AFOSR TR 59-185

THE UNIVERSITY OF MICHIGAN
COLLEGE OF ENGINEERING
Department of Chemical and Metallurgical Engineering

Technical Report

COMBUSTION EXPERIMENT WITH A 50,000-CURIE GOLD SOURCE

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UMRI Project 2288

under contract with:

DEPARTMENT OF THE AIR FORCE
AIR RESEARCH AND DEVELOPMENT COMMAND
CONTRACT NO. AF 18(600)-1218
WASHINGTON, D.C.

administered by:

THE UNIVERSITY OF MICHIGAN RESEARCH INSTITUTE ANN ARBOR

November 1959

engl

UMR 1014

PREFACE

This research was supported by the United States Air Force through the Air Force Office of Scientific Research, Air Research and Development Command.

Personnel of the University of Michigan Aircraft Propulsion Laboratory assisted in the construction of the equipment and in the experimental work.

Mr. C. C. Palmiter and other personnel of the University of Michigan Radiation Control Service assisted in the design of the experiment, directed the transfer of the radioactive gold from the Materials Testing Reactor to the laboratory, and monitored the source during the experiment.

The radioactive gold was transported from the Materials Testing Reactor to Ann Arbor by an Air Force flight arranged for by the Office of Scientific Research.

ABSTRACT

The data obtained in a previous experiment in which the flame zone and preflame zone of premixed propane and air were both irradiated with a 12000-curie gold source have been reworked and reinterpreted for journal publication [Ind. Eng. Chem. 49, 1419-22, 1423-28 (1957)]. The previously reported effects of irradiation on the flame speed and emission spectra of the flames were reconfirmed, but the uncertainty in the recomputed OH-rotational temperature was found to be greater than the observed variation with source strength and hence was not determinable from these data.

A new experiment was designed to irradiate the flame and preflame zones separately under essentially the same conditions. An accidental explosion in the test chamber shortly after installation of the radioactive gold damaged the burner and the optical transmission system used for remote observation of the flame, preventing collection of the intended data. The details of the experimental design and the history of the experiment are nevertheless reported as a guide to future research of a similar nature.

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BIBLIOGRAPHICAL CONTROL SHEET

1. Originating agency and monitoring agency:

   O. A.: University of Michigan, Ann Arbor, Michigan
   M. A.: Office for Advanced Studies, Air Force Office of Scientific Research

2. Originating agency and monitoring agency report number:

   O. A.: UMRI Technical Report 2288-9-T
   M. A.: AFOSR Technical Report 59-185

3. Title and classification of title:

   COMBUSTION STUDIES WITH A 12000-CURIE GOLD SOURCE (UNCLASSIFIED)

4. Personal authors: Stuart W. Churchill, Alexander Weir, Jr., Leroy F. Ornella, Roy L. Gealer

5. Date of report: November, 1959

6. Pages: 20

7. Illustrative material: 4 figures

8. Prepared for Contract No.: AF 18(600)-1218

9. Prepared for Project No.: Task 37507

10. Security classification: UNCLASSIFIED

11. Distribution limitations: None

12. Abstract: The data obtained in a previous experiment in which the flame zone and preflame zone of premixed propane and air were both irradiated with a 12000-curie gold source have been reworked and reinterpreted for journal publication [Ind. Eng. Chem. 49, 1419-22, 1423-28 (1957)]. The previously reported effects of irradiation on the flame speed and emission spectra of the flames were reconfirmed, but the uncertainty in the recomputed OH-rotational temperature was found to be greater than the observed variation with source strength and hence was not determinable from these data.

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INTRODUCTION

In the course of investigations of the utilization of gross fission products at the University of Michigan, flames were irradiated with gold and palladium sources. These investigations, which were carried out at low intensities, indicated some effect upon the flame stability (1) and no effect upon flame speed (2, 3).

Consequently an experiment was designed to study the effect of more intense radiation upon flames. The results of this study were reported in a Technical Note (4) and subsequently in two journal articles (5,6). The data were reworked and reinterpreted in the preparation of references 5 and 6, and these two articles should be considered to supercede completely the Technical Note.

