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Calorimetric Assay of HTGR Fuel Samples

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OAK RIDGE NATIONAL LABORATORY
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Contract No. W-7405-eng-26

**METALS AND CERAMICS DIVISION
ENGINEERING TECHNOLOGY DIVISION**

**HTGR Fuel Recycle Development Program
(189a OHO45)**

Fuel Refabrication — Task 500

CALORIMETRIC ASSAY OF HTGR FUEL SAMPLES

E. J. Allen, S. R. McNeany, and J. D. Jenkins

Date Published: April 1979

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CALORIMETRIC ASSAY OF HTGR FUEL SAMPLES

E. J. Allen, S. R. McNeany, and J. D. Jenkins

ABSTRACT

The fissile content of fuel samples will be measured in High-Temperature Gas-Cooled Reactor (HTGR) fuel recycle plants for quality control and to satisfy material accountability requirements. One candidate assay technique is active calorimetry. This report describes the evaluation of this nondestructive assay technique as applied to HTGR fuel.

In active calorimetry, a fuel sample is exposed to a neutron source. Fissions induced in the sample release heat, resulting in a measurable increase of temperature in the calorimeter. By use of a calibration curve, the fissile mass content of the sample is determined from the magnitude of the temperature increase.

A calorimeter was designed and fabricated by Mound Laboratory, according to specifications provided by Oak Ridge National Laboratory. A calibration curve of the device for HTGR standard fuel rods was experimentally determined. The precision of a single measurement at the 95% confidence level was estimated to be $\pm 0.8 \mu\text{W}$. For a fuel sample containing 0.3 g ^{235}U and a neutron source containing 691 μg ^{252}Cf , this represents a relative standard deviation of 0.5%. Measurement time was approximately 5.5 h per sample.

Use of the calorimeter is limited by its relatively poor precision, long measurement time, manual sample changing, sensitivity to room environment, and possibility of accumulated dust blocking water flow through the calorimeter. The calorimeter could be redesigned to resolve most of these difficulties, but not without significant development work. Because of the significant effort required to improve the calorimeter's performance and the availability of another device to satisfy the needed applications, development work on the calorimeter is being discontinued.

Keywords: nondestructive assay, calorimetry, ^{233}U measurement, ^{235}U measurement, HTGR reactors, californium-252, fissile materials, neutron sources, material accountability, quality control.

INTRODUCTION

Included in the High-Temperature Gas-Cooled Reactor (HTGR) Fuel Recycle Program is an effort to evaluate nondestructive assay (NDA) techniques for use on recycle fuel.¹ The final objective of this work is to demonstrate instrumentation that can be used for quality control and material accountability purposes within an HTGR fuel recycle facility.

Several NDA techniques have been evaluated on an experimental basis. This report addresses the evaluation of active calorimetry. Specifically, active calorimetry is examined for use as a high-precision method of fissile determinations on small fuel samples. Its application would be in a sample inspection station, where it would furnish quality control information, supply calibrated samples for use in other NDA instruments, and provide material accountability information for the sample inspection station. Further details on instrumentation and operating requirements for NDA equipment are reported elsewhere.¹

¹ J. D. Jenkins, S. R. McNeany, and J. E. Rushton, *Conceptual Design of the Special Nuclear Material Nondestructive Assay and Accountability System for the HTGR Fuel Refabrication Pilot Plant*, ORNL/TM-4917 (July 1975).

APPLICATION OF CALORIMETRIC ASSAY TO HTGR FUEL

Calorimetric methods for nondestructive assay can be divided into two types: passive and active. In passive calorimetry, radioactive decay heat generated in the sample is measured and correlated with fissile material present. In active calorimetry, the sample is exposed to an external radioactive source such as ^{252}Cf . Fissions produced in the sample increase heat output by an amount that is proportional to the fissile content.

The properties of the sample material help determine which calorimetric method, either passive or active, is best suited for fissile material assay. In HTGR recycle fuel, the fissile isotopes are primarily ^{233}U and ^{235}U . The uranium isotope ^{232}U is present in small quantities and is important in this discussion because its decay heat is significant.

