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OAK RIDGE Y-12 PLANT

MARTIN MARIETTA

Y-12 DEVELOPMENT DIVISION
TECHNICAL PROGRESS REPORT

Part 8 - Instrumentation and Evaluation Technology (U)

Period Ending August 1, 1987

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2 DUAL CAPACITY
3 LOAN TO ANOTHER DIVISION
4 ACTING
5 LOAN FROM ANOTHER DIVISION

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STACK MONITORING OF URANIUM PARTICULATES AT THE OAK RIDGE Y-12 PLANT

S. A. Wallace and R. J. Reagan

Summary

A prototype system for evaluating the response of a sodium iodide-based detector for observing enriched uranium contaminated filter papers was built. Spectra were taken for characterizing the response to standards, contaminated filters, and thoron daughters. An algorithm was written and tested for triggering an alarm if an excessively fast rate of buildup of uranium contamination occurs.

Introduction

The Y-12 Plant manufactures for the Department of Energy (DOE) precision-machined enriched uranium parts. The room air in areas processing the uranium is vented with fans through ductwork that exits through roof stacks out of the various process buildings. Airborne uranium particulates are trapped in filters in the ductwork. These filters are changed out periodically and burned to recover the trapped material. In the past, the stacks have not been monitored adequately to verify the proper operation of the filters. As a result, the Y-12 Engineering Division has undertaken an extensive program for placing into operation a monitoring system to assure that any aberrant uranium losses from the exhaust stacks would be detected shortly after their occurrence. A one-year fast-track engineering project was approved and funded for designing and erecting the supporting steel platforms for attaching air samplers, purchasing monitors, installing and placing into operation alarm circuits, and turning the system over to the operational divisions.

Prior Work

A report of an evaluation of silicon alpha detection for on-line measurement of the stack losses is presented in ref. 1.

Presentation of Experimental Work

Detection System. A turnkey contract was awarded to Nuclear Measurements Corporation of Indianapolis, Indiana, for furnishing 26 on-line monitors. Loss of enriched uranium up the stacks is measured by pulling a 1-scfm sample stream through a 47-mm Whatman 41 cellulose filter paper. Facing this paper is a beryllium-windowed sodium iodide scintillation detector enclosed within a 3-in.-thick lead pig. For enriched uranium the uranium-234 isotope is present at a 1% assay. With a 248,000-year half-life, this is the most active emitter, having an activity about two orders of magnitude larger than the uranium-235 component. Twenty-eight percent of the uranium-234 decays are accompanied by the emission of L-shell X rays. A 1.5-mm-thick sodium iodide detector captures a fraction of these X rays, and the quantity

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accumulated over time is the determinant of excessive uranium loss. Figure 6 illustrates the layout of the monitoring system. Figure 7 illustrates the response of the detector for americium-241. The higher peak is located at 59.5 keV, and the lower peak is due to L-shell X rays from neptunium in a 12- to 22-keV band. Figure 8 is the detector response due to a 3077 disintegration-per-minute enriched uranium standard. The standard is a plated film on stainless steel obtained from the Oak Ridge Gaseous Diffusion Plant. Although ambient radon fluctuations were thought to present the monitor with an interference spectrum, this has not been a significant problem. A means of correcting for radon is part of the monitor's capability. This can be accomplished by using spectrum windows. Three energy windows can be used. The system gain is set by selecting the high voltage across the photomultiplier such that the 60-keV peak from an americium check source lies at the same channel number as that obtained from pulses from a precision pulser set to a 1-V amplitude with a 2-μs rise time and a 5-μs fall time. Variation in cable length from the photomultiplier output to the pulse-height discrimination input results in each monitor having a different high-voltage setting. The uranium window is selected as those pulses occurring between 0.133 and 0.667 V. This corresponds to an 8- to 40-keV energy window. A second window accepting pulses between 0.833 and 1.667 V is used for radon correction; this corresponds to an energy window of from 50 to 100 keV. A correction algorithm presumes that the spectrum of the enriched uranium and the interferent is known. Figure 9 illustrates two known spectra. Two regions, having window areas 1 and 2, are labeled. The lower solid profile, labeled t, is taken to be that seen after t seconds of measurement. Similarly, the upper solid profile, labeled 3t, is that which would be seen at 3t seconds. If a flat background is present, a shift up in the profile, such as is noted by the profile 3t plus background, would be seen. Background should be constant per unit of time and its profile characterized so that its presence can be removed. The important point is that the ratio of the counts in region 1 to that in region 2 stays constant and independent of the sample interval. Now, given that:

