

RADIOACTIVITY PRODUCED BY A LINAC

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Summary

Over 40 different materials have been irradiated in the 100 MeV bremsstrahlung beam from the NBS linac. The major contributions to the gamma ray spectrum from the radioactive products have been measured using a Ge(Li) detector. Measurements have been made of gamma rays of concern in the 30 minute to 30 day time interval but this includes in some cases gamma rays from nuclides with half lives as long as 30 years. Eighteen of the samples were pure materials and 25 were commonly used structural, shielding, electrical and mechanical components. In several important cases nuclides produced by multiple particle emission dominate the picture.

Introduction

When electrons or their resultant bremsstrahlung x-rays from a high intensity accelerator strike any material, neutrons and protons may be knocked out leaving nuclides which are radioactive. The detailed study of these interactions is fair game for the field of nuclear physics. From a slightly different view point it is also a necessary study for the field of health physics. The gamma rays from the residual radioactive nuclei as a function of time are of prime concern from a health view point. A study has been made using a number of pure materials and common materials to throw light on this problem using the NBS linac operated in the 90 to 140 MeV range.

Yield

The relative yield of products with X nucleons removed from an initial nuclide of atomic mass A and charge Z may be expressed as:

$$Y(Z, E_0, A, X) = \int_0^{E_0} B(E, E_0) \sigma(Z, A, X) dE \quad (1)$$

where the bremsstrahlung spectrum  $B(E, E_0)$  characteristic of electron energy  $E_0$  is incident on a target. When  $E_0$  is low, in the giant resonance region, only processes where  $X=1$  are possible but for higher energy accelerators spallation processes where  $X>2$  are not only possible but significant. The actual values for these cross sections are in general not at all well known though the dipole sum rule  $\int_0^{E_0} \sigma dE = .06NZ/A$  MeV-b allows for reasonable estimates of the total cross section  $\int_0^{E_0} \sigma dE$  for a combination of all photonuclear processes.

In Figure 1 integrated cross section data for three cases, low, medium and high Z are given. The triangles representing the data of Walker and Morton<sup>1/</sup> are for processes leading to the production of <sup>18</sup>F. In this case their data has been plotted against the number of nucleons emitted reduced by the number of spontaneous protons to be expected in the decay chains. The medium Z data

for <sup>141</sup>Pr was obtained at NBS. (I am indebted to Dr. Frank Schima for much of this work.) At high Z the <sup>209</sup>Bi data covers a very broad range of daughter nuclides<sup>2/</sup>. The data have been normalized at  $(\gamma, n)$  and  $(\gamma, 2n)$  points in order to obtain the straight line which may serve as a rule of thumb for calculation of the yield to be expected. This yield may be expressed by the empirical expressions

$$Y(\gamma, Xn) = 0.2Y(\gamma, n)(1.8)^{1-X} \text{ for } X>2 \text{ or} \quad (2)$$

$$Y(\gamma, Xn) = 0.5Y(\gamma, 2n)(1.8)^{2-X} \text{ for } X>2 \quad (3)$$

if the  $(\gamma, n)$  or  $(\gamma, 2n)$  yields are known.

The gamma rays from any particular nuclide are dependent on the decay scheme and half life as well as its population.

In order to demonstrate the significant contributions of the spallation processes 18 pure materials and 25 common materials were irradiated for 10 minutes each in the thick target 90 to 140 MeV bremsstrahlung beam produced by a series of tungsten plates totaling 1.22mm thickness. The decay of each sample was measured using a Ge(Li) detector at several times. The total number of gamma rays from each of a number of nuclides was measured, introducing geometry and detector efficiency corrections. The data from this initial experiment was not complete enough to assign relative intensity numbers to the nuclide decay rates. However it was in general possible to establish the order of importance of nuclides at each particular time. It is this order number that is listed for each measured nuclide in Table 1. A (3) in the  $(\gamma, 2n)$  column indicates, for example, that that nuclide was third in importance at the time of measurement. When several nuclides are present in the stable elements there may be some ambiguity in the assignment of a production process but the nuclide identified by its gamma ray signature is specified.

The important conclusion to be drawn from the data is that while the  $(\gamma, n)$  process most often produces the dominant nuclide, this is by no means universally so. The multineutron spallation processes just cannot be ignored!

