculated at the Hartree Fock level, [14] and activation energies at the HF level of accuracy without scaling are less accurate. If the combined energy of the products is less than the combined energy of the reactants it may show that the reaction is also likely to be spontaneous at higher temperatures. This paper uses the atomic unit of energy, the hartree, [12]. Charges are in units of the electronic charge.
1h = 627.5095 kcal.mol^{-1}.
1h = 4.3597482 x 10^{-18} J

3 Problem Solution

3.1 Total Energies (hartrees)

Metal.porphins may form a variety of complexes, [15], [16]. Carbon monoxide may chelate with the ferrous ion of ferrous porphin, [17], [18], [19],[20], [21], [22] which has been taken as a possible catalyst, to form a charge transfer complex where the charge on the ligand is positive and the charge on the porphin molecule is negative. The enthalpy of formation of the complex is small but it appears stable [9]. The Mg.porphin catalyst has five sites used to form charge transfer complexes enabling the formation of pentoses [8], [9]. The Fe.porphin catalyst can accommodate six charge transfer adducts and thus lead to the hexoses. Here the complexes are neutral without unpaired electrons, and the molecule is assumed to have D_{4h} symmetry, [17], [18]. The following charge transfer adducts are essential to form the initial reactant for hexose synthesis, and are the counterparts of those previously detailed, [9].

Fe.porphin + CO →
(1)

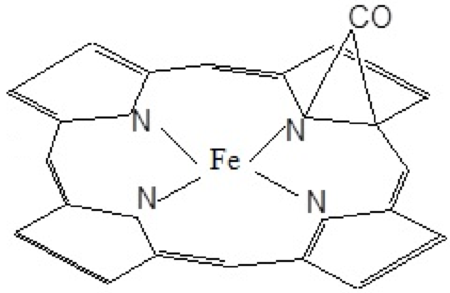


Fe.CO.porphin (2)

Δ H = -0.08259 h

The charge on the adduct of the neutral complex is -0.023
If the Fe.porphin (triplet state) is in the triplet state, the enthalpy change is less favourable.
It is postulated that when the complex becomes excited from photolysis the higher energy complex is formed according to the equation,

Fe.porphin + CO →
(1)



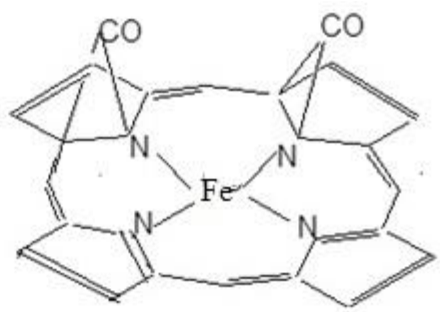
Fe.porphin.CO (3)

Δ H = 0.22846 h

The charge on the adduct is -0.187.
The CO group is able to move through a transition state to the porphin ring where it forms an excited, but stable bridged aziridine-2one ring [9], [15] ,16] at a pyrrole unit. The excitation energy is more than the activation energy for the transformation [9].

This process of binding a carbon monoxide molecule to the ferrous ion and then migrating the CO group to the periphery may be repeated several times.
The following are further adducts needed to form the first adduct complex containing six bound carbon monoxide molecules.

Fe..porphin. + 2 CO →



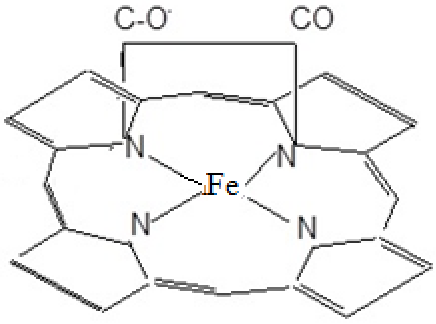
Fe.porphin.2CO (4)

ΔH(= 0.09656 h

The charge on the adducts is 0.168.
Further excitation of this complex in which both CO groups occupy peripheral ring positions may cause the adducts to bond, indicated as Fe.porphin.$(CO\text{-})_2$.

