

# Experiences and Lessons Learned at CARIBU with an Open Californium-252 Source

S. Baker, J. Greene, T. Levand, R. Pardo, G. Savard, R. Vondrasek, and L. Weber

**Argonne National Laboratory** 

June 19, 2012



## Outline

- Cf-252 sources
  - Properties
  - Description
  - Limits
- Source preparation and transfer
- <u>Californium Rare Ion Breeder Upgrade (CARIBU) shielding cask</u>
- Airborne effluent monitoring
  - Off-site calculated doses and mitigation
  - Filtration
- Contamination
  - Cover foil failure
- Lessons Learned
  - Value of weaker sources
  - Importance of monitoring

#### **Properties of Cf-252**

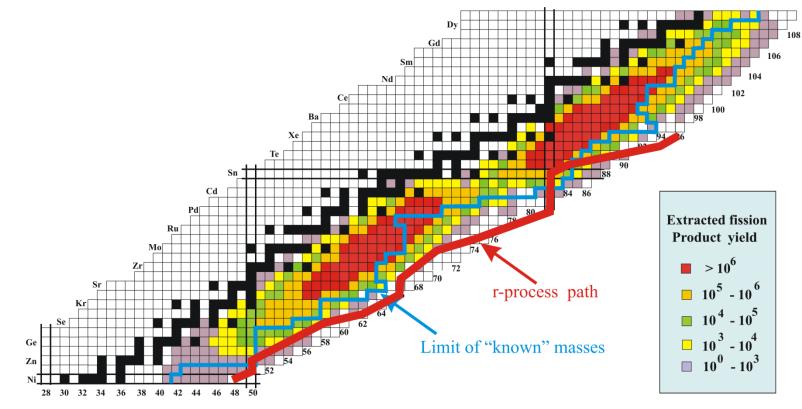
- Half life = 2.645 y
- Spontaneous fission branch = 3.09%
  - $\rightarrow$  1.1 x 10<sup>9</sup> fissions/sec/Ci
- Neutron output = 4.4 x 10<sup>9</sup> n/sec/Ci
- "Prompt" gamma output = 2.3 x 10<sup>10</sup> γ/sec/Ci
- External radiation from a 1 Ci open Cf-252 source :
  - Unshielded neutron dose rate:
    - 46 rem/hr @ 30 cm
  - Unshielded photon dose rate:
    - 2.8 rem/hr @ 30 cm



## Californium Fission Source for ATLAS CARIBU Project

#### <sup>252</sup>Cf fission products are neutron rich.

#### <sup>252</sup>Cf spontaneous fission yield from 1 Ci source



#### **Source Description**

- Electrodeposited source
- Cover foil to stop californium recoils but allow fission fragments to pass through
- Source width limited by diameter of transfer tube between hot cells at ORNL to less than 2.5 cm
- Three sources prepared in succession for gaining experience:
  - 2 mCi
    - Contact handled (100 mrem/hr at 30 cm)
  - 100 mCi
    - Used to meet project commissioning goals
  - 500 mCi
    - Prepared, but not in use yet
    - Close enough to 1 Ci requested strength for experiments

## **Source Preparation and Transfer**

- 2 mCi source prepared in glovebox at ORNL and transferred at Argonne without remote handling for use as a fission fragment ion source at the Argonne ATLAS heavy ion accelerator
- CARIBU shielding cask designed for 1 Ci source used to gain experience with 2 mCi and 100 mCi sources
- Effluents and hold-up characterized with 2 mCi source
- 100 mCi source prepared in ORNL hot cell and transferred to CARIBU cask from Argonne hot cell
- 500 mCi source recently prepared in ORNL hot cell and awaiting transfer until CARIBU is ready
  - Readiness review scheduled for July 17.

