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# HYDROGEN DETECTION INSTRUMENTATION FOR THE SLAC HYDROGEN RECOMBINERS

## Summary

We have examined and tested four commercially available hydrogen detection systems for application in the SLAC hydrogen recombiners. The instruments were particularly examined for linearity, reproducibility, temperature stability, zero drift, response time, and behavior for continuous exposure to hydrogen. Two of the systems, manufactured by Johnson-Williams, Inc., and Davis Instruments, were found to be useful for our specific application.

## I. INTRODUCTION

Large quantities of water are used as the coolant and primary energy absorbent in the SLAC energy absorbers. The electromagnetic cascade shower is primarily developed in water, particularly in the high-power beam dumps. The water molecules are excited and high concentrations of free radicals are formed during the beam pulse dissipation. This process is referred to as radiolysis and hydrogen, hydrogen peroxide and others are formed as the net result. Since the solubility of hydrogen in water is only about $0.8 \times 10^{-3}$ moles/liter, there will, after a certain time, be a net evolution of free hydrogen.

Catalytic recombiners for recombination of hydrogen and oxygen to form water have been developed at SLAC and are presently being built.

To assure safe operation of these units, protect other associated equipment, and guarantee personnel safety, hydrogen sensing instrumentation and alarm systems are needed. Hydrogen will evolve continuously during beam operation. The sensing system must, therefore, be capable of operating continuously in presence of $H_2$. Moreover, this instrumentation must have a fast response and it must be highly reliable.

Four commercially available detection systems were tested and the results are summarized below. The following list of criteria and specifications was assembled to aid in the selection of a suitable sensing system which would guarantee safe operation of the SLAC $H_2$-recombiners.

## II. CRITERIA FOR A HYDROGEN SENSING SYSTEM

General Requirements

1.0. The hydrogen sensing system shall sample the gases to be analyzed continuously and locally (where $H_2$ evolves) only by means of diffusion and natural convection of the gases to the sensing elements. Systems that depend upon the taking and analysis of successive, individual gas samples are inherently slow and must be ruled out.

1.1. The sensing system shall consist of at least two parts: The local sensing element and a separate remote control unit containing indicating meters, indicating lights and alarms.

1.2. The system shall contain no moving mechanical devices, and it shall be essentially a low voltage and low power system.

1.3. Sensing and indication shall be continuous and simultaneous on all channels or systems.

1.4. The zero drift shall not exceed 5% of full-scale for continuous operation over a period of 30 days.

1.5. The zero drift for slow temperature cycling from 0° to 50° C shall not exceed 10% of full scale or 5% of the lower explosive limit, short LEL (approximately 4% of hydrogen by volume in air), whichever value represents a lower hydrogen concentration by volume.

1.6. Temperature shock corresponding to changes of the ambient temperature at the rate of 3° C per minute shall not result in a system drift of more than 5% of the LEL.

1.7. After a proper warm-up period, the sensing element shall respond to 2% hydrogen by volume with 90% of the final reading in 15 seconds and 100% in 2 minutes.

1.8. Reproducibility shall be to within 1% of full scale or 1% of the LEL, whichever is less.

1.9. Span loss shall be 5% or less in 7 days' continuous exposure to 1% by volume of hydrogen in air.

1.10. Humidity fluctuations from 0 to 100% r.h. and condensation shall not result in a system drift of more than 5% of the LEL.

## The Sensing Element

2.0. The sensing element shall detect the presence of hydrogen by means of a catalytic reaction, utilizing the resulting temperature change as the primary signal.

2.1. The sensing head shall respond to hydrogen and deuterium.

2.2. The element shall respond to deuterium in terms of LEL, accuracy, reproducibility and zero drift exactly as for hydrogen (after due allowance has been made for the difference in thermal conductivity between hydrogen and deuterium).

2.3. Low temperature operation (< 500° F) of the sensing element is desirable but not a must.

2.4. A reference element for temperature compensation must be provided.

2.5. The gas volume surrounding the elements shall be small and it shall be separated from the ambient atmosphere (to be analyzed) by sintered metal flame arresters.

