

## **By sun's early light: discovery of new chemical reaction provides clues to primordial gases in early solar system**

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As recipes go, this could be one of the oldest. Add a measure of ultraviolet radiation from an infant sun to a primordial gas consisting of one part carbon and two parts sulfur. The result: a solid aerosol particle of carbon disulfide.

Though not likely to win any bake-offs, this simple formula nevertheless may have cosmic implications, according to a team of chemists from the University of California, San Diego (UCSD). If carbon disulfide particles were created by the sun's early light, the researchers say, it could offer new insights into how and where organic chemicals and planetary atmospheric gases were trapped and carried to their final destinations in our solar system...and beyond.

"If our notion is right, then you have a very good idea of how and where this all happened," said Mark Thiemens, professor of chemistry and biochemistry, who along with UCSD colleagues report their findings in today's issue of Science. "It's a big step in answering some fundamental questions."

It all began about 4.5 billion years ago when the solar system was but a swirling mass of gases and dust. During these formative years, the research suggests, light from the new sun created the energy to turn carbon disulfide vapor into aerosol particles. Highly absorbent and resilient, these spheroid-shaped structures served as a kind of cosmic sponge, soaking up many other gases in the solar system's primordial nursery, and trapping them in a non-reactive molecular matrix. From here, the carbon disulfide particles and their hitchhikers were transported to their new homes across the solar system. Many of these gases wound up in planetary atmospheres. Meanwhile, the researchers suggest, the carbon partner in the carbon disulfide bond went on to become the carbon found in organic matter and various life- forms found on earth.

"In the past, the questions that been asked is: do you deliver these molecules pre-made to the earth and other planets and then start assembling things or do you make it all locally," said Thiemens. "No one knows. But this is a good way to begin to sort all this out."

The new UCSD study came as a spinoff of another unrelated experiment that sought to explain the role of carbon disulfide in the buildup of an atmospheric chemical--carbonyl sulfide--which has been implicated in the cascade of reactions leading to the destruction of ozone on stratospheric particulates.

That experiment--conducted by chemistry graduate student Jonah Colman--did shed some light on the ozone issue. But it also yielded an unexpected discovery: by simply shining light on gaseous carbon disulfide, Colman was able to create a solid aerosol polymer of carbon disulfide. This was surprising since scientists previously thought that polymers of carbon disulfide-- dubbed "Bridgman's black" for its discoverer, P. W. Bridgman-- could only be formed under special conditions and intense pressure, the equivalent of 40,000 atmospheres.

Colman, a student in the lab of UCSD chemistry professor William Trogler, subjected his polymer to a variety of harsh tests to see how it would compare to Bridgman's black. The spheroid-shaped particles did not easily react with other chemicals and they were virtually indestructible when exposed to strong acids or bases. From a physical and chemical viewpoint, Bridgman's black and the new polymer were identical.

"It was interesting that we could make this chemical at ambient conditions in the gas phase by simply shining a light on it," said Trogler. "And sunlight, by itself, will do it. You take a sample, put it in a tube on the roof, and it reacts."

Though chemically interesting, several months passed before the cosmic implications of these results were fully appreciated. It was a study appearing in the March 3, 1995 issue of *Science* that caught Colman's attention. Essentially, the study described unexpected atmospheric observations resulting from the collision of Shoemaker-Levy 9 with Jupiter. These included the appearance of large amounts of carbon disulfide, disulfur and carbon monosulfide, in addition to the presence of dark brown aerosol particles kicked up into the Jovian atmosphere.

Of particular interest was the fact that the distribution of these particle fragments resembled a pattern that Colman and Trogler had observed after they subjected their polymer to intense heat. It suggested that their polymer and the chemical fallout in the Jovian atmosphere were the same.

When Trogler described these observations to Thiemens, a cosmochemist, he became intrigued because he knew carbon disulfide was commonly found in comets, asteroids and meteorites. If the new polymer matched the chemistry of extraterrestrial carbon disulfide, the results would help answer how the polymer was formed on these cosmic travelers, and where it was produced.

Thiemens was particularly fascinated by meteorites because many contain samples of primordial gases that can't be found in any other source. Since several meteorites have crashed into the Earth, scientists have studied the chemical makeup of these trapped gases, with each chemical offering a potential clue into the solar system's early history.

Such studies, for example, have shown that sulfur-bearing organic chemicals found in the most pristine meteorites called carbonaceous chondrites--stony meteorites with a high carbon content--bear a unique chemical signature: an excess amount of isotopic sulfur-33.

"I got interested in Bill's work because we measure sulfur isotopes in meteorites," said Thiemens. "We've always found these sulfur anomalies interesting and when Bill started talking about this, I realized maybe this was the sort of reaction that might occur prior to building planets."

Together, the researchers decided to see if the darkened polymer produced in Trogler's lab shared the same sulfur signature as the crashed meteorites, including a fragment of the Allende meteorite--the largest carbonaceous chondrite available for scientific study that crashed into Mexico in 1969.

"The short answer is yes," said Thiemens. "We don't know what's causing this sulfur signature. It's a brand new isotope effect. But it clearly is the right signature for the critical isotope you see in the meteorites."

Further analyses suggested that the polymer manufactured in Trogler's lab may be a chemical precursor to a black substance left behind after the most pristine meteorites are dissolved in acid. This substance, dubbed "Q" (for quintessence) by its discoverers at the University of Chicago, is particularly noteworthy because it contains all the noble gases of the periodic table: helium, neon, argon, krypton, xenon and radon. Presumably, these gases were trapped by carbon disulfide polymers shortly after its creation in the presolar nebula.

These results, the UCSD chemists say in the *Science* paper, suggest that the photochemical process that created the polymer in Trogler's lab could be at least partially responsible for the manufacture of Q, in addition to the cosmochemical environments of the presolar nebula, asteroids, comets and planetary atmospheres.

"It's amazing someone didn't observe this photopolymerization 50 years ago," said Trogler, "It's really a simple reaction."

Turning to the observations that sparked their investigation, the researchers said that the dark, red-brown debris kicked up in the Jovian atmosphere following the collision with Shoemaker-Levy 9 may have been the result of charred carbon disulfide byproducts created at the impact site. An analysis, using a technique called spectroscopy, suggested that this debris also resembled the laboratory-created carbon disulfide particles when subjected to intense heat.

Trogler noted that gaseous carbon disulfide is stable only in a reducing atmosphere, which is characterized by little or no oxygen.

"Many of the planets when they initially formed had reducing atmospheres," said Trogler. "The meteorites formed in reducing chemical environments and Jupiter, with its hydrogen atmosphere, is highly reduced. This allows enough time for photopolymerization to occur.

"Only earth is unusual because of photosynthesis. So we don't see a lot of carbon disulfide on this planet because we have an oxidizing atmosphere."

Also participating in the study was Xianping Xu, a researcher in Thiemen's laboratory.

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