Relative yield, equation (1) can be generalized to give the nuclide population P reflecting the duration of irradiation T of a sample and variations in energy  $E_0$  and intensity I of the accelerated electrons.

$$P(T, Z, E_0, A, X) = \frac{1}{\lambda T} (1 - e^{-\lambda T}) \int_0^{E_0} \max Y(Z, E_0, A, X) I(E_0) dE_0 \quad (4)$$

where  $\lambda$  is the decay constant for any particular nuclide.

At any time  $t$  after the end of the irradiation the decay rate  $D$  may be expressed as

$$D = \frac{dP}{dt} = \lambda P = \frac{1}{T} (1 - e^{-\lambda T}) \int_0^{E_0} \max Y(Z, E_0, A, X) I(E_0) dE_0 \quad (5)$$

However the energy and operating schedule of an accelerator can only be projected in a crude way and will certainly vary from machine to machine so the NBS studies are centered on the more fundamental parameters of equation (1).

Any single gamma ray measurement is fairly complicated as the relative number of counts under each photo peak plotted against channel number in Figure 2 for a bismuth sample illustrates<sup>3/</sup>. The tables are an attempt to establish the relative importance of a complex family of gamma rays and as such must be taken with some reservation. Statistical uncertainties in some cases could modify the assignments. Gamma rays below 100 keV in energy were not measured in many cases. In a number of cases some gamma rays, as represented by some unidentified lines on Figure 2, went unassigned. Also the major importance of annihilation radiation for many nuclides led to some ambiguity in assignment of nuclides responsible.

#### Special Cases of Interest

The 15h half life <sup>24</sup>Na nuclide dominates the gamma ray spectrum from a wide variety of lower Z materials including concrete.

The 53 day half life <sup>7</sup>Be produced by <sup>12</sup>C( $\gamma, n\alpha$ ) for energies above the 27.4 MeV threshold and the <sup>16</sup>O( $\gamma, n2\alpha$ ) process for energies above 32 MeV is

the most important long half life daughter product for many organic materials. The <sup>7</sup>Be is absorbed in the ion exchange resins of closed loop filtered water cooling systems but remains a key health physics problem when ground water or drain water may be irradiated, or in food materials.

The long half life <sup>58</sup>Co, <sup>57</sup>Co, <sup>56</sup>Co series produced by <sup>63</sup>Cu bombardment are the key activities at long times in a magnet system or accelerator using copper conductors. Eventually the 5.2 year half life <sup>60</sup>Co will become the dominant daughter product in a system in continuous use.

The praseodymium data which has been analyzed more thoroughly than most illustrates the importance of daughter products growing in due to the decay of nuclides originally produced.

#### Acknowledgements

I am indebted to Dr. George Lutz and Mr. Frederick Lundgren and the crew of the NBS linac for the sample irradiations. Mr. Sherman Fivozinsky has been of great assistance in the latter stages of analysis of the data for relative gamma ray intensities. Dr. Frank Shima did most of the praseodymium data analysis. Dr. James E. Leiss and Dr. Samuel Penner have been most helpful in many discussions of the program and treatment of the data.

#### References

1. T. G. Walker and W. T. Morton, Proc. Phys. Soc., 75, 4 (1960)
2. J. M. Wyckoff (To be published)
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TABLE I

