EFFECT OF GEOSYNCHRONOUS ALTITUDE RADIATION ON PERFORMANCE OF Ni/H_2 CELLS

C K DYER

Bell Laboratories, 600 Mountain Avenue, Murray Hill, NJ 07974 (U S A)

Summary

Nickel/hydrogen cells are under consideration as eclipse season power sources for long-life communications satellites in geosynchronous orbit (GEO). There is concern that damage to polymers in key components of these cells may arise from irradiation with high energy protons and electrons at the fluxes present at GEO altitudes. Nickel/hydrogen cells have been subjected to fluences of electrons and protons which simulate exposure to the GEO environment for more than 10 years. The cells show promise for considerable radiation tolerance in this new application.

Introduction

A new application for Ni/H_2 cells is to power communications satellites during their eclipse seasons, which last for 44 days each, during the Spring and Fall equinoxes. The satellites, which are in geostationary orbit (GEO) over the equator, experience eclipses of the sun, by the earth, for up to 70 minutes each day during these periods.

Until 1983, Ni/H_2 cells had not been flown in GEO satellites and although ground testing shows that they should have a much longer life than Ni/Cd cells, they may be more susceptible to space radiation than Ni/Cd because of two of their key features — Teflonated negative electrodes and polymer pressure seals (Fig. 1). Failure of either would cause the cell to become inoperable through leakage of hydrogen or loss of hydrophobicity of the negative electrode, leading to a high cell polarization.

The Teflon or nylon pressure seals have proved stable under Co-60 irradiation (β^- and γ emissions at 0.3 and 1.5 MeV, respectively), but this does not give the appropriate radiation. High energy electrons and protons exist at GEO altitudes and have been recently modeled by Stassinopoulos [1]. His radiation model was used in the present experimental work to simulate conditions that Ni/H_2 cells would experience over 10 years on GEO duty.

Elsevier Sequoia/Printed in The Netherlands

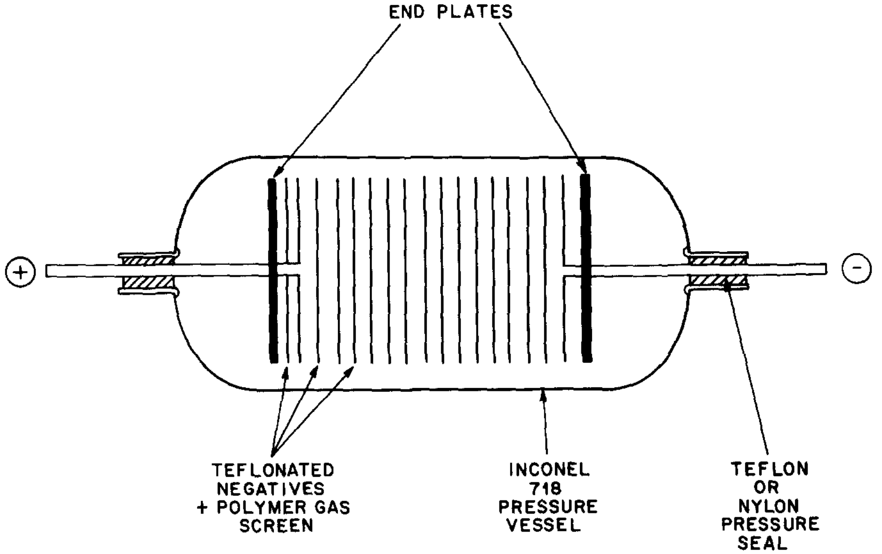

Fig 1 Schematic design of a Ni/H_2 cell

Experimental and results

The GEO environment consists of electrons and protons trapped in the Van Allen radiation belts (energies up to 5 and 4 MeV, respectively) together with solar flare protons (to >100 MeV). Trapped protons are not energetic enough to penetrate the Inconel 718 casing of Ni/H_2 cells and so only trapped electrons and solar flare protons were considered. In Table 1, the energy/integral-fluence spectra are presented, where the fluence for a given particle energy is the flux integrated for the time specified. Figure 2 shows the range, R, in Ni of protons (p) and electrons (e) [2, 3].