#### 500 mCi Cf-252 Source Electrodeposited at ORNL



HIAT 2012

7

## 1 Ci Source Limits

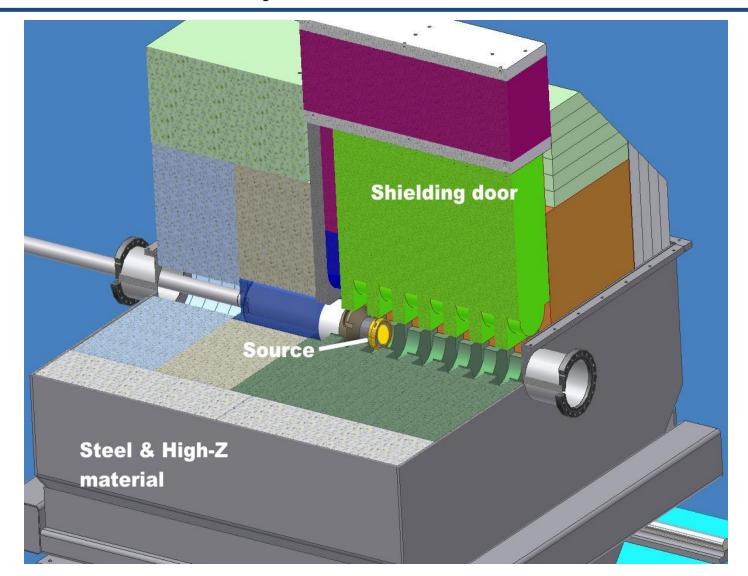
- Cf-252 source strength less than Hazard Category 3 nuclear facility limit of 3 Ci (DOE STD-1027)
  - CARIBU: 1 Ci source with DOE-approved Safety Analysis Document (SAD) safety envelope of 2 Ci
- Dose to nearest member of public off the site less than 0.1 mrem/year from airborne effluents
  - Radionuclide NESHAP (40 CFR 61 Subpart H) limit 10 mrem/year with continuous monitoring, 0.1 mrem/year without continuous monitoring
  - CARIBU: 1 Ci source offsite calculated dose 0.02 mrem/year based on 100 second hold-up time, 15 meter high stack, and no filtration
  - Continuous monitoring selected for operational control of effluents
  - Filtration added to remove any Cf-252 or fission fragment particulate and to remove volatile iodine

## **Limits Continued**

- Design exposure rate on high voltage platform less than 5 mrem/hour
  - Radiation Area between 5 mrem/ hour and 100 mrem/hour at 30 cm
    - Shielding cask with shield door for transfer of source to gas catcher
    - Gas catcher with gate valve to receive source •
- Design exposure rate outside high voltage platform interlocked area less than 0.5 mrem/hour
  - ALARA design requirement
- Design exposure rate for removal of stuck source from gas catcher less than 100 mrem/hour
  - High Radiation Area above 100 mrem/hour at 30 cm
    - Stuck source can be freed manually