The source in the above experiment consisted of 30 grams of irradiated gold with an initial strength of approximately 12,000 curies. The gold was in the form of coils of 0.005-inch wire. Premixed propane and air were passed through a compacted mass of these coils and burned above a screen as shown in Fig. 1. Thus both the preflame mixture and the flame zone were irradiated. The ionization produced by beta radiation exceeded that from gamma radiation and was estimated to be in the order of 3×10^{13} and 8×10^{9} ion pairs/$(cm)^3$(sec)(curie of Au-198 or Au-199) in the region of the gold and in the flame zone, respectively. The spectral emission at a series of elevations through a flat flame and the rate of propagation of bunsen-type flames were measured for a series of propane-to-air ratios as the source strength decreased. Increases in the rate of propagation of up to 50%, and increases in the emission due to CH and C_2 of up to 30% and 150%, respectively, and vertical displacement of the

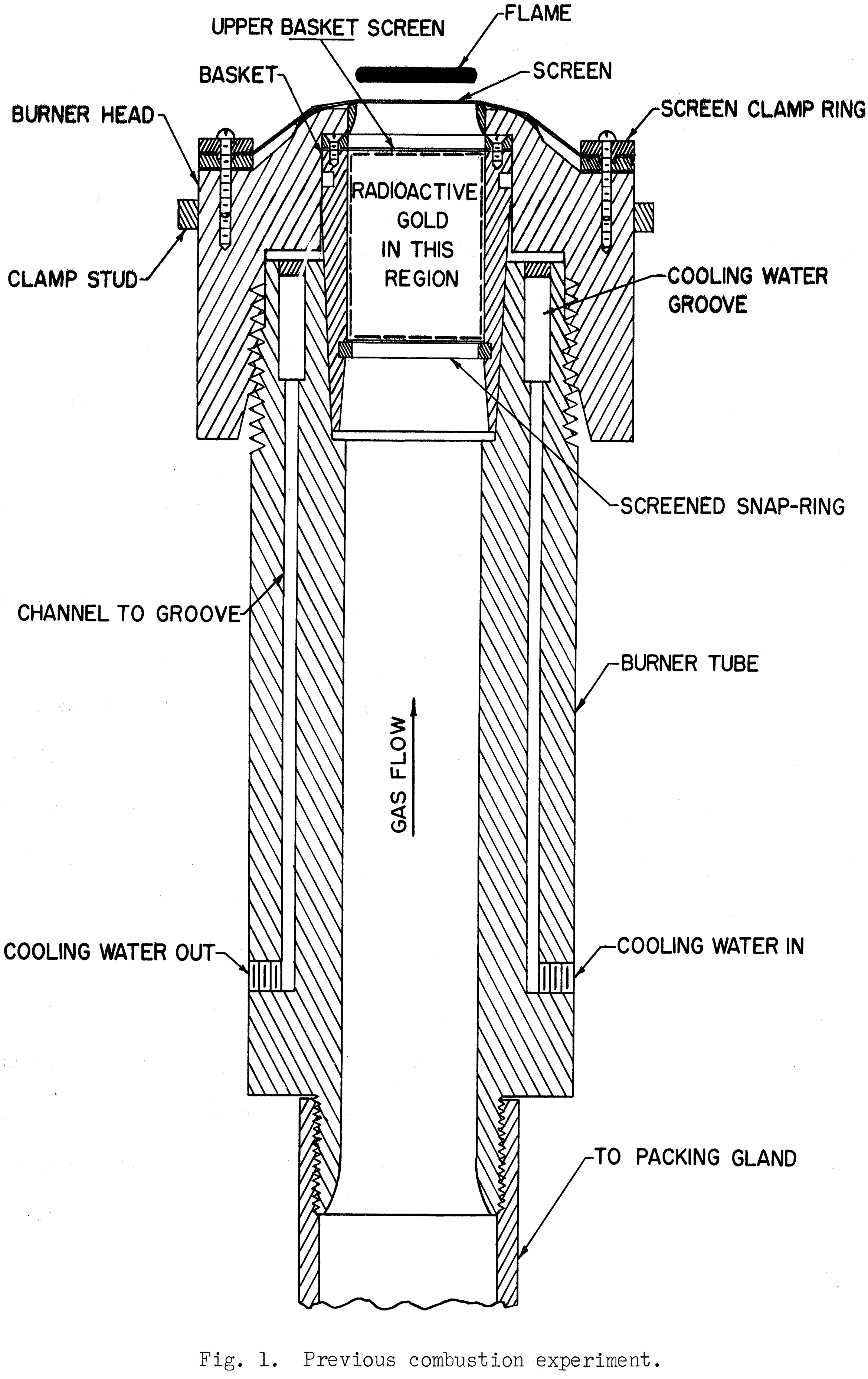

Fig. 1. Previous combustion experiment.

maximum in the spectral emission were observed to result from the irradiation. OH-rotational temperatures were computed from the spectral emission and are reported in the Technical Note (4). However, subsequent reanalysis of these computations revealed several significant errors. The uncertainty in the recomputed OH-rotational temperatures was greater than the variation with source strength. Hence these values are not considered to have significance and were not included in the journal articles (5,6), which supercede the Technical Note.

It was not possible to determine from the above experiment whether the observed effects were primarily due to irradiation of the flame zone, or of the preflame mixture, or of both. This report presents the results of a further experiment which was designed to permit separate irradiation of different zones of the propane-air and flame mixtures. It was intended to attain essentially the same maximum level of ionization as in the previous experiment and again to measure the emission spectra of the flame, the flame speed, and such performance characteristics as blow-off velocity and specific air impulse.

