tween aspartate and glutamate have been seen in Aplysia.

In Aplysia, the receptors activated by iontophoretic application of serotonin and acetylcholine appear to be identical to those existing at serotonergic and cholinergic synapses, respectively (14, 15). Our results suggest that aspartate may also function as a neurotransmitter since there are specific receptors for aspartate which can selectively change the membrane permeability of some neurons to Na<sup>+</sup> or Cl<sup>-</sup> (or both).

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21 December 1975; revised 24 February 1976

# Hydrogen Cyanide Formation over Automotive Catalytic

## **Converters**

In a report in Science (1) Voorhoeve et al. discussed laboratory experiments in which substantial amounts of HCN were generated when reactor gas was passed over Pt oxidation catalysts. This topic is significant since many late-model automobiles use Pt alloy oxidation catalysts for the control of CO and unburned hydrocarbon compounds. In addition, the authors point out (1): "Future application of catalytic converters for the reduction of  $NO_x$  (oxides of nitrogen) is a strong possibility, especially in the remainder of this decade. . . . In some proposed devices for  $NO_x$  reduction, a Pt catalyst is used in a reducing exhaust mixture as an oxygen scavenger upstream from the proper  $NO_x$  catalyst."

Gould, Inc., manufactures an NO<sub>x</sub> reduction catalyst which is similar to the preceding description (2). The system is designed to operate at carburetion slightly rich of stoichiometry to provide a net reducing atmosphere. One element of the catalyst system is a Pt alloy, oxygenremoval catalyst situated upstream of the base metal  $NO_x$  catalyst bed.

Several comments on the relevance of the data of Voorhoeve et al. to an actual automotive exhaust environment can be made. Their reactor feed gas contained 0.5 percent  $H_2$ , 5 percent CO, 0.1 to 0.5 21 MAY 1976

percent NO, 0 to 0.006 percent SO<sub>2</sub>, and 0 to 3.5 percent H<sub>2</sub>O. Although this gas is an approximation to automotive exhaust with very rich carburetion, the correlation is not good. Vehicle exhaust typically contains additional components: 10 percent H<sub>2</sub>O, 12 percent CO<sub>2</sub>, and 0 to 1 percent O2. Laboratory data obtained from gas containing these additional com-



ponents have shown that H<sub>2</sub>O, CO<sub>2</sub>, and  $O_2$  in the exhaust gas can significantly affect the amount of HCN formed over a Pt catalyst. Moreover, the Pt catalyst tested by Voorhoeve et al. promoted complete NO conversion with high selectivity toward NH<sub>3</sub> formation. This particular situation does not occur in a catalyst system where the oxygen-removal catalyst has been treated to eliminate NH<sub>3</sub> formation. The decreased selectivity toward NH<sub>3</sub> formation is observed to influence HCN formation.

We have carried out simulated exhaust gas and actual vehicle testing to check for HCN formation over the NO<sub>x</sub> reduction catalyst system. In laboratory testing we used bottled gas with N<sub>2</sub> as the carrier gas. The exhaust gas delivery rate equaled a space velocity of 100,000 hour<sup>-1</sup>. We measured the HCN by bubbling a measured volume of gas through a 0.1M KOH solution; the solution was titrated with 0.1M AgNO<sub>3</sub> with the use of a KI indicator. This technique was insensitive to feed gas moisture and detected HCN in exhaust gas concentrations as low as 0.5 part per million (ppm) with an accuracy of  $\pm$  5 percent of the measured value.

The oxygen-removal catalyst used with the  $NO_x$  catalyst was an Engelhard PTX-IIB monolith. In some of the experiments we used a PTX catalyst to which a thermochemical treatment had been applied that was effective in suppressing  $NH_3$  formation. The actual  $NO_x$  reduction catalyst was the Gould GEM 68 formulation.

