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January 22, 1954

A NEW CONSTANT MONITOR FOR RUTHENIUM AND IODINE IN STACK GASES
(STATUS REPORT)

INTRODUCTION

Present methods for monitoring Ru¹⁰³, Ru¹⁰⁶, and I¹³¹ in stack gases involve sampling by filtration and scrubbing and analysis by chemical separation and counting or by gamma spectrometry. Filtered samples, which are ordinarily collected over a complete day, are analyzed for total ruthenium and iodine by gamma spectrometry and for the individual isotopes by chemical separation and counting. The present scrubber monitors the sampler gas for total activity.

As an aid to Radiological Sciences' evaluation of stack gas hazards and to operations' control of radioactive gas emission, Methods and Instrument Development collaborated on the planning of a continuous monitoring system. This memorandum has the purposes of outlining the plan, reviewing progress since conception in October, 1953, and reporting work still to be done.

SUMMARY

A theoretically feasible plan for continuously monitoring and making individual records of the concentration of Ru¹⁰³, Ru¹⁰⁶, and I¹³¹ in stack gases has been devised. Ru¹⁰⁶ will be monitored via beta particles of energy greater than 1 Mev; Ru¹⁰³ will be monitored by its photoelectric distribution about 0.5 Mev and I¹³¹ will be monitored by its photoelectric distribution about 0.36 Mev. "Interferences" will be resolved by instrumental corrections.

PLAN

For brevity and clarity, the plan will be outlined with the aid of Table I showing the radioactive emissions of the isotopes involved. (1)

- (1) Data taken from Hollander, Perlman, Seaborg "Table of Isotopes" Rev. Mod. Phys. 25, 469 ff (1953).

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TABLE I

Radioactive Emissions of Ru¹⁰³, Rh¹⁰⁶, and I¹³¹

Ru ¹⁰³				Rh ¹⁰⁶				I ¹³¹			
Beta		Gamma		Beta		Gamma		Beta		Gamma	
Energy	%	Energy	%	Energy	%	Energy	Ratio	Energy	%	Energy	%
0.217	99	0.498	99	3.53	68	0.513	100	0.815	0.7	0.080	2.2
0.698	1			3.1	11	0.624	53	0.608	87.2	0.163	0.7
				2.44	12	0.87	3	0.335	9.3	0.284	5.3
				2.0	3	1.045	8	0.250	2.8	0.364	80
				Others	6	1.55	2.5			0.637	9
						2.41	1			0.722	3

Beta particles of energy 2 Mev and greater accompany at least 94% of the disintegrations of Rh¹⁰⁶. The other two isotopes emit no beta particle of energy greater than about 0.82 Mev and no significant percentage of beta particles more energetic than 0.608 Mev. Rh¹⁰⁶ therefore lends itself to monitoring without interference from the other isotopes if all beta particles less energetic than 2 Mev are excluded from a measurement, either by shielding or instrumental discrimination.

Gamma photons of about 0.5 Mev accompany 99% of the disintegrations of Ru¹⁰³ and some fraction (sometimes quoted as 17%) of Rh¹⁰⁶ disintegrations. A spectrometer channel centered on 0.5 Mev will yield a counting rate equivalent to 100% Ru¹⁰³ plus 17% Rh¹⁰⁶, as a first approximation. Subtracting about 18% of the Rh¹⁰⁶ beta count from the 0.5 Mev channel count leaves Ru¹⁰³ (omitting corrections for counter geometry).

Gamma photons of 0.36 Mev are emitted with 80% of I¹³¹ disintegrations. A spectrometer channel centered on that energy will yield a counting rate equivalent to a fraction of the Compton spectrum of Ru¹⁰³, Rh¹⁰⁶, the higher energies of I¹³¹, and the 0.36 Mev photoelectric peak. By subtracting the appropriate fraction of the count observed in the 0.5 Mev channel from the 0.36 Mev channel count, a correct index of I¹³¹ will be obtained.

PROGRESS

Studies of sampling methods, radiation detection and counting are now in progress. Observations and notes of significance are recorded in the following three sections.

