Sensitivity of Bremsstrahlung Activation Analysis For Iodine Determination[1,2,3]

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INTRODUCTION

The measurement of iodine in concentrations which exist in biological samples is a relatively complex and time-consuming procedure by present chemical techniques, and the methods are subject to contamination by other materials, such as mercury. Activation analysis with thermal neutrons has been used to eliminate the final step in the chemical procedure or for the determination of serum protein bound iodine (1, 2). However, other elements normally present in biological samples, particularly sodium, are activated by the (n, γ) reaction and may have to be removed before the ^{128}I, which is produced, can be measured. Such contamination may not be a problem if the (γ, n) reaction can be utilized to produce iodine-126. Previous work on this reaction has been directed towards determining cross-section values and threshold levels (3-8). In the present study, the sensitivity of the reaction and the possible interference by contaminating elements in normal beef thyroid tissue and pooled human serum have been evaluated.

METHODS

Sample preparations: Dowex 1-x8 (50-100 mesh) resin in the chloride form was washed first with tripled-distilled H_2O followed by ammonium acetate until the Cl^- in the resin was replaced by the acetate ion. The excess ammonium was removed by frequent H_2O washings. The resin column of 0.3 ml volume was placed in a graduated Tomac[1] lcc polypropylene insulin syringe. The flow rate through the resin was determined primarily by the resistance of the resin bed and the glass wool plug which was placed in the bottom of the column to support the resin bed.

In one experiment to evaluate sodium contamination, 10 ml samples containing stable KI (5 μg to 20 μg) in either tripled-distilled H_2O or 0.9% NaCl were prepared. Duplicate 5 ml aliquots were pipetted onto two resin columns. To determine whether the inorganic iodide was getting through the resin column, ^{131}I standards were prepared in both H_2O and 0.9% NaCl, and various amounts of stable KI (5 μg to 1 mg) were added. The fraction of the added ^{131}I in the effluent, after passage through the resin column, was less than one per cent of
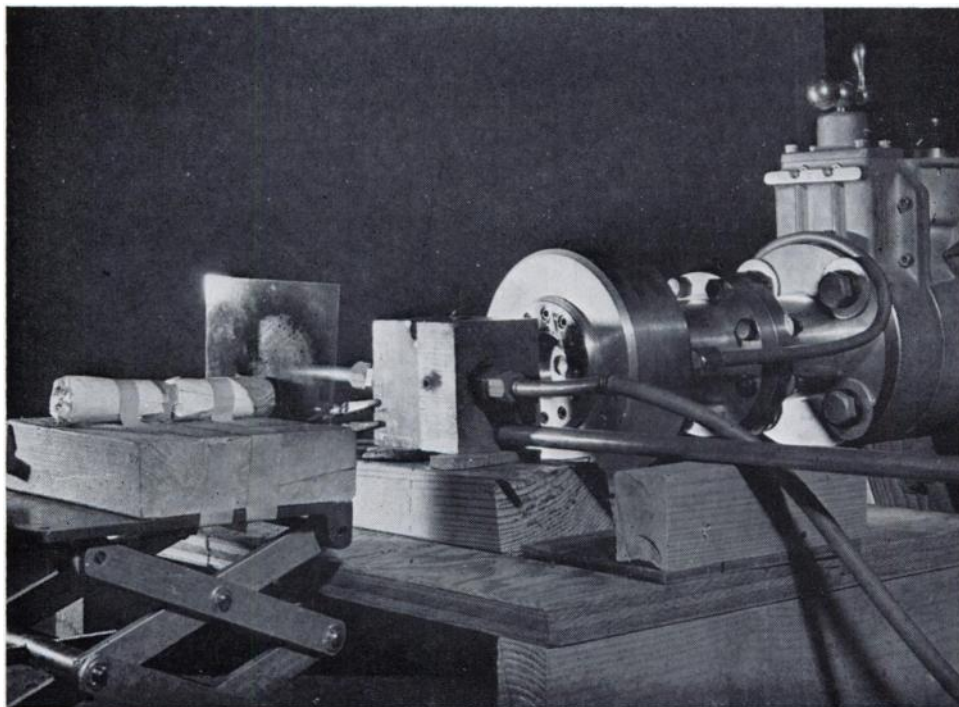

Fig. 1. Sample positioning for activation analysis. Figure shows relationship between end-window of the electron beam, the H_2O cooled aluminum block, and the plastic holders containing 10 samples each.

