

HIGH INTEREST TOPIC AND QUESTIONNAIRE PWR ALARA Association Key West, FL January 20-22, 2015

What is Your site's Dollars per Person Rem Seved for Englineern Modes. Topic: NSSS Plant Contact (Name) 2LW Ginna 2LW Kewaunee **Point** 2LW Beach 1,2 Prairie 2LW Island 1,2 2LW Ringhals 3LW 2,3,4 Beaver 3LW Valley 1,2 3LW Farley 1,2 3LW Harris North 3LW Anna 1,2 INTO Quantite 4 x 20,000 = 80,000 3LW Robinson 3LW Surry 1,2 Turkey 3LW Point 1,2 VC 3LW Summer Braidwood 4LW 1,2 4LW Byron 1,2 4LW Callaway Catawba 4LW 1,2 \$ 20K Comanche 4LW Peak 1,2 4LW Cook 1,2 Diablo 4LW Canyon 1,2 Indian 4LW Point 2,3 McGuire 4LW 1,2 4LW Salem 1,2

2013 ISOE Information Sheet on U.S. \$/Person-Rem Saved Values by NPP Site

2015

	7010	
Dresden		\$60,000
LaSalle	901000000000000000000000000000000000000	\$60,000
Limerick		\$60,000
Wolf Creek		\$40,000
Byron		\$40,000
D. C. Cook	32,632	\$32,632
Crystal River		\$30,000
South Texas Project		\$30,000
Calvert Cliffs		\$25,000
WNP-2		\$25,000
Sequoyah		\$25,000
Watts Bar		\$25,000
Ginna		\$25,000
Farley		\$25,000
Browns Ferry		\$25,000
Surry		\$25,000
Callaway		\$25,000
Conn. Yankee (Haddem Neck)		\$20,000
Nine Mile Point		\$20,000
Vogtle		\$20,000
Palo Verde		\$20,000
Hatch		\$20,000
Susquehanna		\$20,000
Kewaunee		\$20,000
Clinton		\$20,000
Oyster Creek		\$20,000
San Onofre		\$20,000
Prarie Island	ir.	\$15,000
Vermont Yankee		\$15,000
		\$14,000
Fort Calhoun		\$12,500
Millstone		\$12,500
McGuire Catawba		\$12,500
		\$12,500
North Anna		\$12,500
Oconee Characteria		\$12,500
Shearon Harris		\$12,000
Monticello		\$12,000
Cooper		\$10,000
Brunswick		\$10,000
Comanche Peak		\$10,000
Diablo Canyon		\$10,000
Duane Arnold		
Fermi 2		\$10,000 \$10,000
H. B. Robinson		
Hope Creek		\$10,000
Maine Yankee		\$10,000
Point Beach		\$10,000
PSE&G		\$10,000
Salem		\$10,000

2013 ISOE Information Sheet on U.S. \$/Person-Rem Saved Values by NPP Site

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\$10,000	\$10,000	\$10,000	\$20,000	\$20,000	\$20,000
\$9,000	\$8,000	\$8,000	\$8,000	\$20,000	\$20,000
	\$18,000	\$18,000	\$18,000	\$18,000	\$32,632
\$25,000	\$25,000	\$25,000	\$25,000	\$15,000	\$30,000
\$10,000	\$15,000	\$15,000	\$15,000	\$15,000	\$25,000
		\$15,000	\$15,000	\$15,000	\$15,000
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2013 ISOE Information Sheet on U.S. \$/Person-Rem Saved Values by NPP Site

