1200 New Jersey Ave., SE Washington, DC 20590



Pipeline and Hazardous Materials Safety Administration

NOV 26 2008

Mr. James M. Shuler Manager, Packaging Certification Program Safety Management and Operations Office of Environmental Management Department of Energy Washington, DC 20585

Ref. No. 08-0245

Dear Mr. Shuler:

This responds to your September 29, 2008 letter requesting clarification of Competent Authority approvals USA/0696/S-96 and USA/0695/S-96 issued by the U.S. Department of Transportation in accordance with the Hazardous Materials Regulations (HMR; 49 CFR Parts 171-180). Specifically, you ask if the competent authority approvals referenced above, are required to specifically list daughter products, which would be present during transportation. If so, you ask if it would be necessary to apply for a modification to the above mentioned approvals to specifically list the additional radionuclides in order for those Competent Authority approvals to be valid.

The answer is no. Competent Authority approvals issued in accordance with § 173.476 of the HMR require a detailed description of the contents of a special form capsule. However, the approvals do not have to identify daughter products that are part of the natural decay chain of the parent radionuclide. Therefore, it is not necessary to apply for a modification of the above referenced competent authority approvals to identify daughter products.

I hope this information is helpful. Please contact us if you require additional assistance.

Sincerely,

Charles Betts

Chief, Standards Development

Office of Hazardous Materials Standards



Department of Energy

Washington, DC 20585

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Mr. James Williams
Radioactive Materials Branch
Office of Hazardous Materials Technology
Pipeline and Hazardous Materials Safety Administration
U.S. Department of Transportation
East Building, E21-330, PHH-23
1200 New Jersey Avenue, S.E.
Washington, D.C. 20590-0001

Dear Mr. Williams:

As you know, the NA-21 Office of Global Threat Reduction's Off-Site Source Recovery Project (OSRP) at Los Alamos National Laboratory is a registered user of QSA Global, Inc. Model II and Model III Special Form Capsules under competent authority certificate numbers USA/0696/S-96 and USA/0695/S-96.

As a follow-up to their letter to the U.S. Department of Transportation (DOT) dated August 15, discussing approved radionuclide content in the two special form capsule models referenced above, an interpretation is requested to determine whether or not decay daughter products in equilibrium with the nuclides named in the above certificates must also be specifically listed on the certificates of competent authority.

Thank you for your assistance and cooperation. If you have any questions, please contact me or Justin Griffin in Los Alamos at 505-606-0362.

Best regards,

James M. Shuler

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Manager, Packaging Certification Program

Safety Management and Operations

Office of Environmental Management

cc:

Ioanna M. Iliopulos, NA-211 Abigail B. Cuthbertson, NA-211 Justin M. Griffin, LANL - N3: OSRP Julia Whitworth, LANL - NN



Off-Site Source Recovery Project N-3: International Threat Reduction PO Box 1663, Mail Stop J552 Los Alamos, New Mexico 87545 505-667-4711/Fax: 505-665-7913

Date: August 15, 2008 Refer To: N3: 08-096

Richard Boyle
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East Building, 2nd Floor, PHH-23
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SUBJECT: Special form capsules containing sealed sources with impurities and daughter nuclides

Dear Mr. Boyle:

This correspondence was developed through collaboration with Dr. James Shuler, Manager of the Packaging Certification Program for the U.S. Department of Energy. Since time is of the essence for resolution, Dr. Shuler has provided consent for us to contact the U.S. Department of Transportation (DOT) directly regarding the issue described herein.

As verbally advised by DOT, the Off-Site Source Recovery Project (OSRP) at Los Alamos National Laboratory will not transport QSA Global Model II or Model III Special Form Capsules under certificate numbers USA/0696/S-96 and USA/0695/S-96, respectively, for shipment of sealed sources containing ²³⁹Pu until resolution of concerns related to authorized content including impurities and daughter nuclides is achieved.

The purpose of this letter is to provide more detailed background information on the current situation, a brief chronology of events, a description of how estimation of sealed source content is calculated as requested, and a brief summary of impact to shippers of special form sources.

