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Measurement of Thermal Diffusivity and Flow Resistance for TCAP Materials

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Backgound

SRS uses the Thermal Cycling Absorption Process (TCAP) to separate isotopes of hydrogen. The frequency of thermal cycles is a limit of the productivity of the process and that frequency is largely determined by the thermal diffusivity of the absorbent material. For a given tube diameter, a larger thermal diffusivity decreases the time required for each cycle. In 1998, the Engineering Development Laboratory measured thermal diffusivity and thermal conductivity for three TCAP materials in helium, kieselguhr (aka diatomite), foam copper with kieselguhr filling the interstices and palladium coated on kieselguhr (Pd/k). The nominal helium pressures were 5 torr, 1 atm. and 5 atm. The following table contains the previously measured thermal diffusivities at one atmosphere of helium pressure.

material	thermal diffusivity, ft^2/hr
kieselguhr	0.021
copper foam and kieselguhr	0.048
kieselguhr and palladium	0.031

In the current activity this work was extended to more materials and included measurement of flow resistance. The gas was always helium near one atmosphere.

Activity

Hydrogen Technology Section requested an extension of the thermal diffusivity measurements to a total of seven materials, as follows, and all in helium at one atmosphere.

1. foam copper
2. foam copper coated with finely divided palladium
3. foam ZrO_2 ceramic
4. foam ZrO_2 ceramic coated with finely divided palladium
5. kieselguhr
6. foam copper filled with kieselguhr
7. Pd/k

Some of these materials would never be used by themselves as TCAP materials, but thermal transient measurements were made as an aid to understanding the heat transfer process. Thermal diffusivity and flow resistance were measured for all seven materials in helium, at approximately one atmosphere pressure. Helium was used as the fill gas rather than hydrogen, which would have absorbed on palladium and complicated interpretation of the results. The thermal diffusivities of helium, hydrogen and deuterium are 7.19, 9.17, and 6.73 ft^2/hr, so helium is a reasonable substitute for hydrogen isotopes.

Procedure
The following is a brief procedure. Each of the materials was loaded to a ½" diameter (i.d. = 0.43") stainless steel tube about 5" long. The test piece was connected to a vacuum pump, baked in an oven to drive off moisture and periodically weighed until the mass no longer changed. After drying, the test piece was again evacuated and filled with helium at room temperature and about one atmosphere. The test piece was connected to a pressure transducer as shown in Figure 1. Each test piece was quickly immersed in a stirred water bath maintained at about 80°C, which caused the pressure to increase. There were three or four repetitions per sample. One of the test pieces contained only helium and no TCAP material. An aluminum cylinder having the same dimensions was simultaneously immersed to allow measurement of the heat transfer coefficient. In addition, a thermocouple was simultaneously immersed to mark the moment of immersion. Data from the pressure gage and the two thermocouples were initially recorded every 0.2 seconds. Later in each run the rate of data logging was reduced. Rates of pressure rise were compared for the different samples. Heat transfer coefficient in the stirred bath was computed using the temperature readings at the centerline of the aluminum cylinder.

Later, helium was pumped through four different materials in a test piece and also an empty test piece, for a range of superficial velocities and the pressure drop was measured using the apparatus in Figure 2. Helium flow was measured using a rotameter that had been previously calibrated with helium by displacing water from an inverted graduated cylinder. For the flow resistance tests, there were three repetitions per sample. The pressure drop for the TCAP material was calculated as the pressure drop with the TCAP material in the test piece minus the pressure drop for flow in the empty test piece.

Results
Thermal diffusivity transients were run three or four times per material. Figure 3 shows typical reproducibility, which was excellent. Figure 4 compares the normalized transient pressure responses for the seven materials. Table 1 below shows the time constants, τ, for the transient response. Time constant is defined as the period of time required for a change in pressure that was 63.2% of the total change in pressure. Copper foam with Pd/k and palladium on copper foam gave the fastest response for potential TCAP materials. Palladium on ZrO_2 ceramic foam gave the slowest response for potential TCAP materials.