Seabrook	\$10,000	
St. Lucie	\$10,000	
Turkey Point	\$10,000	
V. C. Summer	\$6,000	
Fitzpatrick		*Note: Site no longer has a \$/p-rem value per advice from legal department
Perry		Currently Under Review
Palisades		*Note: Site no longer has a \$/p-rem value per advice from legal department
River Bend		*Note: Site no longer has a \$/p-rem value per advice from legal department
Zion		Under Decommisioning
Beaver Valley		Currently Under Review
Arkansas Nuclear One		*Note: Site no longer has a \$/p-rem value per advice from legal department
Davis Besse		Currently Under Review
Grand Gulf		*Note: Site no longer has a \$/p-rem value per advice from legal department
Indian Point 2		*Note: Site no longer has a \$/p-rem value per advice from legal department
Pilgrim		*Note: Site no longer has a \$/p-rem value per advice from legal department
Indian Point 3		*Note: Site no longer has a \$/p-rem value per advice from legal department
Waterford		*Note: Site no longer has a \$/p-rem value per advice from legal department
Average	\$23,976	
Median	\$20,000	

NEW 2013Draft Table:

dollarem.xls

2014 Values Requested	2013	2010 ref.
Seabrook		\$10,000
St. Lucie		\$10,000
Turkey Point		\$10,000
V. C. Summer		\$6,000
Fitzpatrick		
Perry		
Palisades		
River Bend		
Zion		
Beaver Valley		
Arkansas Nuclear One		
Davis Besse		
Grand Gulf		
Indian Point 2		
Pilgrim		
Indian Point 3		
Waterford		
Average		\$23,976
Median		\$20,000
updated May 2013		TBD

HIGH INTEREST TOPIC AND QUESTIONNAIRE ALARA Association Key West, FL January 20-22, 2015

P	WR ALARA	Associa	allon Ry West to formation on 11,02
Topic: A	re you	whereg	sted in Received Information on 11,02 sted in Received Information on 11,02 Cavity decon? Including ISOE/ERED Export Comments Group Report Att
	Addition	NSSS	Comments Grosp Report 1474
Contact (Name)	Plant	CCCN	
	Ginna	2LW	
	Kewaunee	2LW	
	Point Beach 1,2	2LW	
	Prairie Island 1,2	2LW	
	Ringhals 2,3,4	2LW 3LW	
	Beaver Valley 1,2	3LW	
	Farley 1,2	3LW	
	Harris	3LW	
RMONUTT	North Anna 1,2	3LW	Yes
	Robinson	3LW	
	Surry 1,2	3LW	
	Turkey Point 1,2	3LW	
	VC Summer	3LW	
	Braidwood 1,2	4LW	
	Byron 1,2	4LW	
	Callaway	4LW	
Darcy Campbe Dana Page	Catawba 1,2	4LW	Yes Yes
	Comanche Peak 1,2	4LW	
David Miller	Cook 1,2	4LW	
	Diablo Canyon 1,2	4LW	
	Indian Point 2,3	4LW	
S. Lisi	McGuire 1,2	4LW	NO Alkady Doing it.
S. Lisi Gens TOFF	Salem 1,2	4LW	Yes

HIGH INTEREST TOPIC AND QUESTIONNAIRE

PWR ALARA Association Key West, FL January 20-22, 2015 Topic: Contact (Name) NSSS Plant **Comments** Seabrook 4LW Sequoyah 4LW 1,2 Sizewell B 4LW South 4LW Texas 1,2 Vogtle 1,2 4LW 405 Watts Bar 4LW Wolf 4LW Creek Millstone 4LW, 3,2 CE Calvert CE Cliffs Ft. CE Calhoun **Palisades** CE J. Gyge S. Williams Yes Palo Verde CE 1,2,3 San Onofre CE 2,3 St.Lucie CE 1,2 Waterford CE CE, ANO 2,1 B&W Crystal B&W River Davis B&W Besse Oconee B&W 1,2,3 TMI B&W Areva VFJ EDF Westinghouse

David Miller Cook RP

Radiological Protection

RADIATION PROTECTION ASPECTS OF PRIMARY WATER CHEMISTRY AND SOURCE-TERM MANAGEMENT

(DRAFT) VERSION 89

This version includes comments and inputs received after February 2013 meeting.

		Implemented
Gilles Ranchoux	14 March	✓
Ivan Smieško	26 March	✓
David E. Miller (inputs for CANDUs)	26 June	✓

NATE - Cook EPRI -EDF -

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ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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Cover photos:

Comment [HBO1]: Any suggestions for the cover photo(s).