Background Information

To better understand the issue at hand, we must consider what we have found to be the initial cause of this misunderstanding. It is necessary to review the text in Item 3 of CoCA No. USA/0696/S-96 and No. USA/0695/S-96, which are provided below in Figures 1 and 2, respectively:

3. Radioactive Contents - The capsule described by this certificate is authorized to contain any one of the following radionuclides or the sole pair of radionuclides, in the chemical forms identified, and limited to the activity shown, in the table below. The radioactive material is limited to solid form in stainless steel capsules, between layers of non-radioactive stainless steel, or affixed to non-radioactive stainless steel by electroplating or other means. The maximum mass of the contents is limited to 2,500 grams.

Figure 1: QSA Global Model II Content Description (USA/0696/S-96)

3. Radioactive Contents - The capsule described by this certificate is authorized to contain any one of the following radionuclides or the sole pair of radionuclides, in the chemical forms identified, and limited to the activity shown, in the table below. The radioactive material is limited to solid form in stainless steel capsules, between layers of non-radioactive stainless steel, or affixed to non-radioactive stainless steel by electroplating or other means. The maximum mass of the contents is limited to 1,000 grams.

Figure 2: QSA Global Model III Content Description (USA/0695/S-96)

The particular wording in question for both cases above is the line, "...authorized to contain any one of the following <u>radionuclides</u>..." As users of these special form capsules and as individuals involved in the initial analysis of the Model II and Model III capsules for certification by QSA Global, we contend that the original intent of this verbiage was to limit contents to a unique type of sealed source based on its primary useful isotope* not just a pure/unique isotope. As you know, it is inherent in the production process of manufactured radionuclides and due to radioactive decay that other radioactive constituents (impurities) will be present.

As Item 3 is currently written in each certificate, it could inadvertently be interpreted to restrict capsule contents to one specific and pure radionuclide listed in the table below Item 3. However, since it is not possible to have a pure radionuclide (due to impurities and daughter products), the line should be read/interpreted as saying, "...authorized to contain any one of the following types of sealed sources (including radionuclide impurities and decay products)...," to fully encompass its original intent. OSRP will ask QSA Global to correct this phrasing in a future revision of these certificates.

A portion of the table below Item 3 in each of the special form certificates referenced above is copied as Figure 3 below. As you can see, for ²³⁹Pu/Be neutron sources (or simply ²³⁹Pu without beryllium), the maximum activity can be 100.0 Ci, but only a total of 350g ²³⁹Pu isotope is allowed.

Plutonium-239	3.7 TBq (100.0 Ci)	Oxide incorporated into a ceramic enamel, metal foil or metal plated to substrate (total 350 grams Pu-239 isotope)
Plutonium-239:Be	3.7 TBq {100.0 Ci}	Oxide mixed with beryllium powder or intermetallic pressed into a solid pellet (total 350 grams Pu-239 isotope)

Figure 3: Portion of CoCA Table Showing Plutonium Content Limits

This indicates that the limiting factor for content of 239 Pu isotope is 350g or about 22 Ci, while the overall maximum activity is 100 Ci. This suggests that the difference (100 - 22 \approx 78 Ci) will accommodate the radionuclide impurities and daughter products with higher specific activities.

Even though this table[†] does not specifically list other commonly known plutonium production impurities or daughter products of ²³⁹Pu, it is understood that all sealed sources contain such impurities and decay products. OSRP was not able to locate any special form certificates where radionuclide impurities present in a sealed source due to isotope production or evolution of daughter products were specifically identified in the certificate. Although all special form certificates

^{*} For example, ²⁴¹Am/Be sources only or ²³⁹Pu/Be sources only – while not allowing ²⁴¹Am/Be sources to be encapsulated along with ²³⁹Pu/Be sources.

