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PRODUCTION OF CYSTEIC ACID, TAURINE, AND CYSTAMINE UNDER PRIMITIVE EARTH CONDITIONS

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PRODUCTION OF CYSTEIC ACID, TAURINE, AND CYSTAMINE  
UNDER PRIMITIVE EARTH CONDITIONS

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PRODUCTION OF CYSTEIC ACID, TAURINE, AND CYSTAMINE UNDER PRIMITIVE  
EARTH CONDITIONS

It is now well established that most of the simple, biologically-important "monomers" (such as amino acids, pentoses, purines, and fatty acids) are formed when various high energy sources are applied to samples of the primitive Earth's atmosphere<sup>1,2</sup>. This atmosphere is assumed to consist mainly of  $\text{CH}_4$ ,  $\text{H}_2\text{O}$ ,  $\text{NH}_3$ ,  $\text{H}_2$  and  $\text{H}_2$  (the last decreasing rapidly with increased age of the earth)<sup>3</sup>. Although the amino acids are among the most prominent products of the irradiations of "primitive-Earth atmospheres," no report has yet appeared in the literature of the appearance of the sulphur-containing amino acids. This has presented no problem, since it can be safely assumed that the primitive atmosphere contained at least traces of  $\text{H}_2\text{S}$ —however, it is always good to have laboratory experimentation to back up assumptions.

Primitive-Earth atmospheres, in the presence of added  $\text{H}_2\text{S}$ , have been studied by Heyns, Walter and Meyer<sup>4</sup>. These authors passed a spark through the mixture and they found essentially no effect of the  $\text{H}_2\text{S}$  on the syntheses of amino acids (non-S containing). They tentatively identified one S-containing product, ammonium thiocyanate. However, a search for S-containing amino acids (by the Stein and Moore method<sup>5</sup>) was unsuccessful, although the authors did report the presence on their chromatograms of six ninhydrin-positive, but unidentified, products. These amino acids may have been sulphur-containing, but their yield was too small to permit identification.

It seemed to us to be worthwhile to search for the "primitive-Earth" synthesis of S-containing amino acids by using  $\text{H}_2^{35}\text{S}$  as one of the

reactants, thus greatly increasing the detection sensitivity for an amino acid product. In this way we have been able to establish the appearance of cysteic acid,  $\text{HO}_2\text{SCH}_2\text{CH}(\text{NH}_2)\text{CO}_2\text{H}$ , and the closely-related compounds taurine,  $\text{HO}_2\text{SCH}_2\text{CH}_2\text{NH}_2$ , and cystamine,  $(\text{SCH}_2\text{CH}_2\text{NH}_2)_2$ . Cysteine and cystine probably were formed (there was evidence for them in our early chromatograms), but they appeared to be oxidized to cysteic acid during subsequent chromatography. We found no evidence for methionine production.

In each of the three experiments reported here, 20 ml of 2 N  $\text{NH}_4\text{OH}$ , 200 mm of  $\text{CH}_4$ , and 100 mm of  $\text{H}_2^{35}\text{S}$  (containing 5 mc) were placed in a pyrex tube and irradiated, with an electron beam, in the manner previously described<sup>6</sup>. No air was present during the irradiations. The electron beam was used for no reason other than that it was a convenient source of ionizing radiation. The radiation dose in each case was about  $10^9$  rads. This is a very high dose; however, our products, as they were formed, were removed from the electron beam (in the condensate dripping off a condenser—see ref. 6) and thus protected from radiation decomposition. The  $10^9$  rads were delivered during 45-minute exposures to the electron beam.

After the addition of chromatographic quantities of unlabeled carriers (cystine, taurine, etc.), the non-volatile contents and washings from the irradiation chamber were transferred to a round-bottom flask and evaporated to a small volume at room temperature. Centrifugation was then employed to remove some polymeric material (in part, elementary sulphur). An aliquot portion (usually 0.1) of the concentrated solution was then placed on Whatman No. 1 chromatographic paper. The solvent in

the first direction was n-butanol:acetic acid: water (100:22:50, v/v/v) and, in the second, isopropanol:methanol:water (7:1:2, v/v/v). Positions and shapes of radioactive spots were determined by autoradiography (Kodak "Blue Sensitive" medical X-ray film). Radioactive spots with Rf values similar to those of added carrier compounds were cut out, eluted, and rechromatographed in the following solvent systems: (1) sec. butanol: formic acid (88-90%):water (15:3:2, v/v/v); (2) sec. butanol: tert. butanol:water (43:48.4:8.6, v/v/v). On the second chromatograms the positions and shapes of the radioactive spots were compared to those of the coloured spots revealed by spraying the chromatogram with ninhydrin solution. The amount of radioactivity in a given spot was determined either (1) directly on the paper by means of a G-M counter or (2) after elution, by liquid scintillation counting. This radioactivity figure could then be compared with the total amount of  $^{35}\text{S}$  (as  $\text{H}_2^{35}\text{S}$ ) that was originally present in the irradiation flask. In this way, the yields of products from the  $\text{H}_2\text{S}$  could be determined (due allowance being made for the half life of  $^{35}\text{S}$ ).

The following  $^{35}\text{S}$ -labeled products (yield from  $\text{H}_2^{35}\text{S}$  given parenthetically) were positively identified: cysteic acid (0.01%), cystamine (0.003%), and taurine (0.01%). We expected to find, and therefore intensively searched for, cysteine, cystine, and methionine; however, if these amino acids are formed, their yields must be well below 0.001%. As was mentioned, cysteine and cystine are probably formed but then disappear by oxidation to cysteic acid. Other compounds for which we searched, but could find no evidence for synthesis, were homocysteine and homocysteic acid.

It is pleasing to record that at least one of the S-containing

$\alpha$ -amino acids (cysteic acid) is formed under "primitive-Earth conditions." We believe that others are also formed (for example, the cysteic acid is probably formed by attack of the  $\cdot\text{OH}$  radical on cysteine). Probably, small changes in the conditions of the "primitive-Earth" experiments (e.g., longer times of irradiation at lower dose rates, presence of catalytic surfaces) will lead to the identifications of all the biologically important S-containing amino acids.

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<sup>1</sup>Horowitz, N. H., and Miller, S. L., Progress in the Chemistry of the Natural Products, Vol. XX, 1962, p. 423.

<sup>2</sup>Miller, S. L., and Urey, H. C., Science, **130**, 245 (1959).

<sup>3</sup>Urey, H. C., The Planets, their Origin and Development, Yale Univ. Press, New Haven, Conn., 1952.

<sup>4</sup>Heyns, K., Walter, W., and Meyer, E., Naturwissenschaften, **44**, 385 (1957).

<sup>5</sup>Stein, W., and Moore, S., J. Biol. Chem., **211**, 893, 907 (1954).

<sup>6</sup>Ponnamperuma, C., Lemmon, R. M., Mariner, R., and Calvin, M., Proc. Natl. Acad. Sci., U.S.A., **49**, 737 (1963).



