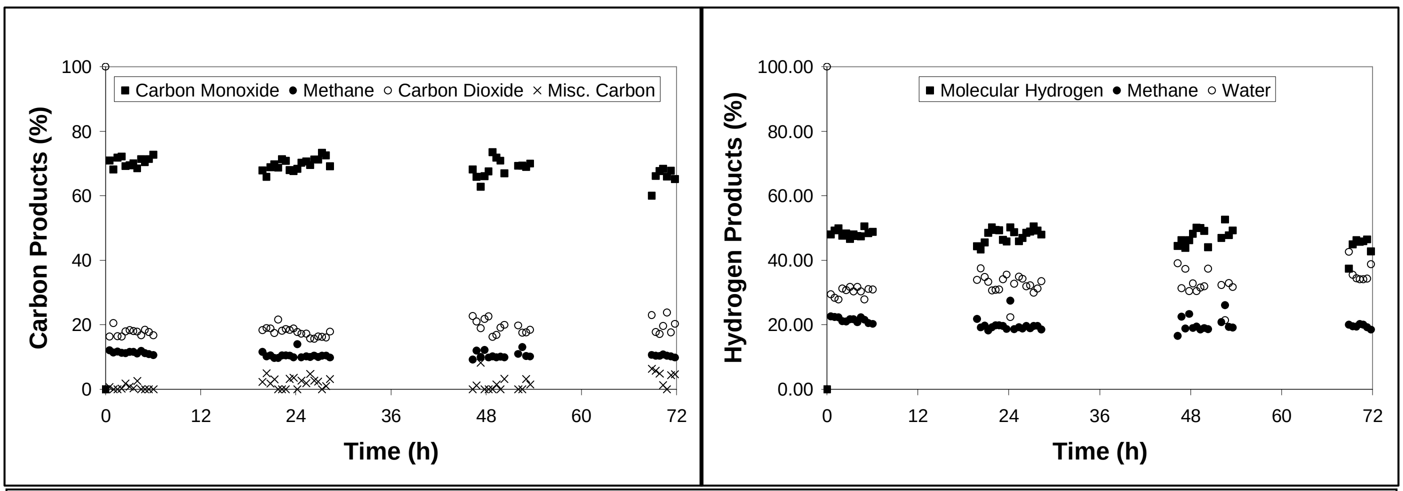
Reformer for Conversion of Diesel Fuel into Synthesis Gas

Summary:
- A fuel reformer designed to transform ultra-low sulfur diesel fuel into synthesis gas.
- A dry, partial oxidation catalyst for improved diesel fuel reforming in Eltron reformers.
- A sulfur-tolerant catalyst thermodynamically stable to 1000ºC under reducing conditions.
- An inexpensive catalyst based on rare-earth and transition metal oxides.

Eltron Research & Development, Inc. is developing a fuel reformer system for use in auxiliary power units to be incorporated into commercial freight liners. The reformer under development transforms commercial ultra low sulfur diesel (ULSD) fuel into synthesis gas (a mixture of hydrogen and carbon monoxide) which can be supplied to a solid-oxide fuel cell system for the production of electricity. This system is predicted to be both more efficient and less-polluting than traditional diesel-fueled electric generators. The resulting technology can also be applied to any system requiring or benefiting from a ready supply of synthesis gas—including NOx trap regeneration.

Eltron has designed and tested partial oxidation catalysts based on electron-conducting perovskites. By design, these oxide catalysts possess high surface and bulk O^{2-} mobility—providing the active and mobile dissociated oxygen essential for the difficult partial oxidation of the polycyclic aromatic hydrocarbons in diesel fuel. The perovskite catalysts are refractory oxides and are thermodynamically stable under reducing conditions to temperatures in excess of 1000℃.