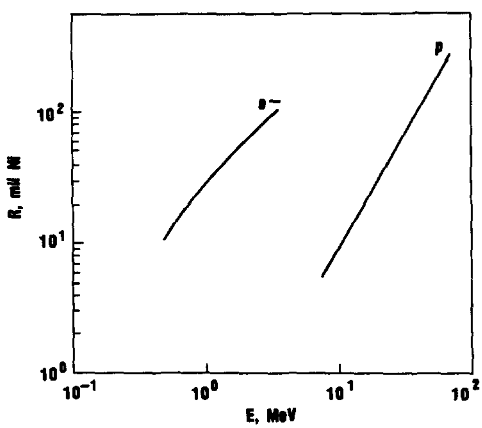

Fig 2 Range, R, in Ni of electrons and protons as a function of their energy

TABLE 1

Radiation environment

A Trapped electrons

Energy E (MeV)	Fluence (per day (> E)) (e/cm^2)
0 04	$2\ 7 \times 10^{12}$
0 10	$1\ 9 \times 10^{12}$
0 5	$2\ 9 \times 10^{11}$
1 0	$6\ 0 \times 10^{10}$
2 0	$3\ 4 \times 10^{9}$
2 25	$1\ 7 \times 10^{9}$
2 50	$8\ 96 \times 10^{8}$
2 75	4.7×10^{8}
3 0	$2\ 6 \times 10^{8}$
4 0	$7\ 8 \times 10^{7}$
5 0	$1\ 6 \times 10^{7}$

For worst case longitude (160° W), orbit inclination 0°, latitude 0°, apogee altitude 35 794 47 km, perigee altitude 35 794 47 km Omnidirectional flux

B Solar flare protons

Energy (MeV)	Fluence (over 11 years) ($protons/cm^2$)
>4	$5\ 5 \times 10^{10}$
>10	$3\ 1 \times 10^{10}$
>30	$9\ 7 \times 10^{9}$
>60	$3\ 1 \times 10^{9}$
>100	$1\ 0 \times 10^{9}$

Inconel 718 is a Ni-rich alloy with 17% Fe, 5% Nb + Ta, 3% Mo. Two designs of Ni/H_2 cell were tested for radiation tolerance at GEO altitudes. The minimum pressure vessel wall thickness for cells of either design was ~0.020 in. around the cylindrical electrode-containing area (Fig. 1). Additional radiation shielding is provided in some cells (made by Hughes Aircraft Co. ("HAC"), Los Angeles, California) by Inconel end-plates (which, in addition to thinner hemispherical end sections 0.012 in. thick, provide at least a 0.020 in. radiation shield through Inconel before reaching the first negative electrode) [4]. In the "COMSAT" design (Comsat Labs, Clarksburg, Maryland) pressure vessel walls were 0.020 in. thick throughout with polymer end-plates. Both designs would be mounted in spacecraft with additional shielding from Al mounting sheaths. Of course, there is additional internal shielding from the layers of Ni sinter of the positive electrodes in the stack. Lower limit energies, however, were selected on the basis of the 0.05 cm (0.020 in.) of pressure vessel shielding (Fig. 2). This lower limit was 0.77 MeV for electrons and 16 MeV for protons. From Table 1, the fluences of all particles above these energy levels over 10 years are $\sim 3.6 \times 10^{14}/cm^2$ for electrons (worst case) and $\sim 2 \times 10^{10}/cm^2$ for protons.