W_1 = counts in region 1 which is the sum of two spectra,

W_2 = counts in region 2 which is the sum of two spectra,

m = ratio of counts due to uranium in region 1 divided by the counts in region 2,

n = ratio of interferent (I) counts in region 1 divided by interferent counts in region 2, then

$$W_1 = mU_2 + nI_2 + Bkgd_1$$

$$W_2 = U_2 + I_2 + Bkgd_2$$

or

$$mU_2 + U_2 = \frac{(m + 1)\ [(W_1 - nW_2) - (B_1 - nB_2)]}{(m - n)} ,$$

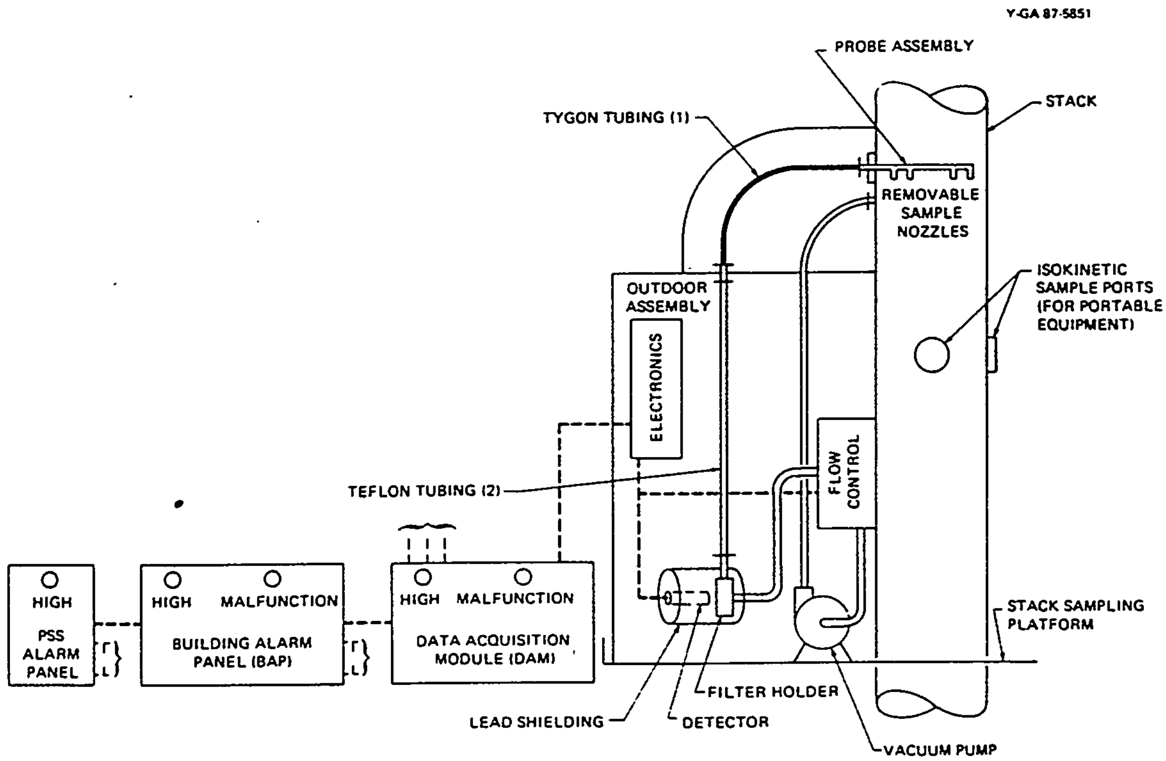

Fig. 6. Schematic of the air-sampling system.