Pure ^{233}U generates about $250\ \mu\text{W/g}$ decay heat. The heating contribution due to decay of ^{235}U in mixtures of ^{233}U and ^{235}U is insignificant. However, the presence of ^{232}U complicates the situation. Pure ^{233}U with 800 ppm ^{232}U generates about $800\ \mu\text{W/g}$. As the decay products of ^{232}U build up, they contribute to the heat generation. For a 100-d decay of initially pure 800 ppm ^{232}U in ^{233}U , over $1100\ \mu\text{W/g}$ is generated. Because of the large amount of decay heat released by ^{232}U relative to ^{233}U and the substantial increase in heat output over relatively short periods of time, accurate information on uranium isotopic contents and chemical purification history is needed to perform accurate passive calorimetric assays. For this reason passive calorimetric methods are considered unsuitable for assay of HTGR recycle fuel.

In active calorimetry, the equilibrium temperature response with the sample in the calorimeter is measured with and without the neutron source present. The incremental increase in heat generation is therefore due primarily to fissions induced in the sample and is independent of decay heat. The fission heat rate depends on the size of the neutron source, the concentration of fissile material in the sample, and the neutron interaction probabilities for the various isotopes present in the sample.

Active calorimetric methods have the advantages that knowledge of the chemical processing history is unnecessary and measurements are less sensitive to uranium isotopic contents than are passive calorimetric methods. A disadvantage of active calorimetric methods is that several standards are needed for calibration. For accurate assays using active calorimetry, the standards and samples must be similar in fissile material content and distribution. Passive calorimetric methods do not require the use of standards since the heat generated by natural radioactive decay can be directly translated into mass of radioactive materials if the isotopic composition is known.

Although the calorimeter evaluated in this report could be used either passively or actively, only active calorimetry was investigated. A ^{252}Cf neutron source, moderated with water and polyethylene, was used to provide a source of thermal neutrons in the sample region. HTGR fuel rods (typically 51 mm in length and 12.7 mm in diameter) with fissile content from 0.15 to 0.45 g were assayed.

DESCRIPTION OF CALORIMETER

The calorimeter evaluated in this report was designed and fabricated by Mound Laboratory according to specifications supplied by Oak Ridge National Laboratory. Basically, the calorimeter is of heat-flow resistance-bridge design with the additional flexibility to allow irradiation of the sample with thermal neutrons. The calorimeter consists of two separate components: (1) a neutron irradiator and (2) an isothermal water reservoir. The samples are loaded into the irradiator, in which the incremental increase in temperature in nickel bridge windings due to fissions in the sample is measured. Water is supplied from the reservoir to remove the heat from the calorimeter and to provide a constant temperature environment.

The irradiator is contained in a 0.19-m^3 (50-gal) water-filled drum, the top of which is pictured in Fig. 1. The water in the drum functions as a reflector and moderator for neutrons released from the neutron source. The neutron source is positioned in a rotatable polyethylene annular region centered in the drum. Inside the polyethylene annulus are two concentric stainless steel tubes between which water from the isothermal water reservoir flows. Inside the inner stainless steel tube are nickel bridge windings and heater windings embedded in an annulus of plastic. A schematic drawing of the irradiator is shown in Fig. 2.

Inside the plastic annular region is a removable column of interlocking aluminum and plastic tubes. The fuel sample fits securely in the bottom aluminum tube. The plastic tubes at the ends of the sample chamber serve to reduce axial heat loss. The interlocking tubes are shown in Fig. 3 along with a typical HTGR fuel rod and its holding sleeve.

The calorimeter has two equivalent thermal elements with two nickel resistance thermometers wound over the chamber length of each thermal element. These provide the sample and reference arms of the Wheatstone bridge shown in Fig. 4. This twin design reduces the effect of environmental temperature fluctuations.² The bridge potential, V , is directly proportional to the temperature difference between the two sides of the calorimeter.

²F. A. O'Hara et al. *Calorimetry for Safeguards Purposes, Determination of Plutonium in Reactor Feed Material*, MLM-1798 (January 5, 1972).

Fig. 1. Top of Irradiator, Showing Sample Loading Access Tube, Rotatable Polyethylene Neutron Source Holder, and Exiting Coolant Water Line.

Fig. 2. Irradiator.

Fig. 3. Interlocking Sample Holder Assembly. Scale is 0.30 m long.

Fig. 4. Bridge Circuit.