[†] Only a portion of the content limit table is presented in Figure 3. Both certificates include several other isotopes as approved content; however, none of these include listings of impurities or daughter products either.

for radioactive materials are affected, of particular importance in terms of isotopic distribution within the source, are sealed sources containing ²³⁹Pu, ²³⁸Pu, ²⁵²Cf, and ²⁴¹Am.

Chronology of Events Leading Up to this Letter

Earlier this year OSRP visited Switzerland to encapsulate five unwanted, U.S.-origin ²³⁹Pu/Be sources into two different QSA Global Model II special form capsules in accordance with certificate USA/0696/S-96 (Rev 3) for threat reduction purposes. The Model II capsules were then packaged into two Type AF containers and prepared for interim shipment to Germany and subsequent repatriation to the United States. This work was performed under direction from the NA-21 Office of Global Threat Reduction, NNSA Office of Defense Nonproliferation.

The German competent authority, upon review of the encapsulation and packaging documentation, requested additional characterization information on each of the sources, with identification of both decay products and impurities. Radioanalysis is not performed on any sources by OSRP; so in order to meet this request, the German authorities were instead supplied with copies of in-house characterization modeling reports. These reports are generated by OSRP using average isotopic breakdown content based on known historical ²³⁹Pu production records, estimated impurities, and evolution of daughter products due to decay. This data is required (and has been accepted) for ultimate disposition of the sources at a transuranic waste disposal facility in the United States.

We suspect that this characterization report was used to document the other radionuclide contents of the capsules besides the primary useful constituent of the sealed sources. Using this additional information, the German competent authority took the position that the radioactive contents were not compliant with the description in the referenced certificate. Understandably, they refuse to accept the packaged sources, under Special Form Certificate USA/0696/S-96 (Rev 3), until they receive additional clarification from the original Swiss applicant.

Since OSRP does not have any regulatory authority over these matters, we advised the Swiss facility where the sources were packaged to directly contact the U.S. competent authority for clarification. As a result, the Swiss facility contacted Mr. James Williams via email to address the situation. Mr. Williams then contacted OSRP by phone to state that without additional information, he was in agreement with the German competent authorities and that OSRP should desist use and transport of QSA Global Model II and Model III Special Form Capsules (USA/0696/S-96 and USA/0695/S-96) containing ²³⁹Pu until the situation is resolved.

Estimation of Sealed Source Content

The OSRP in-house method[‡] used for estimation of all nuclides in a ²³⁹Pu source; and data in the characterization documents provided to Germany is based on modeling information on the isotopic content of ²³⁹Pu materials originally supplied by U.S. production reactors, such as Hanford, to source manufacturers.

Through prior research, records were identified that documented the transfer of ²³⁹Pu material to these source manufacturers (e.g., Mound, Monsanto) which were sufficiently complete to allow an estimation of the total gram quantities of each type of material supplied. The types of material and the characteristic isotopic make-up within these types of material is also documented thus allowing development of a weighted average for all ²³⁹Pu materials provided for the commercial manufacture of sealed sources containing ²³⁹Pu.

[‡] Tables showing EPA approved radionuclide breakdown for ²³⁹Pu ²³⁸Pu, and ²⁴¹Am used for OSRP in-house characterization for waste disposal is provided in Attachment 1.

With documents available showing the mass of materials used in the manufacture of a source, application of that weighted average (when corrected for decay) allows estimation of the isotopic content of ²³⁹Pu sources at a given time, such as upon disposal. This modeling result was fully accepted by the U.S. EPA for disposal of the unwanted sources at the designated waste disposal facility. However, the result is based on the entire documented dataset. No individual records exist which identify the exact type of material used or the isotopic breakdown present in a specific individual sealed source containing ²³⁹Pu; nonetheless, individual sealed sources are reasonably bounded by this modeling result.

Please note that OSRP shippers report content according to the 95% rule on the package label and shipping paper, which for ²³⁹Pu/Be sealed sources usually includes ²³⁹Pu, ²⁴¹Am, ²⁴⁰Pu, and ²⁴¹Pu. This is done for all shipments whether the ²³⁹Pu sources are in a Model II or III special form capsule, or if the sealed sources are left as normal form and shipped in a Type B Packaging.