Unfortunately an explosion occured inside the experimental apparatus soon after installation of the irradiated gold. Damage to the optical system prevented attainment of spectral data and flame speed, and damage to the combustion apparatus itself produced a skewed flame and anomalous performance data. The presence of the radioactive source made adjustments impossible. Therefore no experimental data are presented in this report. The design and history of the experiment are nevertheless reported herein for the guidance of future experimenters in the field.

EXPERIMENTAL APPARATUS

The experimental equipment consisted primarily of a premixed propane-air supply and metering system, a burner head, a movable source of radiation, a vacuum tank and system to control pressure at sub-atmospheric levels, an optical system to transmit an image of the flame outside the source region, instrumentation to analyze the image of the flame, and radiation shielding. The overall experimental arrangement is shown in Fig. 2 and the pertinent details are discussed below.

A. Source and Source Containers

Gold was selected as a source material in the previous experiment because the half-life and strength obtained by irradiation are more suitable for combustion studies than those from other sources of beta radiation. As indicated in the disintegration scheme in Fig. 3, the half-life of Au-198 is 2.69 days while that of Au-199 is 3.15 days (7). This average half-life of about 3 days allows a 24-hour experimental run to be completed with only a 20% decrease in radioactivity. The gamma radiation which is emitted from both Au-198 and Au-199 is of course somewhat disadvantageous because of shielding problems and the possible effect upon the combustion process, but is only a minor contributor to the formation of ion pairs (5). Gold was selected for this experiment because of these reasons and because of the desirability of comparison of the results with the previous experiment.

A detailed analysis of cost, geometry, and mechanical operation indicated that the objective of separate irradiation of a preflame mixture