The enthalpy change to form the carbon-carbon bond is low as shown,

Fe.porphin + 2CO $\rightarrow$

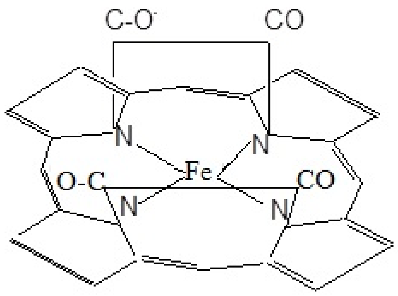


Fe.porphin.$(CO\text{-})_2$ (5)

Δ H = 0.14542 h

The charge on the adduct is -0.23461.
The activation energy to form the carbon-carbon bond is, 0.049 h, when calculated at the HF level by reducing the -CO-CO- bond length.
The formation of this di-adduct may be repeated for two more carbon monoxide groups to be added to the third and fourth peripheral positions and bonded, where the overall reaction is,

Fe.porphin.$(CO\text{-})_2$ + 2CO $\rightarrow$
(5)



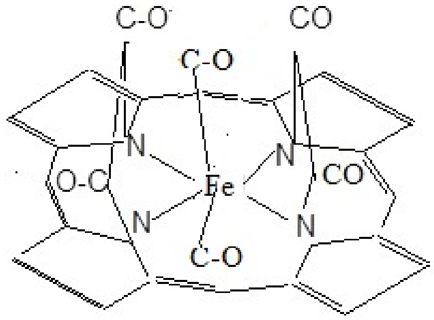
Fe.porphin.$(CO\text{-})_2$.$(CO\text{-})_2$ [6]

Δ H = 0.04230 h

The charge on the adducts is 0.526.

Finally the ferrous atom of the uncharged Fe.porphin .$(CO\text{-})_2$.$(CO\text{-})_2$ may accept two more carbon monoxide molecules as carbonyl adducts, as,

Fe.porphin.$(CO\text{-})_2$.$(CO\text{-})_2$ + 2CO $\rightarrow$
(6)



Fe.2CO.porphin.$(CO\text{-})_2$.$(CO\text{-})_2$ (7)

Δ H = -0.13600 h

The charge on the adducts is 0.150

As shown. This will be taken as the initial reactant for the formation of all the D-hexose pyranosides to be described here. The sum of the preceding chelation reactions may be given as,

Fe.porphin + 6CO $\rightarrow$ Fe.2CO.porphin.$(CO\text{-})_2$.
$(CO\text{-})_2$

Δ H = 0.05172 h

This complex may easily bond to form the backbone of a large variety of D- and L-hexoses, such as D-allose.

Fe.2CO.porphin.$(CO\text{-})_2$.$(CO\text{-})_2$ $\rightarrow$ Fe.porphin.
$(CO\text{-})_6$
Δ H = 0.22865 h

This highly reactive and unstable hexadentate complex has gained considerable energy from photolysis.

Fe.porphin + 6CO $\rightarrow$ Fe.porphin.$(CO\text{-})_6$
(8)
Δ H = 0.28043 h

These high energy compounds may react further to form adduct epimers, or relax to form the more stable van der Waals complexes.

The data for some of the more stable of these molecules are given in Table 1, where peripheral carbon monoxide molecules that are fully bonded are represented as Fe.porphin.$(CO-)_n$, etc.

The total energies and zero point energies for the HF and MP2/6-31G* equilibrium geometries are given in Table 1. ZERO point energies are scaled by 0.9.

Table 1
MP2 /6-31G* total energies and zero point energies (hartrees) for the respective equilibrium geometries