The irradiator controls are shown in Fig. 5. A chart recorder records bridge voltage. The electric resistance heater is regulated by the controls below the chart.

The water reservoir is maintained at a constant temperature and supplies water to the calorimeter irradiator to remove heat and to provide a constant temperature environment for the thermal elements. The temperature of the water reservoir is monitored by a nickel resistance thermometer, which is wound around the inside surface of the reservoir. A similar bridge design is used for the water reservoir and the irradiator except that the amplified bridge voltage output is used to regulate the temperature of the water by controlling the flow of air through a fan located above the water surface. If the temperature of the reservoir changes, the bridge potential increases or decreases, resulting in a change in the forced air flow over the surface of the water. The rate of evaporation of the water increases or decreases and the temperature of the water again approaches the preset value. Water temperature is maintained about 3°C below air temperature.

A motor circulates the water in the reservoir and produces the vortex required to maintain the water siphoning system described later. The top of the water reservoir with the circulation motor and air-supply fan is pictured in Fig. 6.

The water reservoir controls are shown in Fig. 7. The voltage meter registers reservoir bridge potential. The water-reservoir temperature is adjusted by changing the shunt resistance of the bridge. The bridge voltage is amplified and input to the air-supply regulator of the fan.

A functional schematic of the calorimeter is shown in Fig. 8, which illustrates the coupling between the water reservoir and the neutron irradiator. The water flows due to a siphoning action from the top of the reservoir, through the irradiator, and into the vortex of the stirrer.

The calorimeter system except for the irradiator controls, which are located outside the hot cell, is shown in Fig. 9.

Fig. 5. Irradiator Controls and Calorimeter Bridge-Voltage Recorder.

Fig. 6. Top of Water Reservoir Showing the Regulated Air-Supply Fan and the Water-Circulation Motor.

Fig. 7. Water Reservoir Controls.

Fig. 8. Functional Schematic of the Calorimeter.

Fig. 9. Calorimetric Assay Device (Irradiator Controls Not Shown).

OPERATING PROCEDURE

The water reservoir and neutron irradiator must be in thermal equilibrium before a measurement can be made. The water-reservoir temperature must be set below room temperature because the regulating system operates through evaporative cooling. If the temperature is set too near that of ambient temperature, evaporation at minimum fan air supply results in the water temperature fluctuating below the set point. Similarly, if the water reservoir temperature is set too far below room temperature, the air supply at maximum may not be enough to reduce water temperature to the set point. A 3°C difference between room temperature and water-reservoir temperature could be satisfactorily controlled under most humidity and temperature conditions.

Preparation of the calorimeter for a measurement requires about 0.5 d advance scheduling to fill the water reservoir and adjust the temperature controls. After the water reservoir reaches thermal equilibrium, the procedure outlined below, steps 2 through 6, is followed. Measurement time, not including step 1, requires about 5.5 h for a single sample.

1. Load sample and adjust water-reservoir temperature controls. (~12 h before step 2).
2. Adjust electric resistance heater power to be 10 to 20% greater than estimated sample heat release with neutron source present. Start chart recorder. As equilibrium is approached, the shunt resistance is adjusted to locate the trace above the midpoint of chart.
3. When the temperature of the calorimeter reaches equilibrium and a good trace is established (~1 h), readjust heater power to be 10 to 20% less than estimated heat given off by sample with source present.
4. After equilibrium is again reached, turn off heater power, load neutron source into the irradiator, start source rotation motor, and allow the temperature to again come to equilibrium. Allow about 1.5 h to firmly establish the recorder trace.
5. After equilibrium is reached, remove the source and adjust heater power to that used in step 2. Allow time for equilibrium to be reached.
6. Next adjust heater power to be that used in step 3. Allow time for equilibrium to be reached.

The advantage of this procedure is that if the bridge voltage drifts, which often occurs, the repeated heater power measurements allow accurate drift corrections.

CALCULATION OF HEAT OUTPUT

The procedure used to determine heat output of the sample from the recorded bridge voltage is described in this section. Figure 10 shows a representative example of a bridge-voltage trace.

The unknown power of a sample can be determined by Eq. (1), which is obtained from Ref. 3.

$$HS = H1 + (VS - V1)/S1 \quad (1)$$

where

HS = power of the sample,

H1 = power of known source,

VS = bridge voltage with sample,

V1 = bridge voltage with known source,

S1 = sensitivity of the calorimeter at power level H1.