FOREWORD

Since 1992, the Information System on Occupational Exposure (ISOE) has provided a forum for radiological protection professionals from nuclear power utilities and national regulatory authorities worldwide to discuss, promote and co-ordinate international co-operative undertakings for the radiological protection of workers at nuclear power plants. The ISOE objective is to improve occupational exposure management at nuclear power plants by exchanging relevant information, data and experience on methods to optimise occupational radiation protection.

At its meeting in November 2010, ISOE Management Board discussed a new proposal on radiation protects aspects of primary system water chemistry and source-term management. It was indicated that there are many approaches to water chemistry in nuclear power plants with very various results and consequences in terms of radiation protection performance. As such, it was suggested that radiation protection aspects of primary system water chemistry and source-term management should be discussed by an ISOE ad-hoc expert group. The Group is expected to address the experience of various ISOE utilities with various water chemistry regimes to see if experience exchange could help to improve radiation protection performances. Members of the Management Board also noted that water chemistry should not be viewed only from the context of radiation protection issues, and it was proposed to be grouped into a few of the most commonly used water chemistry approaches (e.g. zinc injection, pH control, iron injection, hydrogen water chemistry, etc.) to focus the exchange of experience discussions. For each approaches, it is expected to identify how radiation protection benefits are evaluated with a focus on measurement techniques such as CZT gamma spectroscopy.

The ISOE Management Board welcomed the proposal and decided that the Working Group on Data Analysis (WGDA) should take the lead in managing the work of this group. The Management Board also agreed that the ISOE Technical Centres should participate actively in this body of work, and that the Group should discuss its work with the CRPPH Expert Group on Occupational Exposure (EGOE), as appropriate to build on its experience. It was noted that this activity would benefit from a broad ISOE participation to ensure that the final product would be cohesive and valuable. Thus, it was requested that a call for nominations be sent by the Secretariat to the full Management Board. Following this direction, a call for nominations to the newly established ad-hoc expert group was sent to the ISOE membership in January 2011.

This report reflects the current state of knowledge, technology and experience on primary water chemistry and source-term management issues directly related with radiation protection.

ISOE Network: www.isoe-network.net

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Comment [RDCN2]: This section will be extended by David (EPRI). Referencing cobalt reduction as well.

Consider to move the second paragraph to the next

Comment [HBO3]: Draft by Daniel & David

Note, at the July meeting David Miller committed David Miller and David Doty to building this

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Appendix 1: Typical Filmary Materials for FWKS

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Appendix 4: ISOE Expert Group on Water Chemistry and Source-Term Management

References

List of Acronyms

AAS Aqua-Aerobic System

ALARA As Low as Reasonably Achievable

ANDRA Agence nationale pour la gestion des déchets radioactifs, National Agency for

Radioactive Waste Management

AOA Axial Offset Anomaly BOC Beginning of Cycle

BRAC BWR Radiation Level Assessment and Control

BWR Boiling Water Reactor

CANDU Canada Deuterium Uranium Reactor

CEPN Centre d'étude sur l'Evaluation de la Protection dans le domaine Nucléaire, Nuclear

Protection Evaluation Centre

CIPS Crud Induced Power Shift

CRPPH Committee on Radiation Protection and Public Health

CRUD Chalk River Unidentified Deposits
CVCS Chemical and Volume Control System

CZT Cadmium-Zinc-Tellurium

DCD Dilute Chemical Decontamination

DF Decontamination Factor

DPNGS Douglas Point Nuclear Generating Station

ECP Electrochemical Potential ED Electronic Dosimeter

EDF Electricité de France, French Electricity Utility

EFPY Effective Full Power Year

EGOE Expert Group on Occupational Exposure

EMECC Ensemble de Mesure et d'Etude de la Contamination des Circuits, Assembly of

Measuring and Study of Circuit Contamination

EOC End of Cycle EP Electropolishing

EPRI Electric Power Research Institute FSD Full System Decontamination

HFT Hot Functional Test
HIC High Integrity Container
HTS Heat Transport System
HWC Hydrogen Water Chemistry

