

TITAN'S ORGANIC CHEMISTRY

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ABSTRACT. Voyager discovered nine simple organic molecules in the atmosphere of Titan. Complex organic solids, called tholins, produced by irradiation of simulated Titanian atmosphere are consistent with measured properties of Titan from ultraviolet to microwave frequencies, and are the likely main constituents of the observed red aerosols. The tholins contain many of the organic building blocks central to life on Earth. At least 100 m and possibly kms thickness of complex organics have been produced on Titan during the age of the solar system, and may exist today as submarine deposits beneath an extensive ocean of simple hydrocarbons.

While a case can be made for organic matter on Iapetus (Cruikshank et al., 1983; Bell et al., 1983), perhaps produced by ultraviolet irradiation of methane-containing ices (Squyres and Sagan, 1983), the object in the Saturnian system of greatest interest for organic chemistry is Titan. With a few percent of methane in a mainly nitrogen atmosphere, and abundant sources of high energy photons and charged particles, a complex organic chemistry should be occurring -- and indeed had been anticipated many years ago (Sagan, 1971, 1973, 1974; Khare and Sagan 1973).

In Table I is a recent compilation of the atmospheric constituents discovered on Titan by Voyagers 1 and 2. Nine organic molecules have been detected through the Voyager infrared spectrometer experiment, six of them with triple bonds, and three of them with four heavy atoms. Clearly, there must be a much larger variety of still more complex organic molecules that are present in abundances below the instrumental detectivities. Some idea of the likely abundance of heavier organic molecules in the Titanian atmosphere can be obtained by examining how the relative abundance of other complex nonbiological organics varies with heavy atom number (Fig. 1).

Voyager imaging observations (Smith et al., 1981) show a cloud deck beginning at a planetocentric altitude of about 2825 km, and a detached limb haze at altitudes between about 2850 and 2925 km. In the

Table I.
GASEOUS CONSTITUENTS OF THE ATMOSPHERE OF TITAN

Formula	Name	Abundance
N_2	Nitrogen	≈85-77%
^{36}Ar	Argon	≈12-17% (?)
CH_4	Methane	≈3-6%
H_2	Hydrogen	0.1-0.4%
H_3C-CH_3	Ethane	20 ppm
$H_3C-CH_2-CH_3$	Propane	20-5 ppm
$HC\equiv CH$	Ethyne (acetylene)	2 ppm
$H_2C=CH_2$	Ethene (ethylene)	0.4 ppm
$HC\equiv N$	Methanenitrile (hydrogen cyanide)	0.2 ppm
$HC\equiv C-C\equiv CH$	Butadiyne (diacetylene)	0.03 ppm
$H_3C-C\equiv CH$	Propyne (methylacetylene)	0.03 ppm
$HC\equiv C-C\equiv N$	Propynenitrile (cyanoacetylene)	0.1-0.01 ppm
$N\equiv C-C\equiv N$	Ethanedinitrile (cyanogen)	0.1-0.01 ppm
CO_2	Carbon dioxide	0.01 ppm
CO	Carbon monoxide	60 ppm