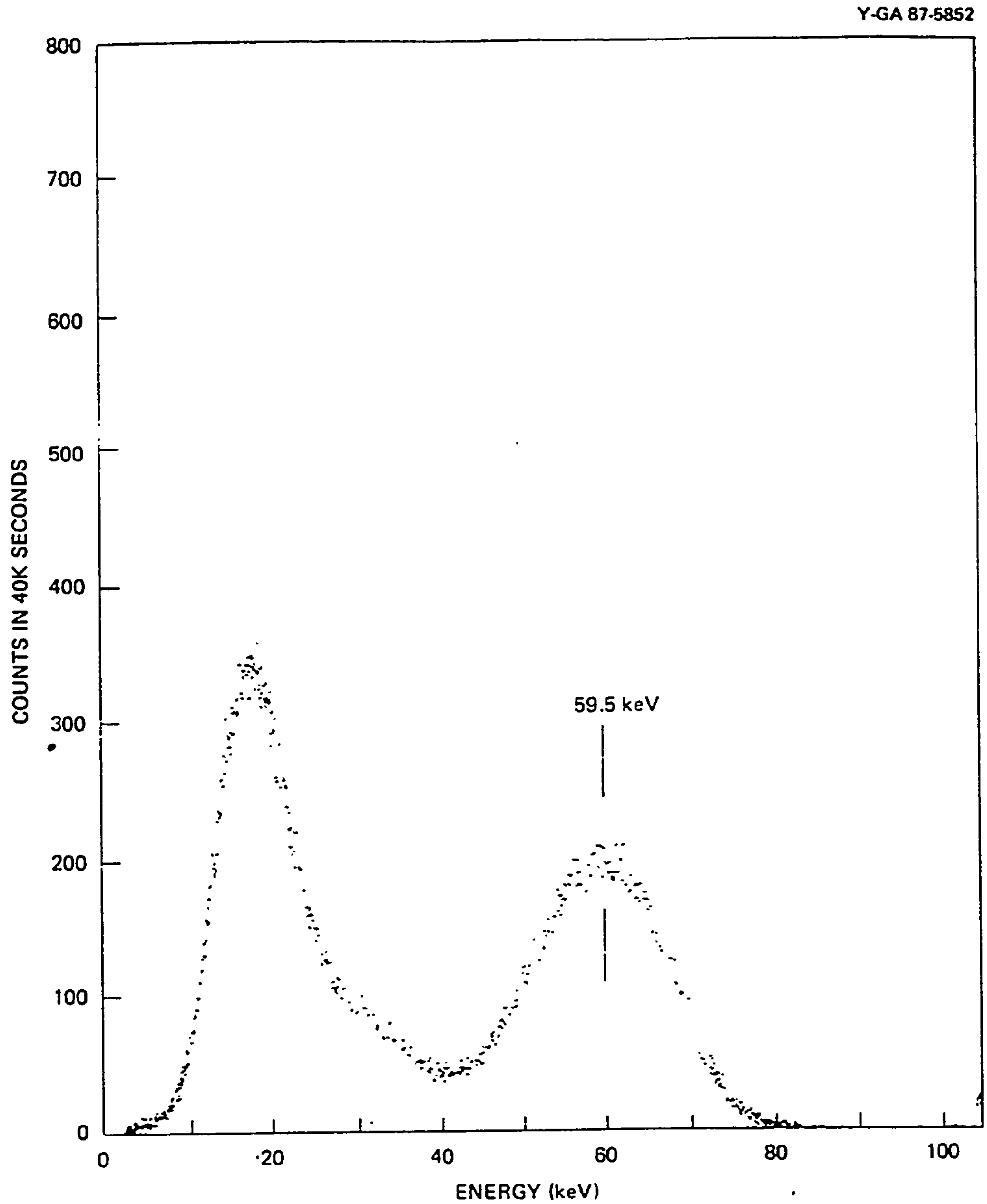

Fig. 7. Spectrum of americium-241 observed with thin sodium iodide detector.

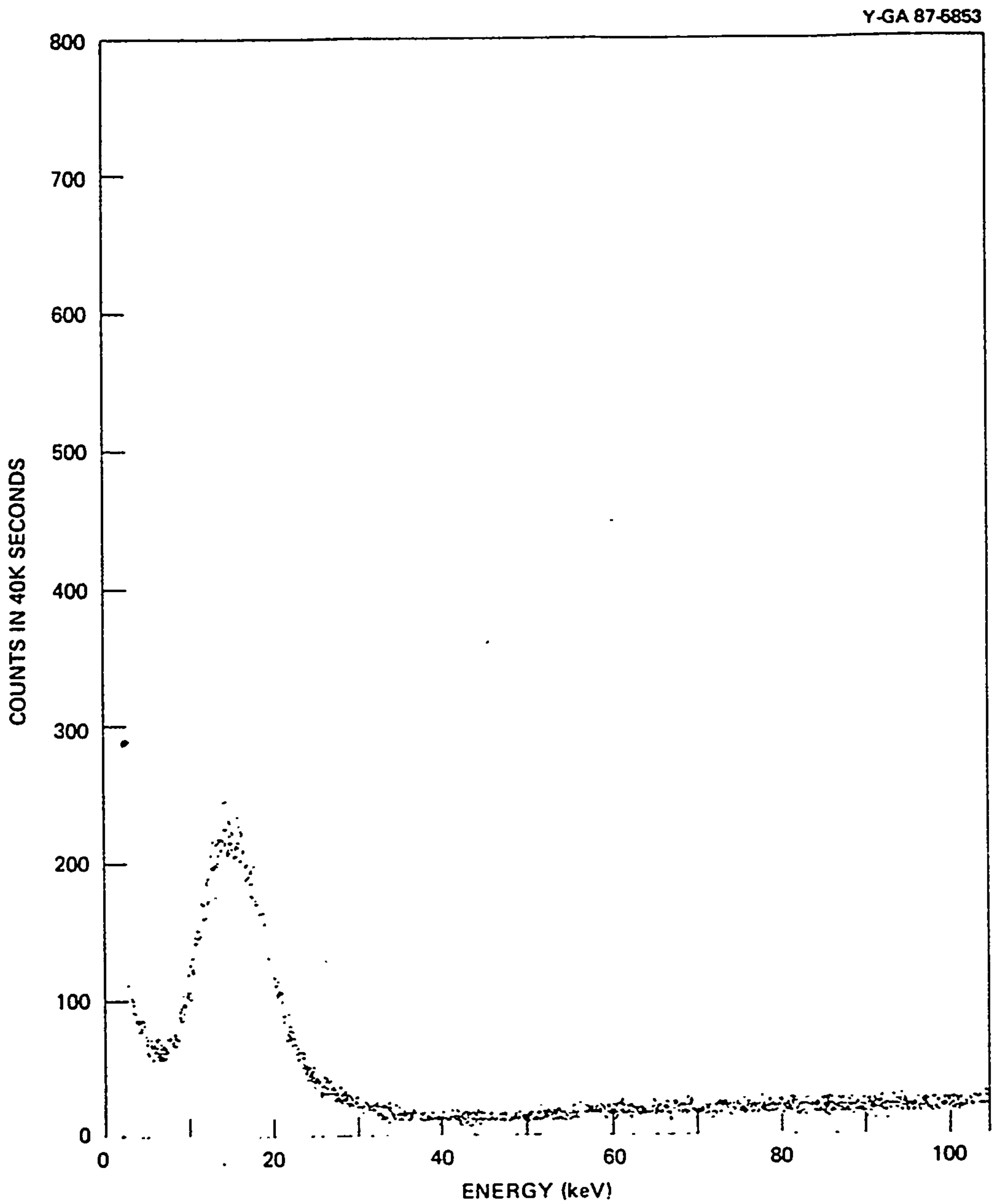

Fig. 8. Spectrum of enriched uranium standard showing L-shell X-ray activity from uranium-234.