2.6. Easy and quick removal of the sensing elements for maintenance and replacement, without the use of special tools, is a requirement.

2.7. The element shall be readily calibratable from a remote location, by means of a calibrating gas of known concentration.

2.8. The system shall not saturate at high hydrogen concentrations (above 4% by volume in air).

2.9. It is desirable that the sensing element transmit the signal to the control unit without intermediate or pre-amplification for cable lengths up to 400 m.

2.10. The change in output signal due to ordinary temperature changes up to 50° C shall not cause an error of more than 5% of full range. This may be achieved by a suitable wire size between sensing element and control volume. (If a local amplifier cannot be avoided, it may not be closer to the sensing element than 8 m because of high radiation.)

2.11. A solid wire of catalyst is preferable to a catalyst-plated surface.

2.12. The minimum life expectancy of the sensing elements shall be one year.

2.13. The sensing elements need not be of radiation-resistant materials if replacement costs are less than $50.00.

The Control Unit

3.0. There shall be one control module for each sensing element.

3.1. The control unit shall display the output of the remote sensing element in a conveniently accessible location, for example, in the Data Assembly Building (DAB).

3.2. The control unit must contain its own power supply.

3.3. The control unit shall actuate and switch an alarm and threshold circuit, which is also to be used in the beam interlock system, using low and high alarm DPDT relay contacts, 3 A at 120 V minimum.

3.4. Each control unit shall have an indicating meter. The scale shall be divided into 50 or 100 units, with 10's numbered.

3.5. The alarm threshold shall be adjustable from 5% to 100% of the LEL. This adjustment shall be made at the control unit. The accuracy shall be to within 0.1% by volume of hydrogen in air minimum (2.5% LEL).

3.6. The meter shall indicate at 2% of the LEL, corresponding to 0.08% by volume of hydrogen in air.

3.7. Zero adjustment shall be located at each control unit.

3.8. Each control unit shall have at least three warning lights: first, a low level alarm indicating a concentration above the set warning level; second, a high level alarm to indicate an unsafe concentration; and, third, a trouble light to indicate failure of the sensing elements, i.e., breakage of the catalyst wire, etc.

3.9. The control unit shall have, in addition, an audio alarm to indicate an unsafe hydrogen concentration.

## III. INSTRUMENTS AND TEST RESULTS

Combustible gas detection systems of the following four manufacturers were examined and models were tested at SLAC:

| | |
|---|---|
| 1. Davis Instruments<br>Division of Davis & Co., Inc.<br>Newark, New Jersey | Model 11-3800 |
| 2. General Monitors, Inc.<br>El Segundo, California | Model 130 |
| 3. Johnson-Williams, Inc.<br>Mountain View, California | Model RHSP |
| 4. Mine Safety Appliances Company<br>Pittsburgh, Pennsylvania | Model 1-500 |

In the following, some of the pertinent observations are recorded:

1. Davis Series 11-3800 Combustible Gas Detector

The sensing element of this unit consists of a spirally-wound catalyst wire filament and a temperature reference filament which form a bridge. An increase in temperature due to recombination of hydrogen and oxygen affects the bridge. The unbalance is proportional to the $H_2$ concentration. The filament is cooled by thermal radiation to the sintered stainless steel flame arrester which encloses it.

The unit was calibrated using a General Monitors calibration chamber. Known quantities of hydrogen are added to a known volume of air, mixed with a fan, and the response of the instrument recorded. A calibration curve is shown in Fig. 1. The behavior is not linear but the curve passes through zero. This lack of linearity is not desirable and it is inconvenient. However, it should not create any unsafe conditions: if the operator should assume a linear relationship, the error is on the safe (i.e., low hydrogen) side. The calibration was repeated after one week and all points were within 2% LEL.

The zero of the unit was unstable to approximately 4% LEL. If no zero adjustment is made, the unit will calibrate within 2% of full scale. The drift appears to be an instability at or close to zero, and the rest of the scale is quite stable.

In a test at the A-Beam Dump temporary recombiner, the unit showed a 15% rise in zero setting as the temperature of the sensing element was quickly raised from approximately 20° C to 50° C. After this was zeroed out, the unit was stable. Subsequently, the unit was tested for temperature stability in an environmental chamber. The temperature was slowly cycled in the range from 0 to 50° C. The unit exhibited a maximum deviation of 7% LEL.

