A MEANS TO MAKE OPEN-FACE CHARCOAL DETECTORS RESPOND CORRECTLY TO VARYING CONCENTRATION RADON FIELDS

Carl H. Distenfeld
TCS Industries Inc.
Harrisburg, PA

ABSTRACT

Ronca-Battista and D. Gray 87, outlined the poor response of open-face charcoal detectors to varying concentration radon fields. At worst, for two day exposures with open-face charcoal canisters, their Table 4 indicated a 75% under-response for radon concentrations that were 10 times higher during the first day of two, 10:1. TCS has made similar measurements with open-face and diffusion barrier detectors in 20:1, 1:20, and 1:1 fields. For the worst case 20:1, measurements indicate TCS two day open-face canisters under respond by 50%, while the Cohen and TCS diffusion barrier devices under responded by about 37%. The reasons for the under response are radon diffusion out of the charcoal due to the forces of lower concentration during the second half of the exposure, and uncompensated radioactive decay of radon gas.

TCS has developed a method to control exposure periods to exactly compensate for the dynamic adsorption characteristic of the two day open-face and diffusion barrier charcoal canisters. The system responds correctly to the average exposure regardless of when the exposure was delivered on the first, second day, or third day for the appropriate type of charcoal and corresponding timing.

INTRODUCTION

Detectors that depend on adsorption of radon gas and subsequent read-out after exposure suffer two radon loss mechanisms. Normal radioactive decay depletes radon starting the instant the gas diffuses into the adsorber and continuing until read-out. For loss by radioactive decay the well known expression is:

$$A=A_0\exp\left(-\lambda * t_{ime}\right)$$

Where A= Activity at time=t
A_0= max. activity at t=0
λ= .693/half-life = decay constant

A second loss occurs when radon gas diffuses out of the exposed adsorber and is lost to the environment. The importance of the two mechanisms depends on detector construction. Considering charcoal detectors, open-face devices tend to have stronger dynamic adsorption characteristics with half desorption times of about a day, Jester 1990. Half desorption time is defined as the time needed for the radon activity in the charcoal to reach half of the initial amount when a canister was moved from high to zero radon concentration. Assuming no radioactive decay and following Cohen et al 1986 or Jester et al 1990:

$$A=A_0\exp\left(-\beta * t_{ime}\right)$$

Where β=.693/half-desorption time and β=diffusion constant

Open-face devices tend to loose more radon by back diffusion during exposure. Conversely, diffusion barrier detectors have adsorption half-times of about 3 days, and the more important loss mechanism is radioactive decay. For radon activity build-up in a constant radon concentration, and accounting for both adsorption and decay after exposure:

$$A = A_0[1 - \exp(-\gamma * t_{time})] * \exp(-\lambda * t_d)$$

Where $\gamma = \lambda + \beta$ (days^{-1})
t_d=time between end of exposure and read-out

It might be assumed that if the adsorption and decay half-time is known, exact corrections could be made. Unfortunately this is not the case since field radon concentrations vary with time in all but carefully constructed exposure test chambers. Some correction is possible by starting the post exposure decay period at mid-exposure rather than at the end of the exposure. For short exposures of 2 days with a diffusion barrier detector, and assuming all the radon was present during the first day, the mid-point system could reduce the underestimate to about 17%. The situation was much worse for open-face detectors. To illustrate, the following data was taken from Ronca-Battista, et al 1987:

Conc. $[C_1:C_2]$	t_1 hrs	C_1 pCi/l	t_2 hrs	C_2 pCi/l	Ratio of Average C pCi/l	Canister Results pCi/l	Delta %
10:1	24	100	24	9	52	24	-54
1:10	24	9	24	97	53	70	+32

METHOD AND DISCUSSION

The salient question was, what technique could be used to improve this problem? The work described here involved varying the exposure time to allow more time in the beginning and less toward the end of exposure thereby compensating for losses. This was accomplished by placing charcoal detectors in a container, and allowing room air to enter at time intervals corresponding to the build-up curve $[1-\exp(-\gamma * t)]$. Compensation for all losses were achieved for open-face and diffusion barrier detectors. The first step was to determine the overall loss characteristic, γ.

γ was determined by exposing canisters to radon levels about 100 times more concentrated than the ambient room values. The uncovered canisters were measured for retained radon several times after removal from the chamber. The following data was developed using TCS open-face devices, TCS 1, style "SG", and TCS diffusion barrier devices, TCS 2, style "G".