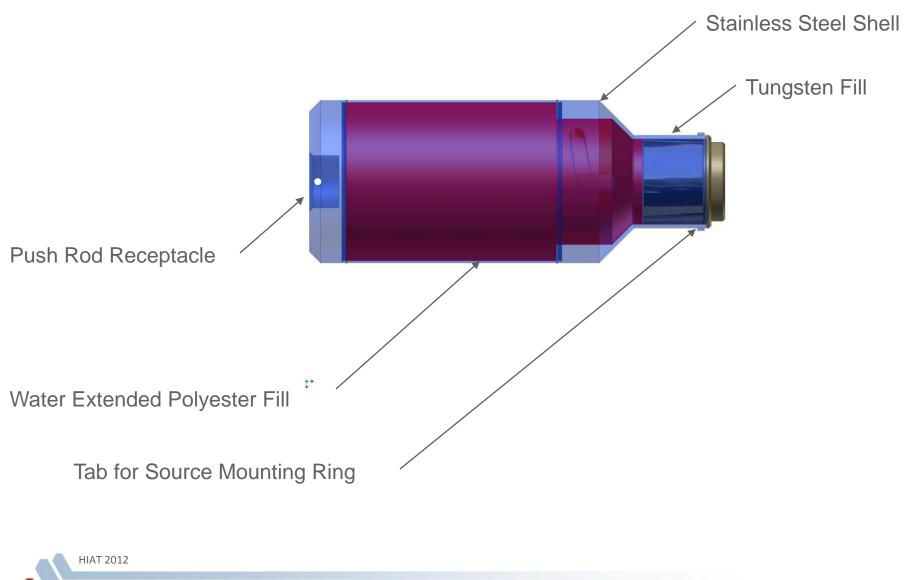


## CARIBU Cask Cutaway View



HIAT 2012

## Source Holder (Milk Jug)



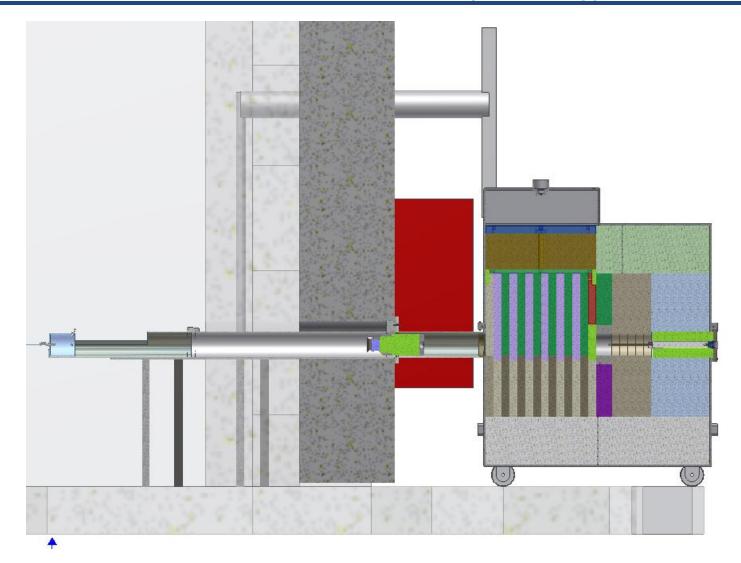
## Shielding Cask in CARIBU Room



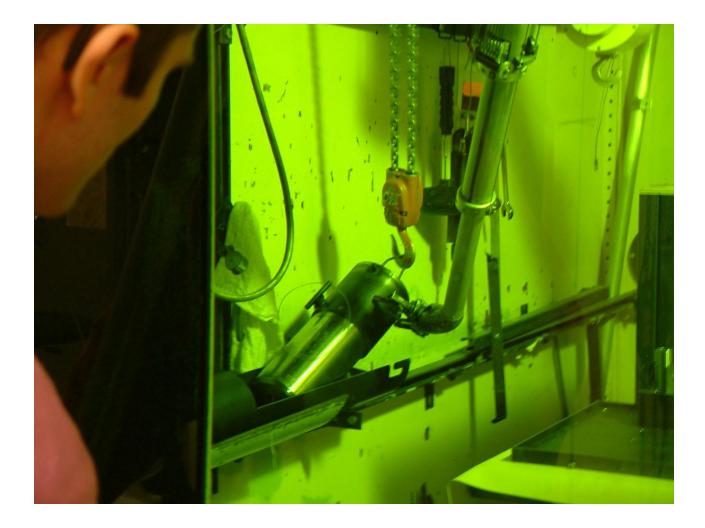
## Cask Positioned at Port in ANL Hot Cell Shielding Door



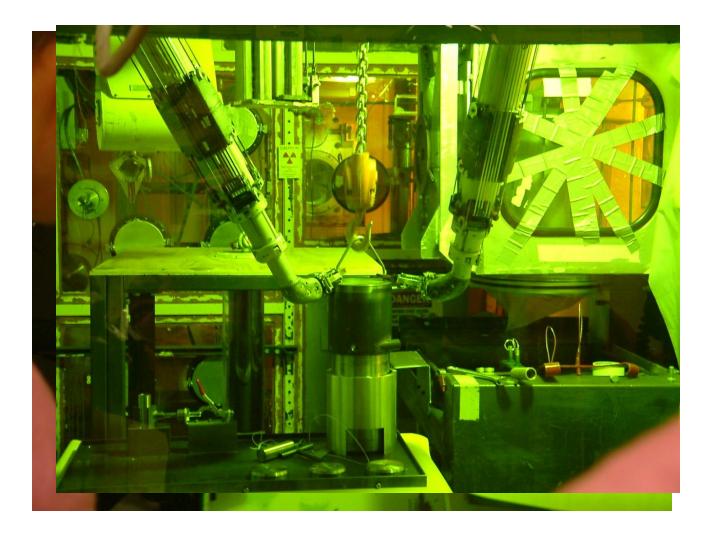
## Use of Port to Transfer Source Holder (Milk Jug)