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[1]Pharmaseal Laboratories, Inc., Glendale 1, California.

the total activity added. The syringes were sealed and placed within a large plastic holder. The holders held two tiers, each containing five samples.

In a second experiment, a 200 mg slice of calf-thyroid tissue was placed directly into a plastic capsule and irradiated.

In a third experiment to evaluate possible mercury contamination in human serum, 1 ml of Mercuhydrin (containing 39 mg of Hg/ml) was injected into a patient, and two hours later 10 ml of blood withdrawn. Five ml of serum was then passed over 0.3 ml of cation resin (Dowex 50W-x8, H^+) and the column prepared for irradiation.

In a fourth series of experiments, 20 ml of pooled human serum was passed over 1.5 ml of anion resin, the column sealed and irradiated.

Activation: Each plastic holder was irradiated for 90 minutes in air using a 22 MeV linear accelerator. The average current during the irradiation time was 250 microamps. The plastic holder was placed 5 cm behind either a 6 cm thick H_2O cooled aluminum block (Fig. 1) or 10 cm behind a H_2O cooled thin tungs-
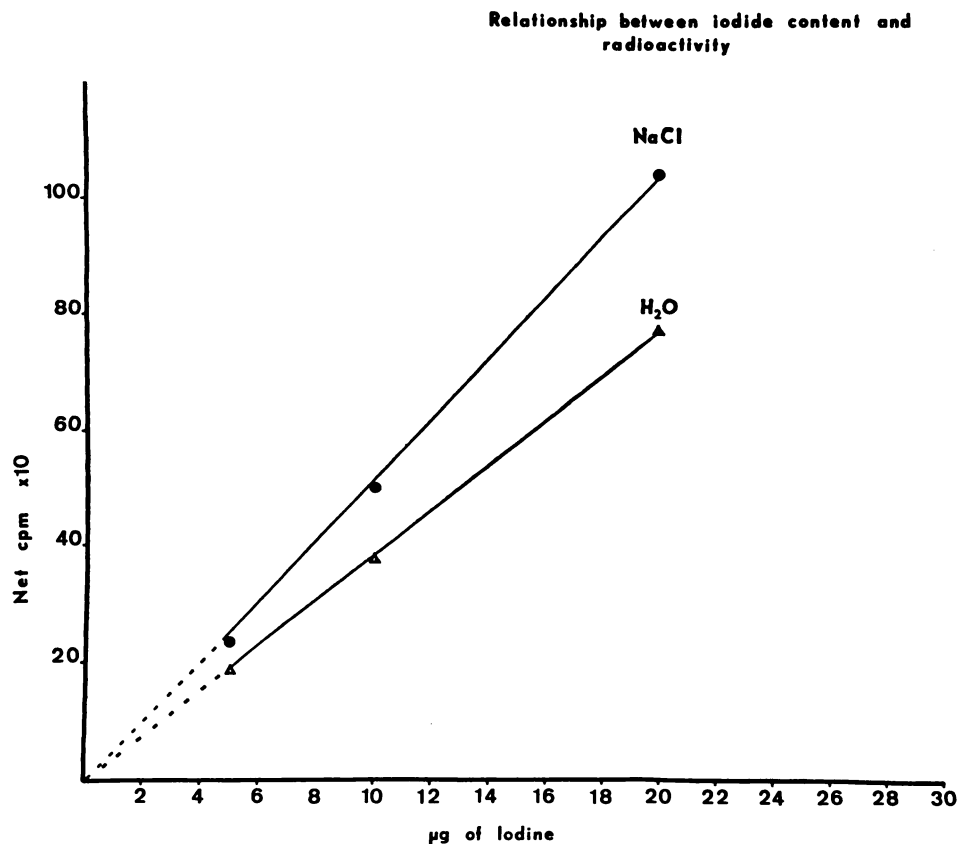

