P80-11-321

NOVEMBER 20, 1980

FIELD TASK PROPOSAL/AGREEMENT SEPARATION AND PURIFICATION OF RADIOISOTOPES FOR RESEARCH

W. R. WILKES AND R. E. EPPLEY

MOUND FACILITY
Miamisburg, Ohio 45342

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

for the
U. S. DEPARTMENT OF ENERGY
Contract No. DE-AC04-76-DP00053

Monsanto

MLM-ML-80-51-0004(F)

Monsanto

MOUND FACILITY
Operated for the United States
Department of Energy

December 4, 1980

Mr. Harry N Hill, Area Manager
U. S. Department of Energy
Dayton Area Office
P. O. Box 66
Miamisburg, Ohio 45342

Dear Mr. Hill:

Enclosed for your review and transmittal to the Department of Energy are 21 copies of the Field Task Proposal/Agreement entitled "Separation and Purification of Radioisotopes for Research," No. P80-11-321. The proposals should be directed to Roger K. Heusser, Director, Materials Processing, Office of Nuclear Materials Production and to the appropriate person in the Office of Basic Energy Sciences.

The Finance Department has participated in the preparation of the financial data and concurs with its presentation as given here.

Very truly yours,

W. T. Cave

W. T. Cave, Director
Nuclear Operations

Vallée:mg
Enclosures 21

cc: H. N Hill (2)

bc: R. E. Vallée, w/enc.
W. R. Wilkes, w/enc.
R. E. Eppley, w/enc.
File, w/enc.

PROPOSAL No. P80-11-321

FIELD TASK PROPOSAL/AGREEMENT
SEPARATION AND PURIFICATION OF RADIOISOTOPES FOR RESEARCH

PREPARED BY: W. R. WILKES AND R. E. EPPLEY

SUBMITTED BY: R. E. VALLÉE

APPROVED BY: W. T. CAVE

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DOE Form 5120.2
(12-78)

U.S. DEPARTMENT OF ENERGY

FIELD TASK PROPOSAL/AGREEMENT

1. WP BIN NUMBER	2. TASK NO.	3. REV NO.	4. PROJECT NO.	5. DATE PREPARED (mm dd yy)	6. CONTRACTOR NUMBER
		0		11/20/80	P80-11-321

7. TASK TITLE	8. WORK PACKAGE TITLE
Separation and Purification of Radioisotopes for Research	

9. BUDGET AND REPORTING CODE	10. TASK TERM Begin: (mm dd yy)	End: (mm dd yy)	11. CONTRACTOR NAME	12. CODE (see instructions)
	070166	Continuing	Monsanto Research Corp. Mound Facility	

13. CONTRACTOR TASK MANAGER (Name, FTS No.)	14. PRINCIPAL INVESTIGATORS
William R. Wilkes 774-3893	Richard E. Eppley 774-3163

15. WORK LOCATION (See instructions): Name of facility, City, State, ZIP Code	16. Does this task include any management services efforts?
Mound Facility Miamisburg, Ohio 45342	☐ YES ☒ NO

17. TASK DESCRIPTION (Approach, relation to work package, in 200 words or less)

The FY-1981 purpose of this task consists of work in two main areas:

I. Production of Separated Isotopes:

A. Uranium-234, Separated from Aged Plutonium-238

B. Polonium-209, Separated from Aged Polonium "Foils" Remaining from the Production of Polonium-210

This material is shipped to ORNL for distribution through their Isotope Sales Group where it is available to both DOE and non-DOE customers.

As described later in the WPAS, there is a continued demand for uranium-234 which warrants its production at a 7-10 gram per year level. It is still too early to characterize the demand for polonium-209 since it just became available during FY-1980. Within the scope of the basic program funding, 10 grams of uranium-234 can be produced or polonium-209 can be substituted for up to 3 g of the uranium-234 (i.e., produce a minimum of 7 g uranium-234). For an equivalent cost, 12 μCi of polonium-209 could be produced for each gram of uranium-234 not produced.

For the uranium-234, the technical approach is to use a process similar to the one developed and used at Mound since 1966. Details of this process are given in Attachment 19.E.