Hours Post Expos.	295 kev	352 kev	609 kev	Avg.	total rel. rel. raw loss	Decay Corr. loss
		(net relative events)				
			SG Style (Open-face)			
0		1	1	1	1	1
6.5		0.70	0.66	0.68	0.95	0.71
26.75		0.29	0.29	0.29	0.82	0.35
48.55		0.11	0.11	0.11	0.69	0.16
55.33		0.08	0.08	0.08	0.66	0.12
			G Style (Diffusion Barrier)			
1.03	1	1	1	1	1	1
7.4	0.85	0.93	0.94	0.91	0.95	0.96
23.4	0.80	0.80	0.83	0.81	0.84	0.97
31.1	0.73	0.73	0.79	0.75	0.79	0.94
47.0	0.61	0.62	0.63	0.62	0.70	0.89
55.3	0.61	0.58	0.60	0.60	0.66	0.91
72.4	0.50	0.50	0.50	0.50	0.58	0.86
79.3	0.47	0.44	0.46	0.46	0.55	0.83
96.4	0.40	0.40	0.40	0.40	0.48	0.83
104.4	0.37	0.36	0.38	0.37	0.46	0.82

The logarithm of the average raw and the decay corrected data are depicted against linear time, Fig. 1 and Fig. 2, for SG and G style canisters as follows:

Regression analysis was performed on the logarithms of the average raw losses vs. time for the two data sets to derive the best values for the loss constants. The corresponding regressed R Squared values were 0.9988 and 0.9977 indicating excellent fits. The calculated loss half-times,(.693/γ), were 15.5 and 72.5 hours for the SG open-face and G style diffusion barrier detectors. It should be noted the maximum, best, loss half-time was set by the radioactive decay of Radon-222. Assuming Zero loss due to desorption the "best" loss half-time was (3.84 days X 24 hrs)= 92.2 hrs. For the G canisters this led to a separate desorption half-time of 14.1 days. The desorption half-time could be calculated by remembering that $\beta = \gamma - \lambda$ and the desorption half-time was (0.693/β).

A 2.6 liter programmable exposure chamber, or cell, was assembled to contain a mini-fan and fittings leading to two external vinyl plastic tubes each 1.26 cm inside diameter and 1 meter long. The mini-fan was powered by a programmable controller. When activated the fan provided a cell ventilation rate of about one air change in 5 minutes.

Unless the mini-fan was activated, G or SG style charcoal detectors placed within the cell registered no measurable radon when the cell was in a radon environment of about 25 pCi/l. Clearly diffusion through the ventilation tubes, and door and other seals was small. The minimum detectable value for three day exposures was about 0.1 pCi/l. Radon was introduced into the cell by switching on the mini-fan. The programmable timer provided twenty-five exposure periods over one to 7 days. The exposure cycle times were based on the measured loss half-times. To illustrate the following calculation provided the cycle times for the three day exposures for the G style detectors:

Eq. 1	$A*\exp(-\gamma*t_1)$	for A steps on day 1, t_1=2.5 days
Eq. 2	$B*\exp(-\gamma*t_2)$	" B " " " 2, t_2=1.5 "
Eq. 3	$C*\exp(-\gamma*t_3)$	" C " " " 3, t_3=0.5 "
Eq. 4	A + B + C = 25	Where γ = 0.229 day^{-1}

Setting equations 1, 2 and 3 to be equal for equal weighting at the end of the exposure, the simultaneous solution was rounded off to 11, 8, and 6 cycles for the first, second, and final day of exposure.

In similar ways the cycle distribution was determined for the SG style open-face canisters. For two day exposures in the cell, 17 cycles are required for the first day and 8 for the remaining day. The following Figure 3 depicts step timing for the SG cans:

One parameter and a few other variables still must be considered. Variables included alteration of adsorption rates due to drafts within the cell caused by the mini-fan. This may have been a two part effect. Radon adsorption by charcoal was enhanced by drafts since air mobility was increased, Gray et al 1988. However during off periods, this effect was reversed. The effective response was an overall reduction in radon loss for the open-face detector, longer effective loss half-time than determined earlier for unrestricted loss to room air. The G style canisters indicated a increase in loss, shorter loss times.