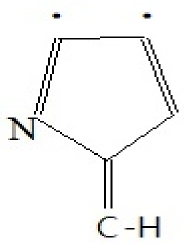
Molecule	MP2 hartree	ZPE (HF) hartree(x0.9)
Fe.porphin (1)	-2247.73860	0.28713
Fe.CO.porphin (2)	-2360.83651	0.29615
Fe.porphin.CO (3)	-2360.53061	0.30130
Fe.porphin.2CO (4)	-2473.67405	0.30655
Fe.porphin.$(CO-)_2$ (5)	-2473.62808	0.30944
Fe.porphin.$(CO-)_2$.$(CO-)_2$ (6)	-2699.61503	0.32608
Fe.2CO.porphin.$(CO-)_2$.$(CO-)_2$ (7)	-2925.78075	0.34325
Fe.porphin.$(CO)_8$ (8) (for allose)	-2925.55185	0.34300
diacetylene cyanide (9)	-245.00678	0.03758
2-methinyl 4,5-didehydro pyrrolenine (10)	-244.79501	0.03700
octa-dehydro dehydroporphin (11)	-979.78567	0.17170
dehydro porphin (12)	-985.14611	0.28801
porphin (13)	-986.48502	0.31154
L-ascorbic acid (14)	-682.80242	0.14976
D-glucose (15)	-685.15052	0.21165
D-amylose dimer (16)	-1294.13940	0.40341
D-cellulose dimer (17)	-1294.13659	0.40388
Fe.$(CO-)_2$.porphin.$(CO-)_2$ $(CO-)_2$ (18) (for ascorbic)	-2925.42161	0.33862
Fe.$(CO-)_2$.porphin.$(CO-)_2$.$(CO-)_2$ (19)	-2925.40006	0.34909
Fe.(CO-)(C-O-).porphin.$(CO-)_2$.$(CO-)_2$ (20)	-2925.38591	0.34118
D-glucose furanoside (21)	-685.16294	0.21305
CO	-113.02818	0.02188
H_2	-1.14414	0.00931
H_2O	-76.19924	0.02068
Fe	-1262.16244	
Fe^{++}	-1261.35698	
OH^{-1}	-75.51314	0.00797
H^- (HF)	-0.49823	
methoxide^{-1} (HF)	-114.38447	

3.1.1 Total Energies (hartrees)

The synthesis of the catalyst Fe.porphin

A prebiotic synthesis of porphin from 2-methene pyrrolenine has involved the simple gases, diacetylene, carbon monoxide, and ammonia presumed to be present in prebiotic times, [8]. However, if hydrogen cyanide was prevalent, a similar synthesis may have involved diacetylene cyanide, [23], which forms a stable high energy cyclic compound with charges conducive to form a cyclic tetramer, as shown,

H-C≡C-C≡C-CN →
(9)

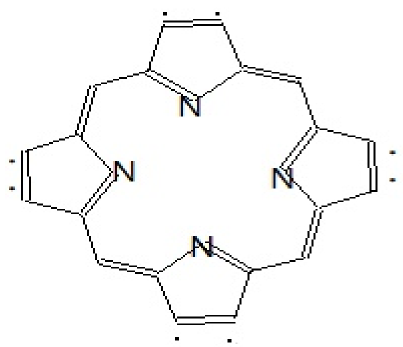


2-methinyl 4,5-didehydro pyrrolenine (10)

Δ H = 0.21120 h

The formation of a cyclic tetramer is as shown,

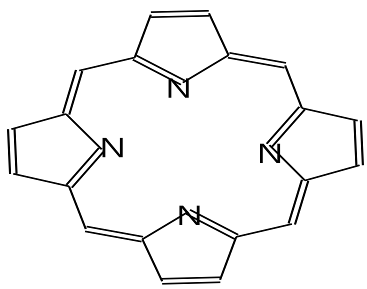
2-methinyl 4,5-didehydro pyrrolenine →



octa-dehydro dehydroporphin (11)

Δ H = -0.58194 h

This cyclic porphin may easily hydrogenate as, octa-dehydro porphin + $4H_2 \rightarrow$ dehydroporphin



dehydroporphin (12)

$\Delta H = -0.70480$ h

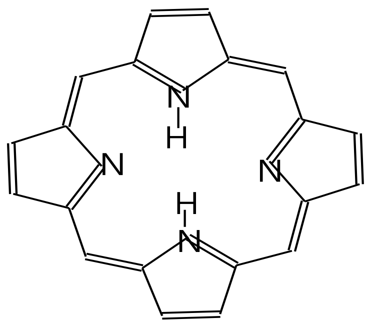
This molecule could react with metallic iron, as,

Fe + porphin $\rightarrow$ Fe.porphin (1)

$\Delta H = -0.43093$ h

However, a more energetic reaction occurs if the porphin first hydrogenates, as

dehydroporphin + $H_2 \rightarrow$ porphin
(12)



porphin (13)
$\Delta H = -0.18055$ h

Disscociation of this porphin with hydroxide anion, followed by insertion of the ferrous ion then yields the desired catalyst, Fe.porphin, as,

Fe^{++} + porphin + $2OH^{-1} \rightarrow$ Fe.porhin + 2 H_2O

$\Delta H = -1.26873$ h

3.2 The formation of chiral saccharides

Previously it has been shown that the photochemical excitation of the A_{1u} to $E_{1g,}$ [24] transition may produce an in-plane electronic transition moment, and concomitantly a magnetic transition moment perpendicular to the ring. Such a transition may impart a diamagnetic response where the higher energy orbital is expected to polarize the molecule to add a hydrogen radical and thereby become fixed as a D-sugar, as shown in Fig.1.

