MASTER

AECU-4441

ISOTOPIC METHOD
FOR AGE DETERMINATION
OF INDUSTRIAL PRODUCTS

Quarterly Progress Report
July - September 1959

CONTRACT AT (11-1)-719

Reported by: T C Gregson
S W Waisbrot

Approved by: S D Gehman

THE GOODYEAR TIRE AND RUBBER COMPANY
1144 East Market Street
Akron 16, Ohio

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

<u>SECTION</u>	<u>PAGE</u>
I. INTRODUCTION.	1
II. MATERIALS AND METHODS.	2
III. DISCUSSION OF RESULTS.	4
IV. FUTURE WORK.	6
APPENDIX I - Tables and Figures.	8
APPENDIX II - Error in Age Determined From an Isotope Mixture Due to Decay of Long-Lived Nuclide in Dating Pair.	21
APPENDIX III - Equations for Determination of Statistical Errors in Experimental Transmission Data.	24
APPENDIX IV - Observations to Date on the Hard Radiation Contamination of the Technetium-99 Beta Spectrum.	26

I. INTRODUCTION

It has been shown in the previous quarterly progress report that the isotopic method developed for age determination is both feasible and operable. This was demonstrated with several different isotope mixtures. However, these results have been confined to pairs containing very short-lived timer isotopes. Moreover, as a rule the short-lived isotope has also been the hard radiation emitting component of the pair, although an age equation was developed for the alternate situation (Appendix II, equation (34), April-June Report). In most cases the nuclides have been confined to pure beta emitters. Since the decay interval was too short at that time to evaluate longer half-lived species, the mixtures thought to be suitable for product application and which had been prepared and were being shelf aged for this purpose were held for future evaluation.

Other nuclide pairs differing from the beta emitters which have been used so far are possible. The applicability of a few of these alternate types were tested. These include beta and gamma, gamma and gamma, beta-gamma and beta, beta-gamma and gamma, and beta-gamma and beta-gamma emitting nuclide pairs.

Each of the three long-lived reference nuclides used previously has a specific shortcoming. The Cl-35 and the C-14 as carbonate are affected by acid fumes. The Tc-99 contains a hard energy component. Some observations are included in this report relative to this hard energy radiation. However, a more precise study will be required before a complete explanation for it is possible.

Other intermediate range nuclides are yet to be investigated with respect to their use as mixture reference materials. Since the half-life of the standard species in the pair needs only to appear long when compared to the timer species half-life, the use of several other standard species may be entirely feasible in pairs destined for relatively short age determinations. This possibility will be evaluated by studying the effect which some decay of the standard has on the accuracy of the measured ages.

The very principle upon which a Geiger tube operates gives rise to errors at high counting rates, the magnitude of which increase with the counting rate. The total count observed is always lower than the total count which should have been recorded due to the tube's lack of response to those pulses which are coincident with, or arrive during, that period of time in which the system is recovering from a pulse. This resolving time, or dead time, is a function of the GM tube's design and characteristics. It can be measured. Once this has been done a coincidence loss correction factor can be applied to the observed count. Up until this quarter, coincidence losses had been considered negligible. These have now been evaluated as insurance against the introduction of significant errors in age calculations.

178 3

I. INTRODUCTION (continued)

Statistical errors resulting from the random nature by which radioactive decay occurs are also a source of error in ages computed from experimental data. These are independent of the total time required for a counting determination but are functions of the natural background and the total counts accumulated during a particular observation. As filters are added to determine transmission curves, the counting rate decreases quite rapidly so that long counting times are required for accuracies comparable with those secured in measuring the unfiltered activities. Unfortunately, our normal method of counting was not set up on a pre-set count basis. This source of errors has now also been investigated.

The objective of this study is the development of a method for the age determination of industrial products. The final proof of the method's utility will, therefore, lie in its application to and demonstration on a representative products. This requires selection of representative products on which to try out the technique. Since products for which this method is suitable will be intended for consumer applications, the amounts of radioactivity in connection with such articles must be minimal so as not to exceed those concentrations specified by Federal Regulations. It is thus certain that product applications will require low level counting equipment for evaluation of the method. There are two types of equipment currently available which may prove suitable for this purpose. Studies will have to be undertaken with both types to determine which shows the most promise and the greatest versatility.

II. MATERIALS AND METHODS

The materials and methods utilized in this quarter for the long-term nuclide pairs are essentially the same as those previously described. The results of measurements on these samples as well as on those previously prepared are listed in Tables I-VII. These measurements have been substituted in equation 29 (April-June Report, p. 4) so as to compute the elapsed time. These results are shown in Table VIII.