Sampling

- Ruthenium isotopes are removed from stack gases with dry filters (90% efficient) or caustic (0.1N NaOH) scrubbers (80% efficient). I¹³¹ can be collected with caustic scrubbers (90% efficient) and filters impregnated with AgNO₃. Impregnated filters were about 95% efficient for collecting I¹³¹ from air but collected as little as

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50% of the isotope from synthesized dissolver off gases during some tests. Since filters on which samples are collected for short periods (e.g. 15 minutes) are easy to handle and lend themselves to individual analysis should the need arise, filtration is the preferred method of sampling. It has other advantages: contamination of the counting system is unlikely and auxiliary solution pumping equipment is unnecessary.

An adequate scrubber, now in use for constant monitoring, will be used should the scrubber sampler be chosen. A satisfactory filter sampler, now in use for special studies of particulates, will be used should the filtration method prove satisfactory. In either event, no more than slight adaptation of existing apparatus will be required.

Radiation Detection

Scintillation counters will be used for both beta and gamma counting. Rh^{106} beta particles will be monitored with a thin or powdered organic crystal; gamma photons will be monitored with a NaI crystal. Low energy (< 2 Mev) beta counts will be eliminated by shielding or discrimination in the electronic system.

Counting

The counting system was designed, tentatively, and elements of it were procured or developed. Testing is now in progress. Except for the recorder, all parts of the system are now on hand or on order.

Counting will be done by integrating pulses until the equivalent of 0.01 Curie of any isotope has accumulated. At that time, the recorder will be pulsed. The chart record will consist of three inked lines, one for each isotope, upon which pulses will be drawn. Emission rate will be indicated by the frequency of recorded pulses; accumulated total will be measured by the number of pulses. For an immediate measure of the total output since a reference time, a mechanical register will show the total number of 0.01 Curie amounts discharged.

FUTURE WORK

Studies which must be completed before the overall system can be tested involve certain critical parts of the equipment.

Sampling

The sampling system must be selected after accumulating enough data to reach a conclusion. Liquid scrubbers have high, consistent efficiencies, but were found likely to become contaminated. They are not especially desirable as beta emitter samplers since the liquid limits the countable sample volume. Filter samplers, since they do not have such limitations, would be excellent if their efficiencies remained constant. Investigation of means for maintaining uniform filter efficiency is in progress.

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Radiation Detection

The optimum beta counting arrangement has yet to be determined. Little work has been done on this phase of the problem because of the status of the sampling study. The detector will be a thin organic crystal; discrimination against low energy beta particles will be by shielding or instrument sensitivity, depending on the outcome of evaluations of the two methods.

The gamma counting arrangement, which will be a scintillation counter, has to be designed along with the sample container and beta counter.

Counting

Subtractions necessary to obtain the desired results may be made by digital or analog methods. Evaluation of both devices, now in progress, must be completed. Elements of the overall system need to be tested before the instrument is laid out and assembled.

DISCUSSION

The sampling line now enters the stack at the 50 foot level. Gases removed at that point are probably representative of stack discharge under normal operating conditions. However, during wash-down, there appears to be a fair chance of sampled gases being somewhat different in activity from those discharged to the atmosphere. This uncertainty could be eliminated by sampling near the mouth of the stack.

Gas flow through the stack is changed from time to time, notably during wash-down (when it is halved), but the sampling rate is constant. Unless the discharge rate is determined fairly accurately, monitoring results can be only qualitative. The most desirable sampling system would be one which withdraws a constant fraction of the exhaust stream. Radiological Sciences measurements could be shown to be quantitative if such a proportioning system were installed.

The recording method outlined is open to some question. It represents a radical departure from the usual philosophy of recording a quantity by deflecting the indicating line away from a baseline. In this case, since a range of 10^3 or 10^4 is required to cover the activities expected, a linear counting rate meter cannot be used and, since subtractions must be performed, circuit elements preceding the recorder drive must be linear. A logarithmic recorder chart does not permit integrating the total emitted activity without re-plotting the data. The method proposed is a compromise which shows both rate and total emission, but requires unconventional recording. It is intended to be useful to both operations to whom rates are of primary importance, and to Radiological Sciences, to whom a total measure of emission is also necessary.

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