Impact to Shippers of Special Form Sources

Most users and shippers of sealed sources do not realize that the special form source they ship contains radionuclide impurities and daughter products. They only consider the useful component (a.k.a., major constituent such as ²⁴¹Am) of the radioactive source when filling out shipping papers and labeling not realizing there are other nuclides in the source.

As previously stated, OSRP was not able to locate any special form certificates where radionuclide impurities present in a sealed source due to isotope production or evolution of daughter products were specifically identified in the special form certificate.

The impact of this recent verbal interpretation effectively questions the compliance of the following:

- OSRP source recovery actions via Type A shipments when compliance is based on the use of a QSA Global special form capsule under USA/0696/S-96 or USA/0695/S-96
- All shipments using any other special form certificate for sources known to contain impurities and decay products which are not specifically included as authorized content in the certificate
- Any shipments of sealed sources, including shipments of unwanted and at-risk sources, to
 the WIPP facility for final disposition when the special form certificate for the sources
 involved does not address any decay products and impurities

Conclusions

Impurities due to isotope production and evolution of daughter products are present in all special form and other radioactive sealed sources but are not commonly listed individually on the certifications.

In addition to affecting OSRP recovery of disused sealed sources, potentially including shipments of sources to WIPP for disposal, this question also implies that any sealed source transported under a special form certificate not specifically authorizing the presence of known impurities or decay products is non-compliant.

In our specific case, the wording in Item 3 of special form certificates USA/0696/S-96 and USA/0695/S-96 could inadvertently be interpreted to restrict capsule contents to one specific and pure radionuclide. As previously stated, the line should instead be read to say, "...authorized to contain any one of the following types of sealed sources (including radionuclide impurities and decay products)...," to fully encompass its original intent. OSRP will ask QSA Global to correct this

phrase in a future revision of these certificates; however, in the meantime we request your office provide efficient evaluation and resolution of this issue with these certificates.

If you have any questions or require additional information pertinent to this issue, please contact me at 505-606-0362 or send email to – jgriffin@lanl.gov.

Thank yo

ustin M. Griffin, P.

JMG

Attachments: Tables of Sample Approximations for Radionuclide Distribution in 239Pu, 238Pu, and

²⁴¹Am sealed sources.

Cy: James Shuler, U.S. Department of Energy, EM-60

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Ioanna M. Iliopulos, NA-211 Abigail B. Cuthbertson, NA-211

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Julia Whitworth, NN, E521

Lori Podolak, QSA Global

N-3 File, E541

OSRP File, J552

Attachment 1

By no means are the following tables meant to imply that all 239Pu, 238Pu, and 241Am sealed sources contain the exact radionuclide distributions shown. Actual content of individual sealed sources will vary.

Nuclide	Mass fraction of Plutonium	Grams of Nuclide per Gram of Plutonium	Specific Activity (Ci/g)	Curies of Nuclide per Gram of Plutonium
Pu-238	1.479E-04	1.48E-04	1.73E+01	2.56E-03
Pu-239	9.321E-01	9.32E-01	6.29E-02	5.86E-02
Pu-240	6.503E-02	6.50E-02	2.30E-01	1.50E-02
Pu-241	2.435E-03	2.44E-03	1.04E+02	2.53E-01
Pu-242	3.269E-04	3.27E-04	3.97E-03	1.30E-06
Am-241	2.500E-04	2.50E-04	3.47E+00	8.68E-04
U-234	3.403E-09	3.40E-09	6.32E-03	2.15E-11
U-235	4.254E-07	4.25E-07	2.19E-06	9.32E-13
U-238	1.697E-04	1.70E-04	3.40E-07	5.77E-11
Cs-137	2.313E-07	2.31E-07	8.80E+01	2.04E-05
Sr-90	1.035E-07	1.04E-07	1.38E+02	1.43E-05

Table 1: Sample Approximation of Radionuclide Distribution in Pu-239 Sources

Note: Plutonium created by neutron activation of ²³⁸U never occurs in an isotopically pure form. The isotopic impurities for plutonium range from mass numbers 238 to 242. Activities of these Pu isotopes, plus ²⁴¹Am, must be evaluated by specific material type and decayed to date. The same concept also applies to other radioisotopes.