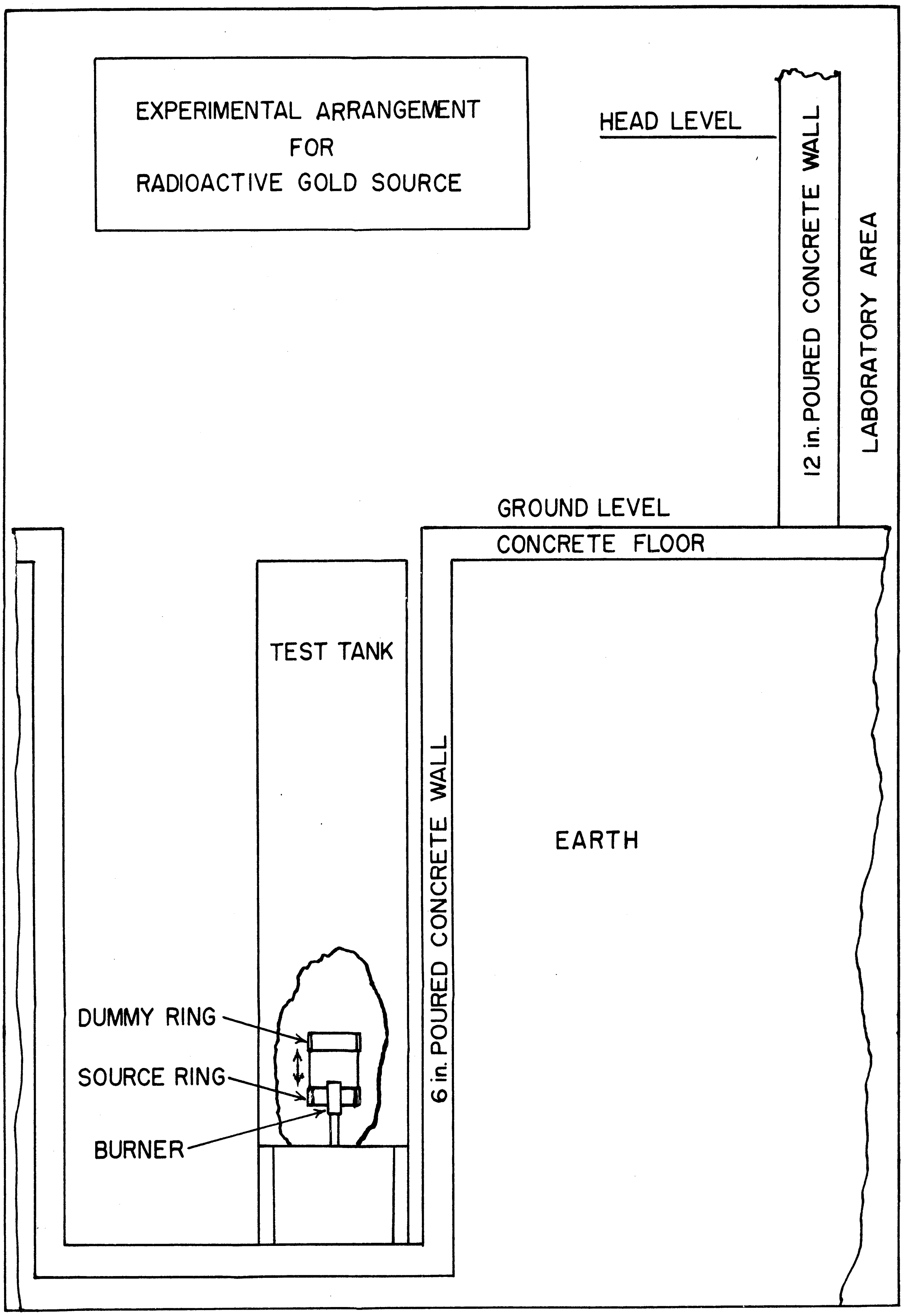

Fig 2. Experimental arrangement.