Table 1 Summary of Uncorrected Time Constants for TCAP Materials with Helium

material	time constant, seconds	mass, grams	sample length, inches
empty, He only	1.4	0.001	4.50
Pd/k	5.7	8.278	4.00
Pd on Cu foam	3.1	14.227	5.12
Pd on ZrO2 foam	10.4	16.512	5.05
Cu foam and kieselguhr	7.2	Cu = 8.337, k = 5.948	4.95
ZrO2 foam only	16.9	12.765	5.00
Cu foam and Pd/k	3.3	Cu = 8.337, Pd/k = 8.409	4.95
Cu foam only	2.4	8.337	4.95
kieselguhr only	8.0	4.970	4.00

It was desired to calculate the thermal diffusivity for the TCAP materials, however, the presence of the stainless steel tube was a complicating factor. The tube absorbed some heat which delayed the heat-up of the TCAP materials inside the tube. From the previous report, the following equation was used to compute thermal diffusivity, α, when the effect of the stainless steel tube is negligible and R is radius.

$$\alpha = 0.111\ R^2 / \tau \qquad (1)$$

Compensation was made for the presence of the stainless steel tube using the thermal transient with only helium inside the tube. As was mentioned before, helium has a high thermal diffusivity, 7.19 ft^2/hr. By comparison, TCAP materials with helium in the interstices have a much lower thermal diffusivity. For example, in the previous measurements the thermal diffusivity of Pd/k was 0.031 ft^2/hr. The method for compensation took advantage of the fact that the thermal diffusivity of helium is more than two orders of magnitude greater than the TCAP materials but the response time in Table 1 was only about a factor of five less. The response time for helium, 1.4 seconds, was due primarily to the presence of the stainless steel tube. Equation 1 and the literature value of thermal diffusivity, 7.19 ft^2/hr, were used to back calculate the helium response time if the tube had not been present, 0.0178 seconds. As compensation, the difference between 1.4 seconds and 0.0178 seconds was subtracted from all of the time constants in Table 1.

Results for raw and corrected time constant, thermal diffusivity and bulk density are listed in Table 2. Among actual TCAP materials, palladium on copper foam and Pd/k with copper foam had the highest thermal diffusivities. Copper foam also had a high thermal diffusivity, but by itself, is not a TCAP material. Pd/k and kieselguhr had about the same measured thermal diffusivities in the current study and the 1998 study. However, copper foam plus kieselguhr had a lower thermal diffusivity in the current study compared to the 1998 study. This may be the result of different bulk densities for material from different batches or different vendors. The bulk densities of just copper foam were 0.61 g/mL and 0.708 g/mL in 1998 and the present, respectively. The bulk densities of copper foam plus kieselguhr were 0.98 g/mL and 1.213 g/mL, in 1998 and the present, respectively.

The thermal transient for the aluminum cylinder was used to measure heat transfer coefficient in the agitated bath. Initially, it was planned to use this information in the data analysis. However, this was never done. For the record, the heat transfer coefficient was 400 btu/ft^2 hr F. Heat transfer coefficient was computed in the following way. Figure 5 was extracted from the previous report [2] and plots dimensionless temperature at the centerline of a cylinder as a function of Fourier number, $\alpha\ \tau / R^2$, with Biot Number, h R/k, as a parameter. The time constant for the 63% response of the aluminum cylinder was 3.8 seconds. Using a radius of 0.25" and a thermal diffusivity of 3.38 ft^2/hr for aluminum gives a Fourier number of 8.22. The parametric value of the Biot number that corresponds that percentage response and that Fourier number is 0.07 ± 0.01. Therefore, the heat transfer coefficient was 400 ± 60 btu/ft^2 hr F.

Table 3 lists pressure drops measured for helium flows through TCAP materials. Pressure drop was proportional to velocity over the range tested, 6 cm/sec to 33 cm/sec. Figure 6 plots pressure drop as a function of helium velocity for Pd/k and kieselguhr. Figure 7 plots pressure drop for copper foam with palladium and for ceramic foam with palladium. For flow through a porous bed, pressure drop is proportional to viscosity. The viscosities of helium, hydrogen and deuterium at ambient conditions are 0.019 cP, 0.0088 cP and 0.012 cP, respectively. Therefore, pressure drops would be less with hydrogen isotopes than with helium.