Adsorption rate played a part. It was usually assumed that adsorption was instantaneous. If this were the case, the activity retained by the charcoal would be directly proportional to the cycle dwell time. Measurements suggested little difference between 5 and 10 minute flow times which may lead to adsorption rates longer than the time noted. Finally cell exposures reduced water vapor adsorption about as much as radon adsorption. Longer field exposures were therefore possible with cell housed charcoal canisters. Typically, the SG style canister exposed in the cell for 25 cycles over 2 days adsorbed about 75% ± 5% of a simultaneous open face exposure. It should be remembered 25 cycles total 6.25 hours of mini-fan operation in 48 hours of exposure.

The parameter was the length of each cycle. Since internal cell volume was large compared to canister volume, about 8.4 to 1, the adsorption process proceeded well beyond a ventilation cycle. Prichard 1985, found that one gram of charcoal adsorbed about 2 liters of air at room temperature. Since SG style open-face canisters contain about 85 g of charcoal, at saturation a can could process 200 liters of air. Twenty-five cell air changes accounted for 65 liters, about 1/3 of the maximum. For this reason cycle dwell times were selected to be longer than required to flush the cell.

The dwell times were 15 and 30 minutes for the SG and G style detectors respectively. The longer time for the G style corresponded to a greater charcoal loading.

As expressed partly above, an explicit solution was difficult to formulate due to the variables, would depend on a quantitative understanding of all physical processes, and would have to be verified empirically. Since quantitative understanding of the variables was not yet available, an iterative solution was used to determine the effective cell time constants for SG and G detectors. To illustrate for the SG style, the established steps/day were used as a starting point and cell exposures were made in radon fields of 0.5:20, 20:0.5, and 20:20 for day 1:2. A matrix solution was used for each step to calculate the amount of residual radon at the end of the 48 hour exposure period. When the sum of all steps for the first day equals half of the sum of all 25 steps the effective loss half-times, ($0.693/\gamma$) was established. The following solution matrix was used to establish the loss half-time for SG canisters in the exposure cell:

For unit radon concentration exposure per step:

Step No.	hrs/step	C total hrs.	exp(-C*.693/24) activity at @ t=48 hrs	cumulative activity @ t=48 hrs
0	0.00	0.1	0.25	0.25
1	1.00	1.1	0.26	0.51
2	1.00	2.1	0.27	0.77
3	1.00	3.1	0.27	1.05
4	1.25	4.35	0.28	1.33
5	1.25	5.60	0.29	1.63
6	1.25	6.85	0.30	1.93
7	1.25	8.1	0.32	2.25
8	1.5	9.6	0.33	2.58
9	1.5	11.1	0.34	2.92
10	1.5	12.6	0.36	3.28
11	1.5	14.1	0.38	3.66
12	1.5	15.6	0.39	4.05
13	2.0	17.6	0.42	4.46
14	2.0	19.6	0.44	4.90
15	2.0	21.6	0.47	5.37
16	2.0	23.6	0.49	5.87
17	2.5	26.1	0.53	6.40
18	2.5	28.6	0.57	6.97
19	2.5	31.1	0.61	7.58
20	3.0	34.1	0.67	8.25
21	3.0	37.1	0.73	8.98
22	3.0	40.1	0.80	9.78
23	3.5	43.6	0.88	10.66
24	3.5	47.1	0.97	11.63

The cumulative activity after step 16 to the total was 0.504=(5.87/11.63). This value was in good agreement with the empirical results that are depicted below. The established loss half-time was 24 hours for the SG, and in similar ways, 60 hours for the G style canisters.

RESULTS FOR CELL EXPOSED CANISTERS

Ratio Conc. $[C_1:C_2]$	t_1 hrs	C_1 pCi/l	t_2 hrs	C_2 pCi/l	t_3 hrs	C_3 pCi/l	C Expected pCi/l	Canister Results pCi/l
SG STYLE								
20:1	24	20	24	0.5			10	10.5±0.3
1:20	24	0.5	24	20			10	9.1±0.5
G Style								
20:1:1	24	20	24	0.5	24	0.5	6.7	6.2±0.4
1:1:20	24	0.5	24	0.5	24	20	6.7	7.3±0.4
1:20:1	24	0.5	24	20	24	0.5	6.7	6.2±0.4

The uncertainty associated with the canister results were combined standard deviation of the challenge concentration and the cell measurement.

CONCLUSIONS

As can be seen, the programmed cell overcame the combined effect of radioactive decay and desorption for 2 and 3 day exposures. This resulted by controlling the radon feed to allow charcoal canisters to function as pseudo true integrators for varying radon fields.

