LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-TR-219454

Rate Of Oxidation Of Plasma Polymer (GDP or CH)

Bob Cook, Steve Letts, Evelyn Fearon, Kelly Youngblood

March 1, 2006

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

Target Area Technologies Program

Mail Station L-481

Ext: 2-3117
Date: October 26, 2005

To: Distribution

From: Bob Cook, Steve Letts, Evelyn Fearon, Kelly Youngblood

Subject: Rate of oxidation of plasma polymer (GDP or CH)

Summary: There has been concern and uncertainty about the level of O incorporation in plastic shells prepared by coating mandrels with plasma polymer (GDP) and the Ge-doped analogs. In FY05 we undertook a controlled study of the rate of oxidation under various conditions, both to quantify the levels and to determine methods for restricting the levels. Our results are shown in the Figure below. In summary, the level can be kept to less than 0.2 atom % by pyrolyzing the shells at 300 °C (which is necessary for the removal of the PαMS mandrel), and then restricting the exposure to air to less than about 100 hours, which is easy to do. Based on this result we believe the NIF capsule specification for O can safely be lowered to 0.2 atom %, and this level should be used in future design calculations.

Experimental: To measure CH and CHGe oxidation we used PαMS shells that were weighed and then coated for about one week to deposit approximately a 75-μm thick layer. After coating the vacuum system was vented to 1 atmosphere pressure and allowed to purge with dry nitrogen for 16 hours. The coated shells were next divided into three groups and quickly weighed. The pan used to hold the shells was also weighed. The first group continued in air exposure and was weighed approximately every 30 minutes. The second and third groups were placed in tube furnaces and heated to 300°C in flowing nitrogen or in a flowing argon-hydrogen mixture (5% hydrogen or deuterium). The experimental results (Fig. 1) show that CHGe (about 1 atomic% germanium) oxidizes faster than CH. Earlier studies showed that CHGe contains Ge-H bonds that are highly reactive with air. Both the CH and CHGe pans, which have coatings that are thicker than the shell, showed similar oxidation rates. This shows that oxygen diffusion does not limit the oxidation rate. Both CH and CHGe after heating at 300 °C oxidize at a much lower rate than CH. We believe that heating CH and CHGe films increases chain mobility and allows the reactive fragments in the polymer film to react together rather than with oxygen. We also found that hydrogen or deuterium react with the CH material when heated to 300°C. FTIR showed incorporation of deuterium in CH films. The films heated in deuterium showed a very low rate of oxidation when exposed to air indicating that the free radical content was reduced.