ORDER OF IMPORTANCE FOR GAMMA RAY EMISSION  
FOLLOWING PURE MATERIALS ACTIVATION

Z	MATERIAL	TIME AFTER IRRADIATION	PROCESS					OTHER			
			$\gamma, n$	$\gamma, 2n$	$\gamma, 3n$	$\gamma, 4n$	$n, \gamma$				
6	Carbon	2h37m	1	$^{11}\text{C}$				2	$^7\text{Be}$	$\gamma, n\alpha$	
		4h49m						1	$^7\text{Be}$	$\gamma, n\alpha$	
		42h55m						1	$^7\text{Be}$	$\gamma, n\alpha$	
13	Aluminum	2h51m						1		$\gamma, n2p$	
		14h11m						1		$\gamma, n2p$	
		15d15h						1		$\gamma, 3n2p$	
22	Titanium	5h12m	2	$^{41}\text{Ti}$							
								1	$^{47}\text{Sc}$	$\gamma, p$	
		20d15h						3	$^{44}\text{Sc}$	$\gamma, np$	
27	Cobalt	2h54m	1		2	4				$\gamma, n2p$	
		5h18m	1		2	4	5			$\gamma, n2p$	
		12h45m	1		2	3	5			$\gamma, n2p$	
		18h19m	1		2	3	4			$\gamma, n2p$	
		3d11h	1		2	3					
		53d	1		2	3					
28	Nickel	2h21m	2	$^{57}\text{Ni}$	3	$^{56}\text{Ni}$			1	$^{57}\text{Co}$	$\gamma, p$
		1d17h	1	$^{57}\text{Ni}$	2	$^{56}\text{Ni}$			1	$^{57}\text{Co}$	$\gamma, p$
		11d16h	2	$^{57}\text{Ni}$					3	$^{56}\text{Co}$	$\gamma, np$
29	Copper	37d							1	$^{58}\text{Co}$	$\gamma, n\alpha$
									2	$^{57}\text{Co}$	$\gamma, 2n\alpha$
									3	$^{56}\text{Co}$	$\gamma, 3n\alpha$
33	Arsenic	2h 2m	1		2						
		4h28m	1		2			3			
		11h54m	1		2	4		3			
		18h13m	1		2	4		3			
		37h26m	1		2	4		3			
		54d16h	1								
45	Rhodium	1h 6m	5		2	1	4		3	$\gamma, 6n$	
		1h37m			3	1	2				
		2h31m	3			1	2				
		4h37m	3			1	2				
		14h48m	3			1	2				
		38h33m	3		2	1					
46d12h	1		2								
46	Palladium	1h19m			2	$^{99}\text{Pd}$			1	$^{107}\text{Rh}$	$\gamma, p$
		1h50m	1	$^{109}\text{Pd}$				3	$^{106}\text{Rh}$	$\gamma, np$	
			2	$^{101}\text{Pd}$							
		13h25m	1	$^{109}\text{Pd}$				3	$^{105}\text{Ru}$	$\gamma, n2p$	
	2	$^{101}\text{Pd}$									
47	Silver	56m	1	$^{106}\text{Ag}$			2	$^{103}\text{Ag}$			
		2h29m	1	$^{106}\text{Ag}$	3	$^{105}\text{Ag}$	2	$^{103}\text{Ag}$			
		42h 4m	1	$^{106}\text{Ag}$	2	$^{105}\text{Ag}$			4	$^{105}\text{Rh}$	$\gamma, 2p$

TABLE I (continued)

Z	MATERIAL	TIME AFTER IRRADIATION	PROCESS					OTHER
			$\gamma, n$	$\gamma, 2n$	$\gamma, 3n$	$\gamma, 4n$	$n, \gamma$	
53	<u>Iodine</u>	2h 5m	1			2	3	
		3h 7m	1			2	4	3
		4h53m	1			2		3
		18h23m	1			2		4
		44d18h	1					3
								2
55	<u>Cesium</u>	2h29m	1					
		5h25m	1			2		
		12h40m	1			2		
		140h30m	1					
		51d15h	1				3	4
								2
59	<u>Praseodymium</u>	33m			1			2
		2h		3	1	2		3
		10h		2	1			1
		2d						3
		14d12h						2
73	<u>Tantalum</u>	1h30m	2	<sup>180</sup> Ta				1
		2h 5m	1	<sup>180</sup> Ta	3	<sup>178</sup> Ta		2
		2h53m	1	<sup>180</sup> Ta	4	<sup>178</sup> Ta		3
								2
78	<u>Platinum</u>	5h	1	<sup>195</sup> Pt	3	<sup>194</sup> Pt		
			2	<sup>197</sup> Pt				
		6d18h	1	<sup>195</sup> Pt	3	<sup>194</sup> Pt		4
			2	<sup>197</sup> Pt				
		20d11h	1	<sup>195</sup> Pt	2	<sup>194</sup> Pt		
79	<u>Gold</u>	53m	1				2	3
		1h22m	1					2
		1h55m	1					2
		2h43m	1					2
		3h33m	1					2
		13h44m	1					2
		16h20m	1					2
		42h50m	1					2
		47h 4m	1					2
		43d21h	1		2			
82	<u>Lead</u>	1h39m	1	<sup>203</sup> Pb	2	<sup>202</sup> Pb	3	<sup>201</sup> Pb
		2h46m	1	<sup>203</sup> Pb	2	<sup>202</sup> Pb	3	<sup>201</sup> Pb
		64h50m	1	<sup>203</sup> Pb	2	<sup>202</sup> Pb	3	<sup>201</sup> Pb
83	<u>Bismuth</u>	43m			3			1
								2
								4
		2h 3m			1	4		2
								3
								5
		10h 8m			1			2
								3
								4
		3d11h			1	3		5
		36d		3	1	2		2
60d		2	1	3				