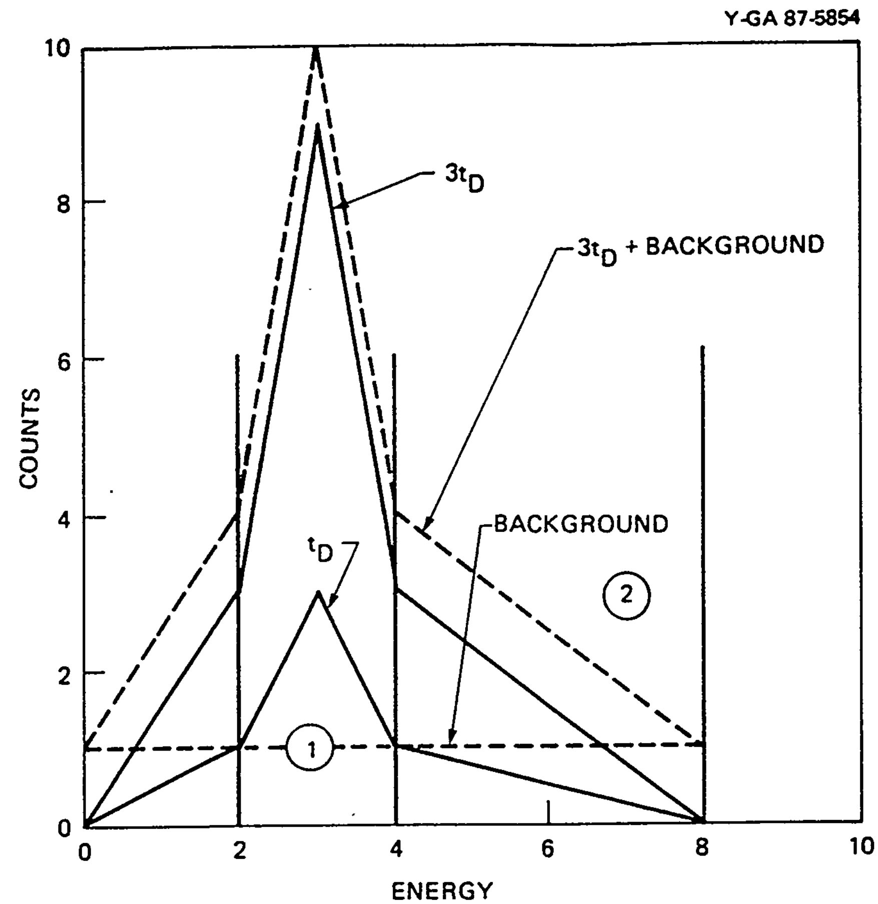

Fig. 9. Spectra at t_D and $3t_D$, L-shell X-ray type.

which is the total uranium counts. Note that if no uranium is present, then W_1 is $Bkgd_1$ and W_2 is $Bkgd_2$, resulting in the total uranium counts being equal to zero.

An experimental verification of this design was implemented by using two single-channel analyzers (SCAs) and two counters, as is seen in the schematic of Fig. 10. An HP-41CX calculator using an IEEE-488 interface to the counters allowed programmed data acquisition to be made. The program (designated NCTR) is given as **Appendix A** to this report. By using the built-in statistical functions of the calculator, a rate-of-rise algorithm is included for signaling an alarm condition. It is derived by calculating a straight line through the last 15 measurements. Since the filter paper is observed for 1-min periods, the algorithm alarm level is based upon that rate of rise which can be allowed in the same period. The algorithm is a running window in that

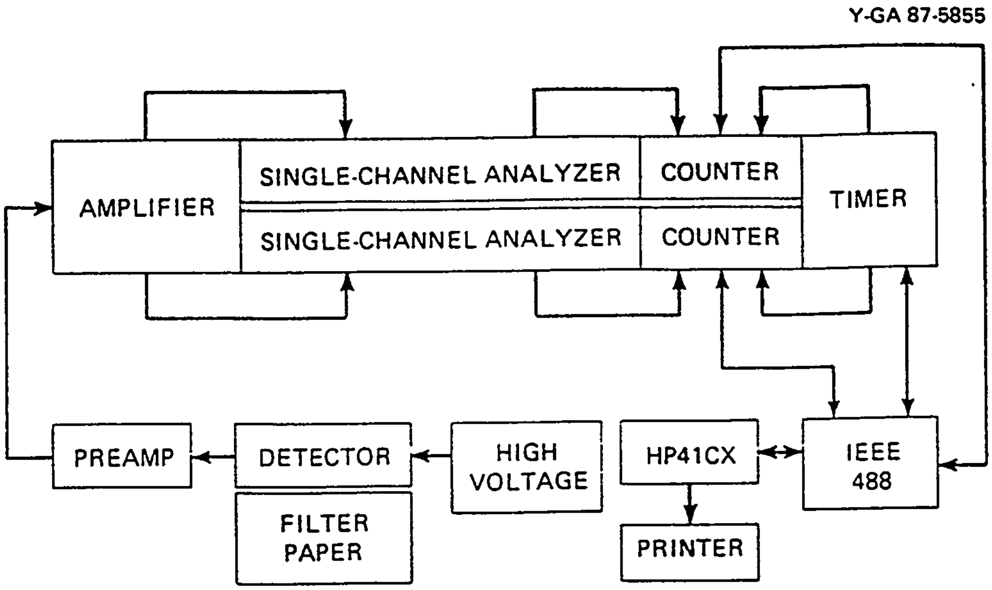

Fig. 10. Schematic of data-acquisition system.