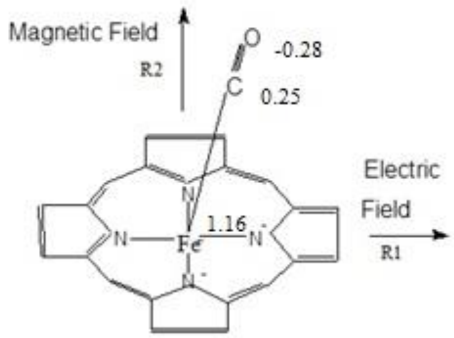


Fig.1. Electrical and Magnetic Transition Moments in a Fe.porphin complex.

For Fe.porphin, the orbital diagrams for the HOMOa and LUMOa have some similarity to the corresponding orbitals for Mg.porphin, as shown in Fig.2 and Fig.3. The adducts may influence the orbitals.

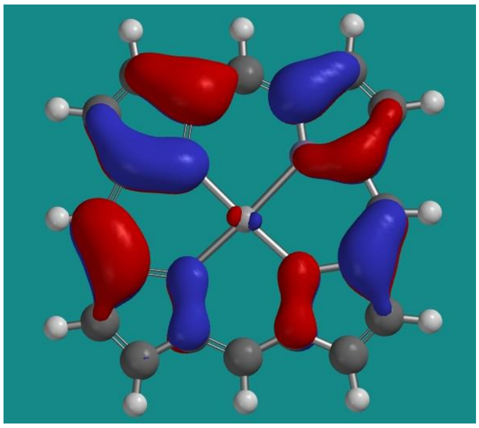


Fig.2. Fe,porphin. HOMOa

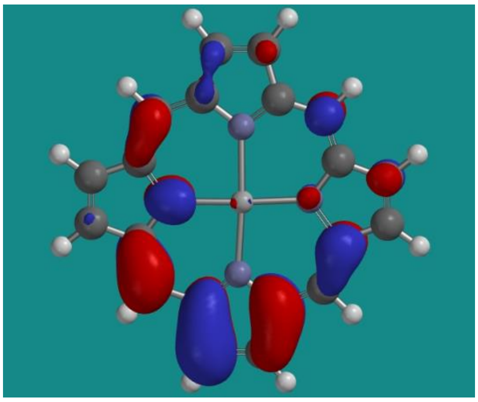


Fig.3. Fe.porphin. LUMOa

For all the complexes the total charge was taken as zero with zero unpaired electrons. The complexes may also exist at other multiplicities, [21].

3.3 The overall stoichiometry for the formation of D-sugars.

Although Fe.porphin is here taken as the catalyst for the reactions, the overall stoichiometry to form the L-ascorbic acid, D-glucose, D-amylose, and D-cellulose are as follows,

$$6CO + 3H_2 \rightarrow C_6.H_6O_6$$
L-ascorbic acid (14)

$$\Delta H = -0.06957 \text{ h}$$

$$6CO + 6H_2 \rightarrow C_6.H_{12}O_6$$
D-glucose (15)

$$\Delta H = -0.09206 \text{ h}$$

$$12CO + 12H_2 \rightarrow C_{12}.H_{22}O_{11} + H_2O$$
D-amylose dimer (16)

$$\Delta H = -0.22093 \text{ h}$$

$$12CO + 12H_2 \rightarrow C_{12}.H_{22}O_{11} + H_2O$$
D-cellulose dimer (17)

$$\Delta H = -0.21764 \text{ h}$$

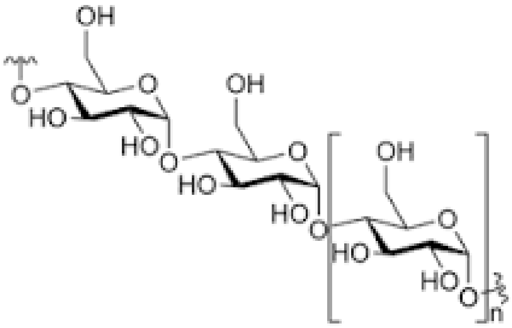


Fig.4 D-α(1-4) amylose.