TABLE 2

Voltage (V) and pressures (P) of an HAC cell before and after ırradıatıon

1 Charge 6 A, 11 °C, 21 5 A h

Cycle no	V(Volts) (5 A h)	V (10 A h)	V (15 A h)	V (17 5 A h)	V (20 A h)	V (21 5 A h)
1	1 434	1 450	1 467	1 484	1 537	1 533
2	—	1 452	1 469	1 485	—	1 534
3	—	1 453	—	1 492	1 539	1 535
4	1 435	1 451	1 467	1 484	1 533	—
After electron ırradıatıon						
5	1 448	1 460	1 472	1 481	1 519	—
6	1 440	1 457	1 470	1 484	1 537	1 534
7	—	—	—	1 486	1 537	1 532
8	1 442	1 458	1 469	1 481	1 534	1 534
After proton ırradıatıon						
9	1 441	1 460	1 472	1 484	1 533	1 528
**10	1 437	—	—	1 489	1 542	1 539
11	1 434	—	1 469	1 487	1 535	1 537
12	1 438	1 454	1 468	1 485	1 536	1 533
13	1 435	1 455	1 469	1 485	1 537	1 530

**Temperature fluctuatıons durıng cycle

2 Dıscharge 12 A, 11 °C, to 1 0 V

Cycle no	V(Volts) (5 A h)	V (10 A h)	V (15 A h)	V (17 5 A h)	V (19 A h)	CAP (A h)	P_{EOC} (psı)	P_{EOD} (psı)
1	1 316	1 295	1 267	1 241	1 102	19 67	435	220
2	1 315	1 295	1 266	1 240	—	19 60	425	220
3	1 316	—	—	1 246	1 114	19 75	—	210
*4	1 306	1 285	1 256	1 154	—	18 33	390+	215

After electron irradiation								
5	1 322	1 306	1 279	1 234	1 054	19 27	410	200
6	—	1 303	1 274	1 243	1 050	19 26	405	200
7	1 321	1 303	1 273	1 250	1 121	19 66	410	200
8	1 321	1 303	1 272	1 244	1 039	19 20	405	200
After proton irradiation								
9	1 322	1 304	1 274	1 240	1 016	19 06	400	185
10	—	1 302	1 271	1 250	1 080	19 40	395	185
*11	1 311	1 291	1 260	1 177	—	18 20	370+	180++
12	1 321	—	1 269	1 252	1 137	19 66	390	180
13	1 321	1 300	1 269	1 247	—	19 40	385	175

++No change after 19 h
*After 22 5 h on open circuit at 13 °C
+BOD = Beginning of Discharge, EOD = End of Discharge, EOC = End of Charge

TABLE 3

Pressures (P in psi) and voltages (V) of COMSAT cell before and after irradiation

Cycle no		5 A h	10 A h	15 A h	17 5 A h	20 A h	23 A h	25 A h
1 Charge 6 A, 11 °C, 25 A h								
1	V	1 432	1 448	1 461	1 470	1 487	1 540	1 528
	P	142	206	272	304	337	377	400
2	V	—	1 450	1 462	1 470	—	1 541	1 531
	P	—	203	269	300	—	372	395
3	V	—	1 449	—	1 472	1 487	1 541	1 532
	P	—	202	266	305	332	371	394
4	V	1.431	1 448	1 460	1 469	1 485	—	—
	P	147	210	270	301	—	—	—
After electron irradiation								
5	V	1 440	1 454	1 465	1 472	1 482	1 530	1 533
	P	120	187	257	290	325	365	397
6	V	1 437	1 451	1 463	1 471	1 487	—	1 526
	P	140	204	267	300	335	375	398
7	V	—	—	—	1 471	1 488	1 538	1 528
	P	—	—	—	298	331	367	390
8	V	1 436	1 451	1 462	1 470	1 484	1 538	1 529
	P	129	178	244	284	316	350	369
After proton irradiation								
9	V	—	1 452	1 465	1 473	1 485	1 536	1 528
	P	—	166	228	261	302	334	366
10	V	1 436	—	—	1 473	1 490	1 540	1 530
	P	129	—	—	264	296	336	361
11	V	1 432	—	1 462	1 472	1 489	1 536	1 527
	P	108	—	238	269	301	342	368
12	V	1 434	1 449	1 462	1 471	1 487	1 538	—
	P	101	165	230	261	294	333	362
13	V	1 440	1 450	1 462	1 471	1 486	1 537	—
	P	97	162	226	569	292	333	354