Fig. 2. Relationship between iodide content (KI standards) and induced radioactivity. Note: There is a linear relationship between iodide content and radioactivity expressed as net counts. (The greater radioactivity in the NaCl standard is a reflection of different irradiation conditions rather than an effect of NaCl on induced radioactivity).
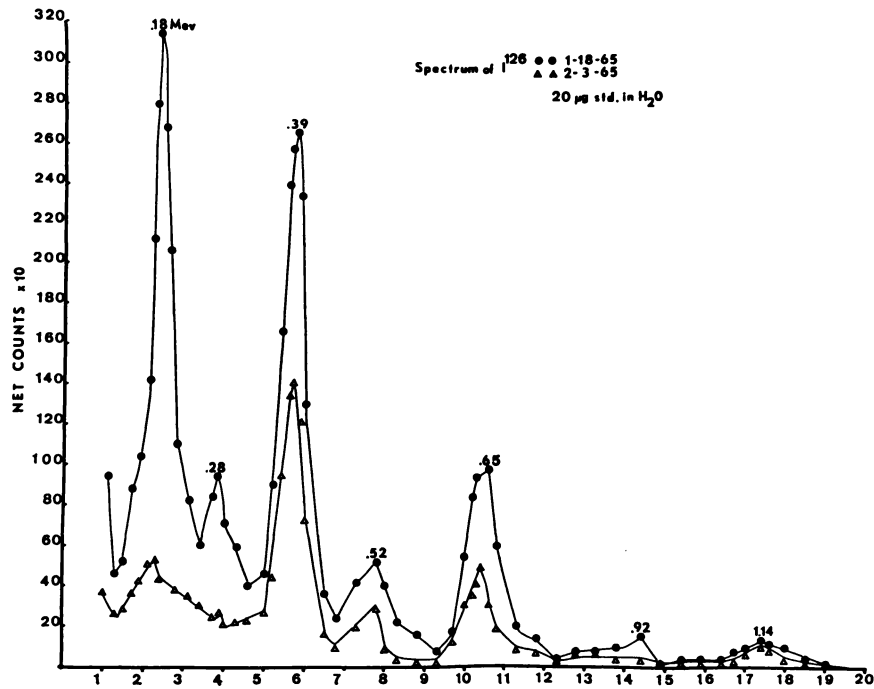
Fig. 3. Spectrum of a 20 μg standard of ¹²⁶I in H₂O. Note: Spectrum indicates the change in the energy peaks as seen on 1/18/65 compared with a repeat analysis taken on 2/3/65.
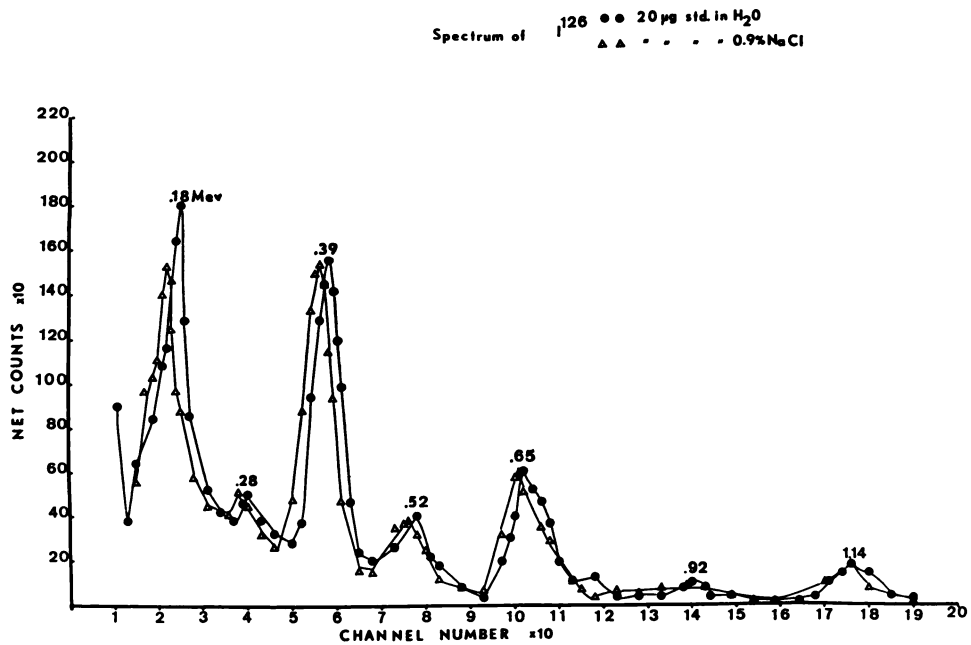

