MONOLITHIC HYDROGEN PEROXIDE CATALYST BED DEVELOPMENT

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ABSTRACT

With recent increased industry and government interest in rocket grade hydrogen peroxide as a viable propellant, significant effort has been expended to improve on earlier developments. This effort has been predominately centered in improving heterogeneous, typically catalyst beds; and homogeneous catalysts, which are typically solutions of catalytic substances. Heterogeneous catalyst beds have traditionally consisted of compressed wire screens plated with a catalytic substance, usually silver, and were used in many RCS applications (X-1, Mercury, and Centaur for example). Aerojet has devised a heterogeneous catalyst design that is monolithic (single piece), extremely compact, and has pressure drops equal to or less than traditional screen beds. The design consists of a bonded stack of very thin, photoetched metal plates - silver coated. This design leads to a high surface area per unit volume and precise flow area, resulting in high, stable, and repeatable performance. Very high throughputs have been demonstrated with 90% hydrogen peroxide. (0.60 lbm/s/in³ at 1775-175 psia) with no flooding of the catalyst bed. Bed life of over 900 seconds has also been demonstrated at throughputs of 0.60 lbm/s/in³ across varying chamber pressures. The monolithic design also exhibits good starting performance, short break-in periods, and will easily scale to various sizes.

NOMENCLATURE

Aₜ = throat area
Cₜₐ = characteristic velocity, actual
Cₜₜ = characteristic velocity, theoretical
mdot = mass flow rate
Pc = chamber pressure
G = throughput, mass flow per unit area
A = cross sectional area

INTRODUCTION

Catalyst beds for hydrogen peroxide have been used for years in the rocket propulsion industry. RCS/ACS thrusters for Mercury and Scout, and V2 gas generators are a few examples of applications. More recently hydrogen peroxide has been used as a steam and oxygen source for vacuum ejectors on high-powered lasers.

Over the last five years Aerojet has applied platelet technology to catalyst bed designs. From this effort a high performing, compact catalyst bed has been developed that offers many improvements over traditional approaches.

This design approach results in significant advantages:

1) More compact - Traditional catalyst beds are typically greater than 2 inches in length. Monolithic catalyst beds operate best when the length is approximately 0.5 inches.

2) Monolithic - Traditional catalyst beds are an assembly of many pieces that must be retained in a structure and are difficult to service in the field and replace. Monolithic designs are single piece, self-contained "cartridges" that allow for simple removal and replacement.

3) Cold Starts - traditional catalyst beds often need heaters or warming pulses to start without flooding. Monolithic catalyst beds cold start on their own.

4) Short Break-in periods - traditional catalyst beds need extensive break-in pulses, typically in the hundreds. Monolithic beds will break-in with full flow in approximately 5 seconds.

Self-starting capability, smaller envelope, single piece construction, and short break-in periods significantly impact system design, weight, and cost. Liquid bipropellant/monopropellant ACS/RCS engines, gas generators, and laser vacuum ejectors all can benefit from these features.

Currently the design was designed and demonstrated with 90% hydrogen peroxide. Designs have been completed that would allow for operation with 98% hydrogen peroxide, but not demonstrated.

BACKGROUND/HISTORY

Hydrogen peroxide is a well-known monopropellant. When hydrogen peroxide comes in contact with a catalyst it decomposes. The reaction is represented as follows:
The temperature of the resulting steam/oxygen mixture is dictated by the concentration of the hydrogen peroxide. Decomposition temperatures of low concentrations of hydrogen peroxide always remain the boiling temperature of water at the conditions of operations (~212°F). This is because the evolved heat is all consumed by vaporizing the water. As concentration increases, the amount of residual water will be reduced. When the concentration of hydrogen peroxide is high enough (~67% H₂O₂ by weight) there is enough heat released to vaporize 100% of the H₂O. At this point the temperature will quickly chain upward, as the excess heat only serves to raise the steam and oxygen temperatures. Some representative decomposition temperatures are shown in Table 1.

<table>
<thead>
<tr>
<th>H₂O₂ Conc.</th>
<th>Decomp. Temp. (°F)</th>
<th>C⁺ (ft/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>70</td>
<td>500</td>
<td>2243</td>
</tr>
<tr>
<td>80</td>
<td>946</td>
<td>2704</td>
</tr>
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<td>90</td>
<td>1390</td>
<td>3080</td>
</tr>
<tr>
<td>98</td>
<td>1740</td>
<td>3341</td>
</tr>
</tbody>
</table>

Table 1 - Hydrogen Peroxide Decomposition Temperatures and Characteristic Velocities

TRADITIONAL SILVER SCREEN DESIGN

A traditionally manufactured catalyst bed has several drawbacks: inconsistent performance, long break-in periods, and poor cold start transients. Aerojet’s monolithic catalyst bed significantly exceeds traditional screen catalyst beds in each area.