Reference: Sagan and Thompson, 1984

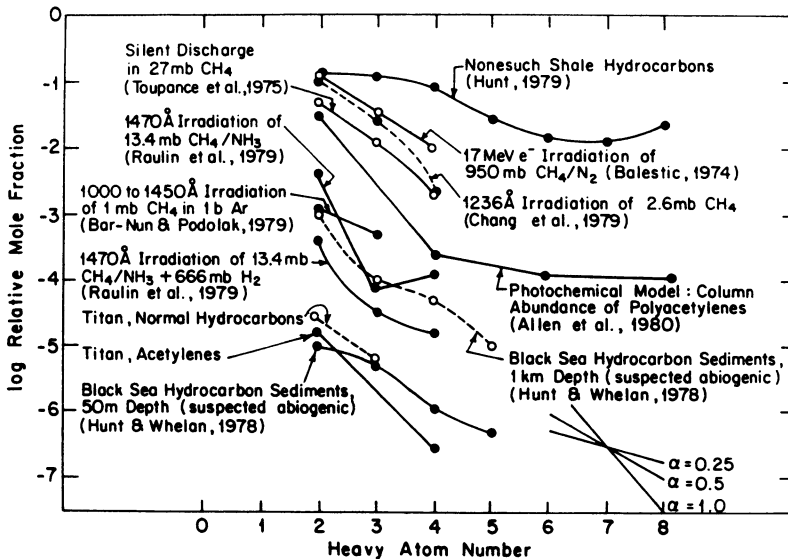


Fig. 1. The abundance of simple organics as a function of heavy atom number for a variety of laboratory simulation experiments, for terrestrial biogenic and suspected abiogenic sediments, and for Titan observations and photochemical models. After Sagan and Thompson, 1984.

3200 to 3600 km planetocentric altitude range there is an additional region of extreme ultraviolet (EUV) opacity (Smith et al., 1982). Fig. 2 shows the altitude and magnitude of energy deposition from the full suite of available energy sources at Titan including electrons and protons in the Saturnian magnetosphere, interplanetary electrons, cosmic rays and solar ultraviolet radiation. We see that none of the energy sources would, in the absence of an antecedent cloud deck and haze, deposit their energy in the 2800–2925 km altitude range. Therefore, the altitude of these aerosols cannot be explained by preferential energy deposition. Preferential condensation of particular atmospheric species is an alternative possibility worth exploring. The EUV opacity region does correspond to a broad maximum in energy deposition rate, produced

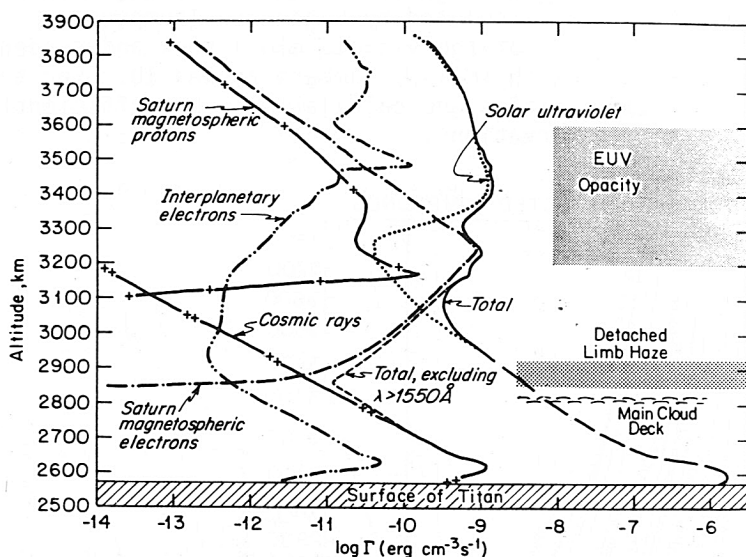


Fig. 2. Variation of disk-integrated, globally averaged energy deposition rate (Γ) with altitude for UV and charged particle energy sources in Titan's atmosphere. Each curve is labeled with the energy source whose deposition rate through the atmosphere it defines. Every energy source shown is sufficient to produce $> 10 \text{ g cm}^{-2}$ of accumulated organic products over geological time. Solar UV absorbed by major atmospheric constituents (primarily $\lambda < 1500 \text{ \AA}$ and above 3200 km), Saturn magnetospheric electrons, and UV absorbed by minor constituents (primarily $\lambda > 1500 \text{ \AA}$ and below 3200 km) are ordinarily the dominant energy sources in Titan's atmosphere. When Titan lies outside the Saturnian magnetopause, the solar wind electron flux can be a factor 20 larger than shown here. The "Total" line is dashed below 2900 km to indicate that, because of scattering and particulate absorption, long-wavelength solar UV will actually be deposited mainly near the top of the main cloud deck, instead of deep in the lower atmosphere as indicated here. (After Sagan and Thompson, 1984).