The response of the unit to the ultimate reading during calibration was very rapid, since it employs a radiation-cooled filament. Approximately 95% of the final reading was reached within 10 seconds, the final reading took approximately one minute.

The meter scale displaying percent LEL is inadequate for a process control unit. The loan instrument had 2% divisions identified at 20% intervals. It is desirable to have 1% divisions with 5's and 10's accentuated, and the 10's numbered.

2. General Monitors Model 130 Combustible Gas Detector

The sensing element consists of two thermistors which form the legs of a bridge. One thermistor has the catalyst plated onto the surface of the tip, the other acts as temperature compensator. The thermistor tips are located in hollow

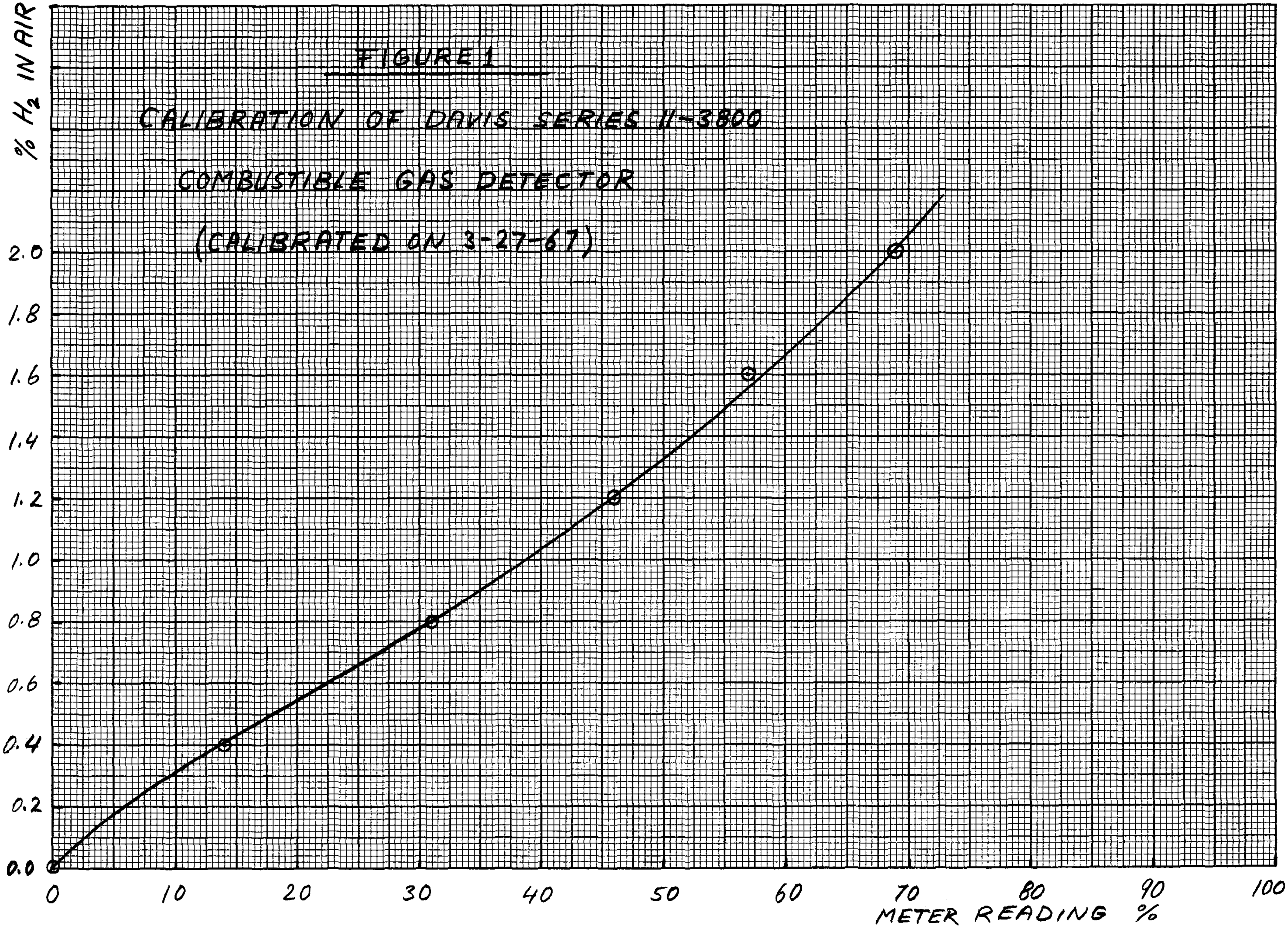

FIGURE 1

CALIBRATION OF DAVIS SERIES II-3800
COMBUSTIBLE GAS DETECTOR
(CALIBRATED ON 3-27-67)

spaces in a sintered bronze cylinder which in turn is surrounded by another sintered bronze flame arrester.

The instrument was calibrated in the same way as already described above. Approximately a dozen sensing elements were tested and a typical calibration curve is shown in Fig. 2. Summarizing all the test results, the following can be deduced:

The calibration curves are in general linear; however, in most cases the system could not be adjusted to include zero, i.e., the functional relationship between hydrogen concentration and indicated percentage level of lower explosive limit would not include the origin. Zero drift of up to 25% of LEL has been observed, with 10% being a rather common figure. None of the sensing elements would give reliable readings below 5% of LEL, the average limit being about 10% LEL. The accuracy for concentrations in excess of 10% of LEL met the criteria as long as the sensing head was functioning properly.

Reproducibility of the sensing element was satisfactory as long as it was functioning. However, in all cases a loss of range was observed, almost from the onset of exposure to hydrogen. This means that for an instrument previously calibrated, the output indicating the percentage level of the LEL is lower than the actual hydrogen concentration present would require. This loss of range accelerates with further exposure to hydrogen; see Fig. 2.

Losses of 50% of range have been observed. The usual way to correct this loss is to recalibrate and adjust the gain on the instrument. Unfortunately, during an experiment one would not know when such a loss of range has occurred. Thus this instrument would, for example, indicate a safe situation, whereas the actual concentration of hydrogen may have approached or exceeded the lower explosive limit.

In the advanced stages of range loss, yet another phenomenon appears. The meter needle starts to oscillate, indicating a fluctuating concentration of hydrogen (which is actually constant). To begin with, the amplitude of the oscillation is only a few percent of the LEL. Shortly before total failure, fluctuations of 80% LEL have been observed. The sensing element finally gives no output at all.

It should be noted that all tests were performed in a relatively dry atmosphere, and hydrogen concentrations never exceeded 4% by volume in air. Moreover, exposure to hydrogen was never longer than 30 minutes at a time.