The suitability of a given standard isotope is dependent on the ratio of its half-life to that of the short-lived timer isotope in the pair. An equation was developed which shows that the error in the measured age due to decay of the long-lived species of the pair is directly proportional to the half-life ratio of the pair species, (see Appendix II). For example, the error from this cause will be limited to 10% even if this ratio is only 0.1.

The "split-planchet" method⁽¹⁾ was used to determine the resolving time of the Tracerlab Superscalar counting apparatus which is currently in use. A planchet was split in two and a sample of nuclide placed in each

II. MATERIALS AND METHODS (continued)

half. These were designated as right and left so as to maintain the same geometry throughout the entire counting procedure. Sample activities were chosen and counted so as to yield optimum results (2). Each half was counted separately and then both were counted together, the same relative positions being used throughout the counts. The values so obtained were then substituted in the following equation (3).

$$T = \frac{n_1 + n_2 - n_{12} - B}{(n_{12})^2 - (n_1)^2 - (n_2)^2} \quad \text{where} \quad (53)$$

T = dead time, seconds
 n_1 = cps of the right half
 n_2 = cps of the left half
 n_{12} = cps of both halves
 B = cps background

This value of T can then be substituted in the equation

$$N = \frac{n}{1 - nT} \quad (54)$$

N = true counting rate
 n = obs. counting rate

so as to correct any observed counting rate for the resolving time loss. These results are shown in Table IX. For convenience equation (54) has been plotted for a 212 microsecond resolution time in Figure 1, Appendix I.

A study was made of the degree to which errors in transmission values obtained after high filtrations could be reduced by longer counting periods. Transmission values of a Sr-89, C-14 mixture were measured in the normal fashion by making three 3-minute counting determinations for each filter thickness. The same sample was then counted for three counting periods during each of which 10,000 counts were accumulated for each of the filter thicknesses. These data are shown in Table V and Appendix III. The deviations were computed from the equations likewise presented there. Since a constant time was not used in determining the transmission of the standard and the concentration is calculated from this standard, these concentrations are not statistically low in error.

In order to incorporate a radioisotope into a rubber sample, the tagged filler should be of such a physical nature that it can be handled with ease. For this reason P-32 was combined in $Mg_2P_2O_7$ before incorporation in an unvulcanized rubber compound. A similarly prepared sample

II. MATERIALS AND METHODS (continued)

contained C-14 as labeled octadecanol. A third sample of rubber contained both the P-32 and the C-14 in the same concentrations as used in the individual samples. After thin sheets (0.035" thick) of these rubbers were vulcanized, 1-1/4" diameter sample discs were clicked out. Both the separate samples and the mixture were counted using the various filter thicknesses. After specific age intervals, additional counts were made. The results are shown in Table X and Figures 2-5.

A few exploratory rubber samples were dated with isotopic mixtures and counted on one of the currently available types of equipment for precision low level counting: a liquid scintillation packaged unit. Rubber samples were prepared by adding the isotopes Ca-45, Cl-36, Nb-95 and Tc-99 to the raw rubber, mixing and vulcanizing. Isotope pairs (Ca-45, Cl-36 and Nb-95, Tc-99) were similarly incorporated in other rubber samples. All samples were pulverized by freezing with liquid nitrogen and grinding in the Micro Wiley mill so as to pass through a 60 mesh screen. A more finely divided sample was prepared by grinding this material with a mortar and pestle under liquid nitrogen. Suspension samples suitable for liquid scintillation counting were prepared by the method of Ott et al. (4). Samples of tagged, ground rubber, were weighed into a vial which was then filled with Cab-O-Sil M-5. Toluene (15 ml) containing 4g/liter diphenyl oxazole was added and the mixture was shaken to disperse and suspend the rubber particles. Activities of these samples were measured with the Tri-Carb liquid scintillation spectrometer manufactured by Packard Instrument Company. Activities of the dry ground rubber were also determined by counting samples with our G-M Superscalar equipment.

III. DISCUSSION OF RESULTS

The results obtained by incorporating radioisotopes into rubber did not show a close correlation between the observed time and the calculated time. As can be seen in Table X, there is some self absorption due to the rubber. This has been corrected for by using as the plot-standard a sample of P-32 in rubber, the size and geometry of which have been kept identical to those of the mixture specimen.

A somewhat cursory examination of the extent to which self absorption may pose a problem leads us to believe that the method which has been used entirely eliminates the need for any consideration on the part of either constituent in the pair.