1. Mononuclidic elements underlined.  
 2. Nuclide follows order number for poly-nuclidic elements.

TABLE II  
ORDER OF IMPORTANCE FOR GAMMA RAY EMISSION  
FOLLOWING COMMON MATERIALS ACTIVATION

Z	MATERIAL	ELEMENT	TIME AFTER IRRADIATION	PROCESS		
				$\gamma, n$	$\gamma, 2n$	OTHER
6	Teflon	Carbon	4h52m	1	$^{11}\text{C}$	
		Fluorine		1	$^{18}\text{F}$	
6	Nylon	Carbon	3h 4m	1	$^{11}\text{C}$	
		Carbon	14h20m		$^{11}\text{C}$	1 $^7\text{Be}$ $\gamma, n\alpha$
6	Rubber ("o"ring)	Carbon	6d19h			1 $^7\text{Be}$ $\gamma, n\alpha$
6	Polystyrene	Carbon	48m	1	$^{11}\text{C}$	
		Carbon	2h36m	1	$^{11}\text{C}$	
			18h14m			
6	Polyethylene	Fluorine	3h36m	1	$^{19}\text{F}$	
		Carbon	18h22m			1 $^7\text{Be}$ $\gamma, n\alpha$
		Carbon	8d			.1 $^7\text{Be}$ $\gamma, n\alpha$
6	Bakelite	Carbon	21m	1	$^{11}\text{C}$	
		Carbon	1h58m	1	$^{11}\text{C}$	
		Sodium	18h22m			1 $^{24}\text{Na}$ $n, \gamma$
		Calcium	8d 9h			2 $^{43}\text{K}$ $\gamma, p$
11	Ceramic (2)	Sodium	4h 2m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	17h 5m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	6d17h	2	$^{22}\text{Na}$	1 $^{24}\text{Na}$ $n, \gamma$
		Cobalt		3	$^{58}\text{Co}$	
11	Glass (with water)	Sodium	2h37m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	5h25m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	15h 8m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	3d13h	2	$^{22}\text{Na}$	1 $^{24}\text{Na}$ $n, \gamma$
11	Glass (ampoule)	Sodium	4h31m			1 $^{24}\text{Na}$ $n, \gamma$
		Calcium	17h31m			2 $^{43}\text{K}$ $\gamma, p$
		Sodium	6d11h	1	$^{22}\text{Na}$	?
11	Glass Spaghetti	Titanium	6d21h			1 $^{48}\text{Sc}$ $\gamma, p$
11	Concrete	Sodium	14m			1 $^{24}\text{Na}$ $n, \gamma$
		Silicon	14m			2 $^{29}\text{Al}$ $\gamma, p$
		Sodium	2h10m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	3h35m			1 $^{24}\text{Na}$ $n, \gamma$
		Sodium	18h57m			1 $^{24}\text{Na}$ $n, \gamma$
		Calcium	18h57m			2 $^{43}\text{K}$ $\gamma, p$
			8d			?
	16d10h			?		
11	Rubber	Sodium	2h45m			1 $^{24}\text{Na}$ $n, \gamma$
		Lead	6h15m	1	$^{203}\text{Pb}$	
26	Steel (Structural)	Iron	1h12m			1 $^{56}\text{Mn}$ $\gamma, p$
		Iron	4h17m	2	$^{52}\text{Fe}$	1 $^{56}\text{Mn}$ $\gamma, p$
		Iron	3d13h			1 $^{52}\text{Mn}$ $\gamma, np$
		Iron				2 $^{54}\text{Mn}$ $\gamma, np$
26	Steel (Drill)	Iron	6d12h			1 $^{52}\text{Mn}$ $\gamma, np$
						2 $^{54}\text{Mn}$ $\gamma, np$
						3 $^{51}\text{Cr}$ $\gamma, n2p$

TABLE II (continued)

