of clear calcite that traverse the rocks. Coulson may well have been referring to the prominent cross sections of long, tabular stromatoporoids, which stand out as very light gray calcite bands in the pale red limestone matrix. In 1957 Kidwai (3) made a plane table map of the hills in order to evaluate the limestone reserves.

Fossil assemblages have also been collected recently from limestone localities in northern West Pakistan as much as 56 kilometers distant from the Nowshera reef. Some of these limestones seem to be correlative with the Nowshera reef, especially the Kala limestone north of the town of Swabi in the Mardan district, which in part represents a reef talus.

These limestones have in the past been assigned partly to the Precambrian and partly to the Carboniferous and Permian. The newly found fossils suggest that all the limestones which cover large areas of Swat state and Mardan district may have been deposited during the Silurian or Devonian, or possibly during parts of both periods, in a sea bordered by reefs or containing reef platforms.

The regional metamorphism in the Peshawar area increases toward the north so that the recrystallization and deformation of fossils in limestones tend to increase northward. Dolomitization of these limestones apparently increases toward the east. It is, therefore, unlikely that fossil assemblages as well preserved as those at Nowshera will be found in the limestones either to the east or to the north, although many less-well-preserved fossils have already been found.

No equivalents of these limestones have as yet been found south of Nowshera, but they may correlate with Paleozoic limestones recently found by A. Schreiber (4), of the German geological mission to Afghanistan, south of the Kabul River, about 70 kilometers northwest of Peshawar.

Both in Swat state and Mardan district these fossiliferous limestones are associated with extensive, black, fetid, fine-grained limestones and with vuggy, highly porous dolomites. The combination of reefs, fetid limestones, and porous dolomites has potential economic implications for the discovery of oil.

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Ultrasound Chemical Effects on **Pure Organic Liquids**

Abstract. Molecular fragmentation of organic liquids was produced by cavitation due to ultrasound waves, even in the absence of water. The sonolysis of acetonitrile under argon yielded N_2 , CH_{1} , and H_{2} ; but under oxygen the products were N_2 , CO, CO₂, and H₂O. Pure nonaqueous carbon tetrachloride also underwent sonolytic decomposition under either argon or oxygen, with the production of elemental chlorine.

Intense ultrasonic waves cause chemical changes in water and in aqueous solutions if a permanent gas is present, by a mechanism involving cavitation (1). In pure water, sonolysis produces hydrogen atoms and hydroxyl radicals, as well as molecular hydrogen, oxygen, and hydrogen peroxide (2, 3). When dissolved substances are present, they may react with the H and OH radicals, causing further chemical changes in a manner similar to the "indirect action" of radiolysis (3, 4). In addition many volatile organic solutes in water (such as alcohols, aromatic compounds, or carbon tetrachloride) undergo a direct sonolysis inside the cavitation bubbles, with fragmentation of the molecule (3, 5).

These facts suggested the possibility that pure organic liquids might be decomposed by intense ultrasound even in the absence of water, although no such observation has been reported. Experiments were performed on highpurity acetonitrile and carbon tetrachloride; both liquids underwent sonolytic decomposition. Acetonitrile was chosen because of its wide use in physical chemistry as a nonaqueous solvent; CCl₄ was picked because it is customarily used for sonochemical studies in aqueous solution.

In the experimental procedure (3), a 10-ml glass bulb served as the sample cell, which was irradiated by an ultrasonic generator (800 kcy) and

cooled by circulating water. Acetonitrile (4-ml samples) was first saturated with argon, then irradiated in a closed system for 2 to 10 hours; dissipation of acoustic power inside the cell was estimated to be 3 watts. Analysis of the gas phase by both gas chromatography and mass spectrometry showed that nitrogen, methane, and hydrogen were formed in molar ratios of 1:2:4. The yields of these gaseous products were about 10^{-8} mole liter⁻¹ min⁻¹, which is about two orders of magnitude less than the yield of hydrogen peroxide obtained from water under the same experimental conditions. An attempt to detect less volatile products in the liquid phase was not successful.

Further experiments were made in which the argon atmosphere was modified by the addition of 10 percent of deuterium gas. Mass-spectrometric analysis showed that the ultrasonic treatment produced HD in substantial yield comparable with that of H₂, an indication that hydrogen atoms are a primary product in the sonolysis.

An oxygen atmosphere was substituted for argon. Comparative experiments showed that the yield of nitrogen from acetonitrile remained unchanged in the oxygen-saturated system, but the yield of hydrogen and methane fell to zero. In their place, new products appeared: namely, carbon monoxide, carbon dioxide, and water, in molar proportions of 6:2:1; the dissolved gas plays a major role in the sonochemical reaction. The mass spectrometry and gas chromatography techniques were here supplemented by O18-tracer experiments of higher analytical sensitivity; parallel experiments were made under O^{18} — O^{18} and O^{16} — O^{16} , and the mass spectra of the gaseous products were compared.

For acetonitrile under argon, the sonolytic formation of methane and nitrogen in 2:1 ratio is consistent with the rupture of the C-C bond, giving methyl and cyanide radicals as intermediates. The production of hydrogen in high yield and the formation of H atoms as intermediates are evidence for efficient cleavage of the C-H bonds also, which is not surprising in view of the bond strengths (6).