 IASCC
 Irradiation Assisted Stress Corrosion Cracking

 IGSCC
 Intergranular Stress Corrosion Cracking

 ISOE
 Information System on Occupational Exposure

 MADAC
 Mobile Analyser for the Detection of Activity in Crud

NDE Non-destructive Evaluation
NIVDS Nuclear Island Drain/Vent System
NPD Nuclear Power Demonstration
NPP Nuclear Power Plant
NSSS Nuclear Steam Supply Systems

NWC Normal Water Chemistry
PHWR Pressurized Heavy Water Reactor

PMUC Products and Materials used in Power Plants

PWR Pressurized Water Reactor

PWSCC Primary Water Stress-Corrosion Cracking

RB Reactor Building

Reaktor Bolshoy Moshchnosti Kanalniy, High Power Channel-type Reactor **RBMK**

RCA Reactor Containment Area

RCPB Reactor Coolant Pressure Boundary

RCS Reactor Coolant System

RHRS Residual Heat Removal System **RWCU** Reactor Water Cleanup System RTD Resistance Temperature Detector SCC Stress Corrosion Cracking SCrP Stabilized Chromium Process

SG Steam Generator

SRMP Standard Radiation Monitoring Program TCLP Toxic Characteristic Leach Procedure TLD Thermo-Luminescent Dosimeter

TTThermally Treated VCT Volume Control Tank

VVER Vodo-Vodyanoi Energetichesky Reactor, Water-Water Energetic Reactor

WGDA Working Group on Data Analysis

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Executive Summary To be included

Comment [HBO4]: Draft by Ludovic (CEPN) by the finalization of the report

1. Introduction

During the 50 years of successful commercial nuclear power plant electrical generation, the importance of water chemistry management and radioactive source term reduction became key aspects of the sustainability and life-cycle management practices for the first and second generation reactors.

An example of the lessons learned in proper water chemistry management and plant component reliability is the failure of PWR steam generators in the 80's, 90's and beyond due to poor water chemistry regimes leading to tube failure and plugging. Replacement steam generators have precluded the degradation of the component based on improved water chemistry controls.

A wide range of annual dose values is observed in the current global fleet of operating PWRs. BWRs, CANDU and VVERs. The report details and explains the cause of water chemistry and source term good practices and lessons learned. Topics addressed include:

- 1. Water Chemistry Controls
- 2. Reactor Shutdown Protocols
- 3. Chemical Decon Experience
- 4. Source Term Removal
- 5. Instrumentation for Source Characterization
- 1. Informed owners and operators
- 2. Sound water chemistry
- 3. Understand materials and fuel limitations

Application of lessons learned and challenges related to source term reduction efforts

Source term is universal

Value of international information exchange

Value of multiple NSSS designs globally

In short, the ISOE expert group report focuses on globally informed life cycle plant management with the goal of asset preservation and low occupational dose and public dose management.

2. Scope

The publication primarily focuses on three topics dealing with water chemistry, source term management and remediation techniques. One key objective of the report is to provide current knowledge regarding these topics and to address clearly related radiation protection issues. In that mind, the report prepared by the EGWC was also reviewed by radiation protection experts. In order to address various designs, PWRs, VVERs and BWRs are addressed within the document. Additionally, available information addressing current operating units and lessons learnt is outlined with choices that have been made for the design of new plants.

Chapter 3 of this report addresses current practices regarding primary chemistry management for different designs, "how to limit activity in the primary circuit and to minimize contamination".

Comment [HBO5]: To be included by David Miller in collaboration with the Sec

Let's update before we leave today!

Updated for comments by the team

General information is provided regarding activation, corrosion and transport of activated materials in the primary circuit (background on radiation field generation). Primary chemistry aspects that are related to radiation field generation are addressed, such as material issues (steam generator, cobalt inventory, surface preconditioning and fuel assembly support structure material) and chemical methods (pH control, zinc injection, shut down and start-up operations and purification) are also addressed. Specific contamination with ¹¹⁰Ag or ¹²⁴Sb is also discussed.