Secondary advantages allowed the use of quality control techniques. Addition of a separate timer, a manual momentary mini-fan operating switch, and dual canisters provided explicit assurance of proper operation. A timer wired in parallel with the mini-fan tracked total ventilation time. The over-ride switch, used at the end of exposure, verified operation of the mini-fan which lent assurance that the timer correctly monitored overall cell operation. Similar readings for the duplicate canisters provided assurance of reproducible results.

If the lab used gamma ray spectrometers, that employed several regions on interest for analysis of the charcoal canisters, a high level of assurance would exist that the canisters responded to radon and nothing else. This level of assurance cannot be achieved by other detector systems. For example machines count events and E-Perms register voltage changes while charcoal retains radon which is determined by unique gamma ray signatures of the daughter products which are in equilibrium with the retained radon. The inherent stability the electo-mechanical components of the exposure cell must match if not exceed typical radon machine mean time to failure.

We recommend the programmed cell as an alternate for short term machine measurements as well as an excellent measurement device for real estate purposes.

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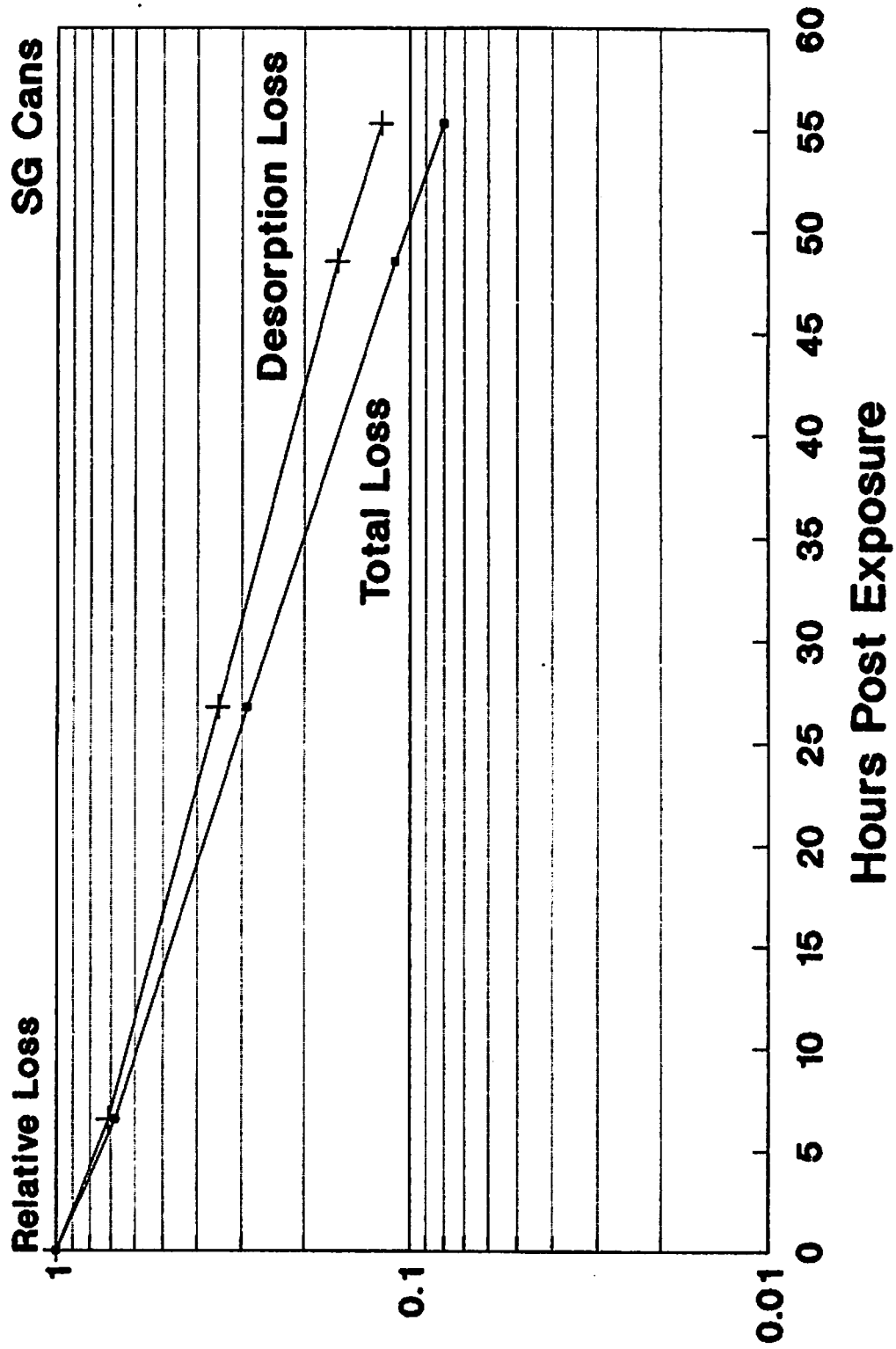
Radiation Protection Dosimetry, 24, 1/4, pp361-365, 1987