## Lifting Milk Jug Locked in Trunion for Transfer to Tray



## Milk Jug Transferred to Tray



## **Airborne Effluent Control**

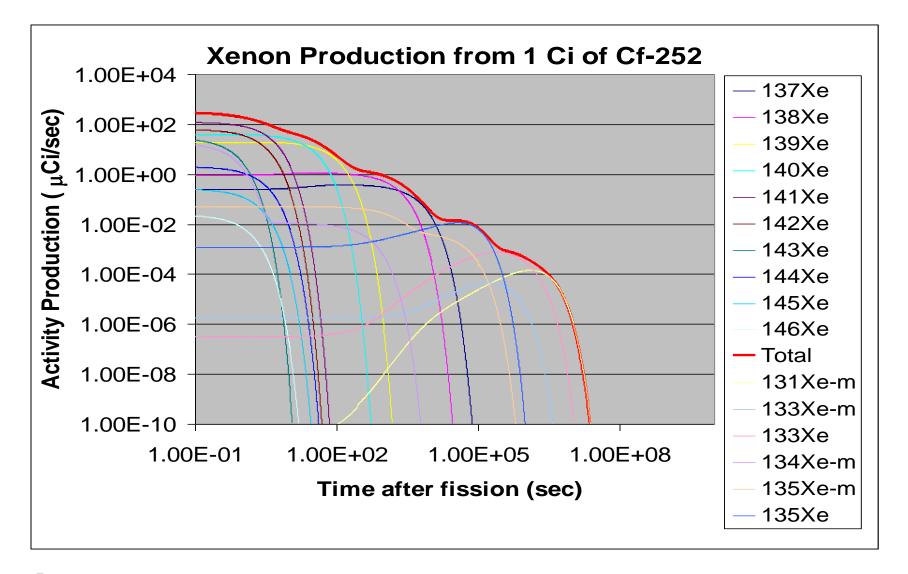
- Californium-252 Source
  - Fission fragment ions collected in gas catcher for acceleration
  - Fission fragment gases released from **15** meter high stack
- Design features for controlling effluents
  - Leak-tight cask for containment during transport and storage
  - Effluent hold-up of at least **100** sec for decay of short-lived radionuclides
  - Particulates and vapors collected using HEPA and charcoal filters
  - Dilution to reduce radionuclide concentration at nearby building air intakes

## **Airborne Effluents**

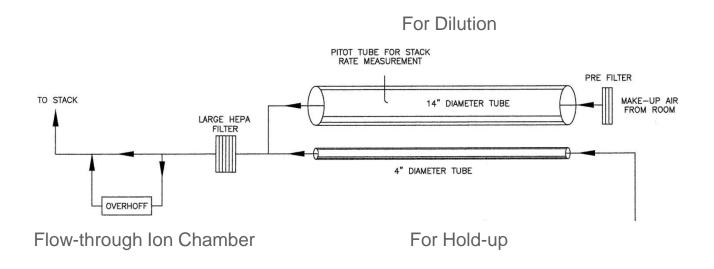
- Airborne effluents
  - Gaseous fission fragments
    - Xenon isotopes
  - Volatiles \_
    - Iodine isotopes
  - Particulates \_
    - Other fission fragments



#### **Noble Gas Production Continued**



## **Effluent Monitoring System**



Hold-up greater than 100 sec through 15 meters of 10 cm (4") diameter tube from the CARIBU cask and gas catcher Flow through cask and through gas catcher also lower than expected Dilution factor greater than 1000

| Radionuclide                  | Half-life<br>(days) | Activity 10 days<br>after effluent flow<br>ceased<br>(microcuries) | Activity 10 days<br>after effluent flow<br>ceased<br>(dpm) |
|-------------------------------|---------------------|--|--|
| I-131<br>(upstream filter)    | 8.04                | 0.0073   | 16,000   |
| I-131<br>(downstream filter)  | 8.04                | 0.00001  | 38   |
| Percentage on upstream filter |                     |  | 99.77%   |

**Conclusion: Carbon filter is very effective in removing iodine.** 

#### **Contamination Measurements**

- 2 mCi source
  - Approximately 1% of activity (50 million dpm alpha) in deposition rinse water, indicating some loose material left on source
  - Source plate smeared near source deposit
    - 50,000 dpm alpha
  - Source covered with thin aluminum foil
    - 300 dpm alpha in foil after 24 hours from Cf-252 recoils (knocked out by 100 MeV fission fragments)
- Scaling up to 100 mCi source
  - Contamination expected near source deposit
    - 2,500,000 dpm alpha expected (approximately 1 microcurie)
  - Source covered with thin aluminum foil
    - 15,000 dpm alpha expected in foil after 24 hours from Cf-252 recoils