Table 3 Pressure Drops for Helium Through TCAP Materials

material	Pressure drop, inch water per foot length, @ 20 cm/sec
Pd/k	30 ± 1
Kieselguhr	16 ± 1
Copper foam with Pd	0.3 ± 0.05
Ceramic foam with Pd	0.1 ± 0.05

Acknowledgement
Jerry Corbett designed, built and tested the apparatus for both the thermal transients and pressure drop measurements. Vern Bush assembled and programmed the data acquisition system.

References

1. Steimke, “TCAP Heat Transfer Measurements”, SRT-ETF-98002
2. Steimke and M. D. Fowley, “Transient Heat Transfer in TCAP Coils”, WSRC-TR-98-00431, January 1999.

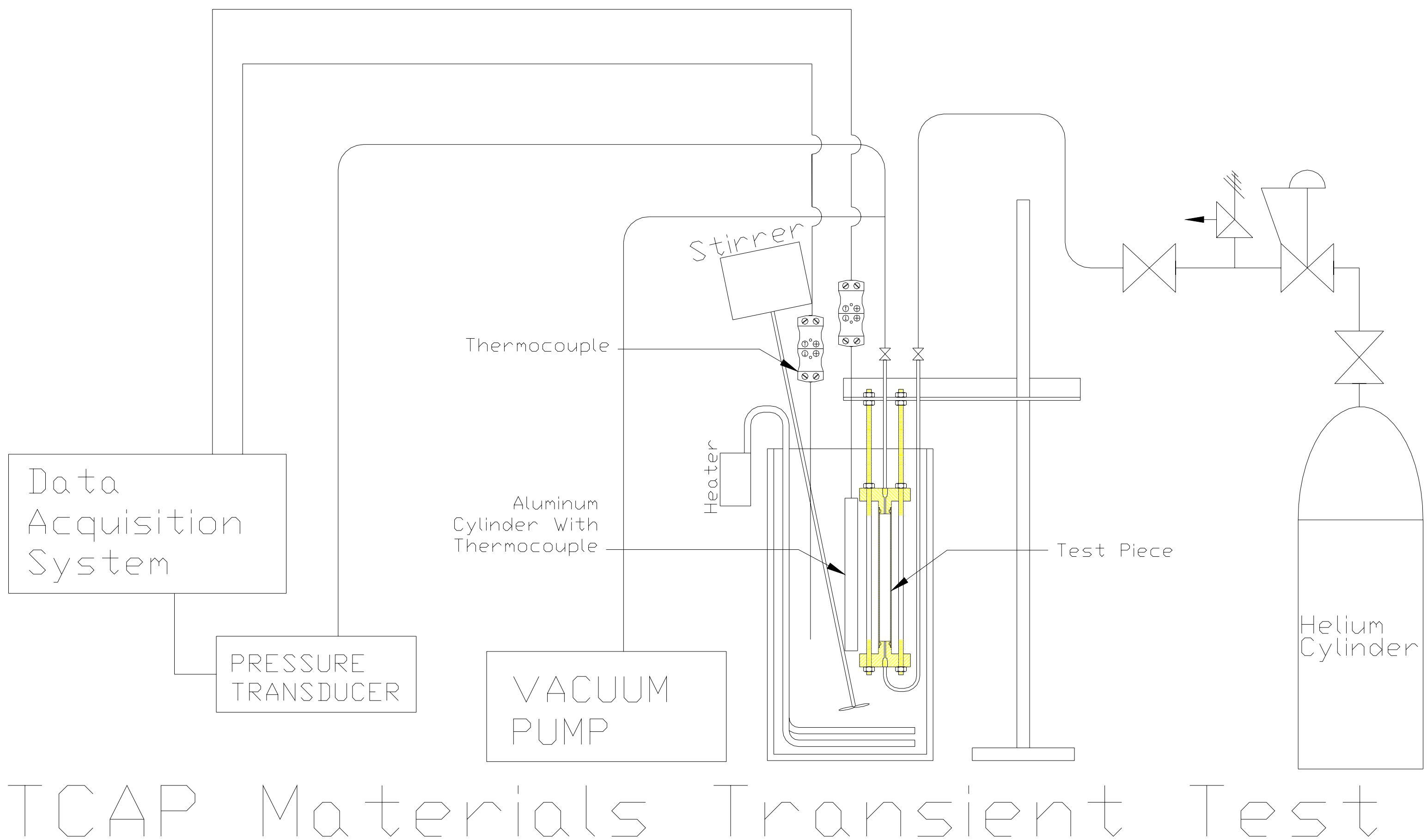

Figure 1 Test Hardware for Thermal Transient Testing

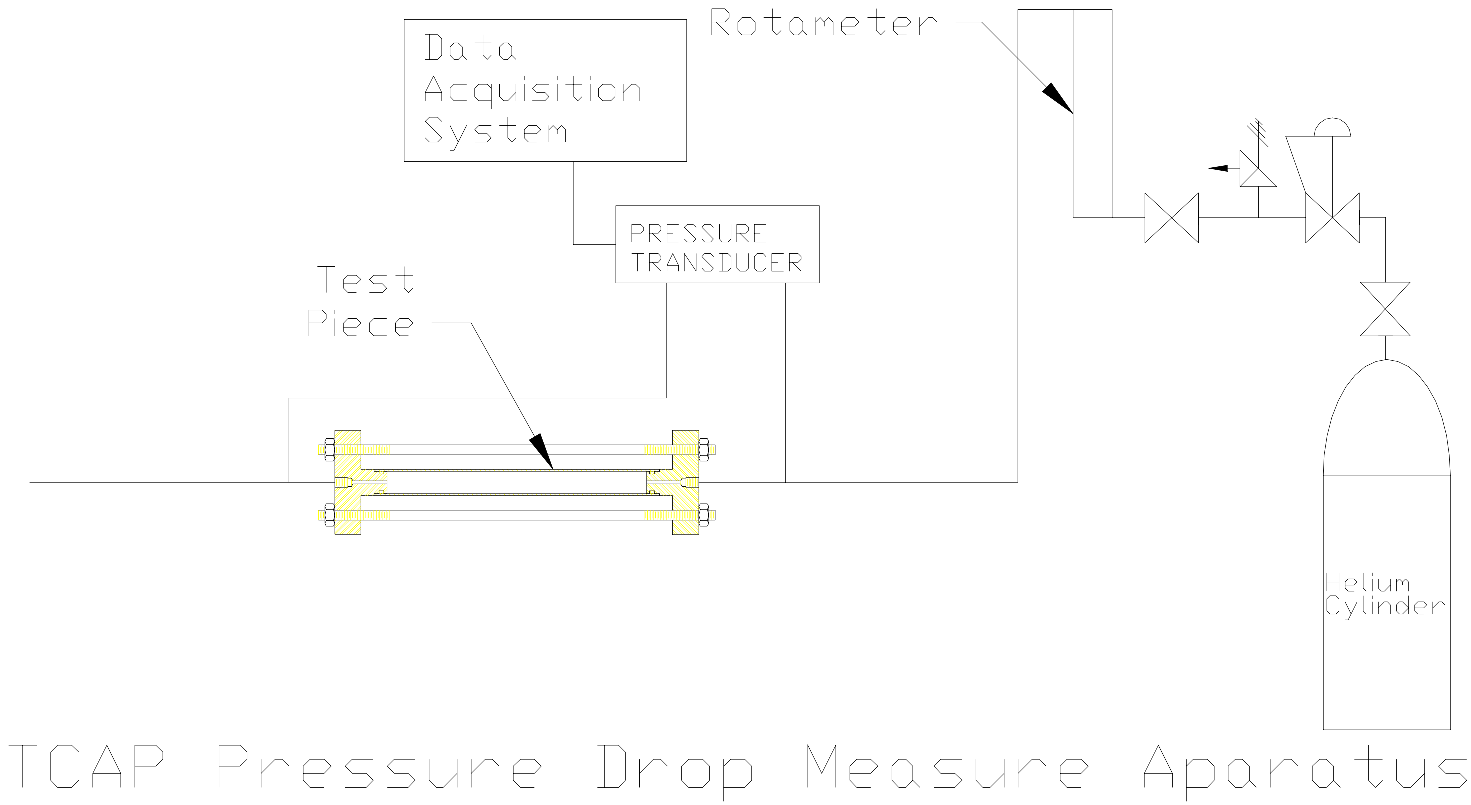

Figure 2 Test Hardware for Pressure Drop Measurement

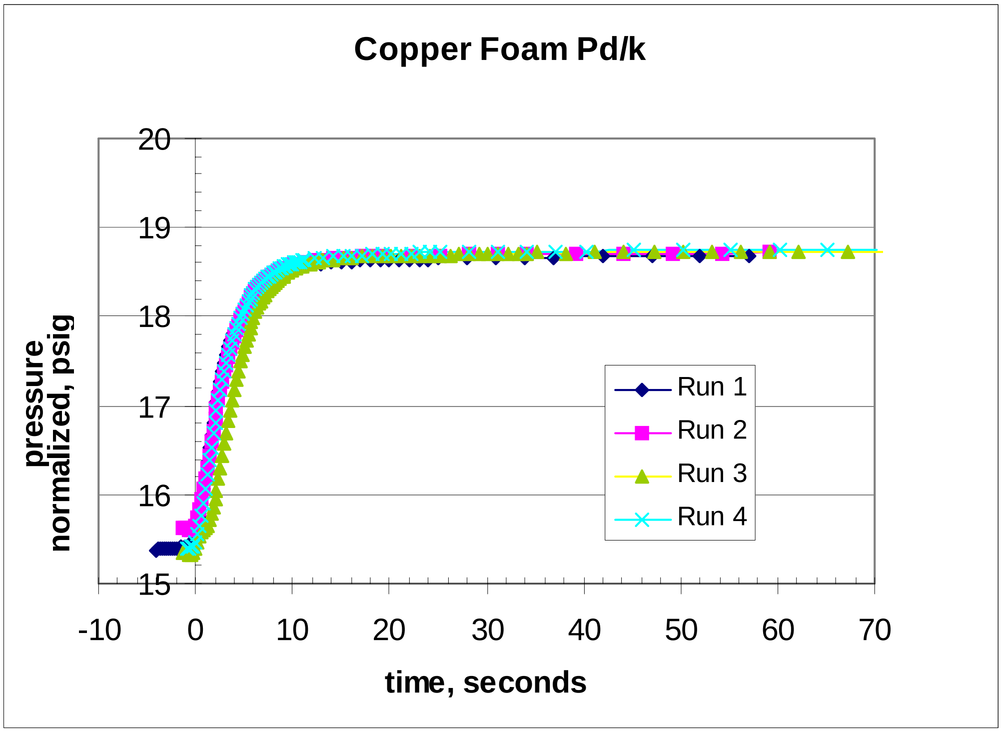

Figure 3 Normalized Transient Pressure Response