In addition, one must also take into account the fact that octadecanol labeled with C-14 tends to migrate to the upper surface of the rubber sample. Since this migration reaches an equilibrium value part of the C-14 radiation is also absorbed and part is brought closer to the G-M tube changing the geometry of the counting apparatus. If we consider

III. DISCUSSION OF RESULTS (continued)

that equilibrium has been reached after 13 days, then the calculated results match the observed results more closely and the method with modifications can be considered applicable to dating of rubber products. Obviously, in any actual dating mixture one must strive to prepare a non-migratory chemical form of the pair constituents.

Contrary to the results obtained previously from pairs of isotopes used in dating, the results summarized in Table IX give poor agreement between observed and calculated time. Pairs containing either Nb-95 or P-32 as the timer species are the only samples which show good agreement. This can probably be attributed largely to the fact that the elapsed age period is still less than one timer species half-life. This is the period where error is greatest. Then, too, an appreciable error arises from the use of octadecanol as the source of C-14. This material not only absorbs some of its own radiation, but also some of the radiation of the second isotope. Since this layer of octadecanol is non-uniform the results will be inconsistent. Additional samples are to be prepared using amorphous carbon as the source of C-14.

The 212 microsecond resolving time observed for the G-M actuated Tracerlab Superscalar system agrees well with values published in the literature. A coincidence loss correction was applied (from Figure 1) to the analysis of the Tc-99 transmission data and to the study of the advantages of high total counts under high filtration conditions. The correction is quite small and can be neglected in all cases where the rates are less than 10,000 CPM.

The data in Figure 6 illustrate the much greater accuracy one can obtain by following a constant preset count method throughout the measurements required to establish a transmission curve rather than by the preset time method which we have unfortunately been following. Such errors quite naturally influence the age determined from transmission data. We are revising our standard procedure to eliminate such deviations in future work.

Analysis of the Tc-99 data (see Appendix IV) has shown that our observations are rendered inconclusive by these very same large statistical errors. One cannot state with any degree of reliability that decay of the hard radiation constituent does or does not occur.

The data gathered during the liquid scintillation counting of suspensions of ground rubber samples containing various isotopic species and mixtures will be presented in the October Progress Letter. As yet we have not been able to draw many conclusions from these data. However, the experience gained in this exploratory study will be of immeasurable help in designing a future evaluation program.

IV. FUTURE WORK

Low level counting studies will be conducted with liquid scintillation counting equipment and with anti-coincidence shielded equipment to determine which type is most advantageous for this work. Equipment will be selected and purchased on this basis.

Low level techniques requisite for product application studies will be developed.

The technetium -99 study will be continued. All determinations will be conducted on a preset count basis.

The suitability as pair standards of isotopes with half-lives shorter than those heretofore used will be demonstrated experimentally.

REFERENCES

- (1) Cooper and Cotton, Science 129, 1360 (1959).
- (2) Beers, Rev. Sci. Inst. 13, 72 (1942).
- (3) ORSORT Reactor Physics Course, Experiment #6 (1953).
- (4) Ott, Richmond, Trujieo, and Foreman, NU 17 #9, 106 (1959).
- (5) Jarrett, AECU-262 (1946).

APPENDIX I
TABLES AND FIGURES

TABLE I

<u>Sample 13A</u>		% Transmission				A standard (t, X ₀)		
Element	Filter mg/cm ² Al	<u>Cl³⁶</u>	<u>← Cl³⁶ + Ca⁴⁵ →</u>			<u>A mixture (t, X₀)</u>		
Time, days		-	<u>0</u>	<u>20</u>	<u>63</u>	<u>0</u>	<u>20</u>	<u>63</u>
	33.7	67.2	34.3	36.4	38.0	.510	.542	.565
	70.7	33.3	14.9	15.4	16.7	.447	.462	.502
	106.9	13.5	5.9	5.96	6.79	.437	.441	.503
	140	4.8	2.2	2.06	2.28	.458	.429	.475
	175.2	1.5	.62	.57	.71	.413	.380	.473
	263	-	-	-	-			
					Avg.	.447	.444	.493

TABLE II

<u>Sample 13B</u>		% Transmission				A standard (t, X ₀)		
Element	Filter mg/cm ² Al	<u>Cl³⁶</u>	<u>← Cl³⁶ + Ca⁴⁵ →</u>			<u>A mixture (t, X₀)</u>		
Time, days		-	<u>0</u>	<u>16</u>	<u>64</u>	<u>0</u>	<u>16</u>	<u>64</u>
	21.4	83.7	50.8			.607		
	33.7	69.4	38.4	37.9	40.4	.553	.546	.582
	70.7	33.4	16.8	16.9	17.3	.503	.506	.518
	106.9	13.9	6.6	7.23	6.95	.475	.520	.500
	140	5.0			2.57			.514
	175.2	1.6			.76			.475
	263	.12			-			
					Avg.	.510	.524	.518

