Feasibility Study for Production of ^{51}Cr

Background

Cr-51, with a half-life of 27.7 days, decays by electron capture to ^{51}V, emitting a ~750 keV neutrino 90.14% of the time and emitting a ~430 keV neutrino and 320 keV gamma ray 9.86% of the time. As the energy of a solar neutrino resulting from electron capture decay of ^{7}Be is 862 keV, Cr-51 provides for a similar source of neutrinos. Interest has been expressed in the production of 5-10 MCi Cr-51 source, which meets the special form requirements, as described by the International Atomic Energy Authority (IAEA) document "Regulations for the Safe Transport of Radioactive Material." This study assesses the feasibility of producing such a source in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL) and describes the proposed methods and associated costs for producing the source.

Target Material

In the 1990s the GALLEX collaboration used 36.5 kg of 38% enriched Cr-51 (which was also depleted in the relatively high neutron absorbing isotope Cr-53) to produce a 1.7 MCi source by irradiating the material in the Siloè reactor in Grenoble, France. They later repeated this operation with the same material and produced a 1.8 MCi source. This project will be using the original enriched GALLEX source material for production of the MegaCurie level Cr-51 source. ORNL will provide the drawings and specifications for the enriched Cr target rods, as well as for two natural Cr test targets, described later in this study. However, fabrication of the natural and enriched Cr target rods will be by an external supplier, and the responsibility for this fabrication lies solely with the customer. ORNL will be supplied with all documentation necessary to approve the Cr target rods for placement in the HFIR. ORNL will fabricate any liners or other removable fixtures necessary for placement of the target rods in the HFIR.

Target Design

The preliminary design chosen for the Cr target rods is a single solid cylindrical target per irradiation position. Use of a single solid target per position both maximizes the amount of Cr that can be loaded into each position, and simplifies the target handling requirements during target loading, unloading, and packing, in turn allowing for faster shipment, and a larger source delivery to the customer. The height of the target will be approximately 50 cm, the active height of the HFIR core. The end of each target will include a tapped hole, or other fixture such that a lifting attachment may be fitted to one end of each target, to allow for pick-up and movement with handling tools using the bridge crane over the HFIR pool. The lifting attachment will be shipped with the targets, to be removed during source installation. The solid target rods may further be placed inside a thin target liner inside the small VXF positions, to provide greater control over the flow of coolant through the channel. If used, the target liner would be an independent fixture, and would not be shipped with the Cr targets. Use of a target liner would also simplify target design/acceptance in the event that the targets are re-irradiated. A fully scoped target design, including preparation of all associated drawings, welding specifications, inspection plans, and other technical documentation is included as part of this proposed work, assuming use of a target liner. The design phase of this work will also assess the impacts on the target loading on the HFIR beam tube fluxes, and adjust the design as necessary to minimize any effects.

Target Irradiation

Irradiation of the Cr target rods was simulated assuming the following parameters:

Table 1: Target parameters

Target height	50 cm
Target radius	1.82 cm
Cr density	7.0 g/cm^3
Cr volume	521 cm^3
Cr mass	3.65 kg
# Targets	10

The 10 targets were modeled placed in the 10 available positions in the inner small vertical experimental facilities (VXF), including a 0.1 inch thick Al liner. Irradiation was simulated assuming 85 MW operation and a cycle length of 24 days. Based on this target design and irradiation conditions, it is expected that 3.13 MCi can be produced in a single irradiation cycle of the 10 targets.

If desired a second irradiation cycle can be performed, with a reduced downtime between cycles. Typical downtime for refueling and maintenance is a minimum of 18 days. Depending on the activities scheduled during this downtime, it may be possible to reduce this downtime to a minimum of 6 days; however, the actual downtime will vary. The following figure shows the estimated Cr-51 activity following two cycles of irradiation, based on the downtime between cycles. This figure shows that with an 18 day downtime, the source activity is increased by 38% to 4.21 MCi. With a 6-day downtime, the shortest that is currently considered feasible, this activity increases to 4.69 MCi, a 9% increase over the 18-day downtime scenario, and a 50% increase over the single cycle irradiation.