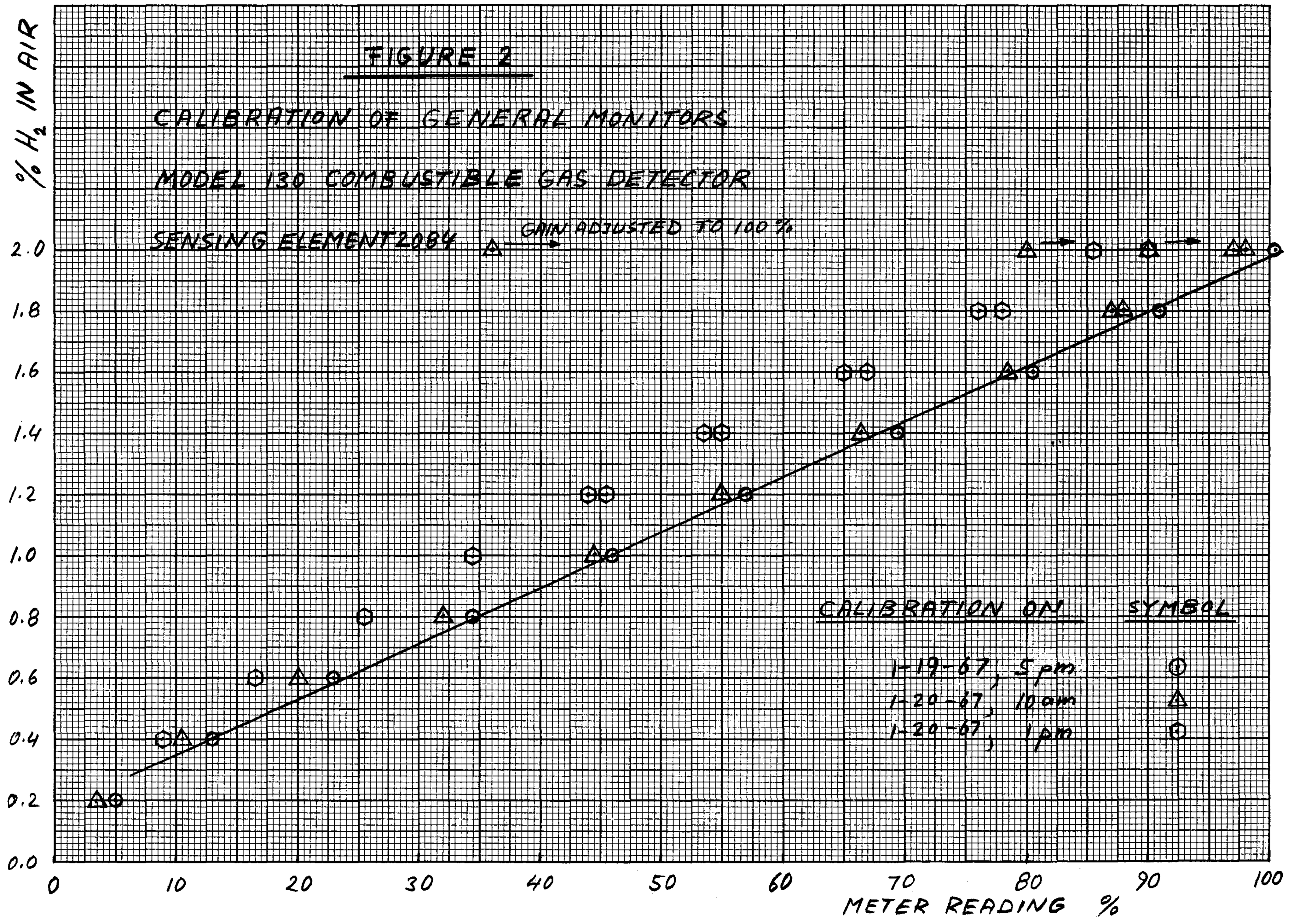

FIGURE 2

CALIBRATION OF GENERAL MONITORS
MODEL 130 COMBUSTIBLE GAS DETECTOR
SENSING ELEMENT 2084

It appears that for continuous exposure to hydrogen, the initially active, oxidized catalyst surface of the thermistor is reduced to an inactive state. After presentation with these observations, the General Monitors technical personnel agreed that their system is not suitable for our application of continuous exposure to hydrogen.

The results indicate, though, that this sytem is probably acceptable and safe in applications where hydrogen has to be detected only occasionally for short periods of time, and in low concentrations. In such an environment, the catalyst is expected to remain in an active state.

### 3. Johnson-Williams Model RHSP Gas Analyzer

The sensing element of this system is unique in that the filaments are inside glass tubes. The tube for the reference filament is closed at one end and open to the environment at the other. The tube for the active filament is open at both ends and thermally pumps the gas through and by the filament, provided the tube orientation is vertical. This chimney action cools the filament by natural convection in addition to cooling by thermal radiation. The filaments are spirally-wound solid catalyst wire.

The calibration curve is linear to approximately 50% of the LEL and includes the origin; see Fig. 3. Essentially no zero drift was detected within a two-day test period, and during a 10-hour experiment a shift of 1% of LEL was observed. Zero adjustment did not affect the span, as this was the case with other models, notably the General Monitors system. The reproducibility was ± 1% of full scale (LEL).

A test in the environmental chamber resulted in zero drift equivalent to 15% of LEL for a sudden temperature change from 0 to 50° C. The humidity was increased to 100% r.h. and did not have any additional effect on the drift. For slow temperature cycling in the range between 0 and 50° C maximum zero drift of 5% LEL was observed.