The clouds of Titan are distinctly red in the visible -- a fact noted from groundbased observations many decades ago (Harris, 1961), and confirmed by Voyager observations. In addition, high resolution spectroscopy of the Titanian clouds exists, as do near ultraviolet spectra from the International Ultraviolet Explorer, groundbased microwave observations, and Voyager infrared continuum spectroscopy. Do the complex organic products of irradiation of simulated Titanian atmospheres have properties appropriate to explain this by now quite elaborate data set?

To approach this question, glass and alkali halide plates were coated by passing a continuous d.c. electrical discharge through a 0.9 N₂/0.1 CH₄ gas mixture at a pressure of 0.2 mb, corresponding to a planeocentric distance of 2825 km: just at the top of the main cloud deck. Through transmittance microscopy, specular reflectance, interferometry, Brewster angle measurements, and ellipsometric polarization techniques, the real (n) and imaginary (k) parts of the complex refractive index of the complex organics produced in these experiments were measured (Khare et al., 1984). The measurements, as displayed in Fig. 5, range from 250 Å in the soft x-ray region to 1 mm

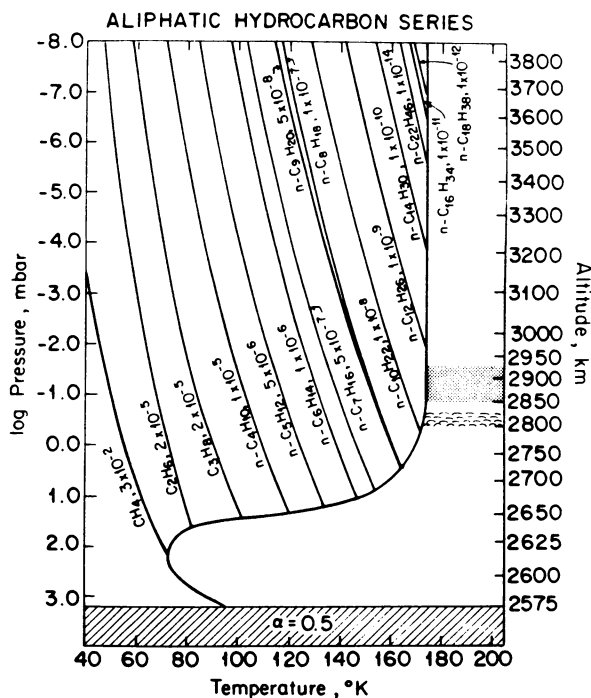


Fig. 4. Vapor saturation temperature profiles for normal hydrocarbons up to C₂₆. This particular calculation is performed for $\alpha = 0.5$, where α is the common logarithm of the abundance change for unit decrease in heavy atom (C + N) number. Detectable condensates at the altitudes of the observed upper cloud hazes can be formed only if $\alpha \leq 0.5$ (cf. Fig. 1). (After Sagan and Thompson, 1984.)

in the microwave region, and apparently represent the most extended analysis of the complex refractive index of any organic material ever performed. The solid organic product produced by irradiation of simulated Titanian atmospheres is a reddish powder that we call Titan tholin. (Tholins are a general class of complex organic solids produced by the irradiation of cosmically abundant, generally reducing, gases.) The steep decline in k from short to long visible wavelengths corresponds to its reddish coloration. Among the other absorption features apparent are the C-H stretching vibration near $3 \mu\text{m}$ and the $\text{C}\equiv\text{N}$ vibration at $4.6 \mu\text{m}$.

Using these optical constants, we have performed two-stream radiative transfer calculations in the UV and visible region (Sagan et al., 1983). A representative calculation is shown in Fig. 6, in which tholin reflectivities are compared with observations of Titan in the visible (Nelson and Hapke, 1978). Because of the presence of abundant