When hydrogen peroxide comes in contact with the catalyst bed, it decomposes and creates free oxygen and heat. To date, heterogeneous catalyst bed designs are many loose parts, compressed to significant pressures, to get an acceptable mixture of pressure drop, capacity, and have been a mixture of science and art, leading to many design iterations and scalability problems.

Traditionally catalyst beds have consisted of inert metal wires electroplated with a predetermined thickness of silver, although solid silver wires have been used. The wires are stacked in layers. The mesh or open area of the screen will change along the length of the bed, with the front/inlet portion less open than the back/exit portion. The wire screen layers are compressed, typically to several hundred psi or greater, to remove voids and generally increase tortuous nature of the flow path. An example of a traditional silver screen catalyst bed is shown in Figure 1.
Figure 1 - Traditional Screen Bed Design

By their nature, actual flow paths through screen based beds are unknown. The bed has an aggregate flow direction and assumptions are often made, but the reality is that the bed has unknown local variances. These variances lead to non-uniform flow distributions. Non-uniform flow is especially unwanted in the liquid portion of the catalyst bed (typically the first 1/3 of the bed length). Localized zones or low restriction allow the flow to "core" and the bed will "flood". Flow can also run down the sides of the bed and effectively "short circuit" the catalyst bed. Anti-channel baffles and bed compression somewhat limits this, although only partially. The cause of many historically encountered difficulties (unit to unit repeatability, scalability, and poor cold start transients/flooding) can be explained by non-uniform flow distribution.

MONOLITHIC DESIGN DESCRIPTION

Aerojet’s design and manufacturing experience with chemically etched platelets was an ideal approach to further advance heterogeneous catalyst bed technology. The process is very precise and accurate. Chemically etched platelets allow for greater freedom and control in fluid design. Flow control features, passage shape, open area, and surface area are optimized to promote mixing, heat transfer, and pressure drop.

The initial concept, shown in Figure 2, has been very successful. It consists of through etched holes placed equidistant from each other. The backside of the platelet is depth etched, allowing the fluid to flow sideways or transverse to the holes. This forces the hydrogen peroxide to impact directly on the surface of the next platelet and mix very thoroughly. Thorough mixing greatly enhances catalyst bed performance. In the first portion of the catalyst bed, the reaction is driven primarily by the catalytic action. Once the temperature of the mixture exceeds approximately 400°F, the reaction is primarily thermally driven. Complete and thorough mixing enhances the exposure to the catalyst and heat transfer, and therefore overall catalyst bed performance.

Figure 2 - Platelet Catalyst Bed Flow Path

The diameter of the through holes is varied to change the open flow area. The precision inherent to platelet manufacturing, tolerances typically +/- 0.0005 inch on features of size, eliminates significant variations of flow across the bed cross section. Open area gradients are also very controllable throughout the length of the catalyst bed.

A solid border is left around the perimeter of the platelets. When bonded this border makes for a solid edge that does not allow for leaking or for flow to take the path of least resistance and "channel" down the sides of the catalyst bed.

INSERT SECTION ON SCREEN OPEN AREA

CATALYTIC COATING AND BONDING

Aerojet funded experiments in several catalyst and substrate materials settled on simple silver plating for the initial designs. This allowed for the best initial balance between bond strength and catalytic activity. The plates are stacked together and then bonded into a single piece, monolithic structure. For manufacturing and demonstration of batch production methods, the platelets were designed to make eight beds at a time. These beds were stacked and bonded in a single operation. The resulting "brick" of catalyst
beds can be seen in Figure 3. Post bond, each catalyst bed was removed from the brick by EDM cutting, turned round in a conventional lathe, and then EB welded into a stainless steel flange (Figures 4 & 5). The stainless steel flange add a good sealing surface removed from the reactive portions of the catalyst bed, eliminates a possible leak path, and makes for simple, easy to remove cartridge/filter type assembly.

Figure 3 - Bonded Catalyst Beds

Figure 4 - Catalyst bed Assembly, Inlet Side

Figure 5 - Catalyst Bed Assembly, Exit Side
TEST METHODS AND EXPERIENCES

Catalyst bed operation, start transients, and decomposition efficiencies are typically measured in a simple heat sink chamber. A chamber with a rounded throat has a catalyst bed installed on one end. Redundant pressure transducers and thermocouples are installed on the sides of the chamber. Hydrogen peroxide flow is introduced into the catalyst bed. Chamber pressure, temperature, and plume quality is observed. A clear plume (no condensed H2O) typically indicates high decomposition efficiency. Chamber pressure measurements are used to determine accurate decomposition efficiencies and start transients.