³ American National Standards Institute, *American National Standard Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control*, ANSI N15.22-1975 (June 5, 1975).

Fig. 10. Representative Chart Recorder Output.

By assuming a linear response of the chart recorder with voltage level and a second known source measurement, Eq. (2) can readily be derived:

$$H_S = H_1 + \frac{d_1}{d_1 + d_2} (H_2 - H_1), \quad (2)$$

where

H_2 = power of second known source,

d_1 = distance from trace of first known source to trace of sample,

d_2 = distance from trace of second known source to trace of sample.

The known heat output of the sample can be calculated from Eq. (2) after the distances d_1 and d_2 are measured from the record of the bridge voltages. If the bridge voltage is linearly drifting with time, this equation still yields accurate estimates of sample power.

CALIBRATION EXPERIMENT

Several HTGR fuel rods containing known amounts of fresh fuel (93% enriched in ^{235}U) were assayed in the calorimeter, and a calibration curve based on these measurements was obtained. For ease in handling, procurement, and fabrication, experiments with samples containing ^{233}U were not performed. We assumed in assessing the calorimeter's performance that similar results would be obtained for either ^{233}U or ^{235}U fuels since the measured incremental increase in heat generation is due primarily to fissions induced in the sample and is independent of decay heat.

Information on the fuel rods used in the calibration experiment is given in Table 1. Information on the fuel particles of which the fuel rods were composed is given in Appendix A of Ref. 4. The measured heat outputs of the fuel rods in the calorimeter normalized to a ^{252}Cf source mass of 690.6 μg are given in Table 2.

The calibration curve of heat output versus uranium content is given in Fig. 11. Heating in the fuel rod with no ^{235}U present is due mainly to absorption of gammas released by the source. The calibration curve is closely approximated by Eq. (3), which can be derived from the physics of heat generation as an approximate expression relating heat output and fissile content:

$$H = C_0 [1 - C_1 \exp(C_2 U)] \quad (3)$$

where

H = heat output of the sample (μW)

U = ^{235}U content (g)

C_0, C_1, C_2 = constants

Equation (3) can be used to determine the uranium content from the measured heat output once the constants C_0 , C_1 , and C_2 have been determined from calibration standards. For the HTGR fuel rod measurements, C_0 , C_1 , and C_2 were estimated by a least-squares technique to be 408.23 μW , 0.97063 μW , and $-0.6508/\text{g}$, respectively. The agreement between measured heat outputs and those calculated with Eq. (3) is given in Table 2.

⁴P. Angelini and J. E. Rushton, *Uranium and Thorium Loadings Determined by Chemical and Nondestructive Methods in HTGR Fuel Rods for the Fort St. Vrain Early Validation Irradiation Experiment*, ORNL/TM-6562, (January 1979).

Table-1. HTGR fuel rod content

Fuel rod	^{235}U content (g)	Particle contents, g			Matrix content (g)	Total mass (g)
		Uranium	Thorium	Shim		
5009	0.0	0.0	6.0076	4.224	3.142	13.374
1002	0.1508	0.9313	6.0088	3.317	3.123	13.380
1102	0.2715	1.6768	6.0078	2.568	3.084	13.337
1202	0.3016	1.8631	6.0077	2.448	3.119	13.438
1302	0.4524	2.7948	6.0078	1.545	3.165	13.513

Table 2. Measured and calculated heat outputs of fuel rods

^{235}U content (g)	Heat output, μW		Measured - Calculated Measured (%)
	Measured ^a	Calculated ^b	
0.0	11.89	11.99	-0.84
0.1508	49.71	49.42	0.58
0.2715 ^c	76.18	76.82	-0.84
0.2715 ^c	77.47	76.82	0.84
0.3016	83.02	83.32	-0.36
0.4524	114.13	114.01	0.11

^aNormalized to ^{252}Cf source strength of that on Jan. 10, 1978 (1.58 $\times 10^9$ n/s).

^bCalculated with Eq. (3).

^cFuel rod 1102 was assayed twice.

Fig. 11. Calibration Curve for HTGR Fuel Rods.