The filament-containing glass tubes are surrounded by a flame arrester consisting of 5 layers of Monel screen. This proved to be insufficient in strong drafts. For example, a 2% hydrogen concentration by volume in the calibration chamber resulted in a reading of 40 ± 2% LEL with the fan on, and a reading of 49.5% LEL was observed after the fan was turned off. Thus, with the fan on, the needle oscillated by 4% LEL and the reading was much lower, due to forced convection on the active filament. A sintered-bronze flame arrester which was subsequently installed prevented oscillations and made the sensing element insensitive to draft.

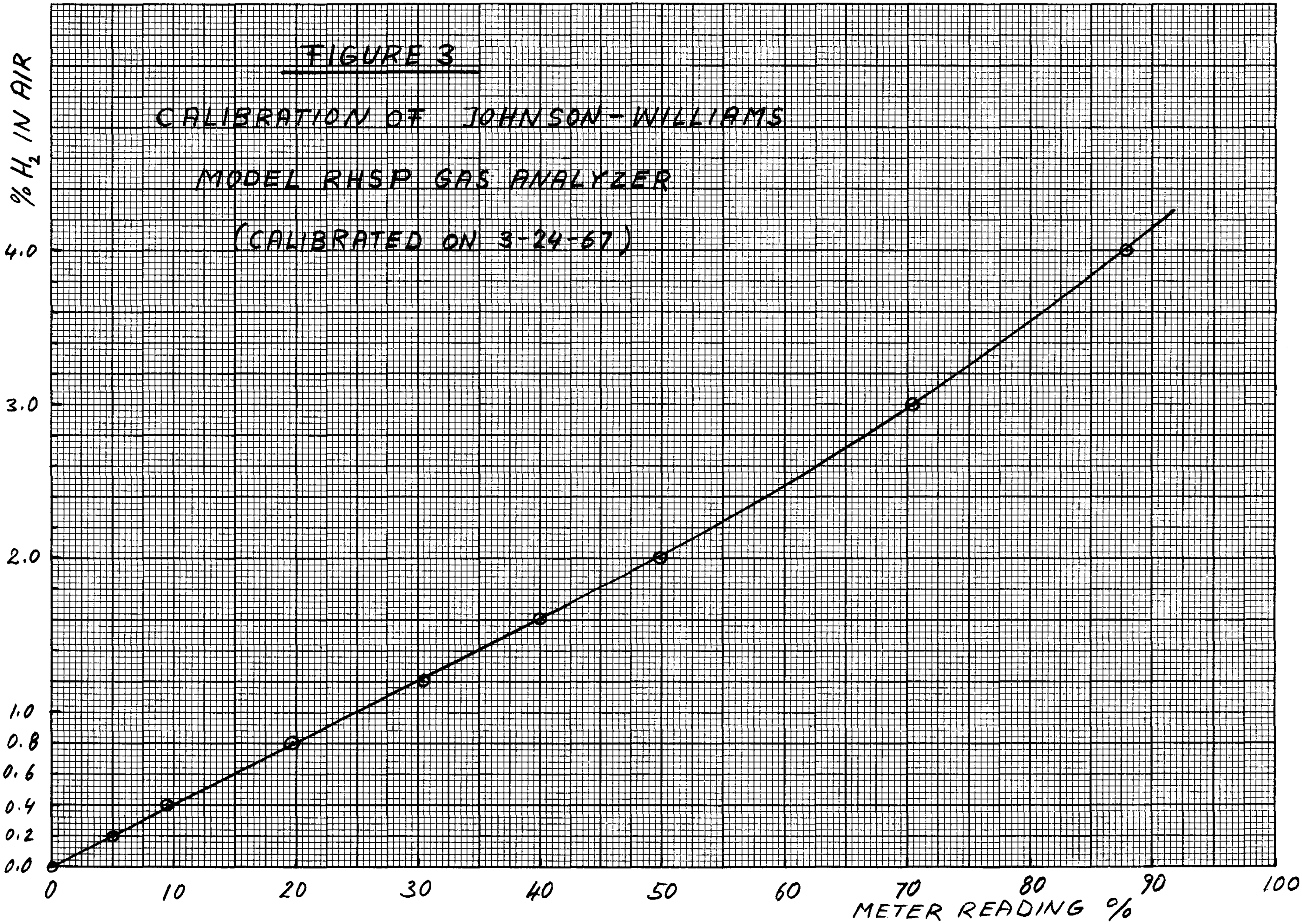

FIGURE 3

CALIBRATION OF JOHNSON-WILLIAMS
MODEL RHSP GAS ANALYZER
(CALIBRATED ON 3-24-67)

Due to the combined convection and radiation cooling, this element should have somewhat lower operating temperature than the one produced by Davis. This should have a beneficial effect on the life span of the element.

The response time of the element was acceptable. A reading of 90% of the final value was typically reached within 10 seconds after exposure, and the final value after one minute.

4. Mine Safety Appliances, Model 1-500 Combustible Gas Detector

The sensing element consists basically of two pairs of resistance wires, each of which has a ceramic bead fused to it. The two active filaments have the catalyst plated to the surface of the ceramic bead.