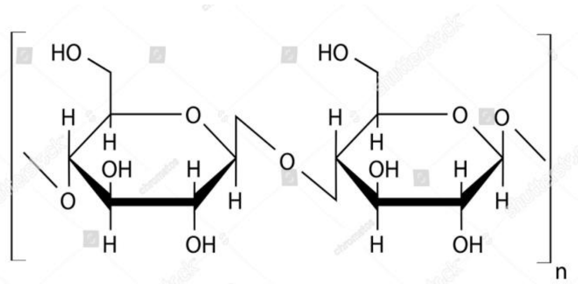


Fig.5 D-β(1-4) cellulose

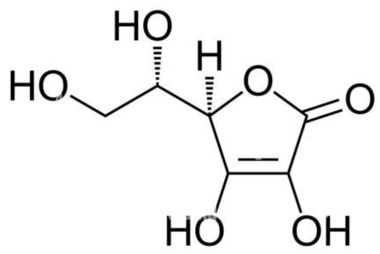


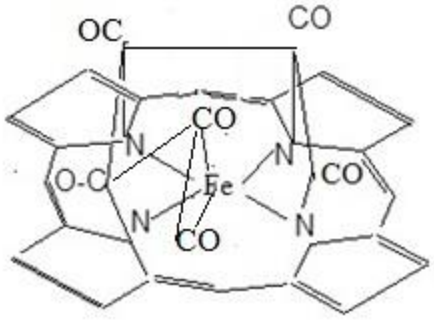
Fig.6 Ascorbic Acid, Vitamin C.

The enthalpy changes are negative indicating that these may be the energetically favourable routes to the initial formation of these hexose monomers and dimers or polymers. The intermediates by which these stoichiometric reactions may have occurred are as follows:

3.4 The formation of L-Ascorbic Acid.

The high energy complex Fe.2CO.porphin.$(CO\text{-})_2$.$(CO\text{-})_2$ (7), may be excited by UV radiation to bond forming a large variety of monosaccarides which are here mainly designated by their number. A prenascent D-ascorbic acid, or L-ascorbic acid may be formed. As the natural Vitamin C is stated as L-ascorbic acid, the bonding here is to form L-ascorbic acid. This does correspond with a diamagnetic response from the adduct complex. Minor variations may produce a large variety of monosacchrides.

$$\text{Fe.2CO.porphin.}(CO\text{-})_2\,.(CO\text{-})_2 \xrightarrow{h\upsilon}$$
(7)



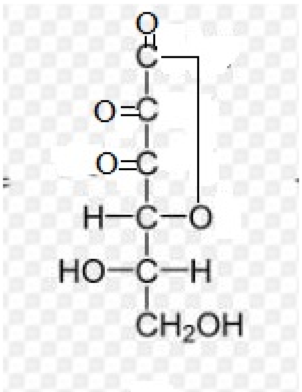
Fe.(CO-)$_2$.porphin.(CO-)$_2$.(CO-)$_2$ (18)

ΔH = 0.35450 h

The charge on the complex is 0.031

The complex may be susceptible to reactant with free radical hydrogen or hydrogen ions from photolysis, [25], as,

Fe.(CO-)$_2$.porphin.(CO-)$_2$.(CO-)$_2$ + 3H$_2$ $\rightarrow$
(18) Fe.porphin +



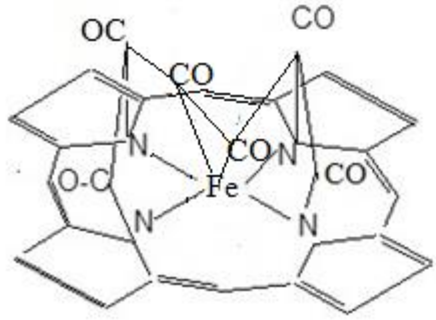
L-Ascorbic Acid Lactone, Vitamin C (14)