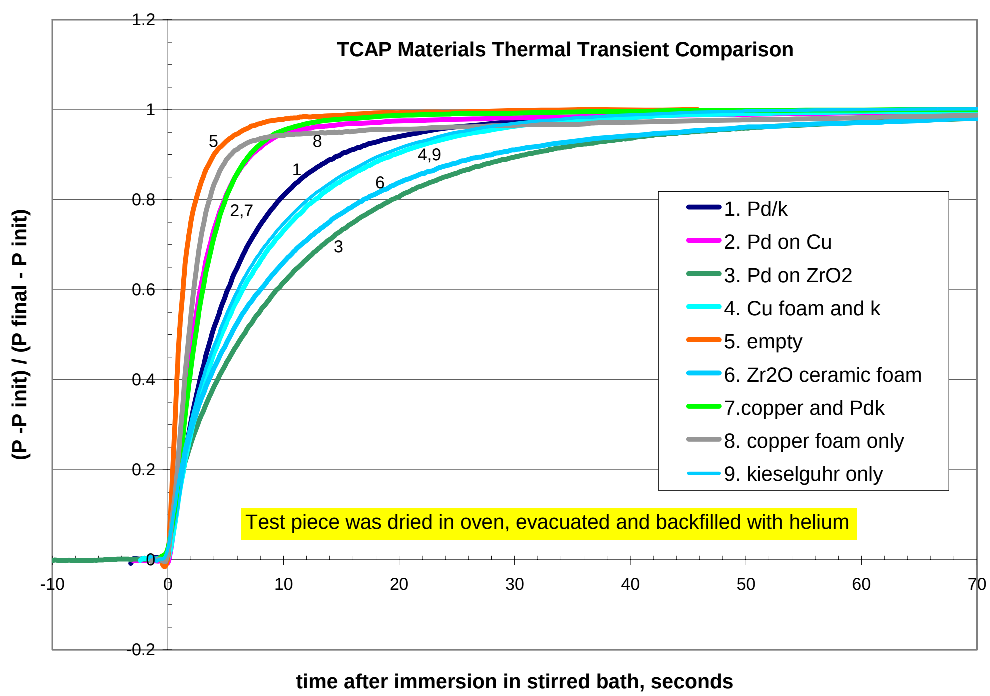

Figure 4 Comparison of Transient Responses for Different TCAP Materials

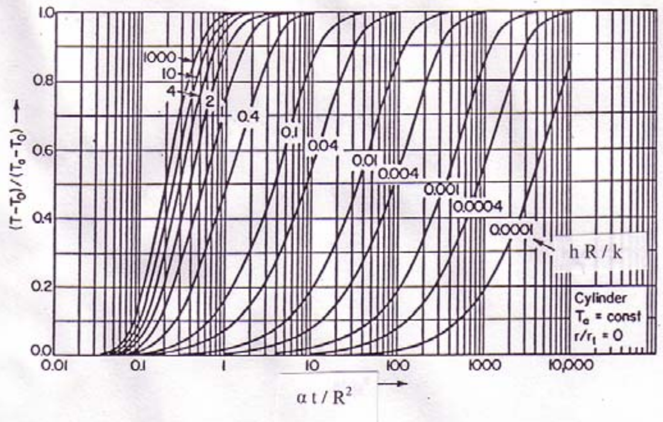

Figure 5 Centerline Temperature Response of Long Cylinder After Sudden Exposure to Uniform Convective Environment