A calibration curve is shown in Fig. 4. The relationship is not quite linear, but continuous and repeatable to ± 1% of LEL. The range required no adjustments during two weeks of testing in concentrations up to 2% hydrogen by volume.

At room temperature, zero drift was within our set criteria. Zero adjustments could be made without altering the span. A zero drift equivalent to 5% LEL was observed when the sensing element was placed in gases at approximately 90° C. Continuous exposure to a mixture of 2% hydrogen by volume in air for a period of 24 hours caused a decrease in span of about 5% LEL. To verify that a span change had indeed occurred, the sensing element was continuously exposed for an additional 16 hours to a mixture of 4% hydrogen by volume in air. The span decreased again by 5% of LEL.

The response time of the instrument was acceptable, although it was slower than the solid wire catalyst type filaments. This is due to the fact that the heat must first be conducted through the ceramic bead before a temperature change of the resistance wire can occur.

## IV. SELECTION OF $H_2$ SENSING SYSTEM

The systems investigated can be divided into two groups:

A. The solid wire filament type sensing element where the whole wire is catalyst. The Davis and the Johnson-Williams systems are of this type.

B. The plated catalyst type of sensing element as employed by General Monitors and Mine Safety Appliances.

We have already ruled out the General Monitors system because of the specific application where the sensing element is continuously exposed to some concentration of hydrogen.