ΔH = -0.47585 h

3.5 The formation of D-Glucose

The formation of D-glucose involves another method of bonding, as shown,

$$\text{Fe.2CO.porphin.(CO-)}_2\text{.(CO-)}_2 \xrightarrow{h\upsilon}$$
(7)

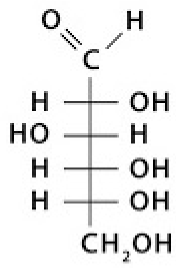


Fe.(CO-)$_2$.porphin.(CO-)$_2$.(CO-)$_2$ (19)

ΔH = 0.38653 h

The D-glucose may be released from the catalyst by hydrogenation as,

Fe.(CO-)$_2$.porphin.(CO-)$_2$.(CO-)$_2$ + 6H$_2$ $\rightarrow$
(19) Fe.porphin +



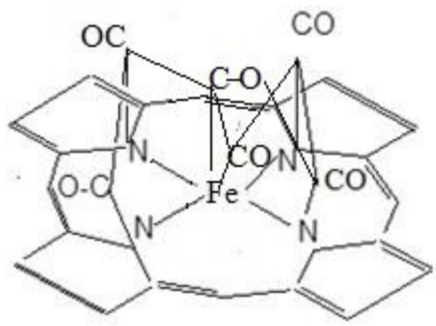
D-glucose (15)

ΔH = -0.53036 h

3.6 The formation of Amylose

To form the polymeric amylose requires that a suitable epimer be formed that can then be initiated to propagate by a nucleophile and then the chain terminated with an electrophile. The bonding here reflects the formation of the monomer, as

$$\text{Fe.2CO.porphin.(CO-)}_2\text{ .(CO-)}_2 \xrightarrow{h\upsilon}$$
(7)



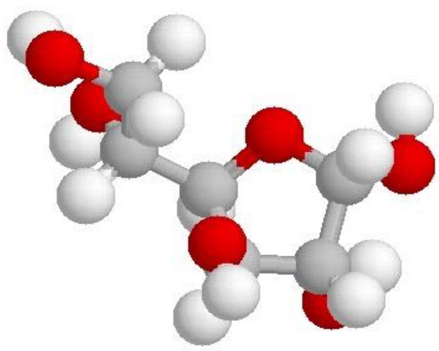
Fe.(CO-)(C-O-).porphin.(CO-)$_2$.(CO-)$_2$ (20)

$$\Delta H = 0.39276 \text{ h}$$

The amylose epimer may be released from the catalyst with hydrogenation, as,

$$Fe.(CO\text{-})(C\text{-}O\text{-}).porphin.(CO\text{-})_2 .(CO\text{-})_2 + 6H_2 \rightarrow \quad (20)$$

Fe.porphin +



D-glucose furanoside (21)

$$\Delta H = -0.54762 \text{ h}$$

This molecule is susceptible to nucleophilic attack and polymerization. The reaction may be simplified and exemplified as the reaction of the nucleophile, methoxide anion at C1 of the monosaccharide, as shown in Fig.7. where the internal coordinates are designated as R1 and R2.

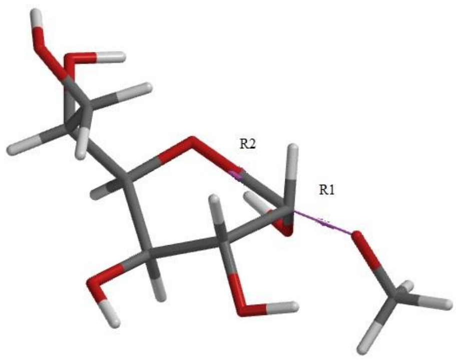
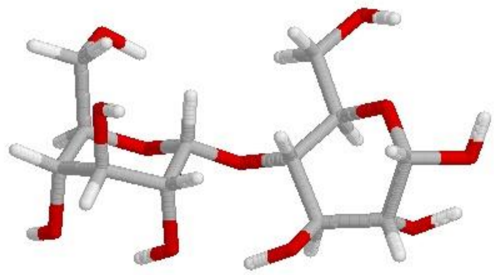


Fig.7. The nucleophilic reaction of methoxide anion on D-glucose furanoside, involving the internal coordinates R1 and R2.