upon receiving a new measurement, the measurement made 15 min back in time is dropped out. The number of samples to calculate the slope is a keyed entry and can be extended to the last 30 measurements. Likewise, the positive slope, which if exceeded results in alarm, is a keyed entry. **Appendix A** is a sample of the hardcopy output generated using this data-acquisition system. The turnkey system installed on-line came with an adaptively weighted, exponential-averaging algorithm that was not amenable to conversion to the HP-41CX calculator. Monte Carlo simulations using rising ramps of Poisson-distributed numbers with both algorithms gave similar alarm times as determined by a Statistical Services study.

Although radon and its daughters have not been an interferent problem, two stacks venting Savannah River material have alarmed because of thoron daughters. Figure 11 shows the spectrum obtained as a result of thoron. All the activity is gone after 5 days as can be seen in Fig. 12 taken from the same 47-mm filter obtained from stack 13.

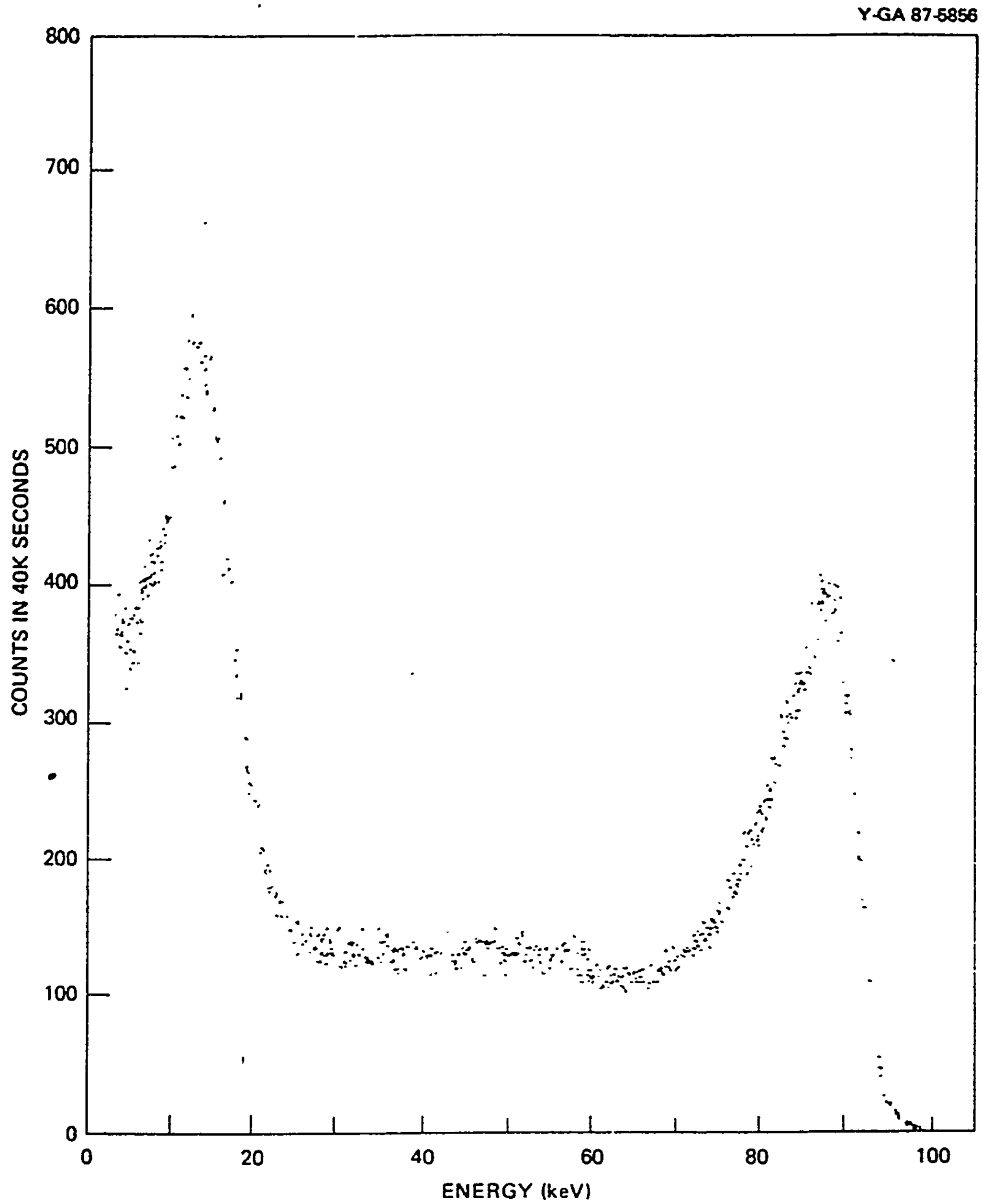

Fig. 11. Spectrum showing fresh-filter thoron daughter activity from stack 13.