The laboratory data are shown in Fig. 1. Curve 1 was measured under the conditions outlined by Voorhoeve et al. to maximize HCN formation, that is, 5 percent CO, 0.3 percent NO, and 0.5 percent H<sub>2</sub>. The superimposed dashed curve (curve B) represents the data of Voorhoeve et al. (1) measured under the same conditions except that the curve 1 space velocity was 100,000 hour<sup>-1</sup> and the curve B space velocity was 14,000 hour<sup>-1</sup>. When 5 percent H<sub>2</sub>O (curve 2) is added to the feed gas, the maximum amount of HCN formed decreases from 300 to 30 ppm. When H<sub>2</sub>O is adjusted to the 10

Fig. 1. Formation of HCN over Pt alloy and base metal catalysts: curve 1, PTX in extreme (to maximize HCN formation) exhaust gas containing no H<sub>2</sub>O; curve B, data of Voorhoeve et al. (1) (sponge Pt-II) under conditions similar to those of curve 1: curve 2. PTX in extreme exhaust gas containing 5 percent  $H_2O$ ; curve 3, PTX in extreme exhaust gas containing 10 percent H<sub>2</sub>O; and curve 4, untreated PTX (●), treated PTX (▲), and GEM 68 (1) in typical synthetic exhaust gas containing 10 percent H<sub>2</sub>O.

percent level typically seen in vehicle exhaust gas, the maximum HCN is further decreased to 2 ppm, as shown in curve 3. Curve 4 represents data measured in typical synthetic exhaust gas (1.5 percent CO, 0.5 percent H<sub>2</sub>, 12 percent CO<sub>2</sub>, 0.1 percent NO, and 10 percent H<sub>2</sub>O). Data are given under these conditions for the untreated PTX, treated PTX (for low NH<sub>3</sub> formation), and the base metal GEM 68 NO<sub>x</sub> reduction catalyst. At typical catalyst operating temperatures (650°C) in simulated exhaust gas, HCN formation is about 0.5 ppm over the  $NO_x$  catalyst system. The data of Fig. 1 clearly indicate that H<sub>2</sub>O inhibits the formation of HCN.

Laboratory experimentation has shown that HCN can be formed over a noble metal catalyst under reducing conditions. However, when the gas approximates vehicle exhaust, under net rich carburetion conditions, very little HCN is formed, typically less than 1 ppm. The exhaust gas components that act as HCN promoters are, in order of increasing effectiveness, NO, NH<sub>3</sub>, CO, and H<sub>2</sub>. On the other hand, the exhaust components that are HCN inhibitors are, in order of increasing effectiveness, CO<sub>2</sub>, H<sub>2</sub>O, O<sub>2</sub>, and SO<sub>2</sub>.

Actual vehicle tests have also been conducted to verify the laboratory data and to assess the possibility of the formation of HCN in catalyst-equipped vehi-

cles. A 1975 California Chevrolet Nova (350 cubic inch displaced) was fitted by Gould with a GEM 68 dual-bed catalyst system calibrated to meet the 1978 standards of the Clean Air Act, that is, 3.4 g of CO per mile (2.1 g of CO per kilometer), 0.41 g of hydrocarbon per mile, and 0.4 g of  $NO_x$  per mile. The vehicle was operated with an air pump to provide about 5 percent O2 to the oxidation catalyst and was driven on a chassis dynamometer according to the 1975 Federal Test Procedure schedule (3), with Indolene containing 50 ppm of sulfur used as the test fuel. The driving cycle is typical of normal urban driving with accelerations and decelerations giving an average speed of about 25 mile/hour (40 km/hour). In addition to the normal driving mode, a worstcase situation was also simulated when the vehicle was driven at high speed (50 mile/hour) with heavy choke. In the worst-case situation the exhaust contained 6 percent CO and 0.06 percent  $NO_x$ . This situation could prevail under heavily loaded driving conditions, for example, a vehicle pulling a trailer uphill. The sampling technique for the vehicle tests was similar to the method used in the laboratory; that is, a measured volume of the tail pipe exhaust was passed through the KOH solution and titrated.

In the normal driving mode, there was no detectable HCN in the vehicle tail pipe emissions. When the car was driven under worst-case conditions, the vehicle exhaust contained 0.5 ppm of HCN, which is the approximate limit of detectability of HCN. These values from the vehicle tests are consistent with the laboratory study indicating a relatively small amount of HCN formed in the routine operation of the dual-bed catalyst system. We did not monitor the HCN concentration of the engine output during the vehicle tests. However, low concentrations of HCN ( $\sim 1$  ppm) have been detected in the exhaust of noncatalyst vehicles (4).

Thus the ability of an automotive catalytic converter to form HCN is strongly influenced by the chemistry of the exhaust gas. Significant amounts of HCN can be generated in moisture-free or lowmoisture gas. When the gas composition is adjusted to approximate typical vehicle exhaust, laboratory data indicate that less than 1 ppm of HCN is formed. Actual vehicle tests have confirmed these laboratory data.

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