18. CONTRACTOR TASK MANAGER

(Signature) (Date)

19. DETAIL ATTACHMENTS: (See instructions)

☐ a. Facility Requirements	☒ d. Background	☒ g. Future accomplishments	☐ j. Explanation of milestones
☒ b. Publications	☒ e. Approach	☒ h. Relationships to other projects	
☒ c. Purpose	☒ f. Technical progress	☐ i. Environmental assessment	☒ k. Other (specify): K.1 - Encapsulation K.2 - Drum Repainting K.3 - Capital Equi

17. TASK DESCRIPTION (Cont'd)

The polonium-209 separation process is also described in Attachment 19.E.

II. Maintenance of Standby Facilities:

Laboratory space is being maintained for the resumption of the following processes:

1. Protactinium-231 and Thorium-230. Separation and purification from natural uranium (Cotter) concentrates.

2. Thorium-229. Separation and purification from aged uranium-233.

The above two processes were put in stand-by at the end of FY-1979 because of rising inventories at ORNL for each of the separated isotopes. These processes can be resumed when the sales/loan levels warrant.

A survey completed in FY-1979 (Ref. 4) indicates that there is a continued interest and a potential increase in demand for these isotopes. Details of the Cotter and uranium-233 processes are given in References 2 and 3, respectively.

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TASK REQUIREMENTS FOR OPERATING/EQUIPMENT COSTS AND OBLIGATIONS

CONTRACTOR NAME
Monsanto Research Corporation

BIN NUMBER	TASK NO.	REV NO.	DATE PREPARED	CONTRACTOR NUMBER
		0	11/20/80	P80-11-321

20. STAFFING (in staff years)	BY-2 1981	BY-1 1982 PRESIDENT'S	BY-1 1982 REVISED	AUTHORIZED	BY-FY 19 83
a. SCIENTIFIC	2.3		2.3		2.3
b. OTHER DIRECT	0.2		0.2		0.2
c. TOTAL DIRECT	2.5		2.5		2.5
21. OBLIGATIONS AND COSTS (in Thousands)					
a. TOTAL COSTS	289/400*		323/447*		355/492*
b. TOTAL OBLIGATIONS					
22. EQUIPMENT (in Thousands)					
a. EQUIPMENT COSTS	145				
b. EQUIPMENT OBLIGATIONS					
23. OTHER COSTS (specify)					
a. Encapsulation Case I**	100K				
b. Incremental Cost: Case II**	101K				
c. Cotter Drum Maintenance (See K2.)	40K				

24. OPTIONAL FIVE-YEAR PLAN (in Thousands) Constant BY dollars	BY + 1	BY + 2	BY + 3	BY + 4
a. TOTAL OPERATING COSTS				
b. TOTAL OPERATING OBLIGATIONS				
c. TOTAL EQUIPMENT COSTS				
d. TOTAL EQUIPMENT OBLIGATIONS				

25. MILESTONE SCHEDULE	PROPOSED SCHEDULE	AUTHORIZED SCHEDULE

*These values show the uranium-234 production cost (NMP) and the total program cost. The difference is the SW laboratory stand-by costs (BES) for the thorium-229, thorium-230, and protactinium-231 production.

**See K1.

19.B. PUBLICATIONS

1. P. L. Keister, P. E. Figgins, and R. M. Watrous, Recovery and Purification of Uranium-234 from Aged Plutonium-238, MLM-2490 (January 20, 1978).

2. M. R. Hertz, W. S. Stringham, and R. M. Watrous, A Solvent Extraction Method for the Recovery of Thorium-229 from Aged Uranium-233, MLM-2151 (January 31, 1975).

3. P. E. Figgins, M. R. Hertz, W. R. Deal, R. M. Watrous, and R. E. Eppley, Concentration of Protactinium-231 and Thorium-230 from Uranium Mill Limed Raffinate Solids (to be published).

4. R. E. Eppley and R. E. Vallée, Reported Needs for the Radioactive Isotopes Uranium-234, Protactinium-231, Thorium-230, and Thorium-229, Report to J. L. Burnett, OBES (June 1, 1979).

5. P. E. Figgins, V. R. Casella, C. T. Bishop, and A. A. Glosby, Polonium-209: A New Source of Analytical Tracer, presented at the 23rd Conference on Analytical Chemistry, Gatlinburg, TN, October 9-11, 1979.