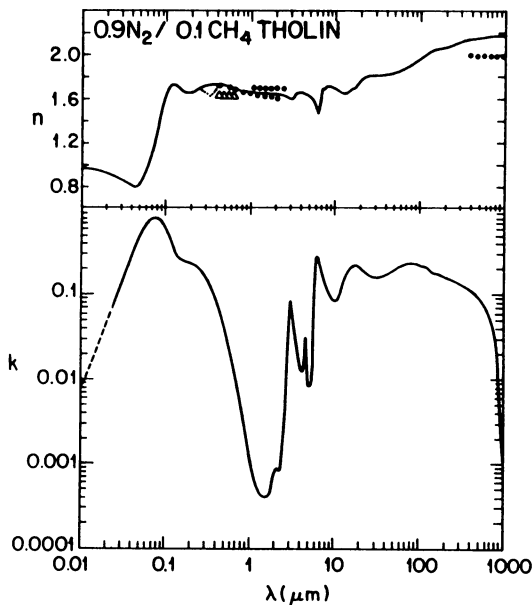


Fig. 5. Real, n , and imaginary, k , values of the complex refractive index of Titan tholins as a function of wavelength (as measured by Khare et al., 1984). Determined by Kramers-Kronig analysis (—). Also shown are n values obtained independently by ellipsometry (o), from interference patterns in transmission (●), from Brewster angle measurements (Δ), and from measured R and k values (●●●).

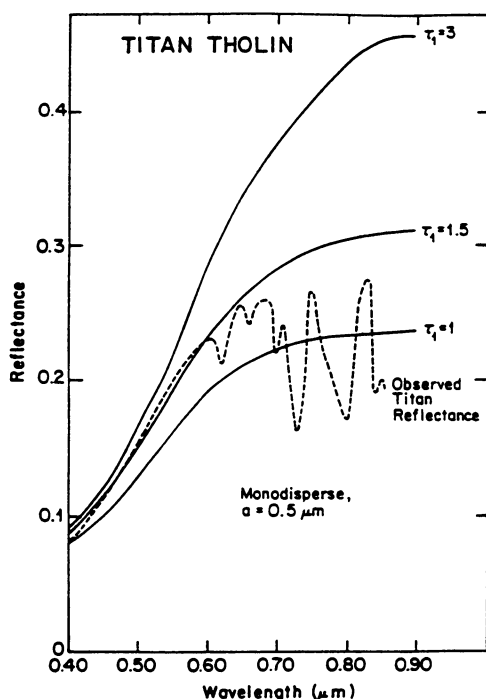


Fig. 6. Two-stream radiative transfer calculations of the reflection spectrum of the proposed tholin aerosols of Titan for mean particle radii $0.5 \mu\text{m}$, slightly larger than photometric and polarimetric observations suggest. (But the mean particle radius is expected to be larger than the sizes of particles in the upper atmosphere of Titan.) Shown for comparison is the groundbased reflectance spectrum of Titan obtained by Nelson and Hapke, 1978. (After Sagan et al., 1983).

methane absorption longward of $0.63 \mu\text{m}$, the predicted reflectivities in the red and near infrared should be greater than those observed on Titan, as shown. Good agreement is obtained for assumed monodisperse particle radius $a = 0.5 \mu\text{m}$ and optical depth $\tau = 1.5$. Recently, more elaborate calculations with Henyey-Greenstein phase functions, condensed methane clouds, and tholin aerosols increasing their particle size with depth show even better agreement with visible and ultraviolet observations simultaneously (Sagan et al., 1984). In addition, the combined sedimentation equilibrium/light scattering models of Podolak et al. (1984) yield optical constants in the visible for the Titan haze virtually identical to those of Titan tholin.

We have successfully fit the full range of available infrared and microwave observations of Titan -- using only surface and atmospheric thermal opacity sources for which there is independent evidence: gas phase N_2 , gas phase CH_4 , gas phase H_2 , condensed CH_4 cloud, and tholin organic haze (Thompson and Sagan, 1984). The dominant sources of