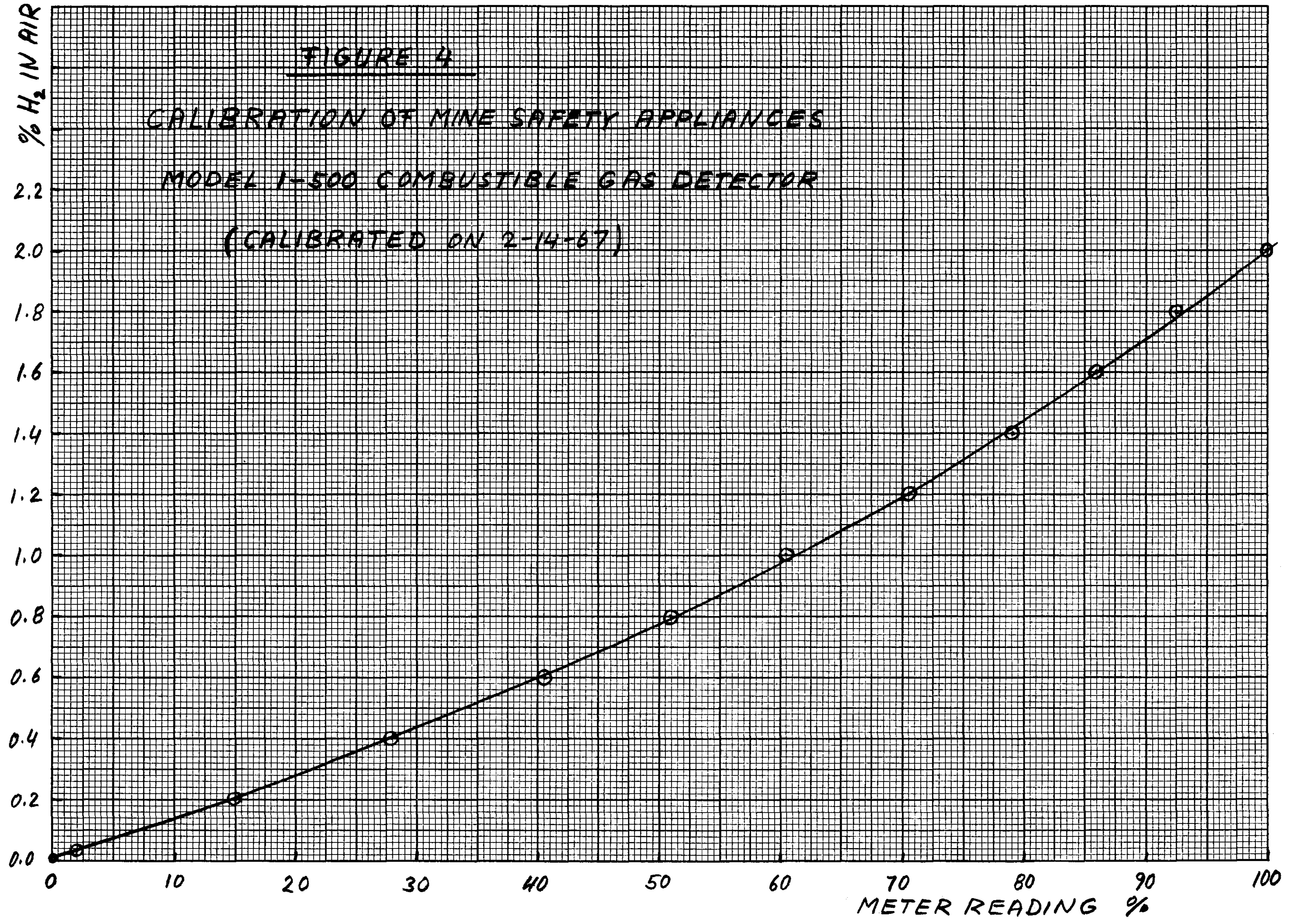

FIGURE 4

CALIBRATION OF MINE SAFETY APPLIANCES
MODEL I-500 COMBUSTIBLE GAS DETECTOR
(CALIBRATED ON 2-14-67)

The plated catalyst type of element as used by Mine Safety Appliances is not acceptable for three reasons: first, the edges of the plating disappear according to the thickness variations of the plating and hence in an unpredictable manner; second, it is possible for areas of plating to fall off, either by spallation of the plating or by fracture of the ceramic bead (due to sudden temperature changes as function of fluctuations in the hydrogen concentration and due to the high modulus of elasticity of the ceramic); the response is proportional to the catalyst surface area and can thus change. Third, there is no failure indication if part or all of the catalyst falls off. The sensing element would indicate very low or no hydrogen concentration, whereas the actual concentration may have exceeded the lower explosive limit.

In contradistinction, the area and properties of a solid wire catalyst will change in a uniform and predictable fashion. A continuity alarm warns of a broken wire.

Both instruments of this type are acceptable and meet the criteria. However, the Johnson-Williams unit is slightly preferable to the Davis unit. In the Johnson-Williams sensing element, the filament is cooled by both natural convection and thermal radiation. It, therefore, probably operates slightly cooler than the Davis element which is cooled by thermalradiation alone. The J-W sensing element is less affected by ambient temperature changes. Furthermore, the calibration is precisely linear in the operating range of the hydrogen recombiners, 0 to 2% hydrogen by volume. The zero drift is somewhat less than for the Davis element.

The Johnson-Williams system has a minor limitation in that the sensing element is orientation sensitive; the glass tube (chimney) must be vertical to result in maximum cooling by natural convection.

The J-W system was adopted with slight modifications for the SLAC hydrogen recombiners.

## Acknowledgement

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The tests in the environmental chamber were conducted by W. Pierce; his contribution is appreciated.