19.C. PURPOSE

The present purpose of this program is to produce high-purity uranium-234 (99%) and polonium-209 for the scientific community, both Governmental and non-Governmental. In addition, facilities for separation and purification of protactinium-231, thorium-230, and thorium-229 are maintained in stand-by condition for the resumption of these processes when conditions warrant. The uranium-234 isotope is separated from aged plutonium-238 material, purified, and converted to solid U_3O_8. This oxide is subsequently shipped to Oak Ridge National Laboratory for distribution through their Isotope Sales Group.

The principal use of uranium-234, which is recovered from aged plutonium-238, is in fission detectors used to monitor reactors. Approximately one-third of the total uranium in a fission detector is uranium-234. The other two-thirds is uranium-235. A typical detector might contain 15 mg total uranium. As the neutron flux in the reactor causes fission of the uranium-235 in the detector, it also converts the uranium-234 to uranium-235. This latter nuclear reaction,

$$^{234}U + n \rightarrow {}^{235}U,$$

greatly extends the life of the detector in the reactor.

Other uses are mostly for research in nuclear and chemical studies. Listed are typical uses:

1. Standard for emission spectroscopy
2. Energy level determinations
   A. Moessbauer spectroscopy
   B. Multi-level analysis of neutron cross sections
   C. Single-level analysis of uranium-233 fission cross section
3. Uranium-234 fission studies
4. Tracer in uranium separation chemistry
5. Uranium-234 proton-induced fission
6. Uranium-234 quadrupole moments
7. Uranium-234 radiometric analysis in continuous in-line monitor for UF_6 enrichment
8. Uranium-234 scintillation counting of normal uranium in water and urine
9. Uranium-234 thermal fission study using gaseous scintillators and plastic track detectors
10. Uranium-234 nuclear reactions studies

This is the only program providing uranium-234 to ORNL for inventory purposes.

Polonium-209 shows potential principally as a tracer for polonium-210 determinations. Currently, polonium-208 is used for this purpose. Polonium-209 should be superior because it is less expensive than polonium-208 and, additionally, has a half-life of 103 yr compared to only 2.90 yr for polonium-208. Interest in polonium-209 has been expressed by several organizations, including NBS. Ref. 5 describes the use of polonium-209 in more detail.

19.D. BACKGROUND

Mound has been separating uranium-234 from aged plutonium-238 since 1966. Prior work with thorium-232, radium-226, and actinium-227 provided the background and experience for this separation.

Until recently, the major source of uranium-234 has been from waste solutions generated during plutonium-238 recovery from heat sources. Various quantities of plutonium-238 (as oxides, metal or alloys) in various encapsulations are now the principal sources of uranium-234. Currently, the program has about 5,000 grams of plutonium-238 in storage. This material is processed at the rate of 150 - 200 g per year for uranium-234 recovery. The production level can be increased in the future should a greater user demand arise.

19.E. APPROACH

Uranium-234 Product

Uranium-234 is recovered from aged plutonium-238. A summary of the process is shown in Fig. 1. As shown in the figure, after a batch of the solid material is dissolved, there are three major chemical steps necessary to complete the separation and purification. These are 1) the initial oxylate precipitation, followed by 2) an anion exchange in nitrate media, followed by 3) an anion exchange in chloride media. At the end of this third step, the uranium-234 is calcined, packaged as U_3O_8 and shipped to ORNL. After the uranium has been recovered, the plutonium is converted to oxide and returned to storage.

Typically, the "cow" material contains about 80% plutonium-238. During the processing, each successive step lowers the level of plutonium in the recovered uranium-234. After the final chloride exchange step, the plutonium-238 level is less than 10 ppm. This procedure normally reduces all impurities to a level of 1% or less, as detected by emission spectroscopy.

All uranium-234 processing is performed in three glovebox lines, located in a single room. The entire procedure is closely monitored by Health Physics for safe operation.

Plutonium-238 has a half-life of 87.404 yr and decays by α-emission to uranium-234. This means that 100 g of plutonium-238 will yield 790 mg of uranium-234 in one year.