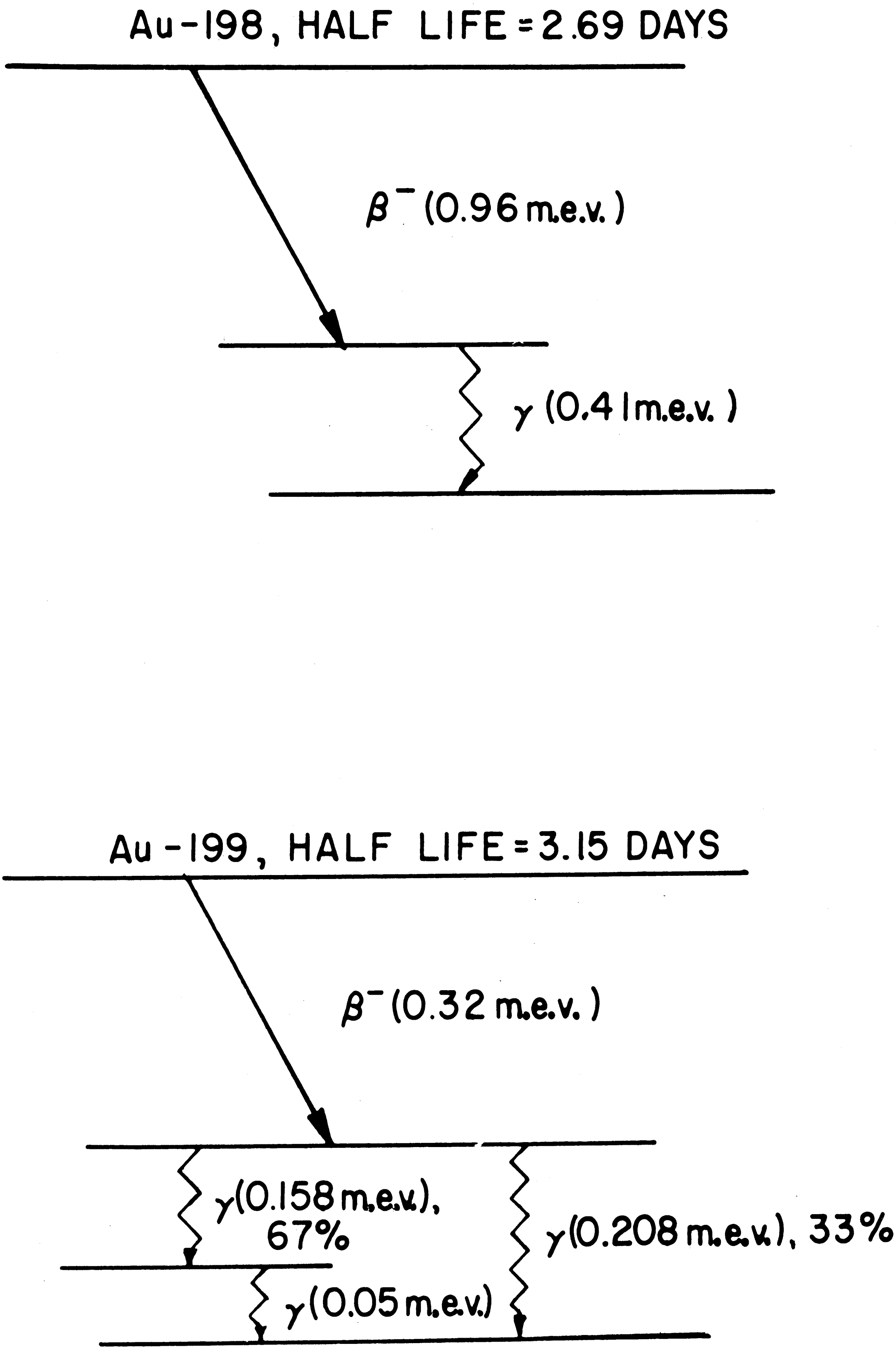

Fig. 3. Disintegration schemes of Au-198 and Au-199.

and a 1-inch-diameter flat flame, at levels corresponding to an energy absorption of 50×10^8 mev/(cm)^3(sec) could be best attained with a cylindrical ring 3 inches high and $8\frac{1}{2}$ inches in diameter composed of 52 1/16-inch-thick by $\frac{1}{2}$-inch wide by 2-7/8-inch high aluminum slabs coated on one side with 0.00266 inch of gold. The gold coating totalled 60 grams and the initial source strength was expected to be about 75000 curies.

The slabs were held in an aluminum carriage consisting of an outer shell, a grooved base, and an inside retaining wire. This carriage ring was then fastened to a hydraulic jack together with a dummy ring spaced 5 inches above the source ring as indicated in Fig. 2. In the uppermost position the source ring irradiated the flame zone, and the burner head shielded the preflame mixture. In the downmost position the source ring irradiated the preflame mixture, the burner head shielded the flame zone, and the dummy ring preserved the same geometrical surroundings for the flame.

The dimensions of the gold-plated slabs were determined by the permissible dimensions of the capsules to be installed in the Materials Testing Reactor as well as by the desired experimental conditions. For irradiation the slabs were placed in four aluminum capsules 3 inches long with an inside diameter of 7/8 inch and an outside diameter of 1-1/8 inch. After irradiation the capsules were placed in a lead shipping container 15 inches high and $14\frac{1}{2}$ inches in diameter with a central cavity just big enough for the four capsules. The lead container was clad in 1/8-inch steel.