Fig. 4. Spectrum of a 20 μg standard prepared in two different media, H₂O and 0.9% NaCl.
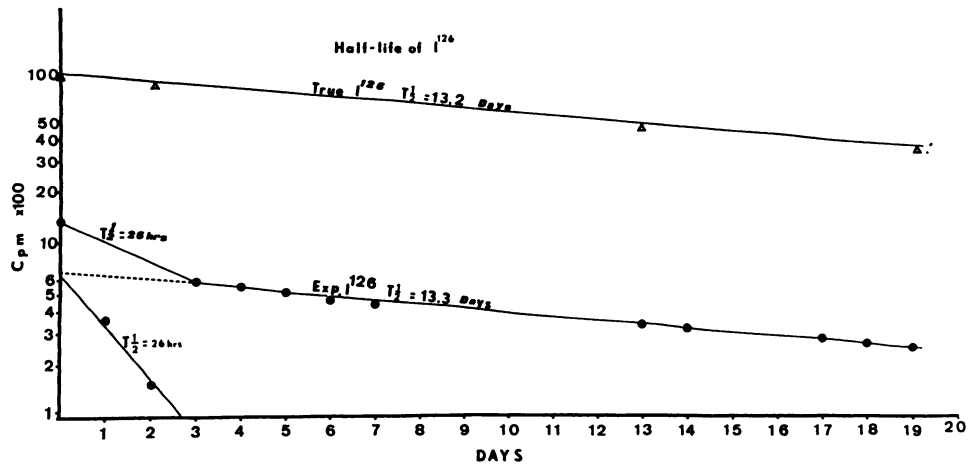
Fig. 5. Half-life of iodine-126. Figure shows the half-lives for both ^{126}I and the low energy contaminants (0.18 MeV and 0.28 MeV).
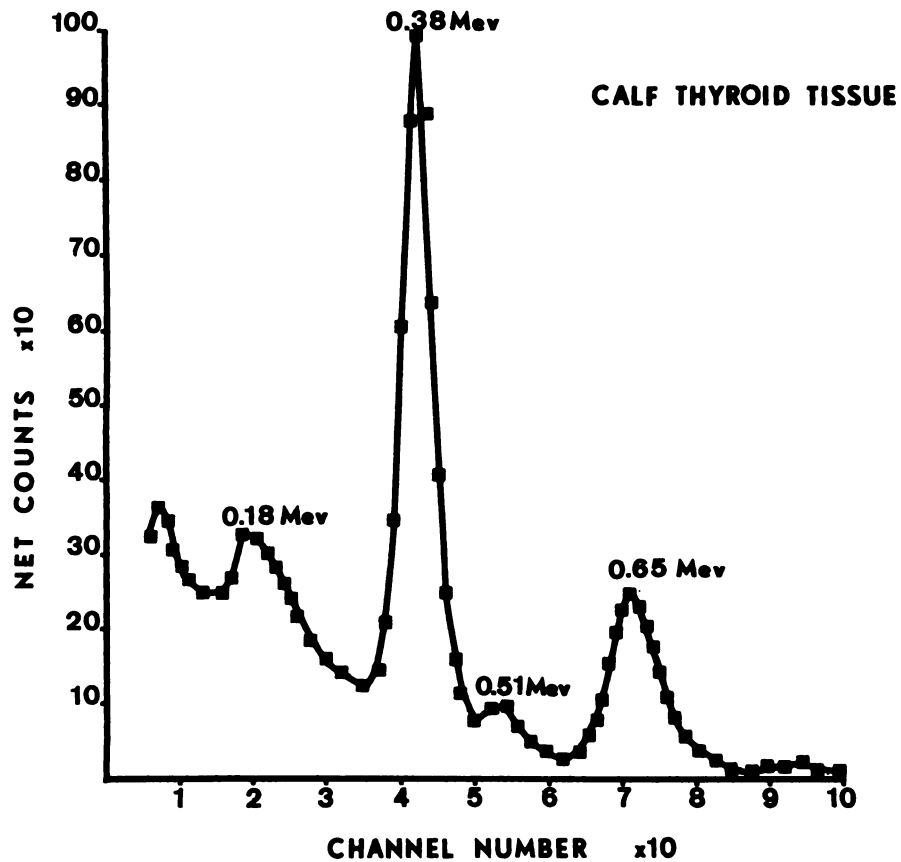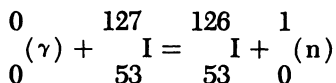

Fig. 6. Spectrum of a 200 mg slice of calf-thyroid tissue. The 0.39 MeV and 0.65 MeV are from ^{126}I while the 0.18 MeV peak has not been identified.

ten converter designed for 0.2 radiation lengths. The electrons from the "linac" beam were captured in the block and the resultant bremsstrahlung reaction

$$^{0}_{0}(\gamma) + \, ^{127}_{53}I = \, ^{126}_{53}I + \, ^{1}_{0}(n)$$

produced iodine-126. There was no apparent diminution in gamma intensity within or between the samples of each tier as shown by replicate samples in different positions in the holder. The threshold energy value for the above reaction was 10 MeV with a cross section value of 0.04 barns (3).