178

11

TABLE III

Sample 36

Element
Time, DaysFilter
mg/cm²AlCa⁴⁵

% Transmission

 $\leftarrow \frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)} \rightarrow$
 $\leftarrow \frac{A_{\text{Ca}^{45}}}{A_{\text{Ca}^{45} + \text{C}^{14}}} \rightarrow$
0 1 27 37 57 98
 $\frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)}$
0 1 27 37 57 98

3.2	74.0	61.9	63.0				
10.5	27.2	18.4	17.5	16.9	16.4	16.5	15.6
24.0	4.7	2.7	2.5	2.8	2.5	2.83	2.33
28.1	2.2	1.4	1.5				
33.7	.86	.52	.37	.54	.60	.46	.52

.836	.851				
.676	.643	.621	.603	.607	.574
.574	.532	.596	.532	.602	.496
.636	.682				
.605	.430	.628	.698	.534	.605
Avg.	.623	.619	.615	.611	.581

Avg.

TABLE IV

Sample 37

Element
Time, daysFilter
mg/cm²AlTe²⁰⁴

% Transmission

 $\leftarrow \frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)} \rightarrow$
 $\leftarrow \frac{A_{\text{Te}^{204}}}{A_{\text{Te}^{204} + \text{C}^{14}}} \rightarrow$
0 41 42 43 125
 $\frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)}$
0 41 42 43 125

21.4						
24.0	54.5	38.8	38.8	38.3	38.3	37.8
33.7	42.7	29.0	30.5	30.0	30.0	30.0
70.7	17.6	12.3	12.2	12.0	12.1	12.2
106.9	6.9	4.8	5.0	4.8	4.8	4.77
175.2	.80	.49	.69	.61	.62	.76
429	-	-	-	-	-	-

	x	x	x	x
.712	.712	.703	.703	.694
.679	.714	.703	.703	.703
.699	.693	.682	.688	.693
.696	.725	.696	.696	.691
.613	.862	.762	.775	.950

Avg.

.680	.710	.696	.697	.695
------	------	------	------	------

TABLE V

Sample 38

Element	Filter	89	% Transmission								A standard (t, X ₀)							
			← Sr ⁸⁹ + C ¹⁴ →								A mixture (t, X ₀)							
Time, Days	mg/cm ² Al	Sr	0	14	35	54	62	63	74	75	0	14	35	54	62	63	74	75
106.9	45.3		24.2	23.0	20.8	17.4	16.9				.534	.508	.459	.384	.373			
175.2	28.5		14.4	13.8	12.7	10.6	10.8				.505	.484	.446	.372	.366			
263.0	13.2		6.9	6.6	6.55	5.5		5.65			.523	.500	.496	.417		.427		
351	6.0		2.9	3.0	3.36	2.9			2.74		.483	.500	.560	.483			.457	
429	2.9		1.4	1.5	1.87	1.5				1.55	.483	.517	.645	.517				.535
618	.57		.32	.35	.30	.29					.561	.614	.526	.509				
843	-		-	-	-	-												
Avg.											.506	.502	.467	.414				

TABLE VI

Sample 39

Sample 39		% Transmission					A standard (t, X ₀)		
Element	Filter	P32	← P ³² + C ¹⁴ →			A mixture (t, X ₀)			
			0	12	23	0	12	23	
	24.0		38.5						
	33.7	90.1	36.5			.405			
	70.7	76.8	28.6	20.8	13.6	.372	.271	.177	
	106.9	63.0	22.1	15.8	9.9	.351	.251	.157	
	175.2	40.7	13.1	9.5	6.2	.322	.233	.152	
	263		6.2	4.4	2.8				
	429	4.08	1.3	.88	.46	.319	.216	.113	
	618	.252	.14	-	-	.556			
Avg.						.341	.243	.150	

TABLE VII

<u>Sample 47</u>					A standard (t, X ₀)	
Element	Filter	Nb ⁹⁵	← Nb ⁹⁵ + Tc ⁹⁹ →		A mixture (t, X ₀)	
Time, Days		<u>Nb</u>	<u>0</u>	<u>18</u>	<u>0</u>	<u>18</u>
% Transmission						
21.4			14.3	15.3		
33.7			6.20	6.08		
70.7			2.70	2.35		
106.9			2.46	2.13		
175.2			2.21	1.93		
429		3.56	1.74	1.46	.489	.410
618		3.20	1.55	1.30	.484	.406
843		3.41	1.49	1.17	.437	.343
1708		2.88	1.23	.93	.427	.323
Avg.					.459	.371