Decomposition efficiency is a ratio of measured C* to theoretical C* (2). Chamber pressure is used to measure C* as a function of chamber pressure, mass flow rate, and area of the sonic throat (3).

\[ \eta_{C^*} = \frac{C^*_{\text{act}}}{C^*_{\text{theo}}} \quad (2) \]

\[ C^* = \frac{P_r A_2}{m} \quad (3) \]

Thermocouple data as a measure of decomposition efficiency is very unreliable. When a catalyst bed is operating in a non-optimum manner, partially decomposed fog/H2O2 vapor exits the catalyst bed. This fog will continue to decompose thermally if it comes in contact with any hot metal surface. If the thermocouple is a shielded thermocouple, it will read full decomposition temperature. This will be in direct conflict with a C* calculation based upon chamber pressure. This can be somewhat alleviated by using an exposed junction thermocouple. Therefore, thermocouple data is a nice reverence point, but chamber pressure measurements take precedence.

SUMMARY OF TEST RESULTS

For initial proof of concept demonstrations, a bed 1.0 inch OD x 1.5 inch length was chosen due to the significant historical database available on similar sized screen beds. Initial testing obtained 100% decomposition of the hydrogen peroxide. Excessive pressure drop illustrated the catalyst bed length should be reduced. Further refinements to catalyst bed length ended with an optimal length of 0.560 inches length. Further refinements were tested on contract with NASA, and length was reduced to 0.512 inches. The beds were scaled in size to an outside diameter of 1.25 inches. All testing was done with 90% hydrogen peroxide.

Break-in periods for these beds are almost nonexistent. Typically the bed is installed in the test fixture, a short warming pulse is applied, and then operated at its design point. Initially chamber pressure has a slight roughness, but after approximately 5 seconds, this stabilizes and the bed is "broken in". A typical example of a break-in run is Figure 6. This basic design has been fabricated and tested for IR&D tests and on contract to NASA with success. Several bed designs have had catalyst bed pressure drop as a function of bed throughput and downstream chamber pressure thoroughly mapped.
Figure 6 - Break-in Run, $P_c=1775$, $G = 0.60$ lb/s/in$^2$, $\Delta P = 175$

Catalyst bed performance is typically measured in terms of flow per unit of cross-sectional area and pressure drop (4).

$$G = \frac{m}{A} \quad (4)$$

The pressure drop ($\Delta P$) of a catalyst bed is a function of the throughput ($G$) and downstream chamber pressure ($P_c$). Obviously, it is desired to maximize throughput while minimizing pressure drop. Pressure drop is driven by internal catalyst bed geometry, speed of reaction, and downstream chamber pressure. High downstream chamber pressure with a low throughput will result in a low-pressure drop, just as opposite conditions will result in a high-pressure drop. A bed with a very low-pressure drop often floods or has chug-like pressure oscillations.

Different monolithic catalyst bed designs and their respective pressure drop at chamber pressures are listed in Table 2. All tests showed full decomposition, with clear plumes, and decomposition efficiencies above 95%. The 5% C* loss is attributed to cold chamber heat sink effects during a short duration firing.

<table>
<thead>
<tr>
<th>Design / L (in.)</th>
<th>$P_c$ (psia)</th>
<th>$\Delta P$ (ps)</th>
<th>$G$ (lb/s/in$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>001/.560</td>
<td>1725</td>
<td>175</td>
<td>0.60</td>
</tr>
<tr>
<td>002/.512</td>
<td>1775</td>
<td>175</td>
<td>0.60</td>
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<tr>
<td></td>
<td>1510</td>
<td>200</td>
<td>0.60</td>
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<tr>
<td></td>
<td>250</td>
<td>500</td>
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</tr>
<tr>
<td></td>
<td>175</td>
<td>335</td>
<td>0.60</td>
</tr>
<tr>
<td>003/.512</td>
<td>1725</td>
<td>225</td>
<td>0.60</td>
</tr>
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Table 2 - Test Results, 90% $H_2O_2$
An exhaustive test series to determine the life of monolithic catalyst beds has not been performed. However, on NASA's ISTAR program, a single catalyst bed was used for the entire test series. The catalyst bed endured 678 seconds of testing at a throughput of 0.62 lbm/s/ft² and 238 seconds at 0.67 lbm/s/ft². There were 55 starts (full thermal cycles) without any measurable change in performance.

SUMMARY AND CONCLUSIONS
Platelet technology results in significant advances to the state of the art in heterogeneous catalyst bed designs. With relatively small total investment compact, high performing, and reliable catalyst beds were fabricated and demonstrated.

Monolithic catalyst beds offer the potential to significantly improve system performance, weight, and cost for many applications, particularly 90% H₂O₂ concentrations and below.

REFERENCES