Chapter 4 - radiation field measurement techniques - provides information regarding measurement techniques and mapping strategies (such as the EPRI methodology or the EDF RB index) that are used in order to precisely follow radiation field evolution within the RB and to detect abnormal elevation of dose rate. Routine measurements with common techniques such as routine dose rate meters are described as well as more complex techniques such as CZT detectors or germanium detector. Advantages and disadvantages of both techniques are discussed. In the follow up of the report, techniques for full system and component remediation are discussed with quantitative data sets "remediation of contamination". Experiences of various sites with respect to source term management are provided, addressing the topics previously discussed in the report in section titled as "radiation protection outcomes".

3. Introduction of Strategies and Techniques

3.1 Background on Radiation Field Generation

The reactor coolant chemistry is complex. It involves soluble and insoluble (colloidal and larger particulates) species in a forced-convective, non-isothermal system. Complex processes control the release of corrosion products to the coolant, resulting in the potential activation from the intense neutron field present in operating reactor cores. Corrosion products undergo a series of processes to reach ex-core surfaces producing the radiation field. These processes include release, transport to the fuel surface, deposition, activation, release from fuel surfaces, and the subsequent uptake on out-of-core surfaces. Through the process of generating energy and by exposing the released corrosion products to the neutron flux, a significant inventory of radioactive corrosion products is created over time, which in turn, can be transported and deposited on ex-core surfaces. This results in the build-up of radiation fields impacting worker dose.

There are essentially three types of activity that the plant chemists and radiation protection professionals / health physicists are concerned with; fission products, coolant activation products and activated corrosion products. Activated corrosion products can then be sorted into two additional groups; corrosion products deposited on fuel surfaces from out-of-core surface corrosion and highly activated corrosion products from fuel and reactor materials.

Section 3.1 covers the basics related to activity release and build-up on system surfaces. It is not intended to provide a detailed discussion of different NSSS designs. As an example, differing chemistry conditions are maintained in light water western style PWR, BWR and VVER designs; combined with the different materials of construction, unique situations for each design are created. These designs are discussed in later sections.

3.1.1 Fission Products

In order to understand the fission sources, one must consider the source of "fissile material". The primary fissile materials in light water reactors after initial start-up are ²³⁵U (natural and enriched), ²³⁹Pu (neutron capture with ²³⁸U), and ²⁴¹Pu (neutron capture with ²⁴⁰Pu). ²³⁵U and ²³⁹Pu isotopes are the primary drivers for nuclear fission while the others support the longer operating cycle supporting

continued fission. Other fissile materials may include 238 Np, 243 Pu, 241 Am, 242 Am, 244 Am and 242 Cm, 243 Cm, and 245 Cm.

Fresh PWR and BWR fuel consists of ~4% enriched ²³⁵U with a balance of ²³⁸U (~96%), ²³⁴U and ²³⁶U (less than 1%); PHWR fuel contains natural uranium. The ²³⁵U is the fissile material in new (fresh) fuel assemblies, while ²³⁹Pu and ²³³U are produced or converted to fissile material by neutron capture after start-up. These fissile materials are essential for the long-term operation of plants. Based on (G Friedlander, 1981), for approximately every 1 megawatt day of reactor operation, 1 gram of fissile material undergoes fission and approximately 0.5 grams of ²³⁹Pu is produced.

Equation 1

$$^{238}U(\eta,\gamma)^{239}U \rightarrow ^{239}Np \rightarrow ^{239}Pu$$

Actinides can create unique challenges for radiation protection personnel. The potential issues related to alpha contamination and system clean-up following fuel failures requires diligence and significant effort to minimize worker dose-related issues. In general, for plants operating without fuel failures, this is not an issue for normal refuelling outage operation, but should be considered in the overall source term discussion.

During the fission reaction, the heavy nucleus is generally divided into two unequal mass nuclei called "fission products" consistent with the equation (2).