TABLE VIII

Sample No.	Short-lived Isotope	Long-lived Isotope	$\frac{A}{\lambda}$ standard (t_0, X_0)	$\frac{A}{\lambda}$ standard (t, X_0)	λ , Days	Time Calc. Days	Time Obs., Days
13A	Ca ⁴⁵	Cl ³⁶	.447	.444	163	-	20.0
13B	Ca ⁴⁵	Cl ³⁶	.447	.493		43	63.0
36	Ca-45	C-14	.510	.524	163	13.5	16.0
			.510	.518		8.7	64.0
			.623	.619	163	5.87	1
			.623	.615	163	8.1	27
			.623	.611	163	12.4	37
			.623	.581	163	41.0	57
			.625	.558	163	63	98
37	Tl-204	C-14	.680	.710	4.0 yrs	NO	41
			.680	.696	4.0 "	CH	42
			.680	.697	4.0 "	ANGE	43
			.680	.695	4.0 "		125
38	Sr-89	C-14	.506	.502	53	1.25	14
			.506	.467	53	12.0	35
			.506	.414	53	28.6	54
39	P-32	C-14	.341	.243	14.3	9.8	12
			.341	.150	14.3	22	23
40B	Nb-95	Cl-36	.624	.421	35	41	21
			.624	.382	35	49.7	41
			.624	.209	55	92.4	90
41	P-32	Pm-147	.990	.959	14.3	-	7
47	Nb-95	Tb-99	.459	.371	35	18.2	18

TABLE IX

Resolving time of Tracerlab Superscaler

Planchet cut in half, Tl^{204} on each half.
Also two half-planchets blank.

Sample 1 - L		R	Av. cpm of 10-3 min counts = 34048 (N_1).
	R	Av. cpm of 10-3 min counts = 32317 (N_2).
	R	Av. cpm of 10-3 min counts = 61002 (N_{12}).
Sample 2 - L		R	Av. cpm of 10-3 min counts = 12098 (N_1).
	R	Av. cpm of 10-3 min counts = 12672 (N_2).
	R	Av. cpm of 10-3 min counts = 23762 (N_{12}).

$$\text{Formula } T = \frac{n_1 + n_2 - n_{12} - B}{n_{12}^2 - n_1^2 - n_2^2}$$

Sample 1 High Count	Sample 2 Lower Count
$n_1 = 568.13$ cps	$n_1 = 202.30$ cps
$n_2 = 539.28$ cps	$n_2 = 211.87$ cps
$n_{12} = 1017.37$ cps	$n_{12} = 396.70$ cps
$B = .67$ cps	$B = .67$ cps
$T = .00021206532$ = 212 microseconds	$T = .0002347788$ = 235 microseconds

Figure 1
Coincidence Loss Correction Curve

Observed Counting Rate, CPM

% TRANSMISSION

FIGURE 3

TRANSMISSION P-32
R5#1 - P-32 IN RUBBER

FILTER THICKNESS mg/cm²
178 19

90% TRANSMISSION

FIGURE 4

TRANSMISSION RS #3
 P-30 + C-14 IN PUGG-10

- --- 0 time
- ⊙ --- 13 days
- X --- 18 "
- Δ --- 13 "

FIGURE 6

1/2 841 TRANSMISSION, $\frac{A}{A_0}$ (RS #1)

Curve 4
 Curve 3
 Curve 2
 Curve 1

Time obs. Time calc.

curve 2	13.0 d.	19.0 d.
curve 3	18.0	25.9
curve 4	35.0	44.0

TRANSMISSION, $\frac{A}{A_0}$ (RS #3)

.391 .556 .578 .716

TABLE X

RS #1

Element	Filter	% Transmission		% Transmission		% Transmission		$\frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)}$			
Time, Days	mg/cm ² Al	p ₃₂	8/27	9/9	9/14	9/23		0	13	18	27
		-	0	13	18	27					
	33.7	89.4	80.1	82.3	83.0	81.9		.896	.921	.928	.916
	70.7	75.9	62.2	63.7	65.2	64.1		.819	.829	.859	.844
	106.9	62.2	49.4	49.5	51.6	51.8		.794	.796	.830	.833
	175.2	40.7	27.9	28.7	28.9	27.4		.686	.705	.710	.673
	263	20.1	12.8	12.9	13.4	12.7		.637	.642	.667	.632
	351		5.05	4.93	5.13	5.56		-	-	-	-
	429	4.14	1.87	1.84	2.07	.98		.451	.444	.500	.237
	618	.28	-	-							
	843		-								