Analysis: The gamma activities of the samples were measured in a shielded 2¾ in x 2¾ in NaI (Tl) well assembly connected to two separate single channel analyzers and amplifiers for the 0.38 MeV and the 0.65 MeV ^{126}I photopeaks. One system was calibrated with a ^{133} Ba source and the other with cesium-137. Spectral analysis of the samples were performed using a shielded 2 in x 2 in NaI (Tl) well assembly connected to either a RCL[1] 256 or a TMC[2] 100 multichannel pulse height analyzer having a digital read-out.
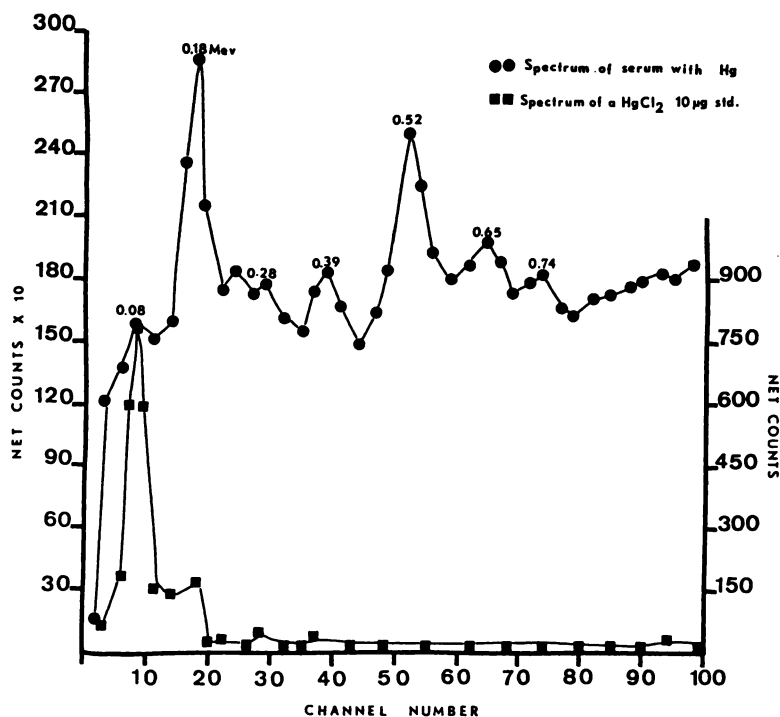

Fig. 7. Spectrum of pooled human serum indicating the two iodine photopeaks, 0.38 MeV and 0.65 MeV along with an 0.18 MeV, and 0.08 MeV and an 0.25 MeV photopeak. The 0.08 MeV and 0.18 MeV photopeaks are in part from ^{197}Hg, judging from calculated half-life values.