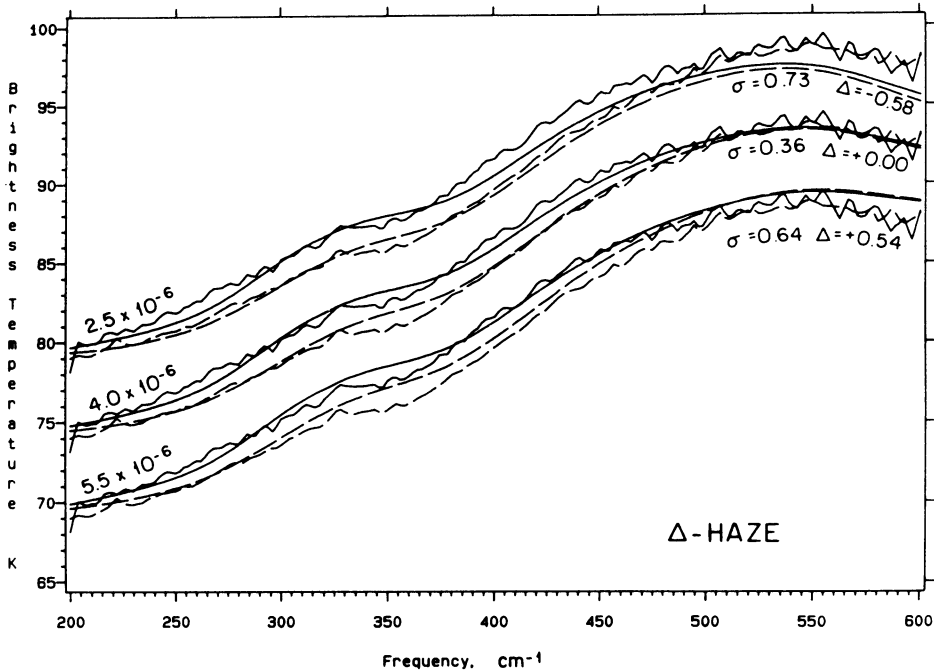


Fig. 7. Computed Titan spectra at 7.3° (smooth solid lines) and 52.7° (dashed lines) emission angles. Lines with detailed structure are Voyager 1 averaged spectra (Samuelson et al., 1981). Top and bottom sets of curves are offset by $+5^\circ\text{K}$ and -5°K , respectively. In all models haze is present only above 2840 km, and H_2 is present at a mole fraction of 0.003. Shown is the effect of varying haze quantity about the optimum value, holding CH_4 cloud abundance and H_2 mole fraction constant. The three sets of curves are labeled on the left by the haze column density in g cm^{-2} . Varying haze quantity by $\pm 40\%$ doubles the average RMS deviation. We determine a Titan tholin haze column density of $4 \pm 2 \times 10^{-6} \text{ g cm}^{-2}$ above the clouds. (After Thompson and Sagan, 1984.)

thermal emission are: the surface for $\lambda > 1 \text{ cm}$, atmospheric N_2 for $1 \text{ cm} > \lambda > 200 \text{ }\mu\text{m}$, condensed and gas phase CH_4 for $200 \text{ }\mu\text{m} > \lambda > 20 \text{ }\mu\text{m}$, and molecular bands and organic haze for $\lambda < 20 \text{ }\mu\text{m}$. In Fig. 7 is shown the model fits to high resolution Voyager observations between 200 and 600 cm^{-1} .

Since the properties of Titan tholin produced in the laboratory agree well with available data on the aerosols of Titan, we believe it is important to examine the chemical nature of the tholin. The low vapor pressure of Titan tholin does not offer many choices for chemical

analysis. Sequential and non-sequential pyrolytic gas chromatography-mass spectrometry allow us to study the volatile components of this tholin, bearing in mind the possibility that some pyrolyzates may be artifacts of the analysis. Towards higher temperatures, where the pyrolysis simulates the processing of tholins by Saturn magnetospheric particles, solar UV, the solar wind, and cosmic rays, the number and complexity of the pyrolyzates increases. The richness of the products is greatest at 200° C and decreases slowly at higher temperatures to 700° C. More than one hundred compounds are released (cf. Khare et al., 1982). The most significant of the compounds are listed in Table II. Infrared spectra of Titan tholin are consistent with the functional groups implied in Table II. Room temperature studies of Titan tholin reveal the presence of