B. Burner

The burner head consisted of two brass blocks 4 inches in diameter and $1\frac{1}{2}$ inches thick with a central passage 1 inch in diameter connected by a 4-inch-long by 1-inch-inside-diameter brass tube with a wall thickness of only 0.001 inch. The upper block contained a sintered bronze plug 1 inch in diameter at the outlet, and water channels for cooling of the plug. This design permitted a high percentage of the radiation to pass through the preflame mixture but shielded the flame zone when the source ring was in the lower position and also shielded the preflame mixture effectively with the source ring in the upper position.

The cooling water lines and the leads from a thermocouple attached to the sintered plug passed out through a length of 3/4-inch pipe connected rigidly to the burner head and serving as a handle for remote installation and removal of the burner head.

The propane-air mixture was ignited by a spark between two 1/8-inch rods connected to a high-voltage transformer. The rods were mounted on a swivel connected to a hydraulic jack which could be used to swing the ignitor away from the burner after ignition.

C. Test Tank

The burner head and source were located in a tank 30 inches in diameter and 9 feet high. A vacuum pump connected to the exhaust line from the tank was capable of maintaining pressures as low as 3 in. Hg. abs. during a combustion experiment. The removable cover of the tank contained an aluminum rupture diaphragm 15 inches in diameter to prevent destruction of the tank in the event of an accidental explosion of accumulated gases. The tank could be filled with water to protect personnel from radiation during installation of the gold source. Storage tanks were connected to the experimental tanks to retain this water.

D. Shielding for Personnel

The test tank was located in a concrete pit as shown in Fig. 2. The walls of the pit, the earth, and a 12-inch-thick concrete wall isolating the experimental apparatus from the laboratory area provided adequate shielding for personnel after lead sheets were placed at a few specific locations indicated by radiation measurements. Personnel entered the test area after installation of the source only when the test tank was filled with water.

E. Optical System

The image of the flame left the tank horizontally through a $\frac{1}{2}$-inch thick quartz window, was reflected downward by a flat mirror, then upward by a 4-foot-focal-length parabolic mirror to a flat mirror, then to a 12-foot focal-length parabolic mirror which focused the image on the entrance slit of the monochrometer. The entire optical system was enclosed in tubing to prevent the entrance of stray light.

F. Instrumentation

The flow rates of propane and air were determined with capillary meters. A thermocouple in the sintered plug in the burner head provided a measure of the temperature of the preflame mixture. A manometer indicated the pressure in the test tank.

A Jarrel-Ash monochrometer was positioned outside the isolation cell with its entrance slit at the focus of the image of the flame supplied by the optical system. The monochrometer scans the spectrum at a speed set by the operator and the monochromatic light falls on a photomultiplier tube at the exit slit. The photomultiplier was connected to a Brown recorder through an amplifier and the result was a plot of relative intensity versus time, with time proportional to wave number. The monochrometer was movable in a direction perpendicular to the plane of the image of the flat flame so that the flame could be scanned vertically, producing a recording of the variation of the intensity at a particular wavelength with height.

The monochrometer could be swung out of the light path to permit the positioning of a photographic film holder at the focus of the flame image. Such photographs were intended to provide a means of determining the flame speed.

HISTORY OF THE EXPERIMENT

After extensive rehearsals of the experiment in the absence of a source, the gold-plated aluminum slabs were sent to the Materials Testing Reactor, Idaho Fall, Idaho for irradiation. After irradiation for approximately five days the capsules were removed from the reactor, installed in the lead container and flown to Ann Arbor. Upon arrival at the laboratory the slugs were removed from the lead container and capsules, and installed in the source ring under water, using specially constructed tools for remote handling from the top of the tank. This operation is shown in Fig. 4. Of the 52 slugs 47 were successfully installed in the source ring. The other 5 were accidentally dropped to the bottom of the tank in inaccessible positions where they remained throughout the experiment.