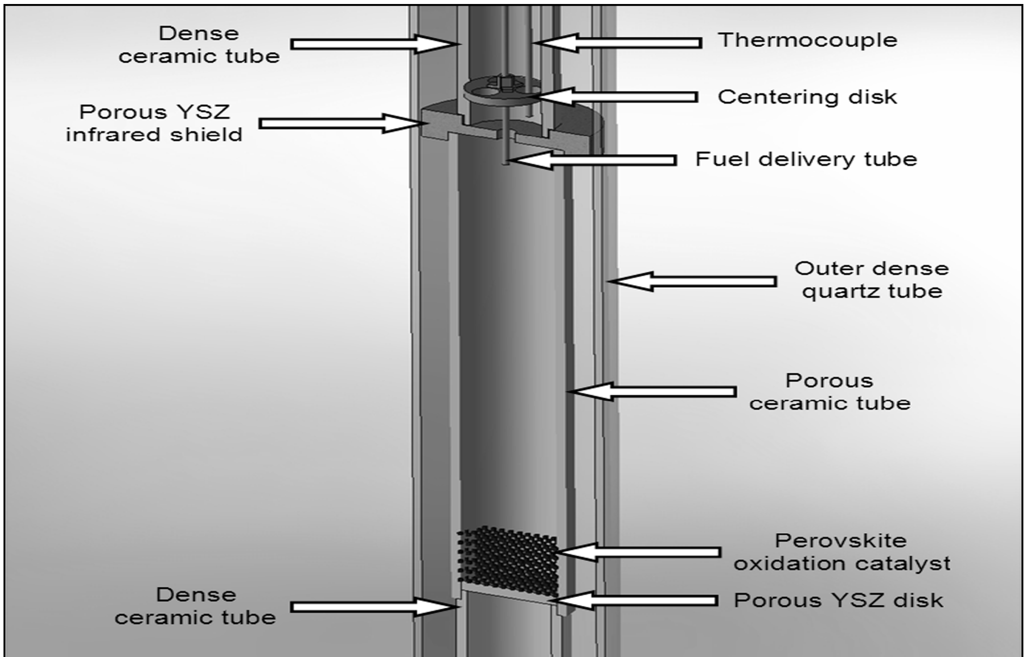


Eltron catalysts produce hydrogen and carbon monoxide with good efficiency from ULSD fuels throughout 72 hour tests with no drop in performance. Tests were conducted with 20% excess oxygen relative to stoichiometric partial oxidation.

Thermodynamic Conditions

A thermodynamic analysis was performed at Eltron to determine the equilibrium conditions most favorable for synthesis gas production while limiting the formation of both deep oxidation products like H_2O and CO_2 and elemental carbon. Calculations are based on a H:C ratio of 1.86:1 (obtained through empirical analysis of ULSD) and predict that H_2 and CO will be overwhelmingly favored if the reformate can be brought to equilibrium at temperatures above 900ºC–1000ºC by the partial oxidation reaction: $C_1H_{1.86} + 0.5\ O_2 = 0.93\ H_2 + CO$.

Moreover, the benzene rings may also act as nuclei for the growth of undesired soot and graphite, which can plug reactor systems. Because of their stability, polycyclic aromatic compounds do not readily react with ground-state molecular oxygen. In order to enhance the reactivity of oxygen, Eltron's catalysts dissociate molecular oxygen into highly active atomic oxygen in order to attack the benzene rings and better reform the polycyclic aromatic compounds present in diesel fuels.



This 3D model of a single-tube reactor is similar to the multi-tube reactor design Eltron has produced for the generation of synthesis gas. When coupled with a solid-oxide fuel cell, the multi-tube design will yield sufficient synthesis gas to generate 5 kW of- clean electricity.

Reactor Design

Eltron's diesel fuel reformer is based on a coaxial design incorporating a porous inner wall and a dense outer wall. Diesel fuel is mixed with air at the desired ratio and evaporated in a thermostatically controlled evaporator assembly. The air-fuel mixture is then admitted through the top of the reactor while additional air is admitted to the lumen formed between the inner and outer reactor walls. The air passing through the lumen serves to cool the reactor—better controlling the internal temperature distribution. The inner tube is sufficiently porous to allow the collection of oxygen within the pore structure (reducing carbon deposition), yet limits the rate of bulk gas transfer across the tube. For improved inertness, internal structures are manufactured from dense and porous zirconia or zirconia toughened alumina. The catalyst is supported in the bottom of the reactor by a highly-porous zirconia plate. Laboratory-scale testing uses an external electric furnace to accurately maintain the catalyst temperature. In this configuration, the cooling air admitted through the lumen is unnecessary; however, a production system, with an increased rate of diesel consumption, would derive sufficient heat from the partial oxidation of the diesel fuel—requiring the cooling air flow to control the catalyst temperature.

System Performance/Stage of Development
Eltron's diesel fuel reformer has demonstrated steady-state (72+ hours) diesel fuel reforming from commercial ULSD fuel under laboratory conditions with minimal carbon deposition. In these tests, the air-to-fuel ratio was maintained at 1.2:1, timer stoichiometric for partial oxidation, x storch for pox. Of the hydrogen present in the input diesel fuel, 50% was reformed to yield molecular hydrogen while an additional 20% was recovered as methane. The remaining hydrogen is thought to exit the reformer as water. A similar analysis based on carbon shows that 70% of the input carbon is recovered as carbon monoxide with an additional 10% obtained in the form of methane. Deeply oxidized carbon dioxide accounts for most of the remaining 20%.

Tests performed in the prototype reformer using 20% (by mass) of the most volatile distillates from ULSD fuel show improved performance. In these experiments, conversion efficiencies of 80% and 90% for molecular hydrogen and carbon monoxide were observed throughout a 72 hour experiment. This improvement is thought to be due in large part to the removal of polycyclic aromatic compounds through the distillation process.

Separate experiments performed using conventional diesel fuel, (before the availability of ULSD), demonstrated stable catalyst performance throughout 200–400 hour tests with continuous operation at 1000° C. Despite the greatly increased sulfur content of the fuel, powder x-ray diffraction indicates that the catalyst was stable under operating conditions with no detectable sulfides, carbonates, or sub-oxides observed in post-reforming catalyst analyses.

Future catalyst work: Efforts are underway to improve the reformer efficiency by reducing the excess oxygen. If the reformer can be run under conditions closer to partial oxidation, the level of deep oxidation products (H2) & CO2) will be significantly reduced resulting in increased hydrogen and carbon monoxide production. Efforts are also underway to enhance the Eltron catalysts to facilitate more efficient reforming of the challenging polycyclic aromatic components in diesel. Both of these improvements are anticipated to increase the overall system efficiency.

Eltron has a patent application filed with the USPTO, *Catalytic Membrane Reactor and Method for Production of Synthesis Gas, application # 11/851,017.*

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4600 Nautilus Court South | Boulder, CO 80301 | Phone: 303.530.0263 | www.eltronresearch.com