Equation 2

$$_{z}^{A}X + \eta_{1}^{0} \rightarrow FF_{1} + FF_{2} + \upsilon + 2\eta_{1}^{0'} + \Delta Energy$$

It is estimated that $3x10^{10}$ fissions per second is required for every watt of power. Noting that during each fission, two fissions products are produced, one can easily show that the fission product inventory is the largest source of radionuclides in the primary system. With intact cladding, fission product release to the coolant is minimized, and coolant activation products and activated corrosion products dominate. If there are high levels of tramp material and/or fuel cladding leakage, the impact of fission products can be a significant contributor to the overall source term that radiation protection must consider in relation to worker dose. In the absence of tramp material and/or fuel cladding leakage, corrosion products, while representing a fraction of the overall inventory, dominate the source term when considering dose to workers.

3.1.2 Coolant activation products

Coolant activation products are those radionuclides come from water activation, impurities contained in water, or chemicals injected into the primary circuit. The principle activation products of concern in the coolant are identified in Table 1. In general, these species are not a concern for worker related dose, but they may be a concern related to effluents. Two exceptions include ⁴¹Ar early in the shutdown for plants injecting argon gas and ³H related to the PHWR design. ³H presents some unique challenges and radiation protection personnel should understand the impact related to dose.

Three radionuclides of concern (¹⁴C, ¹⁶N, and ¹⁸F) are briefly discussed below capturing some of the variables involved in coolant activation products. ¹⁸F is only discussed reflecting the dependence on core design.

 14 C provides some unique challenges due to its long half-life and impact on the environment. The primary production mechanism (outside of interactions within the fuel) is from the (η, α) interaction with 17 O, rather than the 14 N production mechanism, since the coolant is typically degassed and has a low dissolved nitrogen content. However, it should be noted that this is not the case for the VVER fleet injecting ammonia. The 13 N production mechanism plays more of a role in the VVER fleet.

The $^{16}\mathrm{O}$ contained within the water molecule (H₂O) interacts with fast neutrons in a (η, ρ) reaction forming $^{16}\mathrm{N}$. $^{16}\mathrm{N}$ is one of the highest, if not the highest concentration radioactive constituent in the coolant during power operations. The reaction is even more complicated, with the proton recoil reaction leading to the formation of $^{13}\mathrm{N}$. The resulting high energy gamma (\sim 6 MeV) is one of the limiting factors related to containment entries in PWRs and drywell entries and steam dependent entries in BWRs. This high energy gamma is the basis for many system designs that allow for the short-lived $^{16}\mathrm{N}$ decay.

¹⁸F provides an example where a coolant activation product is of little consequence. The production of this isotope is mostly determined by core design and is directly related to the neutron flux. Fast neutrons collide with H₂O molecules, or more precisely interact with the hydrogen atoms producing recoil protons, which in turn react with ¹⁸O forming ¹⁸F. with the combination of the short half-life and low energy gamma of ¹⁸F results in this radionuclide being of little significance to the overall source term.

Table 1 captures some of the more common coolant activation products with the reaction and source for consideration.

Table 1: Origin of the main activation products present in the primary cooling system from the primary

Activation Product	Reaction	Half-life	Source / Notes
¹⁶ N	$^{16}O(n,p)$ ^{16}N	7.13 seconds	Activation of ¹⁶ O in the coolant
13 _N	$^{16}O(p,\alpha)$ ^{13}N	9.96 minutes	Activation of ¹⁶ O in the coolant and the prompt interaction of the proton recoil from the reaction above
¹⁸ F	$^{18}O(p,n)^{18}F$	109.7 minutes	Activation of ¹⁸ O by proton recoil in the coolant
³H	$^{10}B(n,\alpha)$ $^{7}Li(n,n\alpha)$ ^{3}H $^{10}B(n,2\alpha)$ ^{3}H $^{6}Li(n,\alpha)$ ^{3}H	12.3 years	Activation of ¹⁰ B and ⁶ Li injected in reactor coolant to control respectively reactivity and pH Activation and release from secondary start-up sources (antimony - beryllium)
⁴² K		12.36 hours	Activation of K injected in reactor coolant to control pH at VVER reactors
¹⁴ C	$^{17}O(n,\alpha)$ ^{14}C $^{14}N(n,p)$ ^{14}C $^{13}C(n,\gamma)$ ^{14}C	5730 years	Activation of ¹⁷ O contained in reactor coolant and into uranium oxide
⁴¹ Ar	$^{40}Ar(n,\gamma)$ ^{41}Ar	1.83 hours	Activation of ⁴⁰ Ar contained in the reactor pit ventilation air (BWR) or the reactor coolant
³⁸ C1	$^{37}Cl(n,\gamma)$ ^{38}Cl	37 minutes	Activation of ³⁷ Cl contained in coolant as impurity
²⁴ Na	$^{23}Na(n,\gamma)^{24}Na$	23 hours	Activation of ²³ Na contained in coolant as impurity Activation of ⁶⁴ Zn contained in
⁶⁵ Zn	$^{64}Zn(n,\gamma)^{65}Zn$	244 days	coolant as impurity or from natural zinc injection. This may be a significant contributor to shut-down dose rates