2 Discharge 12 A, 11 °C, to 1 0 V

Cycle no		5 A h	10 A h	15 A h	17 5 A h	20 5 A h	CAP (A h)	P_{EOC} (psi)	P_{EOD} (psi)
1	V	1 316	1 295	1 264	1 246	1 203	22 40	400	88
	P	325	255	186	151	113			
2	V	1 316	1 295	1 283	1 245	1 205	22 50	395	83
	P	319	252	182	149	109			
3	V	1 316	—	—	1 246	1 210	22 60	394	82
	P	322	—	—	150	110			
*4	V	1 305	1 281	1 251	1 234	1 117	21 02	357+	93
	P	287	223	167	138	100			
After electron irradiation									
5	V	1 316	1 297	1 268	1 252	1 212	22 18	397	81
	P	326	258	195	170	112			
6	V	—	1 294	1 264	1 247	1 210	22 21	398	91
	P	—	250	185	151	109			
7	V	1 315	1 295	1 266	1 247	1 216	22 32	389	83
	P	319	256	188	155	113			
8	V	1 315	1 293	1 263	1 246	1 208	22 10	369	61
	P	292	228	158	125	83			
After proton irradiation									
9	V	1 315	1 295	1 265	1 246	1 205	21 97	366	57
	P	287	219	152	118	78			
10	V	—	1 295	1 264	1 246	1 210	22 18	361	51
	P	—	215	147	113	73			
*11	V	1 305	1 282	1 250	1 232	1 064	20 67	312+	50
	P	250	188	123	91	51			
12	V	1 313	—	1 259	—	1 191	21 80	361	46
	P	277	—	141	—	65			
13	V	1 315	1 293	1 263	1 245	1 212	22 28	354	43
	P	279	208	140	107	68			

*After 22 5 h on open circuit at 13 °C
+BOD

A COMSAT- and an HAC-built cell were cycled until highly reproducible pressures, voltages on charge and discharge (at the mV level), and cell capacities were reached. The data so generated are shown in Tables 2 and 3. The initial three cycles show highly reproducible values for voltages and pressures during charge and discharge. The fourth cycle was interrupted before discharge for an open circuit stand of 22.5 h. This was to determine the normal rate of recombination or self-discharge of the cells and was to be used to determine whether any slow leaks result from irradiation. Since the normal charge/discharge cycle used took only ~6 h, a slow leak may not have been easy to determine from the pressure readings (the HAC cell was fitted only with a pressure gauge, though the COMSAT cell had a precision strain gauge attached).

(A) Electron irradiation

The cells were taken to a Dynamitron based at the Jet Propulsion Laboratory, Pasadena, CA, for the electron irradiation part of the experiment. They were mounted at an angle of 45° in air, 36 in. from the Dynamitron port (Fig. 3). The maximum beam energy at the source was ~2.7 MeV. The calculated losses in energy due to interposed scatter foils and the air are shown in Table 4 for the two energies used in the experiment. Interpolated values from standard tables were used [2].

The fluxes were measured at the position of the cells by a Faraday cup and the fluences used were both ~25% more than predicted by the worst case model (Table 1), and a factor of approximately 2.5 times and 4 times