The activation energy at the HF level was found to be 0.0085 h. The furanoside is subject to hydrolysis to open the ring, where the enthalpy change wss found to be, -0.01961 h. Similarly, the closure of the ring to form the pyranoside was found to have an enthalpy change of 0.01617 h.
The formation of amylose dimer formed by hydroxide initiation and proton termination may then be written as,

2 D-glucose furanoside → Amylose dimer + H_2O



α (1-4) Amylose dimer (16)

$$\Delta H = -0.01476 \text{ h}$$

3.7 The formation of Cellulose
To form the polymeric cellulose does require the same D-glucose furanoside initiated to propagate by a nucleophile, and then the chain terminated with an electrophile. However the initiation is at C4, as shown,
This molecule is susceptible to nucleophilic attack and polymerization. The reaction may be exemplified as the reaction of the nucleophile, methoxide anion at C4 of the monosaccharide, as shown in Fig.8. where the internal coordinates are designated as R1 and R2.

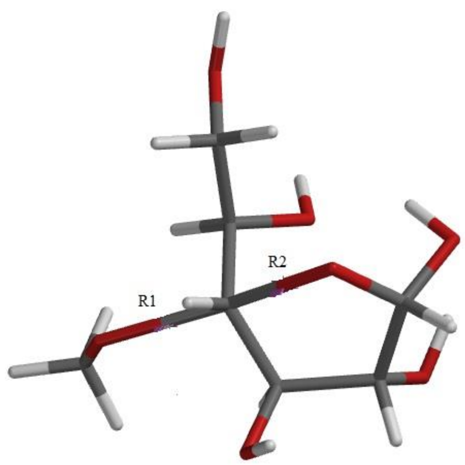
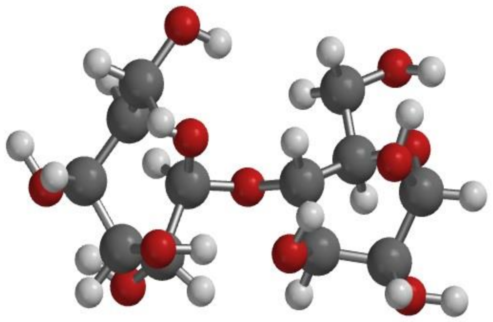


Fig.8. The nucleophilic reaction of methoxide anion on D-glucode furanoside, involving the internal coordinates R1 and R2.

The activation energy at the HF level was found to be 0.0136 h.
The formation of cellulose dimer formed by hydroxide initiation and proton termination may then be written as,

2 D-glucose furanoside → Cellulose dimer + H_2O



β (1-4) Cellulose dimer (17)

Δ H = -0.03924 h

4. Conclusion

The prebiotic synthesis of sugars is usually stated as the oligomerization of the molecule formaldehyde, formed from the reduction of carbon monoxide, activated in aqueous solution by alkali, [6]. However, the predominance of D-sugars enantiomers at the present time does suggest catalysts, as mentioned here. Carbon monoxide is one of the most abundant molecules in the Universe [26], and in condensed phase readily forms both C-C and C-O bonds, [27]. The presence of reactants, carbon monoxide, alkynes and hydrogen in the early Earth's atmosphere together with the catalyst Fe.porphin, suggests that thermodynamically and kinetically viable reactions such as those presented here, could have enabled the synthesis of prebiotic monosaccharides and polysaccharides with photochemical activation. The low activation energy to polymerize glucose furanoside suggests that the molecule or complex could have reacted with any alcohol, amine, or thiol, and be absorbed into a rudimentary prebiotic cell.
Further work at a higher accuracy may alter the values given here.

Acknowledgements

Appreciation is expressed for the use of software from Gaussian Corporation and Wavefunction.

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Contribution of Individual Authors to the Creation of a Scientific Article (Ghostwriting Policy)

The author contributed in the present research, at all stages from the formulation of the problem to the final findings and solution.

Sources of Funding for Research Presented in a Scientific Article or Scientific Article Itself

No funding was received for conducting this study.

Conflict of Interest

The author has no conflict of interest to declare that is relevant to the content of this article.

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