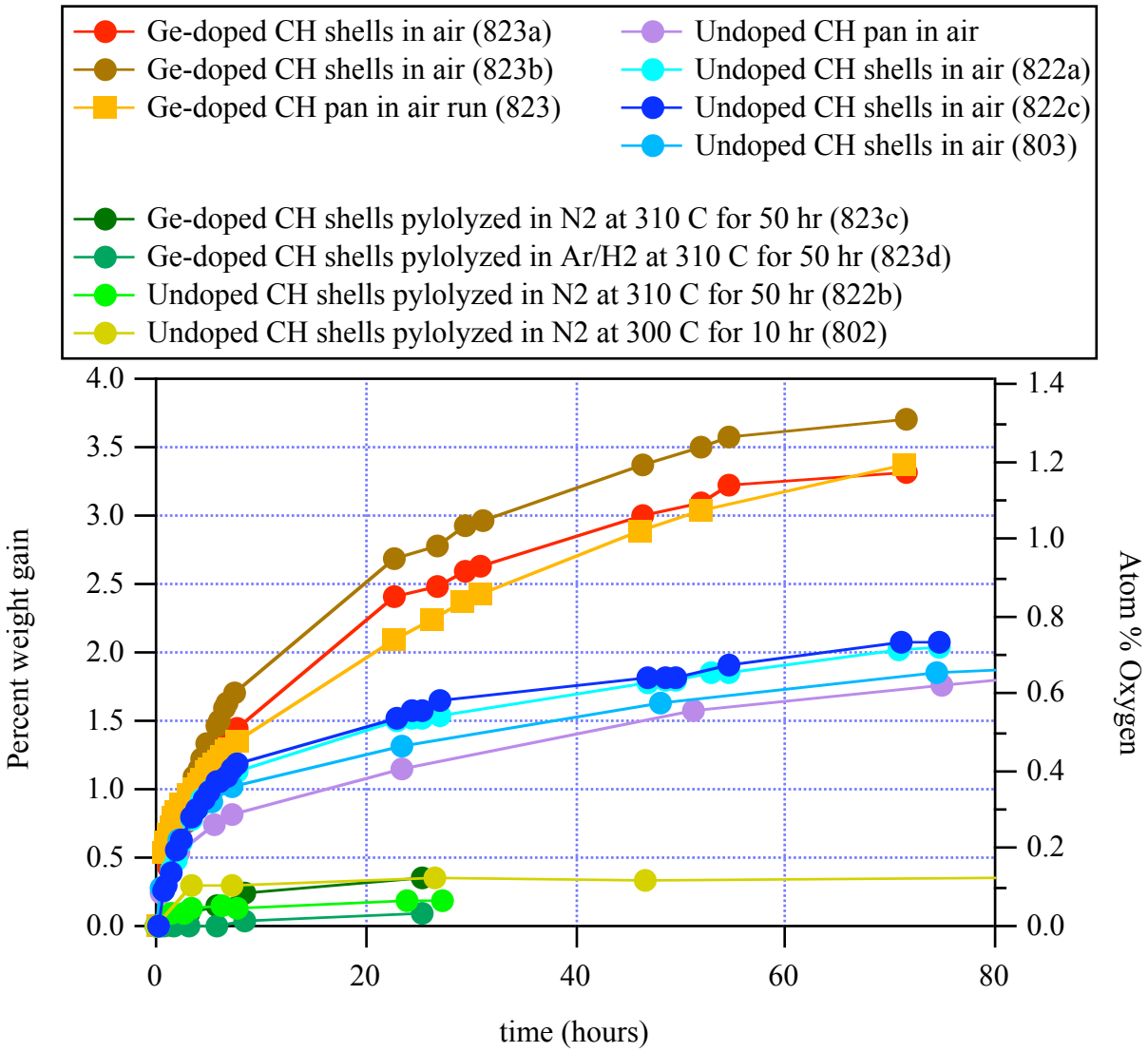

Figure 1. Shown are the oxidation rates for samples of CH and CHGe treated in a variety of ways. A weight gain of 1% represents about 0.35 atom % of O (assumes an empirical formula of $CH_{1.3}O_x$). A level of about 0.5 atom % is reached for the undoped CH shells pyrolyzed in N_2 at 300 °C for 10 hours after 50 days.

Distribution:

Jeff Atherton - atherton1@llnl.gov

Steve Buckley - buckley3@llnl.gov

Gary Deis - deis1@llnl.gov

Tom Dittrich - dittrich1@llnl.gov

John Edwards - edwards39@llnl.gov

Janelle Gunther - gunther2@llnl.gov

Steve Haan - haan1@llnl.gov

Bruce Hammel - hammel1@llnl.gov

Mark Herrmann - herrmann3@llnl.gov

Steve Letts - letts1@llnl.gov

John Lindl - lindl1@llnl.gov

Evan Mapoles - mapoles1@llnl.gov

Marty Marinak - marinak1@llnl.gov

Russell Wallace - wallace11@llnl.gov

Tom Bernat GA - bernat@fusion.gat.com, bernat2@llnl.gov

KC Chen GA - chenkc@fusion.gat.com

Hiabo Huang GA - huang@fusion.gat.com

Abbas Nikroo GA - nikroo@fusion.gat.com

Dave Steinman GA - steinman@fusion.gat.com

Richard Stephens GA - stephens@fusion.gat.com