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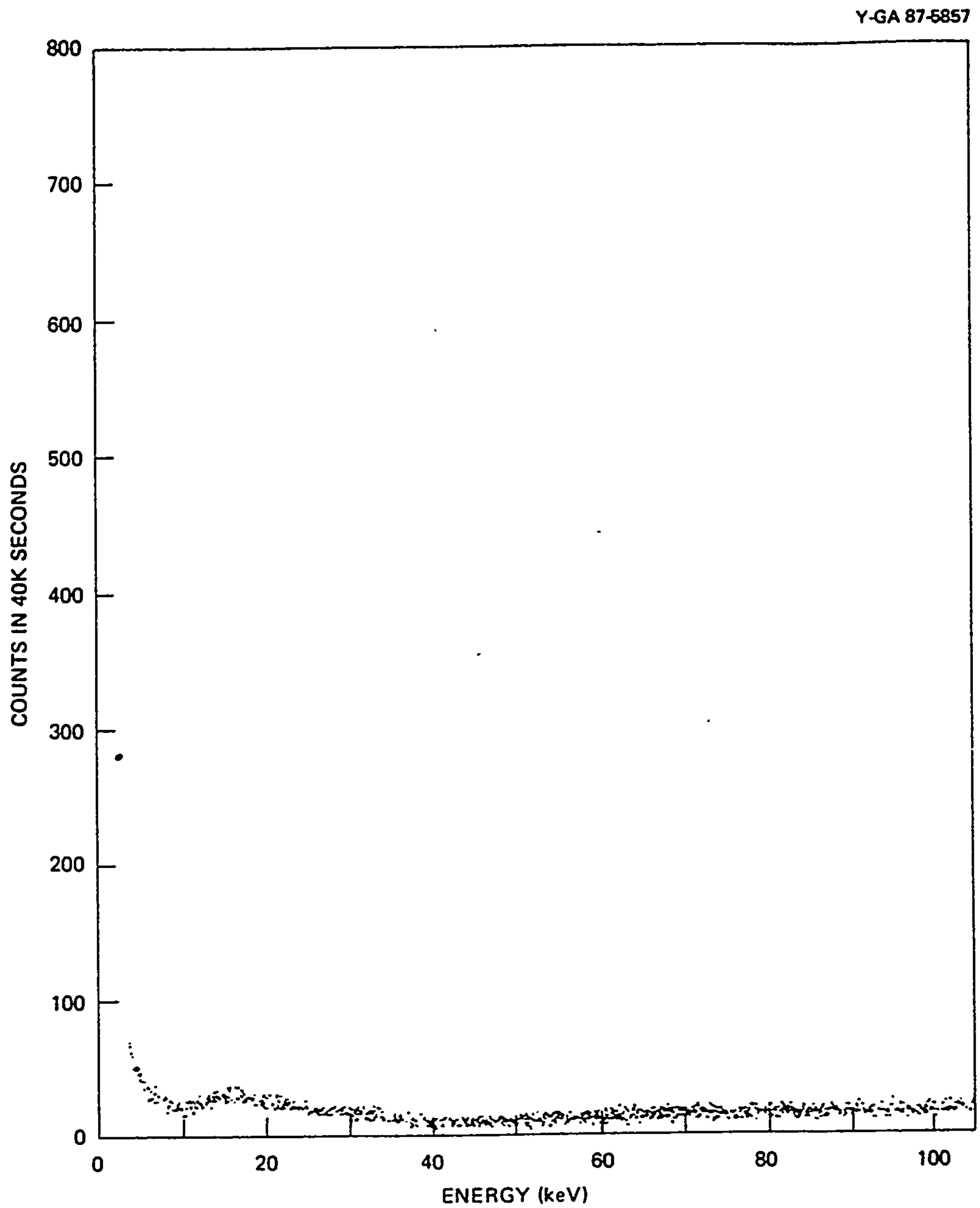

Fig. 12. Spectrum of activity 5-days after that recorded in Fig. 11.

[redacted]

Conclusions

A two-channel data-acquisition system was generated for acquiring measurements of the buildup of uranium on a 47-mm filter paper with the presence of an interferent. A rate-of-rise algorithm was implemented for signaling an alarm in the event of too rapid a rate of rise. The system is generic in the sense that either L-shell X rays or alpha-particle energy can be used as the input into a detector located above the filter, or into a combination of the two detectors. Although this system was developed for monitoring a sample stream taken from vent stacks, it can be used in the floor working area for monitoring transient activity in airborne particulate levels. Since it is projected that the levels of this activity are going to be regulated more strictly in the future, the development of this system is also applicable for future monitoring in this application.

Future Work

The installation and operation of the monitors for airborne discharges from stacks is completed as an Engineering endeavor. Future Development work is to be directed to solving any unique problems that may come up as experience identifies problems. At present no problems have surfaced requiring further development effort. The technology has application to working level airborne measurement and may be pursued for that purpose.

Reference

1. *Y-12 Development Division Technical Progress Report, Part 10 - Material Characterization Processes and Instrumentation and Computer Applications* (U), Y-2362-10, p. 55, Martin Marietta Energy Systems, Inc., Oak Ridge Y-12 Plant, August 11, 1987. SECRET-RD.