PRECISION OF CALORIMETRIC ASSAY

The precision of the assay is primarily limited by fluctuations or noise in the bridge voltage. An accurate determination of the precision is difficult since the chart recorder trace is averaged by visual examination. However, the precision can be estimated.

The trace width fluctuates over about $2 \mu\text{V}$. The true average of the trace is assumed to be within about $0.5 \mu\text{V}$ of the visually estimated average approximately 95% of the time. However, several bridge-voltage trace averages are required to obtain the sample heat output with Eq. (2). Since the sensitivity of the calorimeter is about $1 \mu\text{V}/\mu\text{W}$ ($1/A$) and the sample bridge voltage is approximately the average of the bridge voltages of the two known sources, the estimated 95% error limits on the measured sample heat output are $\pm 0.8 \mu\text{W}$.

This estimate is independent of uranium content or power output of the sample. As the sample power output increases because of a larger source or greater uranium content, the relative standard deviation in the measurement improves.

ACCURACY OF CALORIMETRIC ASSAY

The accuracy of the uranium measurements depends on the precision and on how well the standards represent the samples. Standards must be selected that are characteristic of the samples in chemical composition, volume, and uranium distribution. If the samples or unknowns are well represented by the standards, the accuracy is primarily limited by the precision of the measurements. If the standards are not representative of the samples significant errors can result.

For HTGR fuel samples, whether batches of fuel particles or individual fuel rods, the distribution of the uranium varies from one sample to the next. The fuel particle size and the distribution of particles in the sample vary from sample to sample. The neutron flux from the source will be depressed slightly more in large particles than in smaller particles and in regions of high uranium particle concentration than in regions of low uranium particle concentration. As a result, the fission rate and therefore the heat output will vary slightly per gram of uranium from one sample to the next. If unexpectedly large changes in fissile material distribution occur, significant errors in the uranium measurements can result.

Experiments to determine the effect on accuracy due to changes in fuel particle size or density were not performed with the calorimeter. However, several experiments were completed on a delayed-neutron assay device,^{5,6} which has a similar source-sample geometry to the calorimeter. From these experiments, we estimate that a 20% increase in average fissile particle diameter will result in approximately a 1% decrease in heat output. A threefold increase in fissile particle density will result in approximately a 5% decrease in heat output. Although such large differences in the samples from the standards are not expected, such effects will bias the estimated values of the uranium content unless more representative standards are used.

PROBLEM AREAS

In working with the calorimeter, several problems were encountered. These are listed below.

1. The water reservoir temperature regulating system is sensitive to changes in room temperature and humidity. As a result, the water reservoir temperature setting must be regularly adjusted.
2. Rapid changes in room pressure, such as occur when opening and closing the air lock, significantly affected the response of the calorimeter. The reason for this is unknown and would require additional study.
3. The water reservoir requires daily refilling. If the water reservoir is not maintained above a certain level, air bubbles enter the calorimeter cooling jacket and cause erratic behavior of the bridge voltage. The water also requires filtering to prevent dust from accumulating and eventually blocking water flow through the irradiator.
4. The long measurement time of about 5.5 h decreases the usefulness of the calorimeter as an assay device.
5. Present design of the calorimeter restricts its use to nonremote sample changing.
6. The precision of the measurements is relatively poor. Assuming no systematic errors in the measurements, the relative standard deviation is about 1% for a sample containing 0.1 g ²³⁵U and 0.5% for a sample containing 0.3 g ²³⁵U.

POSSIBLE IMPROVEMENTS

Three possible improvements to the calorimeter are discussed in this section. These changes would improve precision, reduce measurement time, expand the flexibility of the calorimeter to allow remote sample changing, and eliminate the problem of water flow blockage.

The average of the fluctuating bridge voltage is currently estimated by visual inspection of the bridge-voltage chart record. The precision of the measurement is not estimated accurately. Incorporating a mini-computer into the calorimeter system will eliminate this problem and also reduce the measurement time.

⁵S. R. McNeany, R. W. Knoll, and J. D. Jenkins, *Design and Evaluation of a Nondestructive Fissile Assay Device for HTGR Fuel Samples*, ORNL/TM-6610, (in press).

⁶C. V. Bayne and S. R. McNeany, *Statistical Precision of Delayed Neutron Nondestructive Assay Techniques*, ORNL/TM-6569, (in press).