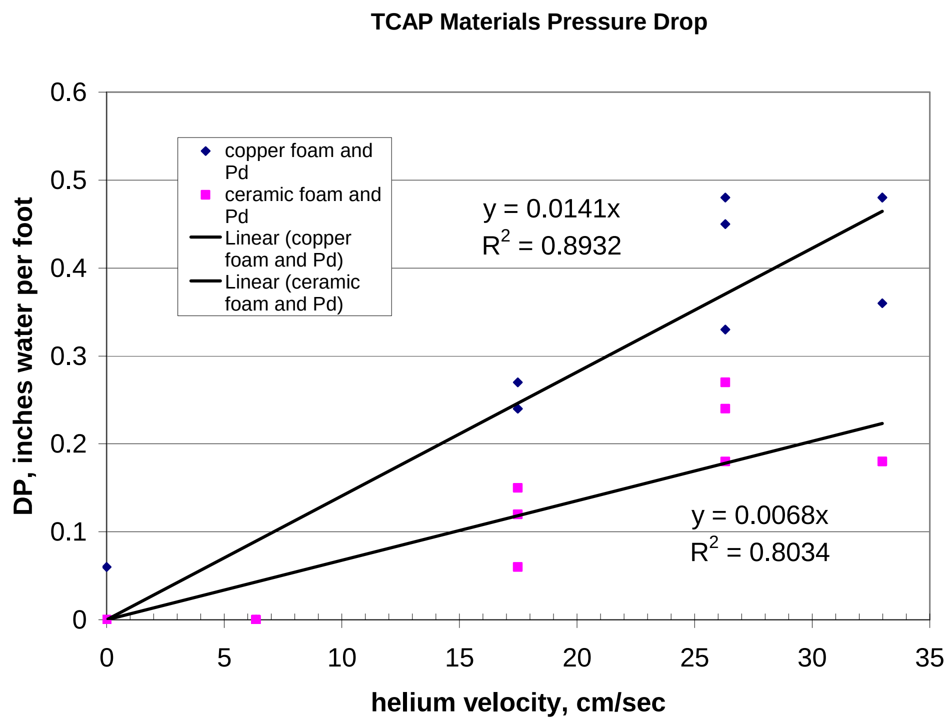

Figure 6 Pressure Drop for Palladium on Copper Foam and Zirconia Ceramic Foam

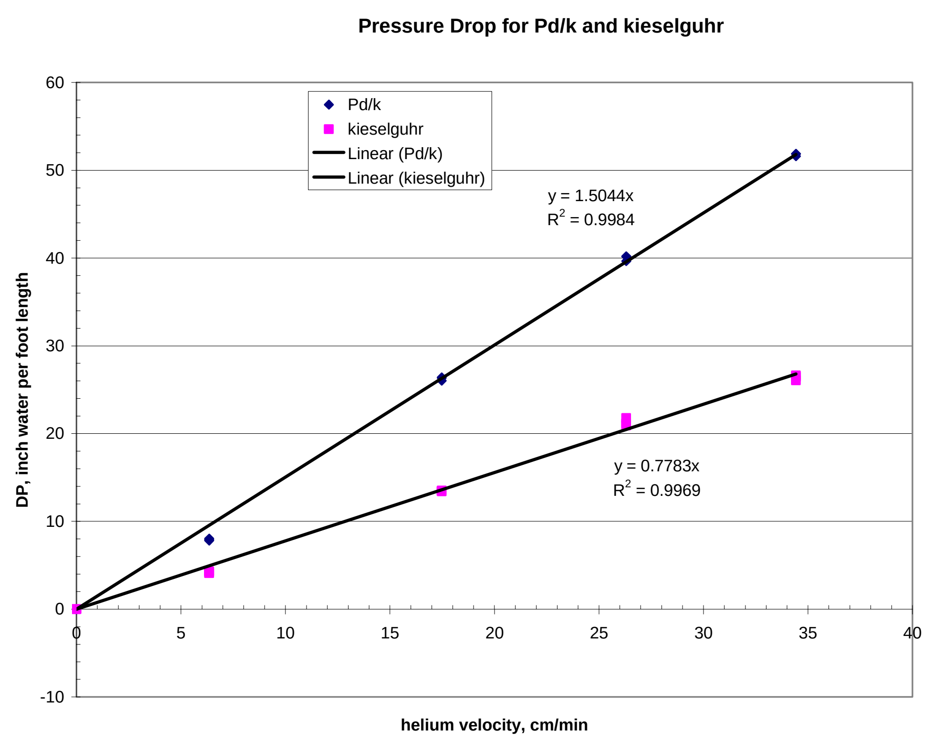

Figure 7 Pressure Drop for Pd/k and Kieselguhr

Table 2 Data and Results from Thermal Transient Tests of TCAP Materials

material	measured time constant, sec	corrected time constant, sec	mass of TCAP material, g	length of TCAP material, inches	current bulk density, g/mL	current thermal diffusivity, ft^2/hr	1998 thermal diffusivity, ft^2/hr	1998 bulk density, g/mL
empty, He only	1.4	0.02	0.001	4.5	0.000	7.190	7.19	
Pd/k	5.7	4.32	8.278	4.001	0.869	0.030	0.031	0.69
Pd on Cu foam	3.1	1.72	14.227	5.115	1.169	0.075		
Pd on ZrO2	10.4	9.02	16.512	5.045	1.375	0.014		
Cu foam and kieselguhr	7.2	5.82	14.285	4.95	1.213	0.022	0.048	0.98
ZrO2 only	16.9	15.52	12.765	5	1.073	0.008		
Cu foam and Pd/k	3.3	1.92	16.746	4.95	1.422	0.067		
Cu foam	2.4	1.02	8.337	4.95	0.708	0.126		0.61
kieselguhr only	8	6.62	4.97	4.001	0.522	0.019	0.022	0.452