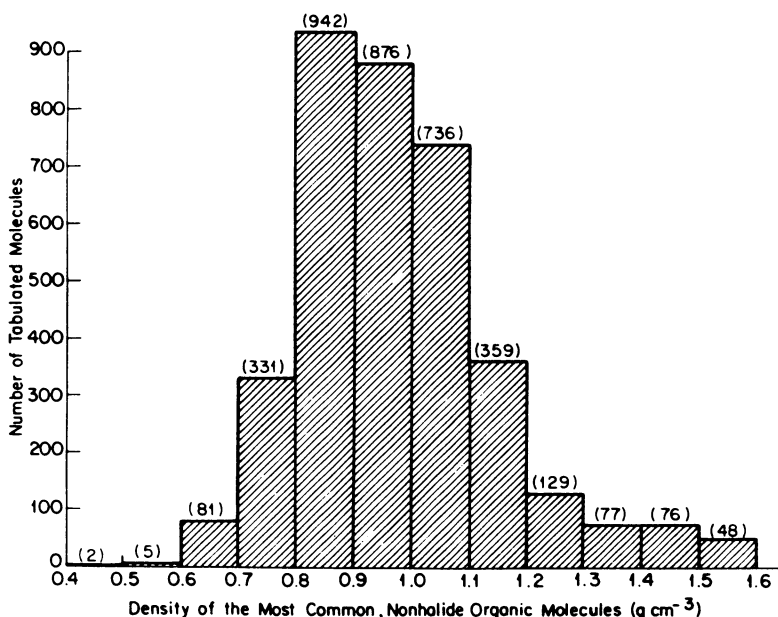


Fig. 8. Histogram of the densities of 3562 non-halide organic molecules (CRC Handbook of Chemistry and Physics 1980). The density of ethene (ethylene), $\sim 0.70 \text{ g cm}^{-3}$ was taken from Lighthart et al., 1979 since the value in the Handbook is in error. Only a few simple organic molecules have densities close to that of liquid CH_4 ($\sim 0.45 \text{ g cm}^{-3}$; Ramaprasad et al., 1978). In the 0.5 to 0.6 g cm^{-3} range fall ethane, propane, propene, 1-butene and 2-methyl-1-butene. Compounds with $\rho > 1.6 \text{ g cm}^{-3}$ are not shown. Even though the presence of C_2H_6 and N_2 will increase the density of the putative Titanian CH_4 ocean to some extent, it does not seem likely that any abundant products of atmospheric organic synthesis will form a substantial crust by floating on the hydrocarbon- N_2 solution.

TABLE II.