RS #2

Element	Filter	% Transmission		% Transmission		% Transmission		$\frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)}$			
Time, Days		c ₁₄	8/28	9/10	9/15	9/30		0	13	18	33
		-	0	13	18	33					
	1.6	68.9	72.7	70.0	72.9	73.2		1.055	1.016	1.058	1.062
	2.7	52.5	57.8	49.4	58.6	57.8		1.101	.941	1.116	1.101
	3.2	45.4	46.5	49.2	53.1	51.6		1.024	1.084	1.170	1.137
	5.2	24.1	26.2	27.5	28.6	28.3		1.087	1.141	1.187	1.174
	6.1	14.3	17.4	16.9	17.3	16.3		1.217	1.182	1.210	1.140
	10.5	4.00	4.63	5.25	5.79	5.17		1.158	1.313	1.448	1.292
	13.6	1.62	2.78	1.79	2.39	1.72		1.716	1.105	1.475	1.062
	21.4	-	.69	-	.76						

RS #3

Element	Filter	% Transmission		% Transmission		% Transmission		$\frac{A_{\text{standard}}(t, X_0)}{A_{\text{mixture}}(t, X_0)}$			
Time, days, Obs.		RS1	8/28	9/10	9/15	10/2		0	13	18	35
Calc.			0	13	18	35					
	33.7	80.1	62.8	45.8	40.5	23.0		.784	.572	.506	.287
	70.7	62.2	49.2	36.3	32.3	18.5		.791	.584	.519	.297
	106.9	49.4	37.2	28.0	24.7	14.1		.753	.567	.500	.285
	175.2	27.9	21.7	15.9	13.8	8.26		.778	.570	.495	.296
	263	20.1	12.8	12.9	13.4	12.7		.720	.520	.525	.284
	351	5.05	3.61	2.94	2.56	1.16		.715	.582	.507	.230
	429	1.87	1.58	1.26	1.47	.74		.845	.674	.786	.396
	618	-	-	-	.26	-					
							Av.	.776	.578	.505	.291

Figure 6

Relative Accuracy of Transmission Data Obtained Under Standard Procedure & Under Long-Counting Conditions

Transmission %

359-60 KEUFFEL & ESSER CO.
Semi-Logarithmic, 2 Cycles X 60 Divisions,
MADE IN U.S.A.

Data for Sample #38, a mixture of Sr-89 & C-14

o - 7/2/59 - Computed from counting data collected in normal fashion

Δ - 8/6/59 - Computed from data obtained during very long count

□ - 8/18/59 - Ditto

95/100 Errors Indicated

Absorber Thickness, mg Al per cm²

178

23

0.05%

APPENDIX II

ERROR IN AGE DETERMINED FROM
ISOTOPE MIXTURE DUE TO DECAY OF
LONG-LIVED NUCLIDE IN DATING PAIR

APPENDIX II

ERROR IN AGE DETERMINED FROM ISOTOPE MIXTURE DUE TO DECAY OF LONG-LIVED NUCLIDE IN DATING PAIR

When the long-lived species chosen as a standard for the dating mixture undergoes a detectable degree of decay during the age period, the age computed from the experimentally determined nuclide ratios will be erroneous. To prevent such an occurrence one quite naturally selects as the standard isotope a species with an extremely long half-life. Such an approach may be unwarranted where the age period of interest is of a relatively short-duration. One may be imposing an unnecessary hardship on the selection of a standard in such instances. Thus, it is of interest to determine the magnitude of the error in age which is introduced by decay of the long-lived constituent as a function of both the half-life ratio of the pair components and the fractional decay which occurs in the short-lived species.

Such an error relationship can be developed for the case where the short-lived material in the pair is the hard emitter through the use of age equation (29).

$$t = 3.317 \tau_s \log_{10} \frac{\left[\frac{Ts^*(t, x_a)}{Tm(t, x_a)} - 1 \right]}{\left[\frac{Ts^*(t_0, x_a)}{Tm(t_0, x_a)} - 1 \right]} \quad (29)$$

Since

$$Tm(t, x_a) = \frac{As(t, x_a)}{A_1(t, x_0) + As(t, x_0)} \quad (5)$$

equation (29) can be transformed so as to read

$$t = 1.442 \tau_s \log_e \left\{ \frac{\left[\frac{As^*(t, x_a)}{As^*(t, x_0)} - 1 \right]}{\left[\frac{As(t, x_a)}{As(t, x_0)} - 1 \right]} \cdot \frac{\left[\frac{As(t, x_0)}{As(t, x_0) + A_1(t, x_0)} - 1 \right]}{\left[\frac{As(t_0, x_a)}{As(t_0, x_0)} - 1 \right]} \cdot \frac{\left[\frac{As(t_0, x_0)}{As(t_0, x_0) + A_1(t_0, x_0)} - 1 \right]}{\left[\frac{As(t_0, x_0)}{As(t_0, x_0) + A_1(t_0, x_0)} - 1 \right]} \right\} \quad (55)$$