Z	MATERIAL	ELEMENT	TIME AFTER IRRADIATION	PROCESS										
				$\gamma, n$	$\gamma, 2n$	OTHER								
26	Steel (Stainless)	Iron	16h40m	1	$^{56}\text{Mn}$	3	$^{52}\text{Mn}$ $\gamma, np$							
				2	$^{51}\text{Cr}$									
				4	$^{54}\text{Mn}$									
26	Steel (Screw)	Iron	1h58m	3	$^{52}\text{Fe}$	1	$^{56}\text{Mn}$ $\gamma, p$							
						2	$^{52}\text{Mn}$ $\gamma, np$							
	Iron	18h23m	3	$^{52}\text{Fe}$	1	$^{52}\text{Mn}$ $\gamma, np$								
					2	$^{54}\text{Mn}$ $\gamma, np$								
4	$^{51}\text{Cr}$ $\gamma, n2p$													
28	Nichrome	Nickel Chromium	5d20h	1	$^{57}\text{Ni}$	3	$^{56}\text{Co}$ $\gamma, np$							
				2	$^{51}\text{Cr}$			4	$^{56}\text{Ni}$ $\gamma, 2n$					
29	Capacitor (Tubular)	Copper	1h50m	1	2	3	165 KeV ?							
							Copper	2h40m	1	2	3	$\gamma, n2p$		
							Copper	15h40m	1	2	4	$\gamma, n\alpha$		
29	Neon Lamp	Sodium Copper Sodium Copper	3h45m	1	$^{64}\text{Cu}$	3	$^{61}\text{Cu}$							
								2	$^{24}\text{Na}$ $n, \gamma$					
								16h10m	1	$^{64}\text{Cu}$	3	$^{61}\text{Cu}$	1	$^{24}\text{Na}$ $n, \gamma$
								2	$^{64}\text{Cu}$	3	$^{61}\text{Cu}$	4	140 KeV	
29	Lamp (Computer type CM345)	Copper	93m	1	$^{64}\text{Cu}$	2	$^{61}\text{Cu}$							
								Copper	2h18m	1	2	$^{61}\text{Cu}$		
								Copper	18h18m	1	2	$^{61}\text{Cu}$		
29	Capacitor (Disc)	Copper	3h42m	1	$^{64}\text{Cu}$	2	$^{61}\text{Cu}$							
								Copper	15h50m	1	2	$^{61}\text{Cu}$		
								Sodium				1	$^{24}\text{Na}$ $n, \gamma$	
29	Resistor (36k Carbon)	Copper	1h43m	1	$^{64}\text{Cu}$	2	$^{61}\text{Cu}$							
								Copper	2h30m	1	2	$^{61}\text{Cu}$		
								Copper	14h 9m	1	2	$^{61}\text{Cu}$		
								Sodium				3	$^{24}\text{Na}$ $n, \gamma$	
29	Metal from (pin jack)	Copper	41m	1	$^{62}\text{Cu}$	2	$^{61}\text{Cu}$							
								1	$^{64}\text{Cu}$					
								Zinc	41m	3	$^{63}\text{Zn}$			
								Copper	2h22m	2	$^{64}\text{Cu}$	1	$^{61}\text{Cu}$	
								Zinc	2h22m	3	$^{63}\text{Zn}$			
								Copper	17h57m	1	$^{64}\text{Cu}$	2	$^{61}\text{Cu}$	
								Zinc	7d17h			2	$^{61}\text{Cu}$	
1	$^{67}\text{Cu}$ $\gamma, p$													
3	$^{58}\text{Co}$ $\gamma, np$													
29	Brass Nut	Zinc	32m	1	$^{63}\text{Zn}$	2	$^{61}\text{Cu}$							
								Copper	32m	1	$^{63}\text{Cu}$			
								Zinc	2h11m	1	$^{63}\text{Zn}$	3	$^{67}\text{Cu}$ $\gamma, p$	
								Copper				2	$^{61}\text{Cu}$	
								Zinc	18h41m	1	$^{63}\text{Zn}$	3	$^{67}\text{Cu}$ $\gamma, p$	
								Copper				2	$^{61}\text{Cu}$	
								Copper	1d17h	1	$^{64}\text{Cu}$	3	$^{61}\text{Cu}$	
								Zinc				2	$^{67}\text{Cu}$ $\gamma, p$	
1	$^{67}\text{Cu}$ $\gamma, p$													
181	Tantalum (Capacitor)	Tantalum	3h58m	2	$^{180}\text{Ta}$	1	$^{178}\text{Ta}$ $\gamma, 3n$							
								Tantalum	16h48m	1	$^{180}\text{Ta}$	2	$^{178}\text{Lu}$ $\gamma, n2p$	
												3	$^{178}\text{Ta}$ $\gamma, 3n$	

