## LSNReviews

**From:** natalex [natalex@telus.net]
**Sent:** Thursday, September 13, 2007 10:42 PM
**To:** SWRI; povetkooleg@hotmail.com
**Subject:** SN radiolysis
**Attachments:** SN radiolysis August.doc

SN radiolysis

Alexei

Return-path: <natalex@telus.net>
Received: from mail.cnwra.swri.edu ([129.162.205.6])
by rogain.cnwra.swri.edu (Sun ONE Messaging Server 6.0 (built Oct 29 2003))
with ESMTP id <0JOC00CT27H29720@rogain.cnwra.swri.edu> for
opovetko@cnwra.swri.edu; Thu, 13 Sep 2007 21:41:26 -0500 (CDT)
Received: from virus89-out.ccf.swri.edu
(virus89-out.ccf.swri.edu [129.162.252.34])
by mail.cnwra.swri.edu (Switch-2.2.6/Switch-2.2.6) with ESMTP id l8E3gkl03021
for <opovetko@mail.cnwra.swri.edu>; Thu, 13 Sep 2007 22:42:46 -0500 (CDT)
Received: from virus256-ag.ccf.swri.edu (localhost [127.0.0.1])
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(8.13.1/8.13.1) with ESMTP id l8E2fKVa017450 for
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by bams.ccf.swri.edu (iPlanet Messaging Server 5.2 HotFix 1.17 (built Jun 23
2003)) with ESMTP id <0JOC00KVP7GVVT@bams.ccf.swri.edu> for
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by priv-edmwes34.telusplanet.net
(InterMail vM.7.08.02.00 201-2186-121-20061213)
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13 Sep 2007 20:40:50 -0600
Received: from radprot (unknown [204.191.232.23])
by priv-edmwaa05.telusplanet.net (BorderWare MXtreme Infinity Mail Firewall)
with ESMTP id BAWTKTCH72; Thu, 13 Sep 2007 20:40:49 -0600 (MDT)
Date: Thu, 13 Sep 2007 20:42:11 -0600
From: natalex <natalex@telus.net>
Subject: SN radiolysis
To: SWRI <oleg.povetko@swri.org>, povetkooleg@hotmail.com
Message-id: <20070914024049.BAWTKTCH72@priv-edmwaa05.telusplanet.net>
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Original-recipient: rfc822;opovetko@rogain.cnwra.swri.edu

#08/22/2007
Examined literature on the neutron and gamma radiolysis:
"Radiolytic Specie Generation from Internal Waste Package Criticality, CAL-EBS-NU-000017 REV 00",

- Химические изменения, которые претерпевают некоторые материалы при облучении, известное как **радиолиз** или **радиолитическое разложение**, наблюдается, когда такие материалы, как пластмасса или резина, подвергаются **облучению**, что **приводит к образованию ряда новых химических соединений**.
- **Радиолиз** также вызывает усиленное выделение некоторых химических веществ, содержащихся в материалах отходов.
- Радиолиз воды и органических соединений вызывает образование водорода, а также других воспламеняющихся или токсичных химических веществ. Такое радиолитическое разложение является одним из основных источников опасности возникновения пожаров и/или взрывов в некоторых емкостях с высокоактивными отходами на предприятиях Хэнфорда и Саванна-Ривер-Сайт.
- От образования и накопления опасных химических веществ в результате радиолиза пострадали хранилище с плутонием в Роки-Флэтс и хранилища с ТРУ-отходами на ряде других объектов.
- Одной из причин было разложение изделий из пластмассы под действием радиолиза на воспламеняющиеся и токсичные газы.
- Фактически, радиолиз может сделать отходы со временем более опасными или сделать опасными отходы, первоначально неопасные.

- Radiation effects on the corrosion of metals and alloys include, among other things, radiolysis of the local gaseous and aqueous environment to produce both oxidizing and reducing radicals.

- Radiolysis processes in moist air environments lead to the fixation of nitrogen as NO, $NO_2$, and especially $HNO_3$ (Reed and Van Konynenburg 1988, pp. 393-404).
- Nitric acid is assumed to be the principal corrosive radiolytic chemical specie and is produced in an irradiated air-water vapor ystem when the hydroxyl radicals generated from the water vapor convert nitrogen dioxides, hat are formed by the radiolytic reaction between nitrogen and oxygen, to nitric acids.
- Radiolysis producing a local depression of the pH resulting in localized corrosion of cladding terial is included in the localized corrosion model as a special feature, YMP FEP NO. .1.02.15.00 (CRWMS M&O 2000d, Section 6.2.5).
- Zircaloy has excellent corrosion resistance to nitric acid and hydrogen peroxide, the concentration of these species can be enhanced by radiolysis during n internal WP criticality, potentially accelerating the corrosion effects in the cladding material.

The purpose of this calculation is to provide a detailed calculation the potential for generation of radiolytic species during a **postulated static criticality event in a WP**.

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Radiolytic production of particular chemical species depends upon

- the radiation environment,
- the chemical components present, and
- the physical environment where the radiolytic reactions are occurring.