When the plot-standard species is chosen identical to the dating mixture standard (as it is in the technique we have developed) we find that

$$\frac{As^*(t, xa)}{As^*(t, xo)} = \frac{As^*(to, xa)}{As^*(to, xo)} = \frac{As(t, xa)}{As(t, xo)} = \frac{As(to, xa)}{As(to, xo)} \quad (56)$$

so that equation (55) reduces to

$$t = 1.442 \tau \log_e \left\{ \frac{\frac{A_1(t, xo)}{As(t, xo)}}{\frac{A_1(to, xo)}{As(to, xo)}} \right\} \quad (57)$$

Now the error in the experimentally measured age resulting from some decay of the long-lived pair species can be estimated by evaluating the following equation:

$$\left\{ \begin{array}{l} \text{The fractional} \\ \text{error in the} \\ \text{measured age} \\ \text{due to decay} \\ \text{of the long-} \\ \text{lived species} \end{array} \right\} = \frac{\left\{ \begin{array}{l} \text{Actual age as} \\ \text{computed without} \\ \text{decay of the long-} \\ \text{lived species} \end{array} \right\} - \left\{ \begin{array}{l} \text{Experimentally de-} \\ \text{termined age where-} \\ \text{in decay of long-} \\ \text{lived species has} \\ \text{occurred.} \end{array} \right\}}{\left\{ \begin{array}{l} \text{Actual} \\ \text{Age} \end{array} \right\}} \quad (58)$$

This reduces to

$$\frac{\Delta t}{t} = 1 - \frac{\left\{ \begin{array}{l} \text{Age with long-} \\ \text{lived decay} \end{array} \right\}}{\left\{ \begin{array}{l} \text{Age without} \\ \text{long-lived decay} \end{array} \right\}} \quad (59)$$

Substituting from equation (57) into equation (58) yields

$$\frac{\Delta t}{t} = 1 - \frac{\left[1.442 \tau \log_e \left\{ \frac{\frac{A_1^*(t, xo)}{As(t, xo)}}{\frac{A_p(to, xo)}{As(to, xo)}} \right\} \right]}{\left[1.442 \tau \log_e \left\{ \frac{\frac{A_1(t, xo)}{As(t, xo)}}{\frac{A_1(to, xo)}{As(to, xo)}} \right\} \right]} \quad (60)$$

Where: $A_1^*(t, xo)$ is the activity of the long-lived species after decay.

Since

$$\begin{aligned} A_1(t, x_0) &= A_1(t_0, x_0) \\ A_1'(t, x_0) &= A_1(t_0, x_0) e^{-0.693 t/t_2} \\ A_2(t, x_0) &= A_2(t_0, x_0) e^{-0.693 t/t_2} \end{aligned}$$

substitution of these relationships into equation (60) yields after simplification

$$\frac{\Delta t}{t} = 1 - \frac{\log_e \left[e^{-0.693 t \left(\frac{1}{t_2} - \frac{1}{t_0} \right)} \right]}{\log_e \left[e^{-0.693 t/t_0} \right]} \quad (61)$$

which likewise reduces to

$$\frac{\Delta t}{t} = \frac{t_0}{t_2} \quad (62)$$

Thus the error in measured age due to decay of the long-lived species of the pair is directly proportional to the half-life ratio of the pair species.

APPENDIX III

EQUATIONS FOR DETERMINATION OF
STATISTICAL ERRORS IN
EXPERIMENTAL TRANSMISSION DATA

APPENDIX III

EQUATIONS FOR DETERMINATION OF STATISTICAL ERRORS IN EXPERIMENTAL TRANSMISSION DATA

The fractional error in an experimentally determined counting rate resulting from both the random nature of radioactive decay of the species being counted and the statistical fluctuations in the background counting rate (5) can be computed according to the following equation

$$\frac{x}{X} = \frac{k \sigma}{A_1} = \frac{k \left(\frac{A_T}{t_T} + \frac{A_B}{t_B} \right)^{1/2}}{A_T - A_B} \quad (63)$$

where: x = error in the directly measured quantity X .

$k \sigma$ = absolute deviation of the observed counting rate from the true mean counting rate.

t = counting interval, time units and subscripts T and B are indicative respectively, of the total activity from both the species being counted plus the background and of the background alone.