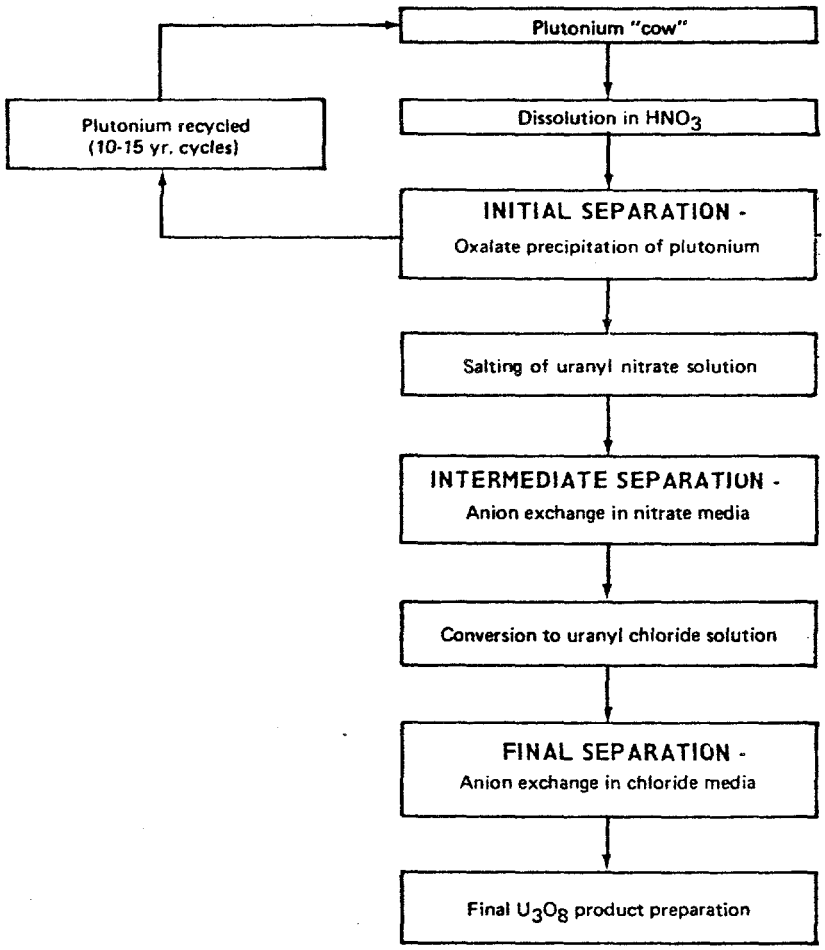

Figure 1. CHEMICAL SEPARATION STEPS FOR RECOVERY AND PURIFICATION OF URANIUM-234.

Clearly, it is advantageous to process well-aged material, in order to increase the yield of uranium-234. In general, material picked for processing is normally 8-10 yr old. Therefore, to separate 10 g of uranium-234 (the FY-1980 level), approximately 150 g of "cow" material must be processed. This is accomplished in a number of batches, normally 25 g each.

Polonium-209 Product

Recently, interest has been expressed for using polonium-209 as a tracer in the analysis of environmental samples. This isotope has an advantage over polonium-208 since its half-life is 103 yr versus only 2.9 yr for polonium-208.

For many years, Mound was involved in the production of polonium heat sources, most of which utilized polonium-210 obtained from the neutron irradiation of bismuth metal. The primary reaction is

$$^{209}Bi(n,\gamma)^{210}Bi \xrightarrow[5.013d]{\beta^-} {}^{210}Po$$

In addition, there are two significant secondary reactions:

$$^{210}Po(n,2n)^{209}Po$$

$$^{210}Po(n,3n)^{208}Po$$

The polonium isotopes from these reactions ultimately were plated on platinum foils and encased in sealed nickel tubes. These tubes, in turn, were enclosed in glass ampoules.

A number of these foils have been preserved at Mound and are now 10-12 yr old. The bulk of the polonium-208 and polonium-210 has decayed, leaving behind polonium-209 which is approximately 99% enriched relative to the other polonium isotopes. No other activities are observed in the foils which have been checked.

To separate the polonium-209, the glass ampoules are first crushed, and the platinum foil is removed from the nickel tube. The foil is leached with hot 4M HNO_3/HCl solution to dissolve the polonium material. The polonium is then separated by plating onto a silver foil.