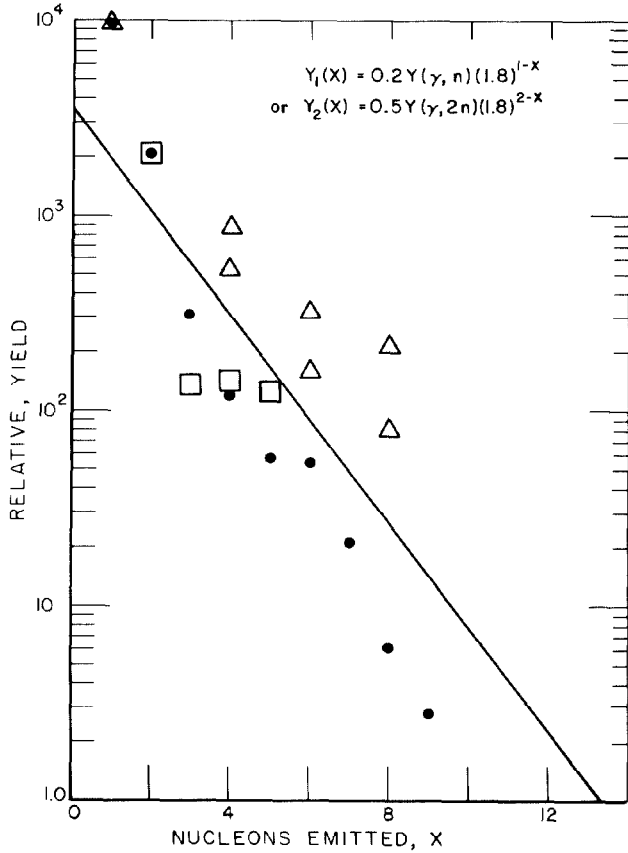


Fig. 1. Relative yield of nuclides from photonuclear processes leading to  $^{18}\text{F}^1$  (triangles), from  $^{141}\text{Pr}$  (squares) and from  $^{209}\text{Bi}$  (circles). For the fluorine case the number of nucleons emitted has been reduced by the number of spontaneous protons expected in the decay chain.

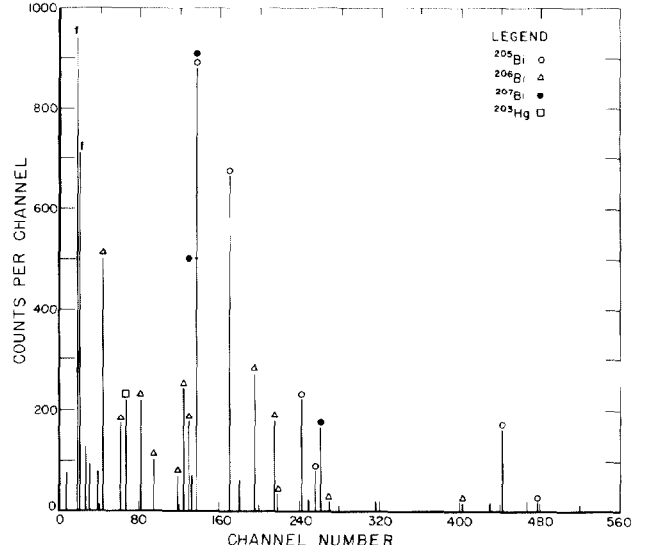


Fig. 2. A characteristic gamma ray spectrum as reduced by the on-line computer. Each line is proportional to the area under a peak in the initial Ge(Li) detector spectrum. This data was taken in a four hour period 55 days after the irradiation of a pure  $^{209}\text{Bi}$  sample. The lines have been identified as due to  $^{205}\text{Bi}$  (circles),  $^{206}\text{Bi}$  (triangles),  $^{207}\text{Bi}$  (crosses) and  $^{203}\text{Hg}$  (squares). The symbol F indicates that the line has been folded in the plotting routine by subtraction of  $1000xm$  counts where  $m$  is an arbitrary number.