The water was then drained from the tank, and the tank and burner dried with air. Propane was introduced and ignited. Shortly thereafter a mild explosion occurred in the test tank blowing out the rupture disk.

The cause of this accident is believed to be as follows. The intense irradiation from the radioactive gold increased the flame speed to the point that combustion occurred in or immediately adjacent to the sintered bronze plug. Solder holding the sintered plug in place melted due to the resulting excessive temperatures in the burner head, resulting in leakage of gas around the edge of the bronze plug. Since the propane-air mixture flowed preferentially through this crack, the region near the center where the spark gap was located probably was too lean to ignite. Unburned gas therefore accumulated in the test tank until eventual ignition created excessive pressure in the tank.

The explosion resulted in misalignment of the mirrors in the optical piping to the laboratory area. It proved impossible to realign the optical system remotely, and it was impossible to enter the test pit even if the gold plated slabs were removed from the source carriage because of the inaccessible slabs which were dropped to the bottom of the test tank.

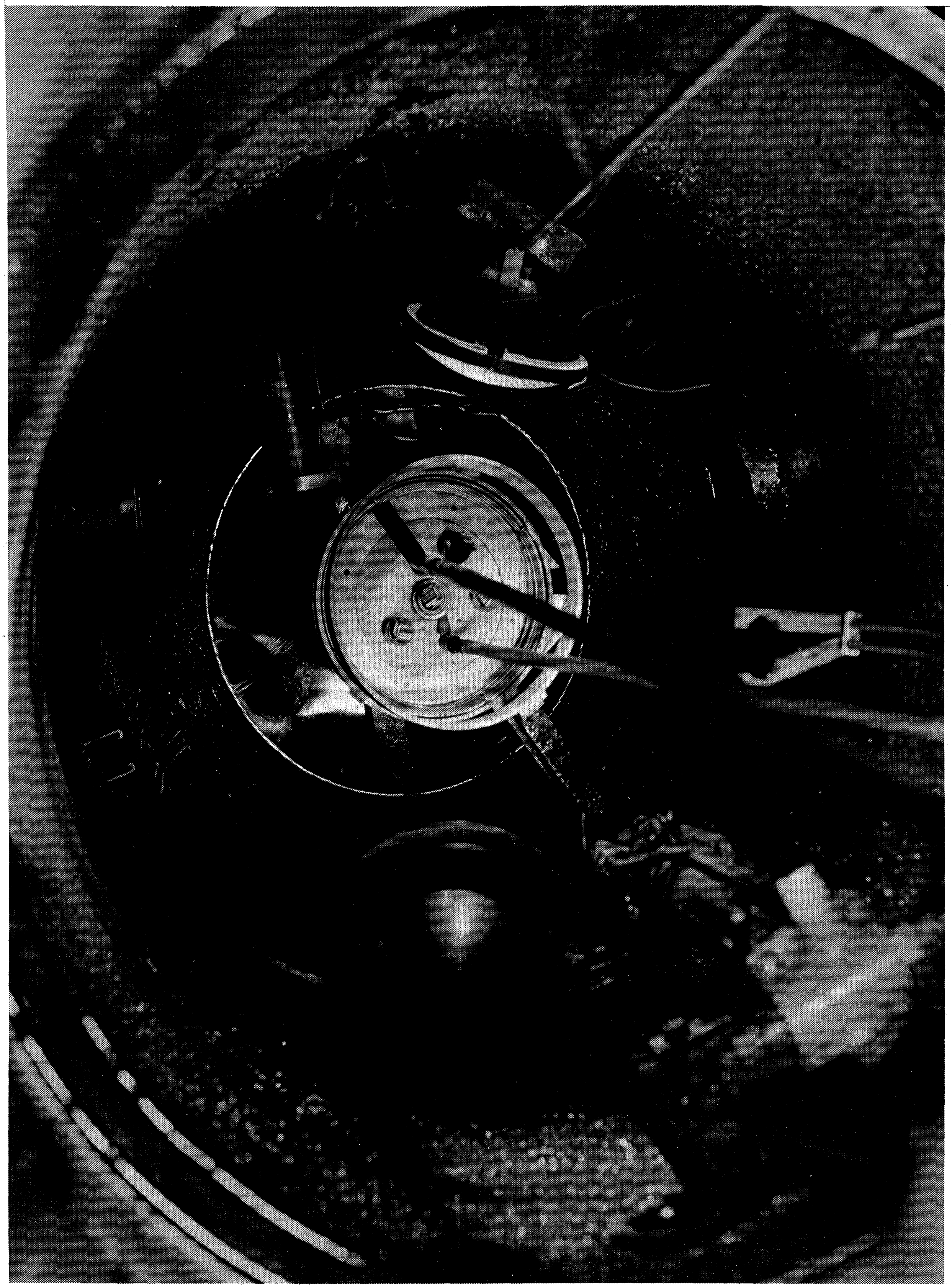

Fig. 4. Installation of gold-coated slabs in carriage.

The top section of the burner was removed from the test tank for repairs. However, the rapid decrease in source strength with time necessitated hasty work, and radioactivity of the burner head due to contamination by the shielding water necessitated remote handling. These makeshift repairs proved to be unsuccessful in that the data which were subsequently obtained for blowoff limits, etc., were erratic and uninterpretable. It is presumed that a leak persisted in the thin tube connecting the upper and low parts of the burner.

CONCLUSION

The only conclusion that can be stated relative to the effect of radiation of the flame is that the radioactive gold did produce gross effects such as were observed in the earlier experiment.