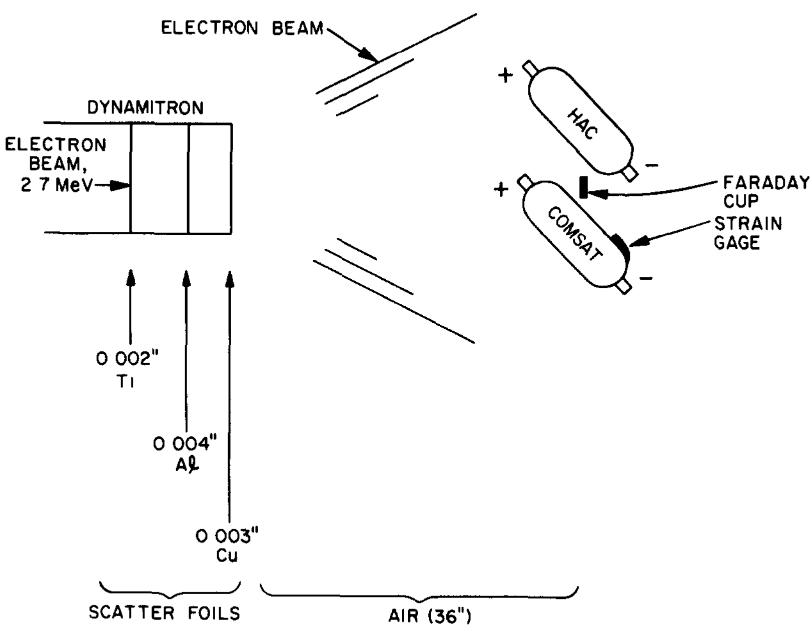

Fig 3 Sketch of the electron irradiation experimental arrangements

TABLE 4

Electron and proton irradiation energies and fluences used (> 10 year simulation)

A Higher-energy electrons

	g/cm^2	Incident beam		Exit beam energy (MeV)
		Energy (MeV)	Range (g/cm^2)	
Ti	0 023	2 700	1 781 - 0 023	2 635
Al	0 027	2 635	1 623 - 0 027	2 592
Cu	0 068	2 592	1 754 - 0 068	2 493
Air	0 118	2 493	1 368 - 0 118	2 287

Energy 2 287 MeV
Flux 1×10^{10} $e/cm^2 s^{-1}$
Fluence used 7×10^{12} e/cm^2

B Lower-energy electrons

	g/cm^2	Incident beam		Exit beam energy (MeV)
		Energy (MeV)	Range (g/cm^2)	
Ti	0 023	1 200	0 743 - 0 023	1 168
Al	0 027	1 168	0 661 - 0 027	1 128
Cu	0 068	1 128	0 714 - 0 068	1 035
Air	0 118	1 035	0 512 - 0 118	0 836

Energy 0 836 MeV
Flux 2×10^{11} $e/cm^2 s^{-1}$
Fluence used $4\ 4 \times 10^{14}$ e/cm^2

C Higher-energy protons
Source beam energy = 153 4 MeV

Beam energy (MeV)	Residual range required (g/cm^2)	Degrader range* required (g/cm^2)	Actual degrader used (g/cm^2)	Fluence required (protons/ cm^2)	(dE/dx) Tissue (MeV g^{-1} cm^2)	Dose used (rads)
100	7 814	8 808	8 836	2×10^9	7 27	233
60	3 128	13 494	13 447	6×10^9	10 76	1033
30	0 893	15 729	15 708	2×10^{10}	18 74	5997

$$\text{Dose} = \text{Fluence} \times \frac{dE}{dx} - 6\ 25 \times 10^7$$

*Range in Lucite (Janni's Tables) for 153 4 MeV protons = 16 622 g/cm^2

higher than the best case model (parking longitude 70° W [1]) for the low and high energy fluences, respectively. The variation with parking longitude at 0° latitude is due to the relative declination of the earth's magnetic field.

In both Ni/H_2 cell designs, the electrochemical stacks are terminated at both ends with the negative electrode and its screen. So, for these electrodes, there was only end-plate shielding. The strain gauge of the COMSAT cell was well shielded from electrons by the whole cell (see Fig. 3).