6, TCS 1: RMP # AC-01056 TCS Radon Detek AC/100

7, TCS 2: RMP # AC-AQ057 TCS Radon Detek AC/200


SG Cans
Desorption Loss
Total Loss
Relative Loss
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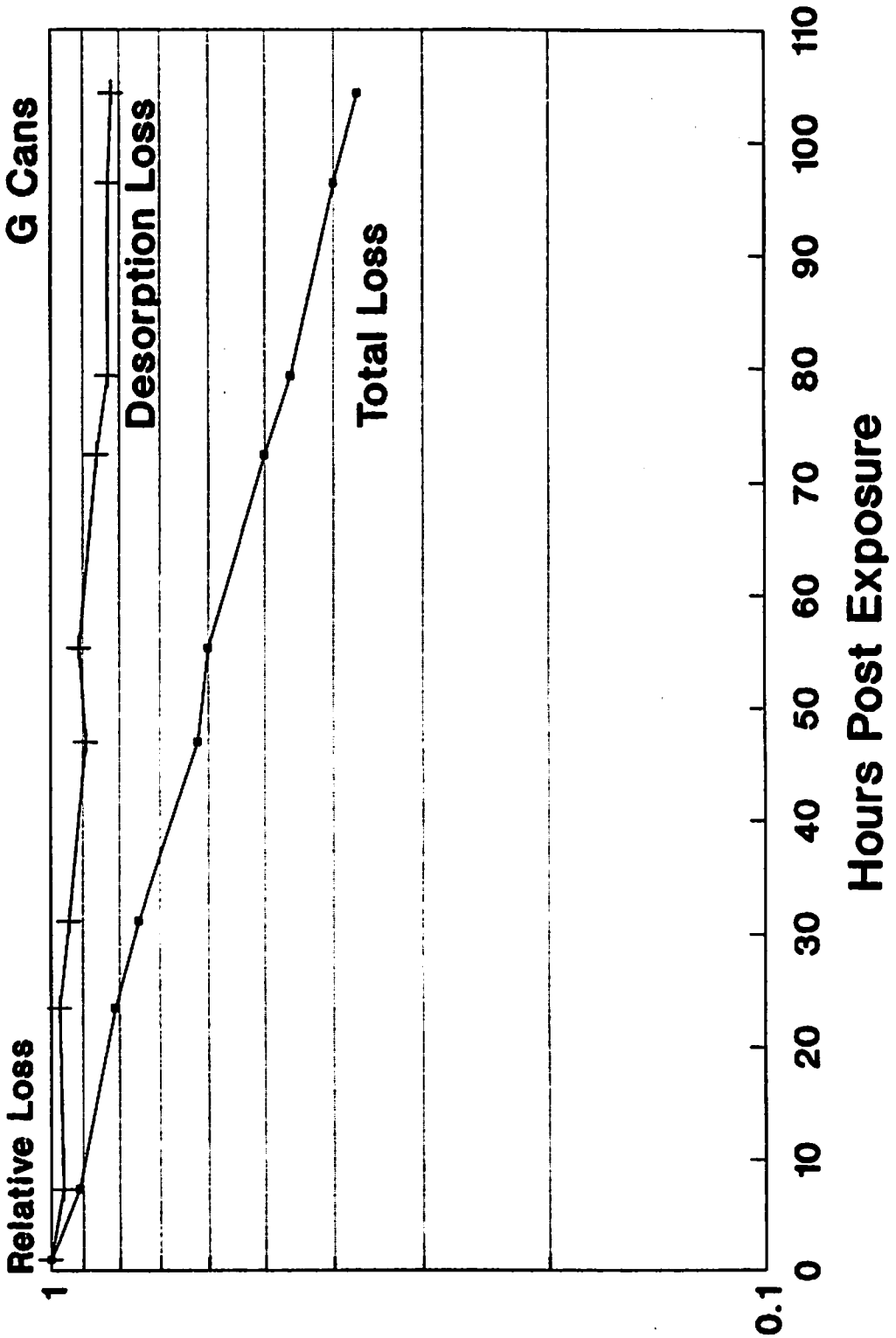
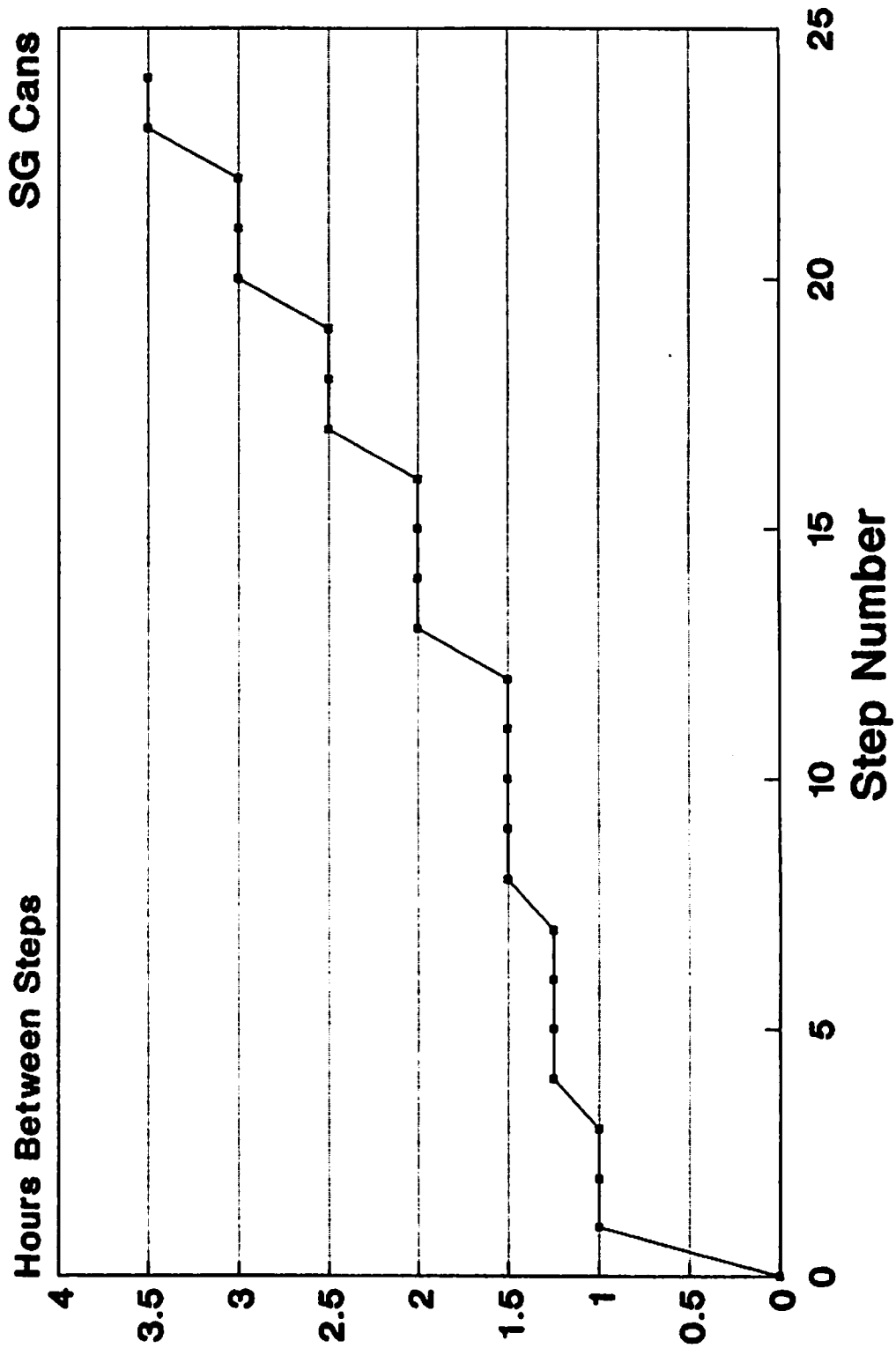
0.01
0
5
10
15
20
25
30
35
40
45
50
55
60
Hours Post Exposure


G Cans
Relative Loss
1
0.1
Desorption Loss
Total Loss
0
10
20
30
40
50
60
70
80
90
100
110
Hours Post Exposure


SG Cans
Hours Between Steps
4
3.5
3
2.5
2
1.5
1
0.5
0
0
5
10
15
20
25
Step Number