During 1980 23 μCi of polonium-209 was separated and sent to ORNL. It is estimated that another 100 μCi or more remains to be separated from foils still on hand. If a larger quantity of this material is needed in the future, it is expected that 700 to 7000 μCi of polonium-209 could be obtained from a SNAP-III heat source.

Ref. 5 of Attachment 19.B contains further details on this process and provides historical background.

19.F. TECHNICAL PROGRESS

Progress in FY-1980

In FY-1980 progress was made in several areas. The following isotopes were shipped to Oak Ridge National Laboratory:

Isotope	Shipped	FY-1980 WPAS Commitment
Uranium-234 (99+%)	10.0 g	10.0 g
Polonium-209 (≈99%)	23 μCi	-

The need for the polonium-209 developed after submission of the FY-1980 WPAS. The work was supported by a purchase order from ORNL.

In addition to this production work, process development work resulted in improved operation in both the initial dissolution/precipitation step and the first uranium-fraction separation step. Semi-automated equipment for the dissolution/precipitation step was designed and installed in a glove line. Part of the FY-1980 production was accomplished using this improved equipment. Decreased personnel radiation dosage and more efficient operation of this step are both expected to result from these changes.

The first anion exchange step was modified by development of a solvent extraction step which is used before the exchange step. The extraction step is carried out over a several-day period in a "continuous" separation process. The solvent in this system is 10% DSBPP (di-sec-butylphenyl-phosphonate) in 1, 1, 2 trichloroethane. The uranium component is continuously stripped with water. This new step has been effective in removing metallic impurities from the uranium-234; and, in particular, the americium-241 content is reduced by a factor of 100-1000. This additional step has reduced the radiation level in the uranium fraction so that the subsequent process steps can be carried out with a lower personnel dose rate.

During FY-1980, substantial maintenance was performed on the tops and fronts of four gloveboxes for improved visibility into the boxes. At the same time, exhaust filters were replaced, and other small changes were made in the boxes. Further work of a similar nature remains to be done in the near future on at least four more boxes.

Laboratory space in SW Building is being maintained in a stand-by condition for production resumption of protactinium-231, thorium-230, and thorium-229.

19.G. ACCOMPLISHMENTS IN FY-1981

The uranium-234 separation and purification will continue at the current rate of production. Seven to ten grams of uranium-234, enriched to 99%, in addition to up to 35 μCi of polonium-209, will be shipped to ORNL. Further process development work will be performed with the goal of improved process efficiency and lowered personnel radiation exposure.

Data on inventory levels and user demand for protactinium-231, thorium-230, and thorium-229 will continue to be reviewed. Laboratory facilities for the processing of these isotopes will continue to be maintained in stand-by condition for start-up when the demand warrants. The Cotter Concentrate material and the uranium-233 "cow" material will remain in storage and will be ready for the resumption of processing.

Encapsulation of the plutonium-238 "cow" material will proceed in accordance with one of the cases described in Attachment 19.K.1. Also, a detailed design and cost estimate for replacement of the uranium-234 glovebox lines will be completed. The design will assume the replacement of all gloveboxes now in use. Attachment 19.K.3 provides more details. If funding allows, work will begin on repainting the Cotter Concentrate drums (see Attachment 19.K.2).

19.H. RELATIONSHIP TO OTHER PROJECTS

Mound is engaged in other isotope separation programs as outlined in proposals dated approximately February 25, 1980, and entitled Separation and Purification of Special Stable Isotopes for Research, Liquid Thermal Diffusion Development for Calcium Isotope Enrichment, Isotope Separation Research and Development, and Chemical Exchange Development for Uranium Isotope Separation.

19.K.1 PLUTONIUM-238 ENCAPSULATION COSTS

Two cases are detailed in Table 1 for encapsulating the plutonium-238 "cow" material in storage at Mound. Case I assumes encapsulation of the existing containers, regardless of the weight of the plutonium they contain (actually between 1 and 190 g). Case II assumes that the "cow" would be subdivided to provide units no more than 25 g each before encapsulation.