<u>ALKANES:</u>		<u>NITROGENOUS RINGS:</u>	
Propane	$\text{CH}_3\text{CH}_2\text{CH}_3$	Dimethylaziridine	
Isobutane	$(\text{CH}_3)_2\text{CHCH}_3$	Azetidene	
Heptane	$\text{CH}_3(\text{CH}_2)_5\text{CH}_3$	Pyrrrole	
<u>ALKENES:</u>		Imidazole	
Propene	$\text{H}_2\text{C}=\text{CH}-\text{CH}_3$	Methylpyrrole (N, 2, 3)	
1-Butene	$\text{H}_2\text{C}=\text{CH}-\text{CH}_2\text{CH}_3$	3-Methylpyrrolidine	
Pentene	$\text{H}_2\text{C}=\text{CH}-\text{CH}_2\text{CH}_2\text{CH}_3$	Dimethylpyrrole	
Hexene	$\text{H}_2\text{C}=\text{CH}-\text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_3$	Ethylpyrrole	
Hexadiene	$\text{H}_2\text{C}=\text{CHCH}_2\text{CH}_2\text{CH}=\text{CH}_2$	Trimethylpyrrole	
Heptene	$\text{H}_2\text{C}=\text{CHCH}_2\text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_3$	Pyridine	
<u>ALKYNES:</u>		Methylpyridine	
2-Butyne	$\text{CH}_3\text{C}\equiv\text{CCH}_3$	2-Aminopyridine	
<u>AROMATIC HYDROCARBONS:</u>		2-Cyanopyridine	
Cyclopentene		2-Ethylpyridine	
Benzene		Dimethylpyridine	
Toluene		Aminomethylpyridine	
Xylene		Trimethylpyridine	
Styrene		Dimethylaminopyridine	
Methyl Styrene		Pyridazine	
Divinyl Benzene		Pyrazine	
Indene		2-Methylpyrazine	
Methyl Indene		Dimethylpyrazine	
Methyl Naphthalene		2,5-Dimethyl-3-ethylpyrazine	
<u>ALIPHATIC NITRILES:</u>		Methylpyrimidine	
Hydrogen Cyanide	$\text{HC}\equiv\text{N}$	2-Aminopyrimidine	
Acetonitrile	$\text{H}_3\text{CC}\equiv\text{N}$	Dimethylpyrimidine	
Propionitrile	$\text{CH}_3\text{CH}_2\text{C}\equiv\text{N}$	Aminomethylpyrimidine	
Methacrylonitrile	$\text{CH}_2=\text{C}(\text{CH}_3)\text{C}\equiv\text{N}$	Triazine	
Isobutyronitrile	$(\text{CH}_3)_2\text{CHC}\equiv\text{N}$	Benzimidazole	
Crotonitrile	$\text{CH}_3\text{CH}=\text{CHC}\equiv\text{N}$	Methylbenzimidazole	
3-Butenenitrile	$\text{CH}_2=\text{CHCH}_2\text{C}\equiv\text{N}$	Indazole	
1-Cyano-1-Butene	$\text{CH}_3\text{CH}_2\text{CH}=\text{CH}(\text{C}\equiv\text{N})$	Ethylindole	
2-Cyano-1-Butene	$\text{CH}_3\text{CH}_2\text{C}(\text{C}\equiv\text{N})=\text{CH}_2$	Adenine	
Cyanopentene	$\text{CH}_2=\text{CHCH}_2\text{CH}_2\text{C}\equiv\text{N}$		
<u>AROMATIC NITRILES:</u>			
Benzonitrile			
Tolunitrile			
Dicyanobenzene			

Reference: Sagan et al., 1983.

Table III.

Net Dissociation and Synthesis Rates

Energy Source	Energy flux ($\text{erg cm}^{-2}\text{sec}^{-1}$)	Synthesis rate [[C+N] $\text{cm}^{-2}\text{sec}^{-1}$]	Column density ($\text{g cm}^{-2}/4.5 \text{ AE}$)
UV, secondary ($\lambda > 1550 \text{ \AA}$)	9.0×10^0	1.8×10^{11}	5.0×10^5
UV, primary ($\lambda < 1550 \text{ \AA}$)	7.3×10^{-2}	4.6×10^9	1.3×10^4
CH ₄	5.0×10^{-2}	3.1×10^9	8.9×10^3
N ₂	2.1×10^{-2}	1.3×10^9	3.7×10^3
Ar	1.8×10^{-3}	1.1×10^8	3.1×10^2
Saturn magnetosphere, energetic electrons	2.4×10^{-2}	4.7×10^8	1.3×10^3
Cosmic rays	9.0×10^{-3}	1.8×10^8	5.0×10^2
Interplanetary electrons	3.2×10^{-3}	6.3×10^7	1.8×10^2
Saturn magnetosphere, energetic protons	2.2×10^{-3}	4.4×10^7	1.3×10^2
Energy deposited			
above 3900 km:			
Solar wind protons	4.8×10^{-2}	9.4×10^8	2.7×10^3
Saturn magnetospheric plasma	1.9×10^{-2}	3.8×10^8	2.7×10^3
Low-Energy interplanetary electrons	1.4×10^{-2}	2.7×10^8	7.6×10^2

Reference: Sagan and Thompson, 1984.

at least 15 biological and nonbiological amino acids present as racemic mixtures (Khare et al., 1983). The richness of products implied by the infrared and amino acid analyses alone suggests that at least many of the GC/MS pyrolyzates are intrinsic to the tholin. The amino acids and the molecules displayed in Table II include many of the organic building blocks of life on Earth.