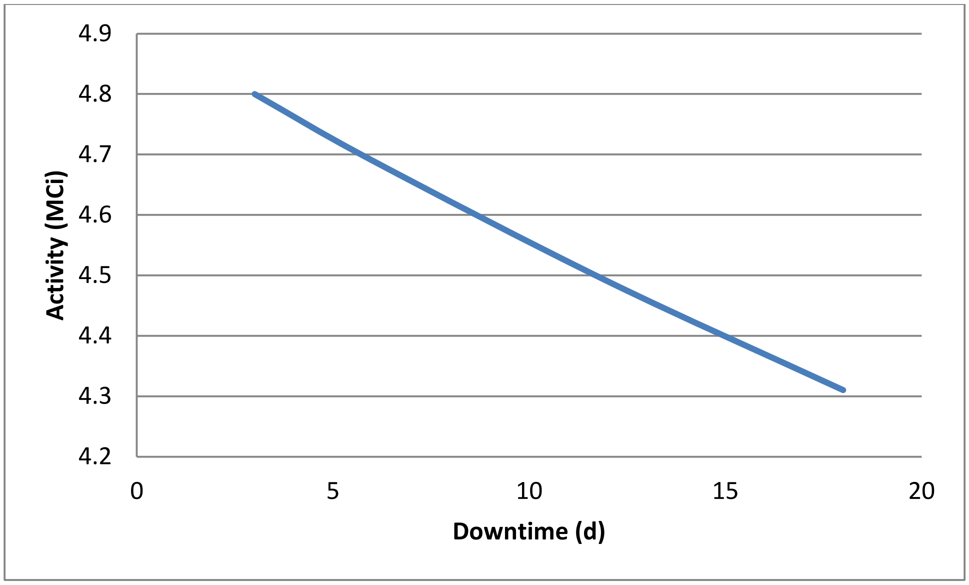

Figure 1: Cr-51 Activity Following 2 Cycles of Irradiation with Varying Downtime

Impurities

Previous irradiation of the GALLEX material to produce a 1.8 MCi Cr-51 source resulted in the following impurities.

Table 2: Impurities from previous GALLEX irradiation

Isotope	Activity at EOB (GBq)	$T_{1/2}$
^{24}Na	0.08	15.0 h
^{46}Sc	.13	83.8 d
^{48}Sc	0.095	1.8 d
^{60}Co	0.03	5.3 y
^{64}Cu	210	12.7 h
^{77}Ge	.5	11.3 h
^{76}As	1.6	1.1 d
^{97}Zr	.4	16.9 h
^{110m}Ag	4.35	250 d
^{124}Sb	.2	60 d

The majority of these radionuclides are short lived, produced directly from a precursor stable isotope, and would be expected to be produced at a level directly correlated with the neutron flux. As such, a bounding level of impurity for these short lived radionuclides would be one order of magnitude greater than the level reported in the Siloè irradiation. For longer lived radionuclides ^{46}Sc, ^{60}Co, ^{110m}Ag, and ^{124}Sb, these are as well bounded by a value one order of magnitude greater than the level reported in the Siloè irradiation. However, these radionuclides would be expected to be lower than this value, due to the greater importance of neutron absorption in the radionuclide, versus decay, in the depletion of the radionuclide.

The exact level of impurities in the GALLEX material is not known at this time, and the impurity levels may be further changed during electroforming of the solid targets. Prior to irradiation of the material, neutron activation analysis will be performed on a sample to determine precise impurity levels, so that expected impurity radionuclides can be assessed in greater detail. This work is included as part of this proposal.

The chromium itself produces only two significant activation products during irradiation. The activation products with the next greatest activity at 6 days post-irradiation are ^{49}V, with an activity of less than 1 Ci and ^{48}V with an activity of 2 mCi. All other activation products from the Cr are less than 1 mCi.

Source Homogeneity

As per discussions with Jonathan Link and Marco Pallavicini, there is no requirement for homogeneity of the Cr-51 source and no provisions are made in this proposal to ensure homogeneity. Specific activity along the Cr rod is expected to vary from 320 Ci/g at the ends of the rod, to 475 Ci/g at the center. Additionally specific activity is expected to vary by ~20% radially through the target, with the highest activity occurring along the edge of the target closest to the core.

Heat Modeling

Each target rod is expected to produce approximately 110 Watts of decay heat. The nuclear heat generation rate of chromium is expected to be similar to that of stainless steel, which is approximately 5 W/g in the inner small VXF (C-HFIR-2012-006). The resulting nuclear heating rate is approximately 18,250 W per target, for a total heating rate of 18.4 kW per target. With a volume of 521 cm^3 the volumetric heating rate is 35,028,791 W/m^3.

A conservative bulk temperature for the coolant during irradiation is 370 K. Applying a convective boundary condition to the outer surface of the Cr rod with a heat transfer coefficient of 22.5 kW/m^2-K and a temperature of 370 K, the maximum internal temperature of the Cr rod with the dimensions listed in Table 1 and a volumetric heating rate of 35,028,791 W/m^3 is 415 K. There are not expected to be any concerns with providing sufficient cooling for the bare Cr targets, though a full study will need to be performed as part of the target design phase of this proposal. As part of this study the total coolant flow through the reflector positions will also be analyzed for any adverse effects on HFIR operation.