APPENDIX A

HP-41 PROGRAM "NCTR"

Appendix A

HP-41 Program "NCTR": Executes two-channel data logging.

```
01LBL "NCTR"
02 .01
03 STO 00        [1]
04 4
05 STO 01
06 5
07 STO 02
08 6
09 STO 03
10 AUTOIO        [2]
11 RCL 01
12 "HCCC"
13 SELECT        [3]
14 REMOTE        [4]
15 OUTA          [5]
16 "CCSC"
17 RCL 02        [6]
18 SELECT
19 REMOTE
20 OUTA
21 RCL 03
22 SELECT
23 REMOTE
24 OUTA
25 CLK24         [7]
26 MDY           [8]
27 CLX           [9]
28 FIX 4         [10]
29 "TIME?HH.MMSS"
30 PROMPT        [11]
31 X=0?
32 GTO 01        [12]
33 SETIME        [13]
34LBL 01
35 CLX
36 FIX 6
37 "DATE?MM.DDYYYY"
38 PROMPT
39 X=0?
40 GTO 02
41 SETDATE       [14]
42LBL 02
43 SF 00         [15]
44 CF 01         [16]
45 CF 02
46 SIGMAREG 31   [17]
47 11.040
48 CLRGX         [18]
49 "NO. PTS?"
50 PROMPT
51 STO 04
52 "U RATIO A/B?"
53 PROMPT
54 STO 05
55 "R RATIO A/B?"
56 PROMPT
57 STO 06
58 "BKGRD A?"
59 PROMPT
60 STO 07
61 "BKGRD B?"
62 PROMPT
63 STO 08
64 "ALARM SLOPE?"
65 PROMPT
66 STO 09
67 ADV           [19]
68 FIX 1
69 "STACK MONITOR"
70 ACA           [20]
71 PRBUF         [21]
72 "M="
73 ACA
74 RCL 05
75 ACX           [22]
76 PRBUF
77 "N="
78 ACA
79 RCL 06
80 ACX
81 PRBUF
82 "A="
83 ACA
84 RCL 07
85 ACX
86 PRBUF
87 "B="
88 ACA
89 RCL 08
90 ACX
91 PRBUF
92 "ALARM="
93 ACA
94 RCL 09
95 ACX
96 PRBUF
97 "NO. PTS="
98 ACA
99 RCL 04
100 ACX
101 PRBUF        [23]
102 RUNSW
103 GTO 06
104LBL 10
105 FC? 49       [24]
106 GTO 05
107 "LOW BATT"
108 OFF
109LBL 05
110 RCL 00
111 RCLSW        [25]
112 X<=Y?        [26]
113 GTO 05
114 OUTA
115 IND          [27]
116 STO 16
117 RCL 03
118 SELECT
119 OUTA
120 IND
121 STO 17
141 FS? 02       [32]
161 /
```

APPENDIX A (continued)

```
122LBL 06
123 "SC"
124 RCL 01
125 SELECT
126 OUTA
127 CLX
128 SETSW        [28]
129 TIME         [29]
130 STO 23
131 DATE         [30]
132 STO 11
133 RCL 14
134 X=0?         [31]
135 GTO 21
136 RCL 16
137 RCL 17
138 *
139 X=0?
140 SF 02
142 GTO 21
143 RCL 14
144 1
145 -
146 RCL 04
147 MOD          [33]
148 STO 13
149 RCL 16
150 RCL 07
151 -
152 RCL 17
153 RCL 08
154 -
155 RCL 06
156 *
157 -
158 RCL 05
159 RCL 06
160 -
162 RCL 05
163 1
164 +
165 *
166 STO 20
167 STO 18
168 39
169 STO 25
170 XEQ 20       [34]
171 RCL 22
172 STO 19
173 RCL 09
174 X<=Y?
175 SF 01
176LBL 21
177 DSE 12       [35]
178 GTO 08
179 FC? 00
180 GTO 24

181 CF 00
182 40
183 STO 12
184 ADV
185 GTO 25
186LBL 24
187 50
188 STO 12
189 12
190 ACCHR        [36]
191 ADV
192LBL 25
193 "DATE  "
194 ACA
195 CLA
196 RCL 11
197 FIX 4
198 ADATE        [37]
199 ACA
200 PRBUF
201 ADV
202 "TIME          "
203 ACA
204 "    POINT    "
205 ACA
206 "  COUNT A "
207 ACA
208 "  COUNT B "
209 ACA
210 "  COUNT U "
211 ACA
212 "  SLOPE U "
213 ACA
214 "  STD DEV "
215 ACA
216 PRBUF
217LBL 08
218 RCL 23
219 FIX 4
220 CLA
221 ATIME        [38]
222 ACA
223 "  "
224 ACA
225 FIX 0
226 RCL 14
227 XEQ 11
228 RCL 16
229 XEQ 11
230 RCL 17
231 XEQ 11
232 RCL 18
233 XEQ 11
234 FIX 1
235 RCL 19
236 XEQ 11
237 FIX 3
238 RCL 38
239 XEQ 11
240 FC? 02

241 GTO 25
242 CF 02
243 " MALFUNCTION"
244 ACA
245 GTO 19
246LBL 25
247 FC? 01
261LBL 11
262 STO 23
263 CLA
264 ARCL X       [40]
265 ALENG        [41]
266 CHS          [42]
267 10
281 1
282 +
283 RCL 25
284 +
285 STO 26
286 RCL IND 26
287 STO 21
```