The yield of any given chemical species is characterized by a single parameter, "**G**", identified as the **G-factor** (Reed and Van Konynenburg 1991, pp. 1396-1403).

***Definition:***
The **"G" value** represents the number of molecules of a chemical species produced per **100 eV** of absorbed radiation energy in the volume containing the irradiated environment.

Measurements of the **"G" factor** for production of **nitrogen dioxide** (one-to-one production ratio for nitric acid) from mixed neutron-gamma radiation range from approximately **0.5 to 2.5** molecules/100 eV of absorbed energy (Reed and Van Konynenburg 1991, p. 1399).

**Assumption:**

1. 21-pressurized water reactor (PWR) WP, containing commercial spent nuclear fuel (CSNF) assemblies, was assumed to have failed and subsequently partially filled with water
2. The steel basket structure was assumed to have fully degraded with the degradation products settling to the bottom of the WP
3. Hematite ($Fe_2O_3$) is assumed to be the only ironbearing degradation product formed from the original basket material (Assumption 3.1)
4. In a separate suite of evaluations, the contribution to the degradation product volume from diaspore generated by oxidized aluminum from the thermal shunt plates is also considered.
5. The packing fraction of the hematite, or the hematite-diaspore mixture, was assumed to be 0.58 (Assumption 3.2), with the remaining space filled with water.
6. For evaluations involving mixtures, complete reaction of the Fe and Al in the donating structures provides a mole fraction of 0.8439 (mass fraction = **0.9350**) for the hematite in the degradation product mixture material.
7. Degradation products were assumed to be present outside the fuel pins in assemblies below the degradation product-water mixture level, but not within the guide tube and instrument tube spaces of those assemblies.
8. The water level above the degradation product-water mixture was assumed to extend sufficiently high to maintain criticality, leaving an air-water vapor space at the top of the WP.
9. The radiant energy deposition in the air-water vapor space was calculated with the MCNP code (Briesmeister, 1997) using the KCODE option and tracking the transport of both neutron and gamma particles.
10. The gamma interactions include photon and electron processes leading to dissociation of the gas molecules and generation of nitric acids in the air-water vapor space.

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**Method**

A series of these tallies have been specified in the MCNP input decks to obtain estimates for the following physical quantities:

1. Total, neutron, and gamma energy depositions, in MeV, in the moist air regions of the waste package
2. Average energy released per fission for the waste package
3. Average number of neutrons released per fission for the waste package
4. $k_{eff}$ for each of the SNF regions: the fuel pins surrounded by degradation products (lower region), the fuel pins surrounded by water (middle region), and the fuel pins surrounded by moist air (top region) (see Figure 5.4 for region definition).

Information is collected for both gamma and neutron events using "f6" and "f4" tally types

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## 3. ASSUMPTIONS

3.1 It is assumed that the steel in the basket assembly and fuel assembly end fittings is fully degraded. Hematite ($Fe_2O_3$) and Diaspore (ALO(OH)) are assumed to be the only degradation products remaining from the steel internals. The rational for this assumption is that these minerals have a very low solubility whereas other degradation products with higher solubilities are more likely to be transported out of the WP. This minimizes the amount of neutron absorber materials in the WP which is conservative. This assumption is used in Sections 2 and 5.

3.2 It is assumed that the porosity of packed particles resulting from degradation of the steel and aluminum internal structure of a 21 PWR WP is 42%. The rationale for this assumption is that measurements of the porosity of compacted granular materials (sand) was limited to approximately 42% before onset of container distortion (CRWMS M&O 1998b, p. 15). This assumption is used in Sections 2, 5, and 6.

3.3 It is assumed that the "G" factor for radiolytic production of nitric acid has the same value for neutron radiation as for gamma radiation. The rationale for this assumption is that radiolytic specie production is proportional to the absorbed energy rather than the effective dose. This assumption is used in Sections 2, 5, and 6.

3.4 It is assumed that the spacing between fuel assemblies in an asymmetric arrangement (resting on the WP) is 0.25 cm. The rationale for this assumption is that degradation products from the basket structure remaining between assemblies will prevent direct contact between assemblies This assumption is used in Sections 5 and 6.

3.5 It is assumed that the stainless steel inner shell of the WP is not degraded. The rationale for this assumption is that it is conservative. Degradation products from the WP shell would increase the total volume of the hematite in the WP, thus decreasing the moist air space available for radiolytic reactions. This assumption is used in Sections 5 and 6.

3.6 It is assumed that the Babcock and Wilcox (B&W) Mark B 15x15 fuel design used for this calculation is representative of the fuel types anticipated for potential disposition in the MGR. The basis for this assumption is this assembly type has been used for WP source term (CRWMS M&O 1999a, Section 3) and radiolysis calculations (BSC 2001b, Section 5.2). This assumption is used in Section 5.

3.7 It is assumed that the instrument tube in a B&W Mark B fuel assembly is the same length as the fuel pins. The rationale for this assumption is that it is conservative allowing slightly more moderator within the assemblies immersed in the degradation products. This assumption is used in Section 5.

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