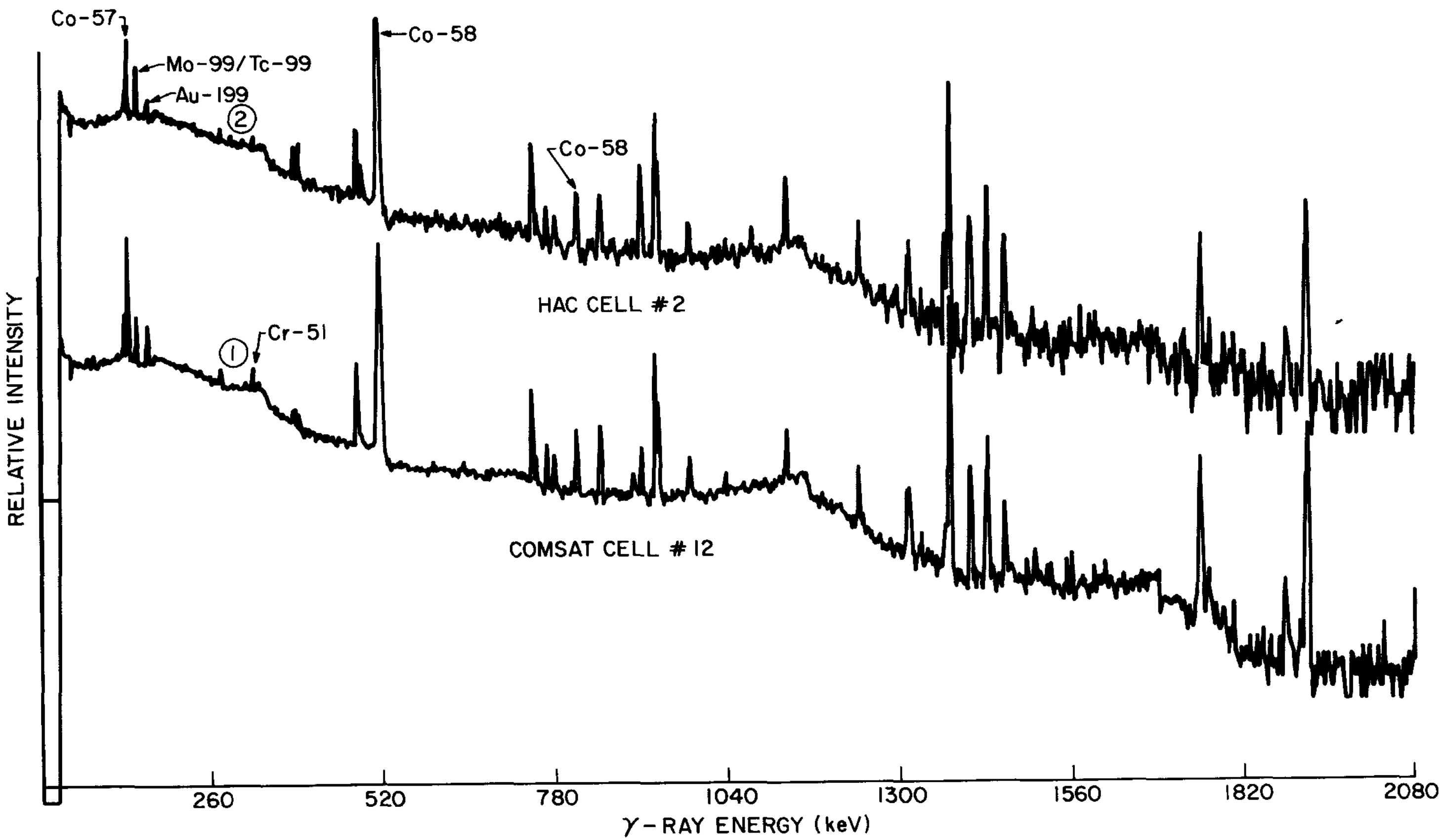

Fig 4 Emitted γ-ray spectrum after proton irradiation of cells

Electrochemical performance of both cells following irradiation was not significantly changed. Examination of Tables 2 and 3 shows that there was, in some cases, a small and similar increase in voltages on charge *and* discharge, so there was no increase in cell polarization. Capacities were marginally lower (by at most 2%). End of charge (EOC) and end of discharge (EOD) pressures for the HAC cell were both lower by ~20 psi (a substantial error of ~10 psi was possible in reading the HAC cell pressure gauge), while COMSAT cell pressures were almost unchanged.