APPENDIX A (continued)

```
248 GTO 26
249 CF 01
250 " ***ALARM***"
251 ACA
252LBL 26
253 1
254 ST+ 14      [39]
255LBL 19
256 PRBUF
257 RCL 02
258 SELECT
259 "RCCC"
260 GTO 10
268 +
269 " "
270 ASTO 24      [43]
271 CLA
272LBL 14
273 ARCL 24
274 DSE X
275 GTO 14
276 ARCL 23
277 ACA
278 RTN          [44]
279LBL 20
280 RCL 13
288 RCL 20
289 STO IND 26
290 RCL 14
291 SIGMA+       [45]
292 RCL 04
293 RCL 14
294 X<=Y?
295 GTO 23
296 RCL 21
297 RCL 14
298 RCL 04
299 -
300 SIGMA-       [46]

301LBL 23
302 RCL 14
303 RCL 04
304 X>Y?        [47]
305 RTN
306 RCL 36
307 RCL 35
308 *
309 RCL 31
310 RCL 33
311 *
312 -
313 RCL 36
314 RCL 32
315 *
316 RCL 31
317 X^2
318 -
319 /
320 STO 22
321 X^2
322 ST+ 37
323 RCL 37
324 RCL 14
325 RCL 04
326 -
327 1
328 +
329 /
330 SQRT

331 SQRT
332 RTN
333 END
```

1. STO nn - Copies contents of X-register into R(nn)
2. AUTOIO - Set interface to Auto mode
3. SELECT - Select device as primary device
4. REMOTE - Set primary device to remote mode
5. OUTA - Output ALPHA string to primary device
6. RCL nn - Copy data from a data register into the X-register
7. CLK24 - Sets a 24-hour clock display
8. MDY - Selects month-day-year date format
9. CLX - Clears X-register
10. FIX n - Selects fixed-pt display format with n decimal plcs
11. PROMPT - Displays contents of the Alpha reg and stops exec
12. GTO nn - Transfers execution to specified numeric location
13. SETIME - Sets clock to time specified in X-register
14. SETDATE - Sets clock to date specified in X-register
15. SF nn - Sets flag nn, 00<= nn <=29
16. CF nn - Clears flag nn, 00<= nn <=29
17. SIGMAREG nn - Two-digit address of six consecutive registers
18. CLRGX - Clears every ii the register from R(bbb) thru R(eee)

APPENDIX A (continued)

19. ADV - Advances paper if printer is present
20. ACA - Accumulate ALPHA register into print buffer
21. PRBUF - Print the print buffer left-justified
22. ACX - Accumulate X-register into print buffer
23. RUNSW - Runs stopwatch
24. FC? nn - Tests flg nn and skips nxt prgm ln unless flg is clr
25. RCLSW - Returns stopwatch time
26. X<=Y? - Skps nxt instrtn unlss numbr in X-reg <= cntnts Y-reg
27. IND - Retrieves ASCII codes of up to 24 chactrs frm pri dev
    IND nn _ Argument of STO, RCL, etc specifying registr address
28. SETSW - Sets stopwatch to strtng time specified in X-register
29. TIME - Returns number for current time
30. DATE - Returns number for current date
31. X=0? - Skips nxt instrtn unless number in X-register = 0
32. FS? nn - Tests flg nn and skps nxt prgm ln unless flag is set
33. MOD - Remainder whn int y in Y-reg is dvded by int x in X-reg
34. XEQ label - Calls spcfd numeric or ALPHA label as subroutine
35. DSE nn - For iiiii.fffcc in R(nn), decrements iiiii by cc and skips next program step if iiiii + cc > fff.
36. ACCHR - Accumulate character into print buffer
37. ADATE - Appends number in X-reg to ALPHA reg in date format
38. ATIME - Appends number in X-reg to ALPHA reg in time format
39. ST+ nn - Adds number in X-reg to number in R(nn) and places result in R(nn)
40. ARCL nn - Appends contents of R(nn) to ALPHA register
41. ALENG - Returns number of characters in ALPHA register
42. CHS - Change sign of the operand in X-register
43. ASTO nn - Copies six leftmost chrctrs in ALPHA reg into R(nn)
44. RTN - Returns execution to line following XEQ instruction that called this subroutine
45. SIGMA+ - Automatic calculation of sums and products for stats
46. SIGMA- - Deletes from statistical function the entry
47. X>Y? - Skps nxt instr unless number in X-reg > numbr in Y-reg

Lines

001-103 Set identifiers of IEEE-488 instruments, clear instruments, set time and date, set flags, input constants, print input
104-108 Battery check
109-113 Primary clock loop used every reading
114-121 Read counters
122-175 Start counters, read current time and date, calculate COUNT U, check for zero readings and flag, calculate modulus of sample number MOD number of points in slope calculation
176-191 Calculation for printer pagination
192-216 Header print control
217-251 Output print control and flag checks for malfunction or alarm

APPENDIX A (continued)

252-254 Sample index incrementer
255-260 Loop logic
261-278 Print format of output
279-320 Slope calculation
321-332 Standard deviation calculation