From the calculated energy deposition rates in the Titanian atmosphere it is possible to calculate (Sagan and Thompson, 1984) the rate at which tholins are synthesized in the Titanian atmosphere, and therefore, in equilibrium, the rate at which such molecules are removed from the atmosphere and accumulate on the surface (Table III). We see that even if ultraviolet radiation at $\lambda > 1550 \text{ \AA}$ does not participate in tholin synthesis, a layer of tholins $> 100 \text{ m}$ thick would still accumulate on the surface of Titan over geological time, were there no further surface processing of the products. If radiation in the range $1550 \text{ \AA} < \lambda < 2400 \text{ \AA}$ can be gainfully employed, the equivalent layer of sedimentary organics produced may be as much as several km thick.

Samuelson et al. (1981) have noted that Voyager data on the state parameters of the Titan surface ($T \approx 95$ K, $p \approx 1.6$ bar) are very near the liquidus in the CH_4 phase diagram. This would be a remarkable coincidence were there no condensed methane on the Titanian surface. In addition, the total present atmospheric abundance of methane on Titan will be lost to photolysis in $\approx 10^7$ years (Strobel, 1982). A substantial methane ocean provides a natural methane replenishment mechanism. Sagan and Dermott (1982) have argued from the methane vapor pressure curve and the tidal evolution of the eccentricity of Titan's orbit that a global hydrocarbon ocean of depth ≥ 400 m is likely; they have also noted the radar detectability of such an ocean. The lower atmospheric temperature gradient on Titan, determined by the Voyager 1 microwave occultation experiment, is approximately 1.38 ± 0.1 K/km -- close to the value calculated for a dry N_2 adiabatic atmosphere. This fact has been used to argue that a global ocean of pure methane is unlikely (Lindal et al., 1983; Flasar, 1983). This apparent discrepancy has been resolved by Lunine et al. (1983), who note that an ocean principally composed of ethane (a CH_4 photodissociation product) can suitably depress the methane vapor pressure; an atmospheric methane abundance of 3 mole percent can coexist with an ocean of 25% CH_4 and 70% C_2H_6 .

In order to minimize the tidal friction that is otherwise fully able over the age of the solar system to circularize the Titan orbit, the hydrocarbon oceans of Titan must either be nearly global or nearly nonexistent (Sagan and Dermott, 1982). Thus, the 100 m or more of Titan tholin produced over geological time has fallen almost entirely on land or almost entirely on a hydrocarbon ocean. The density of a 70% $\text{C}_2\text{H}_6/25\%$ CH_4 liquid is ≈ 0.6 g cm^{-3} . In Fig. 8 is a compilation of the densities of 3562 organic compounds. We see that essentially all possible tholin constituents are more dense than the putative hydrocarbon ocean on Titan and therefore should be present -- if there is such an ocean -- as submarine sediments, not mainly as a surface scum.

The chemistry of immense quantities of Titan tholin in contact with an ocean of liquid hydrocarbons irradiated by cosmic rays over 4×10^9 years, all overlying an icy surface of unknown geological activity remains an unsolved problem. Because the mean surface temperature is so low, it may well be that Titan represents a vast planetary-scale laboratory on which the early events that led to the origin of life on Earth have transpired, with the organic products then deep frozen for all time. On the other hand, it is also possible that, within the very broad confines of our present knowledge, significant further chemical evolution has occurred on the surface of Titan.

It is clear that a major objective for future exploration of Titan should be entry probes and landers (with flotation gear) to examine in situ and in great detail the organic chemistry and other aspects of the atmosphere and surface. Organic matter may well be synthesized in the atmospheres of the contemporary Jovian planets, but

the hydrogen dilution is so much larger and the process of convective pyrolysis so much more efficient there that Titan should be much richer in organic chemistry. Iapetus, other moons and asteroids, and the rings of Uranus may exhibit abundant organic matter, but these environments -- with the possible exception of Triton -- do not have a substantial atmosphere or a possible liquid surface. Of all extraterrestrial environments now known, Titan is by far the most interesting in terms of prebiological organic chemistry.

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