(B) Proton irradiation

The cells were taken to the Harvard Cyclotron Laboratory for the proton irradiation part of the experiment. The cells were mounted in air at an angle of 45° in the same relative position to the proton source as for the electron irradiation experiment (Fig. 3). The proton beam energy from the synchrocyclotron was 154.4 MeV (calculated from an equivalent water range of 16.25 cm). For the required beam energies of 100, 60 and 30 MeV, Lucite degraders of 8.808, 13.494 and 15.729 g/cm^2 were interposed between the beam and the cells. Dosages (Table 4) were based on fluences that were double those predicted (over 11 years) by the radiation model (Table 1) at the three energies selected for the experiment. The beam width was 20 cm, which allowed both cells to be irradiated simultaneously.

At the end of the irradiation, a radiation monitor detected 5 - 20 mR/h from the surface of the cells. This fell to ~1 mR/h after 20 min. The emitted γ-radiation spectrum is shown in Fig. 4. The presence of Au-199, Co-58, and Co-57 were identified from the spectrum.

Electrochemical performance is shown in Tables 2 and 3 and is not significantly changed by proton irradiation. There are no significant changes in electrode polarizations on charge and discharge. Delivered capacities are slightly lower. Cell pressures, P_{EOC} and P_{EOD}, show a declining trend. This does not seem to be due to gas leakage, however, since open circuit stand tests were not followed by lower P_{EOC}'s and P_{EOD}'s but seemed to remain constant for about one day (Cycle 11, HAC cell) or fall by ~5 psi over 2 days (both HAC and COMSAT cells). No hydrogen leaks have been detected externally using a monitor sensitive to 1×10^{-5} standard cc/s (≡50 psi over 10 years).

Discussion

The effect of the electron irradiation on initial cell performance would seem to be small, if any. The voltage, pressure, and capacity data for Ni/H_2 cells normally does change slowly with time [5] and treatment, so that the small changes seen in Tables 2 and 3 may not be due to the irradiation, and certainly do not indicate any deterioration in properties of polymers used in the negative electrodes (which would have immediately resulted in more polarization) or the seals (which would have affected the pressure readings by a general downward trend for both EOC and EOD values).

In relating the conditions of the electron beam test to what a cell might experience in actual orbit, the test conditions were more severe even than indicated by the received fluences because:

(1) The fluxes used in the test were many orders of magnitude greater than the spacecraft would experience in orbit.

(2) The flux was unidirectional in the test though in orbit the cells would experience omnidirectional flux which is less severe.

(3) There would, of course, be additional shielding from the cell mounting sheaths and the spacecraft itself.

(4) The worse case longitude (160° W) was used in calculating the required fluences.

Although only two electron energies were used, the contribution to the fluence of electrons with energies higher than 2.3 MeV is quite small and falls off rapidly with energy (Table 1).

There seems to be no effect of the proton irradiation on the electrochemical polarizations so, as with electrons, the negative electrodes are not susceptible. The downward trend in pressures cannot be linked to the irradiation at this time. Variation (downwards) in both EOC and EOD cell pressures have previously been observed in the absence of gas leakage and is usually due to recombination of hydrogen with residual, undischarged Ni oxyhydroxide, electrically isolated by processes described elsewhere [5].

Acknowledgements

The author thanks Dr B. Anspaugh of JPL for use of the Dynamitron, Dr A. Koehler of the Harvard Cyclotron Laboratory, Dr L. J. Lanzerotti, BTL, for helpful discussions on radiation models for GEO altitudes and for suggesting laboratories where the experiments could be performed, and Dr S. Voris, BTL, for providing the γ-ray spectra (Fig. 4).

References

1 E G Stassinopolos, *J Spacecraft, 17* (1980) 145
2 M J Berger and S M Seltzer, Tables of Energy Losses and Ranges of Electrons and Positrons, *NASA SP-3012*, 1964, NASA, Washington, DC
3 W H Barkas and M J Berger, Tables of Energy Losses and Ranges of Heavy Charged Particles, *NASA SP-3013*, 1964, NASA, Washington, DC
4 D R Warnock, *Proc Symp on Battery Design and Optimization*, Vol 79-1, Electrochemical Society, Inc , Princeton, NJ
5 C K Dyer, *J Electrochem Soc* , to be published