The bridge voltage levels could be read by the minicomputer at discrete time intervals (~ 5 s), and these readings could be continued until the desired precision in the measurement is attained. The measurement time could be reduced by mathematically extrapolating the bridge voltage response to equilibrium before equilibrium is reached.⁷

The bridge voltage, as equilibrium is approached, can be expressed as

$$V_t = (V_e - V_0)[1 - \exp(-\alpha t)] + V_0, \quad (4)$$

where

V_t = bridge voltage at time t ,

V_e = equilibrium bridge voltage,

V_0 = bridge voltage at time $t = 0$,

$1/\alpha$ = longest time constant of the calorimeter.

From Eq. (4), Eq. (5) can be readily derived:

$$\frac{V(t + \Delta t) - V_t}{V_t - V(t - \Delta t)} = \exp(-\alpha \Delta t) = \text{a constant}, \quad (5)$$

where

Δt = time between successive bridge voltage readings

The minicomputer can be programmed to check the ratio of the differences between successive bridge voltages. When this ratio approaches a constant value, the equilibrium voltage can be estimated with Eq. (6):

$$V_e = \frac{V(t + \Delta t) - V_t \exp(-\alpha \Delta t)}{1 - \exp(-\alpha \Delta t)}. \quad (6)$$

This procedure could reduce measurement time by 50%.

Another improvement is to redesign the calorimeter to accept samples from a remote sample loading chamber, which would be required for in-cell use. A pneumatic transfer system similar to that used on the delayed-neutron assay device⁵ could be adapted to the calorimeter. However, an extensive redesign of the basic internal structure would be required, and the potential interference of the transfer system with the measurements would have to be evaluated.

A third improvement is to remove the annular polyethylene region and the outer stainless steel shell from the irradiator and position the irradiator directly in the water reservoir. The neutron source could rotate at a fixed distance from the irradiator in the water reservoir. This modification would simplify the flow of water and eliminate the problem of flow blockage.

⁷Personal communication from C. L. Fellers, Mound Laboratory, to E. J. Allen and S. R. McNeany, Oak Ridge National Laboratory, Nov. 16, 1977.

The precision of the measurements could also be improved by using a larger californium source. More heat would be given off by the sample per gram of fissile material as a result of the higher neutron flux, and the relative standard deviation in the measurements would improve. However, the 691 μg of ^{252}Cf used in the calorimeter is already a moderately large quantity, and further increases in source size would increase the cost of the device significantly.

CONCLUSIONS

The usefulness of the calorimeter is limited for a number of reasons. The accuracy is restricted by the poor precision of the current analysis procedure to about $\pm 1\%$ (95% confidence level) for a sample containing 0.3 g ^{235}U . The measurement time of 5.5 h is relatively long for a nondestructive assay device. The current design requires manual sample changing, restricting use to environments with low radioactivity background. The temperature regulating system requires daily adjustments and is very dependent on room temperature and humidity. In addition, the present design of the calorimeter is inadequate for long-term hot cell use because dust buildup could block water flow through the calorimeter.

An extensive redesign of the calorimeter could resolve most of these difficulties. A microprocessor could be incorporated to analyze bridge voltages to improve precision and reduce measurement time. A pneumatic transfer system could be installed to enable remote sample changing. The calorimeter irradiator containing the sample chamber could be located directly in the water reservoir to avoid flow blockage. However, these modifications would require significant additional work.

A second assay device^{5,6} has been developed concurrently with the calorimeter. This delayed-neutron assay device can be used for the same applications as the calorimeter and appears to have fewer disadvantages.

Because of the significant effort required to improve the calorimeter's performance and usefulness and the availability of another assay device to satisfy the needed application, development work on the calorimeter is being discontinued.

ACKNOWLEDGMENTS

The authors wish to thank G. W. Otto and C. L. Fellers of Mound Laboratory for designing and constructing the calorimeter as well as for providing helpful guidance, J. C. McLaughlin for assistance with the calorimeter measurements, J. E. Mack and R. D. Hurt for reviewing the manuscript, H. R. Livesey and M. A. Patrizio for preparing the figures, S. Peterson for editing and Adroit, Inc. for preparing the report for reproduction.

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