Under oxygen, the sonolysis of acetonitrile results in oxidation to CO and CO_2 and in the formation of N_2 in relatively high yield. The products are different when acetonitrile is subjected to radiolysis (7). This demonstrates again the distinction (3) between sonolysis (mainly a thermal process) and radiolysis (an ionization process).

We next examined the sonolysis of pure CCl₄ under the same experimental conditions. The rate of its decomposition was determined by measuring the yield of elemental chlorine, as analyzed by extraction into an aqueous iodide solution at pH 4.0 (phthalate buffer) and subsequent spectrophotometric determination of the triiodide formed (8). The rate of chlorine formation under oxygen was 5 \times 10⁻⁷ mole liter $^{-1}$ min $^{-1}$. Under argon, however, the chlorine formation reached a steady state value of only about 10^{-8} mole liter⁻¹ because of the back reaction of chlorine with the CCl_3 radicals. When *n*-butyl iodide was added in 0.03M concentration to the CCl₄ to suppress the back reaction, the rate of chlorine production under argon rose to 8×10^{-7} mole liter⁻¹ min⁻¹, as measured by the overall yield of iodine. Inasmuch as CCl₄ (a liquid of low dielectric constant) undergoes sonolysis at a rate only an order of magnitude less than that of water, it is unlikely that electrostatic phenomena (9) are important in sonochemistry, in comparison to the thermal processes which occur (3, 10).

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3 DECEMBER 1965

Strontium Isotopes: Global Circulation after the Chinese Nuclear Explosion of 14 May 1965

Abstract. Strontium-89 and strontium-90, released into the atmosphere by the Chinese nuclear device on 14 May 1965, circled the world in less than a month and caused a sharp peak in the Sr^{89}/Sr^{90} ratio in rain at Tokyo on 12 June.

The second Chinese nuclear test explosion took place in the Lop Nor area (90°E, 40°N), Sinkiang Province on 14 May 1965. Because Lop Nor and Tokyo (140°E, 36°N) are at approximately the same latitude, and also because of the timing of the test explosion, which coincided with the rainy season (Tsuyu) in Japan, it seemed worth while to investigate the fallout pattern of the Sr⁸⁹ and Sr⁹⁰ in individual samples of rain. The radiochemical procedure that has been in use for several years in the laboratory of one of us (P.K.K.) was used for isolation and purification of the isotopes in rain (1, 2). The rain samples were collected at the Meteorological Research Institute, Kōenji, Kita, Suginami-ku, Tokyo. An Aloka radiation counter (model DC4) was used to measure radioactivity. The background of this low-level counting system was about 0.7 count/min.

The data are shown in Fig. 1. The air trajectory in the Northern Hemisphere during May and June 1965, prepared by one of us (J.N.) at the Long-Range Forecast Section, Japan Meteorological Agency, is shown in Fig. 2. The dates in Fig. 2 show the calculated positions of the tropospheric debris. The straight-line aS'_{0} in Fig. 1 illustrates the Sr⁸⁹/Sr⁹⁰ ratio in the stratosphere prior to the explosion of 14 May; it was deduced from data obtained at Fayetteville, Arkansas, after the first Chinese explosion of 16 October 1964 (2). A sharp increase in the Sr⁸⁹/Sr⁹⁰ ratio in rain occurred in Tokyo on 20 May 1965; it coincided with the arrival of the first wave of the debris via the troposphere, shown in Fig. 2. The debris which entered the stratosphere traveled with greater speed and was picked up as dry fallout at Tokyo on 16 May and was also intercepted by an airplane of the Japanese Self-Defense Agency on the same day over Hokkaido Island.

The rapid decrease in the Sr⁸⁹/Sr⁹⁰ ratio during the next 10 days or so was probably caused by the fact that the bulk of the fresh debris moved eastward over the Pacific Ocean. Frequent and heavy rains, which occurred during this period in Japan, may also have contributed to removal of fresh debris from the troposphere. A small peak in the Sr⁸⁹/Sr⁹⁰ ratio in rain observed on 27 and 28 May coincided with the arrival of typhoon No. 6 at Tokyo. This typhoon reached Tokyo from the vicinity of Okinawa, to the southwest.

The pronounced peak in the Sr⁸⁹/Sr⁹⁰ ratio in rain observed during the middle of June shows the arrival of the debris which circled the world. The data are in excellent agreement with the calculation of the air trajectory shown in Fig. 2. Miyake et al. (3) reported in 1956 that the average velocity of movement of debris from the Nevada test site eastward to Japan was 20 m/sec. The tropospheric debris from the Chinese nuclear explosion of 14 May 1965 circled the world from Tokyo back to Tokyo in 22 days, and the average velocity of



Fig. 1. Variation of the Sr⁸⁹/Sr⁹⁰ ratio in rain after the Chinese nuclear explosion of 14 May 1965. The error limit indicated is due primarily to the counting statistics. The straight line aS_0' is the Sr⁸⁹ /Sr⁹⁰ ratio in the stratosphere prior to 14 May. The straight line S_0b indicates the estimated Sr⁸⁰/Sr⁹⁰ ratio in the stratosphere after the second injection of strontium isotopes by the Chinese.