Other than the factors and events which lead to the accidental explosion, the design of the experiment appears to have been satisfactory. The strength of the source turned out to be of the order of 50,000 curies, as predicted, despite early removal from the Material Test Reactor due to reactor operating difficulties. The shielding provided for personnel during the handling of the gold and during the experimentation also proved to be adequate. A different tool for remote handling of the slabs to minimize the chance of dropping a slab is certainly desirable. The use of high-temperature materials in the burner head, and extreme care during the establishment of a flame are obviously necessary due to the abnormal behavior of flames at extreme levels of irradiation.

Recent examination of the gold-plated slabs used in the experiment reveals a significant but not intolerable amount of residual radioactivity due to impurities in the aluminum. It is concluded that these slabs, which were expensive to fabricate, could be irradiated and used again in the same or a similar experiment.

BIBLIOGRAPHY

1. Cullen, R. E., and M. E. Gluckstein, "Effect of Atomic Radiation on the Combustion of Hydrocarbon Air Mixtures", Fifth Symposium (International) on Combustion, p. 569, Reinhold Publishing Corp., N. Y. (1955).

2. Morrison, R. B., R. E. Cullen, and Alexander Weir, Jr., "Utilization of Gross Fission Products; Performance of Combustion Engines under the Influence of Radiation - Jet Engines", The University of Michigan Engineering Research Institute, A.E.C. Contract No. AT(11-1)162, Progress Report No. 2, (January 31, 1952).

3. Morrison, R. B., R. E. Cullen, and Alexander Weir, Jr., "Utilization of Gross Fission Products; Performance of Combustion Engines under the Influence of Radiation - Jet Engines", The University of Michigan Research Institute, A.E.C. Contract No. AT(11-1)162, Progress Report No. 3, (June 30, 1952).

4. Churchill, S. W. Alexander Weir, Jr., L. F. Ornella, R. L. Gealer, R. J. Kelley and M. E. Gluckstein, "Combustion Studies with a 12000-Curie Gold Source", The University of Michigan Research Institute, Report 2288-6-T, AFOSR-TN-56-17, Ann Arbor, Michigan (December 1955).

5. Churchill, S. W., Alexander Weir, Jr., R. L. Gealer and R. J. Kelley, "Rate of Propagation of Propane-Air Flames Irradiated with a 10,000-Curie Gold Source", Ind. Eng. Chem. 49, 1419 (1957).

6. Weir, Alexander, Jr., S. W. Churchill, L. F. Ornella, M. E. Gluckstein, "Emission Spectra of Propane-Air Flames Irradiated with a 1000-Curie Gold Source", Ind. Eng. Chem. 49, 1423 (1957).

7. "Nuclear Data", Nat. Bur. Stds. (U.S.) Circ. 499, Suppl. 3, Washington, D. C. (June, 1952).