RESULTS AND DISCUSSION

The (γ, n) reaction of the [127]I nucleus has been investigated by many researchers. Montalbetti *et al* (5) and Nathans *et al* (6) experimented with the bremsstrahlung of a betatron to determine the cross-section value and threshold level of the [127]I (γ, n) [126]I reaction. MacGregor (9) and Moses (10) produced [126]I by the (γ, n) reaction using large amounts of [127]I (1 gm thick iodide wedges). In the present studies only trace quantities of [127]I were used. The conversion of [127]I to [126]I was carried out with no apparent interference from other primary reactions, especially the production of radioactive sodium. This is presumably because, either the total thermal neutron flux produced during bremsstrahlung activation was below the threshold for the production of a detectable amount of [24]Na by the (n, γ) reaction, or the [23]Na in the system had been largely removed by the resins. In addition, the cross section capture value for the production of [22]Na by the (γ, n) reaction at 22 MeV is only 4.0 mb (milli barn) compared to a value of 80 mb for [126]I (3). It should also be noted that the [22]Na nuclei produced by the gamma irradiation disintegrate with a half-life of 2.6 years. Therefore, a considerable amount of activity was not produced using only 90 minutes for irradiation.
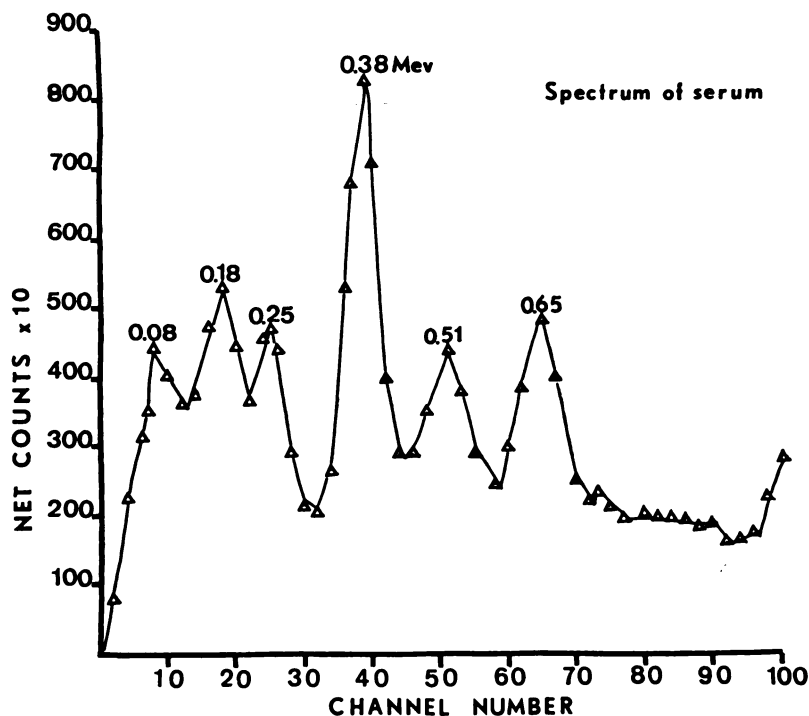

Fig. 8. Spectrum of mercury in samples of human serum taken two hours after a 1 ml dose of Mercuhydrin. In addition, the spectrum from a 10 μg $HgCl_2$ standard is shown.

There was a linear relationship between KI concentration and the net counts per minute of ^{126}I in either water or saline standards (Fig. 2). The induced radioactivity showed a coefficient of variation of 0.56% between the range of 5 μg to 20 μg of KI. With a water-cooled, thin, tungsten converter iodine could be measured in a concentration of 2 μg per cent. This value is lower than that previously reported by Yule et al, who used fast neutrons to produce the (n, 2n) reaction (11).

Spectral analysis of a 20 μg iodine standard in either H_2O (Fig. 3) or 0.9% NaCl (Fig. 4) showed two major gamma rays, 0.386 MeV and 0.65 MeV. The ^{126}I spectrum agrees with Heath's published spectrum (12). The ^{126}I spectrum showed two minor contaminants with energy values of 0.18 MeV and 0.28 MeV. The calculated half-life of these contaminants was approximately 26 hours compared to a half-life of 13.3 days for ^{126}I (Fig. 5). Aside from these low energy contaminants, no other interferring nuclides appeared on the spectrum. In an attempt to determine the source of the contaminants, a series of irradiation experiments were performed using empty plastic syringes as well as syringes containing only the resin. The results indicate that the source of the contaminants is the syringe barrel. This was substantially reduced by transferring the irradiated resin from its original syringe to a nonirradiated tube prior to analysis.

The irradiation of a 200 mg slice of calf-thyroid tissue showed the two major ^{126}I gamma peaks, 0.386 MeV and 0.65 MeV, along with the minor 0.18 MeV peak and without other contaminating peaks (Fig. 6). Results with pooled human serum showed the two major iodine photopeaks along with an 0.18 MeV, and 0.08 MeV and an 0.25 MeV photopeak (Fig. 7). These latter peaks have not been completely analyzed, however, the 0.08 and 0.18 MeV photopeaks are in part from ^{197}Hg, judging from calculated half-life values. Mercury in samples of human serum taken two hours after a 1 ml dose of Mercuhydrin appeared at the 0.08 MeV peak and 0.18 for mercury-197. The peaks as our results have demonstrated were well below the ^{126}I peaks (Fig. 8).

SUMMARY

With bremsstrahlung (gamma photon) activation, trace quantities of ^{127}I at the levels encountered in human serum can be measured. Using the (γ, n) reaction it is not necessary to remove the Na^+ from the system, as it is in neutron activation analysis. In addition, there is no significant intereference from other elements, such as mercury, which are also activated. This technique of measuring trace quantities of ^{127}I may be further developed for biological and medical applications not now possible with available classical techniques.

ACKNOWLEDGMENT

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