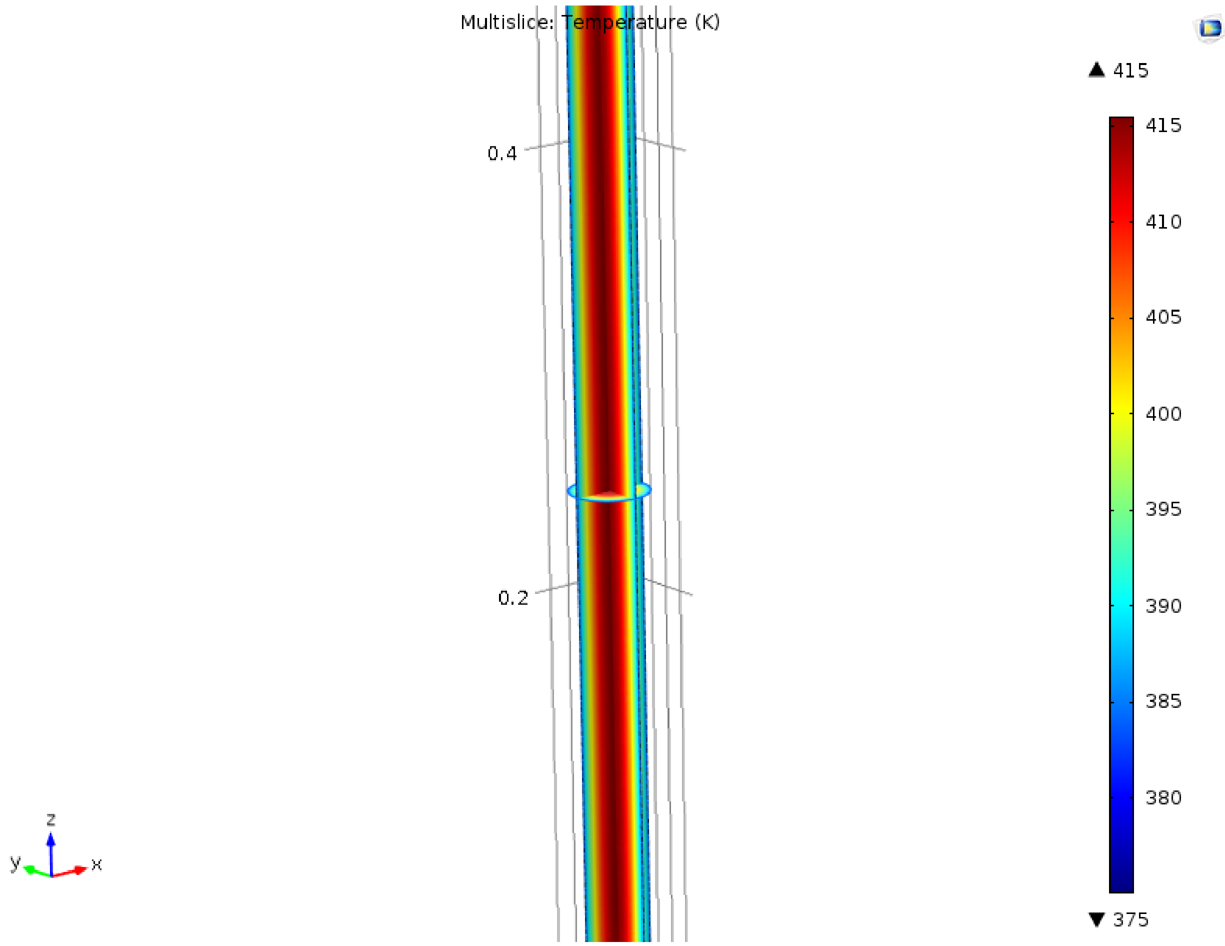

Figure 2: Heat distribution in solid Cr target during irradiation

Gamma Assay

As part of this work a system for performing gamma assay on the irradiated targets, to determine the ^{51}Cr content following removal from the VXF, must be developed. Design of the gamma probe will include assessing the height to which the Cr rods can be raised within the pool, and the design of the collimator and detector shield housing to ensure accurate counts can be made in a short duration, and saturation will not occur. Design of this system will be a cooperative effort between ORNL and the customer team, and the system will be fabricated and supplied by the customer team. ORNL will handle the unit from an operator platform and gamma assay will be performed at predetermined points along the length of target. It is proposed to test the detector efficiency using an irradiated reduced length natural Cr target. The system will be removed from the pool by ORNL following completion of the work, decontaminated, and returned to the customer.