The transmission of activity through a filter of mass thickness x at any time t is calculated according to the relation.

$$T_1(t, x) = \frac{A_1(t, x)}{A_1(t, x_0)} \quad (1)$$

Since transmission is an indirectly measured quantity calculated from the directly measured results of two separate experimental determinations, the accuracy of transmission values can be estimated from the following type relationship (5):

$$\frac{u}{U} = \left[\left(\frac{x}{X} \right)^2 + \left(\frac{y}{Y} \right)^2 \right]^{1/2} \quad (64)$$

where: U = quantity calculated as either the product or quotient of experimentally measured quantities X and Y .

u, x, y = errors in quantities U, X , and Y .

Therefore:

$$\left\{ \begin{array}{l} \text{The fractional error} \\ \text{in the transmission} \\ \text{computed for any fil-} \\ \text{ter thickness } x. \end{array} \right\} = \left[\left\{ \begin{array}{l} \text{The fractional} \\ \text{error in the} \\ \text{activity measured} \\ \text{through filter} \\ \text{thickness } x. \end{array} \right\}^2 + \left\{ \begin{array}{l} \text{The fractional} \\ \text{error in the} \\ \text{activity measured} \\ \text{for the unfiltered} \\ \text{sample.} \end{array} \right\}^2 \right]^{1/2}$$

Combining the above equations and simplifying leads to

$$\frac{\Delta T_1(t,x)}{T_1(t,x)} = k \left\{ \frac{\left[\frac{A_T(t,x)}{t_T} + \frac{A_B(t)}{t_B} \right]}{\left[A_T(t,x) - A_B(t) \right]^2} + \frac{\left[\frac{A_T(t,x_0)}{t_T} + \frac{A_B(t)}{t_B} \right]}{\left[A_T(t,x_0) - A_B(t) \right]^2} \right\}^{1/2} \quad (65)$$

APPENDIX IV

OBSERVATIONS TO DATE ON THE HARD RADIATION
CONTAMINATION OF THE TECHNETIUM-99
BETA SPECTRUM

APPENDIX IV

OBSERVATIONS TO DATE ON THE HARD RADIATION
CONTAMINATION OF THE TECHNETIUM-99 BETA SPECTRUM

This subject along with a preliminary analysis has been treated to some extent in the April-June progress report, Appendix III. Additional transmission measurements have been made since that time, and the accuracies of all transmission data have been computed from equation (66). The data to date are shown in Figures 7 and 8.

Unfortunately the counting procedure which was followed did not provide a sufficiently large number of counts after insertion of the various filters to render statistical errors negligible. The very magnitude of these errors tends to obscure any reductions in activity arising from decay of the contaminant, if such actually does occur.

If one contaminating nuclide species were present, or if the hard radiation is generated either directly or indirectly by Tc-99 itself, a constant slope should be obtained for the transmission curves at various times. Slopes computed by least squares show an appreciable variation.

If there is decay with time, the transmission intercepts (at zero absorber thickness) should show decreasing values with time. This has been noted, with the exception of the data of 5/19/59, but such changes do not follow the expected exponential decrease.

At best, one must conclude that our data are inconclusive, and this is primarily due to large statistical errors.

We intend to continue this study using constant counts for each filter thickness and of such magnitudes as to remove statistical uncertainties.

Figure 7

Effect of Shell Aging on Hard
Radiation Component of Activity
Emanating from Tc-99
(Shipment #2)

Transmission, %

KE SEMI-LOGARITHMIC 359-61G
KEUFFEL & ESSER CO. MADE IN U.S.A.
2 CYCLES X 70 DIVISIONS

75/100 Errors Indicated

* Characteristics of best fitting straight line
(via least squares) for filtrations of $\geq 70.7 \text{ mg/cm}^2$

Absorber Thickness, mg Al per cm^2

Figure 8

Effect of Shelf Aging on Hard
Radiation Component of Activity
Emanating from Tc-99
(Shipment #1)

SEMI-LOGARITHMIC 359-61G
KEUFEL & ESSER CO. MADE IN U. S. A.
2 CYCLES X 70 DIVISIONS

Transmissions, 170

Symbol	Date	Slope*	Intercept*
O -	3/27/59	-0.002103	2.47
Δ -	5/22/59	-0.001429	1.80
* -	6/2/59	-0.005077	1.59
X -	7/27/59	-0.001237	1.54
□ -	9/24/59	-0.001328	1.50

95/100 Errors Indicated

* Characteristics of best fitting
straight line (via least squares) for
filtrations of $\geq 20.4 \text{ mg/cm}^2$

Absorber Thickness, mg Al per cm²

178

34