#### **Operational Failure Modes**

- Flaking source
  - Cover foil contains any loose material as long as cover foil stays intact
  - Filters remove particulates and volatiles
  - Neutron detector alarms if californium reaches filter
- Leaking exhaust system
  - Noble gas release into CARIBU area produces immersion exposure which can be measured by interlocked detectors and provide warning through NARIS interlock system

## **NARIS Interlock System**

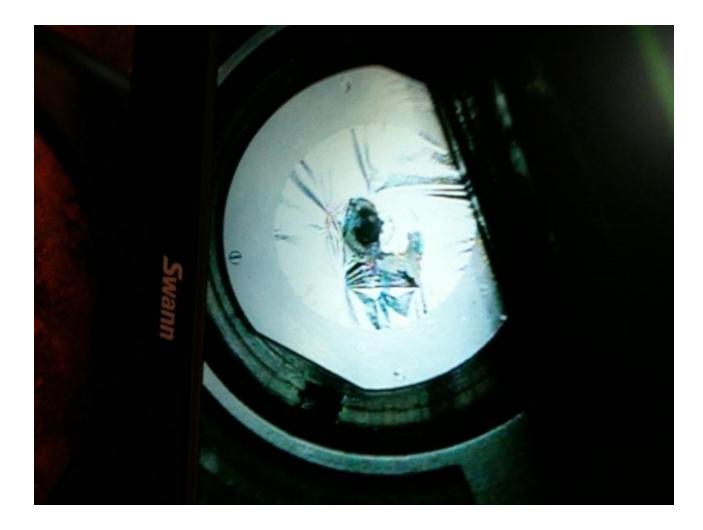
- New Atlas Radiation Interlock System (NARIS)
  - Independent of the existing Atlas Radiation Interlock System (ARIS)
  - Provides monitoring and alarm capabilities for CARIBU
  - Uses physical barriers also used for high voltage isolation
  - Optical fibers used to transmit signals from high voltage platform
- Radiation monitoring at CARIBU
  - Airborne effluent environmental release
    - Flow rate and radiation concentration measurements
    - Logging and alarms
  - Airborne release into CARIBU room
    - Immersion dose monitoring
    - Early warning alarm based on beta-particle detection
    - Monitors room pressure differential
  - Penetrating radiation in CARIBU room
    - Neutron detection from Cf-252 fission
    - Gamma detection from fission and fission fragment decay

## Experience

#### Cover foil failure

- Thin aluminum cover foil for 100 mCi source failed in less than one year
  - Failure from helium buildup?
  - Failure from striking foil with shield door?
- Most of removable radioactivity appears to adhere to cover foil
- Brief noble gas release when gas catcher gate valve was opened with slight overpressure in gas catcher
  - Beta activity in room air detected by thin window geiger counter which alarmed at very low exposure rate

## **Cover Foil Failure**



#### **Foil Failure Consequences**

- Contamination areas (10 CFR 835.2 definitions and Appendix D)
  - Contamination Area: Removable alpha contamination greater than 20 dpm/100 sq. cm.
  - High Contamination Area: Removable alpha contamination greater than
    2,000 dpm/100 sq. cm.
- Alpha contamination in transfer line
  - Before foil failure no alpha contamination was seen in the transfer line between the CARIBU cask and the gas catcher. After foil failure around 10,000 dpm was seen on wipes of those components.
- Alpha contamination at hot cell
  - Around 10,000 dpm (5 nCi) on tray after cover foil replacement
  - Several **mCi** of Cf-252 on cover foil piece sucked into local ventilation filter
- Degrader foil contamination
  - Degrader foil located between the cover foil and the gas catcher to optimize light or heavy fragments
  - Several mCi of Californium-252 on degrader foil after the failure based on neutron exposure rate (probably cover foil piece or pieces)

#### **Foil Failure Corrective Actions**

- Modifications in push rod following cover foil failure
  - Tubular pushrod connector to milk jug replaced by solid connector
  - Push rod shortened to increase distance between source and shield door
- Investigation of other cover foil materials
  - Studies of helium buildup from stopped alpha particles planned
- Improvement of contamination controls
  - Containment tent
  - Personnel protective equipment including respirators
  - Increased monitoring and alarm capabilities

## Summary

- Use of weaker sources
  - Working first with contact-handled source valuable
  - Commissioning with remote-handled source at 10% of final desired source strength valuable
- Effluent release
  - Hold-up sufficient
  - Filtration of iodine excellent
- Contamination issues
  - Cover foil failure needs solution
  - Contamination controls now more stringent