Material Handling/Loading

Material handling of the production targets at ORNL will involve fitting the targets into the target liners, inserting and removing the targets (and liners if used) into the small VXF positions, performing gamma assay of the targets, and loading the targets into the appropriate shipping containers. It is expected that all material handling will take place within the HFIR pool area.

Special Form Requirements

Special form radioactive material shall mean either an indispersible solid radioactive material or a sealed capsule containing radioactive material. Special form radioactive material must be subject to impact, percussion, bending, and heat tests, as laid out in IAEA document "Regulations for the Safe Transport of Radioactive Material." Additionally the material must be subject to a leach or leakage test following each test. ORNL will perform all necessary tests and submit an application for a certificate of approval for the special form radioactive material for the solid Cr rods. It is proposed that these tests be performed on an unirradiated dummy target composed of natural chromium. In addition, a reduced length natural Cr target will be irradiated and subject to additional leach testing, in order to demonstrate how the irradiation of the material may affect the ability of materials to leach from the target. This same dummy target can be used to perform the gamma assay testing discussed in the previous section. This work will involve additional material handling to transfer the irradiated target to a laboratory hot cell facility for performance of the final leach test.

Proposed Scope of Work for Full Scale Irradiation

1. Design and certification of irradiation targets, surrogate targets, required fixtures, and irradiation schemes for the production of a MegaCurie level source of Cr-51. Design, certification, and handling of an in-pool gamma assay system. Development of all necessary procedures, safety documentation, and training for irradiation of targets and loading of irradiated targets into shipping containers.
    a. Supply of the gamma assay system and Cr target rods is the responsibility of the customer. This scope of work covers only design and approval for use.
2. Irradiation of one reduced length natural Cr target rod, to be use for testing of the gamma assay system and evaluation of the leach rate from the target.
3. Perform all required testing to meet the special form regulations for the safe transport of radioactive material. These tests will be performed on surrogate natural Cr rods.
4. Receive 10 fabricated enriched Cr target rods.
5. Fabricate all necessary fixtures to insert the Cr target rods into the HFIR.
6. Receive 3 type B shipping containers, each certified for up to 2.4 MCi of ^{51}Cr and 520 W of thermal power. These shipping containers are intended to be loaded underwater in the pool and they will be designed to ensure that the regulatory limits for radiation dose are not violated when the containers are removed from the pool.
7. Install fixtures and targets in the designated positions in HFIR and irradiate Cr targets using the target configurations and irradiation schemes as determined in task 1.

8. Remove targets from the HFIR. Perform gamma assay at multiple pre-determined points on each target. Determine the activity of each target and provide this information to the necessary persons for preparation of shipment documentation.
9. Load targets into pre-approved shipping containers. Remove closed shipping containers from the pool and hold for pick up by the shipping company, Edlow International.
10. Disposition of surrogate targets, fixtures, and other generated wastes.
11. Decontamination of gamma assay system for return to customer.

Proposed Schedule

The schedule for performance of this work will depend upon the supply of the enriched and natural Cr target rods, the approval of the special form by an IAEA Certificate of Competent Authority (CoCA), and the availability of the HFIR, particularly if a dual-cycle irradiation with reduced downtime is performed. Durations for these tasks have been estimated to be 4 to 6 months each. A tentative schedule is laid out in the attached spreadsheet.

Upon project initiation, detailed target design and approval will be commenced, and the appropriate drawings and documentation will be provided to the supplier of the target rods. These designs will be used for fabrication of the full size and reduced length natural Cr targets, to be supplied for special form testing. Fabrication of HFIR fixtures for the reduced length target may take place concurrently to external fabrication of the Cr targets. Upon receipt of the natural Cr dummy targets, special form testing will begin, including irradiation of the reduced length target for final leach testing. Approval to commence with enriched Cr target fabrication will not be given until the special form CoCA is obtained. Once the final enriched targets are received and accepted, scheduling of the irradiation will be subject to the availability of the small VXF positions in HFIR. A period of six months is allocated for scheduling of irradiation, to allow other experiments in the small VXF to complete their irradiations, or reshuffled other targets as necessary to accommodate the 10 enriched Cr targets.

Year one will involve the design and fabrication of the natural Cr targets, which will be tested and submitted for a special form CoCA in year two. Following issuance of the CoCA fabrication of the enriched Cr targets will begin in year three. Irradiation is expected to take place in year four, with exact timing subject to availability of the HFIR, transportation logistics (arranged by others) and the ability of the customer to receive the source.

Cost Summary

The overall costs for a 1 and 2 cycle irradiation are estimated to be $1,600,000 and $1,900,000, respectively. The cost estimates are based on the project beginning in FY18 using the predicted labor rates. If the project scope is accepted, a formal quotation with a more precise cost estimate would be prepared for the actual planned period of performance and would be the basis for a purchase contract.