Case I provides the most cost-effective approach to encapsulating all of the "cow" material. Each existing container would be handled as a single unit. As can be seen in the table, both labor and material costs would be minimized. In the future, the material would be repackaged into 25-g units as it is processed. The initial encapsulation could be accomplished within 12 mo of authorization to do the work.

Case II, on the other hand, would increase both the labor and material costs. It would require 300 man-days of effort for the encapsulation, which would make it unlikely that the entire job could be completed in a single fiscal year. In addition, the labor estimate does not include the time necessary for process personnel to open existing containers, separate the material into 25-g units, and place these units in new, smaller containers preparatory to encapsulation. These smaller containers would also need to be designed. It is probable that a large fraction of a year would be needed by laboratory personnel to accomplish the encapsulation if it is carried out in this manner. This would impact the year's production of uranium-234 and possibly eliminate production altogether.

Table 1. PLUTONIUM-238 ENCAPSULATION COSTS

Case I: Encapsulate Existing Containers, Regardless of Weight (between 1 and 190 g).

Engineering Design	$ 15K
Installation of Equipment	28K
Containers 70 @ 100	7K
Labor 1.5 man-days/container @ $400/man-day	42K
Contingency (8%)	8K
	$100K

Case II. Redistribute All Material Into 25-gram Lots Before Encapsulation

Engineering Design	$ 15K
Installation of Equipment	28K
Containers: 200 @ 100 (5 kg material/25)	20K
Labor 1.5 man-day/container @ $400/man-day	120K
Contingency (10%)	18K
	$201K

19.K.2. COTTER DRUM MAINTENANCE

The Cotter Concentrate has been stored at Mound for several years in approximately twelve hundred 55-gal drums with TRU liners inside the drums to actually contain the material. The drums are now starting to rust and are in need of repainting. This work should be completed within the next year or two to halt the rust process before it is allowed to create a larger problem. All drums will need to be sanded and then treated with an application of a rust inhibiting paint. The estimated cost of treating the Cotter storage drums is given below. Whether this work is done in FY-1981 or FY-1982, it will require additional funding beyond that shown in Section 21.

Total Labor	$29,150
Equipment	3,000
Paint	1,800
Contingency (≈18%)	6,050
Total	$40,000

19.K.3 DESCRIPTION OF CAPITAL EQUIPMENT ITEMS

The major capital equipment items are the replacement of the glovebox lines used for the plutonium-238 separation process. The boxes now in use are 15-20 yr old and have seen long and hard use. Right now they are safe to use, but it is evident that they will need to be replaced within the next few years. Currently, an estimate of the cost of replacing the gloveboxes is being developed. This estimate will be completed in time to be included in the regular WPAS submission in the Spring of 1981. It is expected that the regular WPAS submission will include a) the cost of a detailed design of the glovebox replacement work; b) an estimated cost of replacing the gloveboxes, including the materials purchased, the tearout and reinstallation costs; and c) a rough timetable for the work.

One of the main points to be made when considering replacement of these boxes is that they almost certainly need to be replaced in groups. There are three distinct groups of gloveboxes, with each group forming a glovebox line for one of the three major uranium-234 processing steps. To minimize contamination of the room, an entire line should be replaced at one time.

For this reason, it is expected that the gloveboxes should be replaced all at once; or at the least, one group per year over a three-year period if budgetary constraints prevent a faster schedule. The estimate will be prepared for the "all at once" option, with an additional estimate of the added cost of doing the work in three phases.

Although no glovebox replacement would occur before FY-1982, even with the "all at once" option, the current lead time of 9-10 mo for gloveboxes requires that purchase of the gloveboxes be initiated at a very early point in the schedule. For this reason, it is requested that $120K be provided to permit designing and purchasing the gloveboxes. In addition, $25K is requested to pay for the detailed design of the entire job. None of these funds would be committed until after the initial estimate has been completed.

Other capital equipment items needed to upgrade the uranium-234 processing can be postponed until the boxes are replaced - provided that this replacement is not delayed for too long a time. It would be desirable to install certain process automation in the new boxes in order to fully implement the development work now in progress; however, the gloveboxes which are being used currently would be difficult to modify for the installation of this equipment. Costs that can be identified now are as follows:

1. Detailed design and cost estimate, $25K.

2. Purchase of gloveboxes, $120K.