Nuclide	Mass fraction of Plutonium (g of Isotope per g of Plutonium)	1	Curie of Isotope per Gram of Plutonium Metal	Grams of	Curies of Isotope per Gram of Pu- 238 Content	Curies of Isotope per Curie of Pu- 238 Content	Grams of Isotope per Curie of Pu- 238
Pu-238	8.03E-01	17.3	1.39E+01	1.00E+00	1.73E+01	1.00E+00	5.78E-02
Pu-239	1.61E-01	0.0629	1.01E-02	2.01E-01	1.26E-02	7.29E-04	1.16E-02
Pu-240	2.63E-02	0.23	6.06E-03	3.28E-02	7.54E-03	4.36E-04	1.89E-03
Pu-241	6.90E-03	104	7.17E-01	8.59E-03	8.93E-01	5.16E-02	4.96E-04
Pu-242	2.33E-03	0.00397	9,25E-06	2.90E-03	1.15E-05	6.66E-0	1.68E-04
Am-241	2.84E-04	3.47E+00	9.84E-04	3.53E-04	1.23E-03	7.08E-0	2.04E-05
U-234	3.40E-09	6.32E-03	2.15E-11	4.24E-09	2.68E-11	1.55E-1	2 2.45E-10
U-235	4.25E-07	2.19E-06	9.32E-13	5.30E-07	1.16E-12	6.70E-14	3.06E-08
U-238	1.70E-04	3.40E-07	5.77E-11	_2.11E-04	7.18E-11	4.15E-1	1.22E-05
Cs-137	2.31E-07	8.80E+01	2.04E-05	2.88E-07	2.53E-05	1.46E-0	
Sr-90	1.04E-07	1.38E+02	1.43E-05	1.29E-07	1.78E-05	1.03E-0	

Table 2: Sample Approximation of Radionuclide Distribution in Pu-238 Sources

Attachment 1

Nuclide			isotope per gram of Am-	Curies of Isotope per Gram of Am- 241	Curies of Isotope per Curie of Am- 241	Grams of Isotope per Curie of Pu- 238	Activity per Gram of source material
Am-241	9.97E-01	3.47	1.00E+00	3.47E+00	1.00E+00	2.88E-01	3.46E+00
Pu-238	6.10E-04	17.3	6.12E-04	1.06E-02	3.05E-03	1.76E-04	Figure 1991 Process
Pu-239	8.19E-01	0.0629	8.22E-01	5.17E-02	1,49E-02	2.37E-01	27 La Esta 1 1 1 1 1 2 2 2
Pu-240	1.65E-01	0.23	1.66E-01	3.81E-02	1.10E-02		6.16-7
Pu-241	1.18E-02	104	1.18E-02	1.23E+00	3,55E-01	3.41E-03	1.74"%[[2.1"], 34 1.
Pu-242	3.55E-03	0.00397	3.56E-03	1.41E-05	4.08E-06	1.03E-03	*** *1 to m.
U-234	3.40E-09	6.32E-03	3.41E-09	2.16E-11	6.22E-12	9.84E-10	2.15E-11
U-235	4.25E-07	2.19E-06	4.27E-07	9.35E-13	2.69E-13	1.23E-07	9.32E-13
U-238	1.70E-04	3.40E-07	1.70E-04	5.79E-11	1.67E-11	4.91E-0	45 4 GENERAL S
Cs-137	7.79E-10	8.80E+01	7.82E-10	6.88E-08	1.98E-08	2.25E-10	The second of the second
Sr-90	3.49E-10	1.38E+02	3.50E-10	4.83E-08	1.39E-08		1 1 Part 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1

Table 3: Sample Approximation of Radionuclide Distribution in Am-241 Sources