3.1.3 Activated Corrosion Products

Section 3.1 previously identified the three general classifications of activity within the primary circuit; coolant activation products, fission products, and corrosion activation products. The activated corrosion products can be further divided into two different sources; out-of-core corrosion products and fuel assembly / materials of fuel construction corrosion products. Section 3.1.3 discusses activated corrosion products.

Metallic non-radioactive corrosion and wear products are affected by coolant chemistry (p H_T , Zn and H_2), as well as local velocity (wall shear forces) and temperature and exist as dissolved, colloidal or particulate species. These species may deposit on fuel rod surfaces by precipitation, adsorption, or particle deposition and activate by absorbing a fast or thermal neutron. Several

processes can cause the re-entrainment or release of the deposited material back into the coolant, including, but not limited to, erosion, thermal hydraulic changes, and chemistry changes such as changes in redox potential or pH. Activation products are transported to ex-core surfaces and can be deposited or absorbed in out-of-core surfaces (oxides) or collected in low flow areas. Figure 1 is an overview of this process related to the pressurized water fleet.

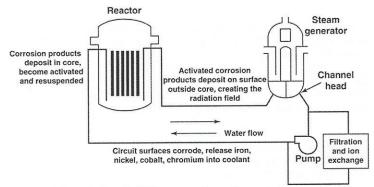


Figure 1: Generic PWR process of corrosion product transport

In early plant designs, site personnel were challenged by material selection issues that gave rise to various activation product source terms. As material and fuel reliability concerns lead to component replacements, NSSS and fuel vendors adopted newer materials to address identified issues. One consideration was in the selection of materials that contained lower cobalt and nickel content to minimize the activated corrosion product inventory. Some examples include;

- Fuel vendors replaced Inconel based fuel grids with Zircaloy-based materials to reduce the amounts of nickel and of associated cobalt impurities.
- The fuel cladding materials was replaced with material having lower cobalt content
- SG tubing was originally replaced with Alloy 600, and for later replacements, Alloy 690 and 800 nickel content and lower cobalt impurity concentrations

Radiation fields tend to build up over a few years following initial start-up before an equilibrium level is reached. Changes in the chemistry regimens or system/component decontaminations can alter these fields over time, but in general these changes have little impact on 60Co dominated radiation fields with the exception of decontaminations. Understanding the processes of release and deposition, combined with the plant limitations, provides personnel with the ability to begin evaluations of the various tools available to manage radiation fields.

Equation 3 shows the basic activation equation accounting for decay as well as build-up. The amount of a radionuclide generated by neutron activation depends on the neutron flux intensity, neutron absorption cross-sections, irradiation duration (the time the species are exposed to the neutron flux).

Equation 3

$$A_i = N\sigma\varphi \Big(1 - e^{-\lambda_i t}\Big)$$

Where:

¹ Inconel is the registered trademark of Special Metals Corporation referring to group of austenitic nickelchromium-based superalloy.

A = Activity

N = Number of atoms available for activation

 σ = Neutron cross section

 φ = Neutron flux

 λ = Decay constant $(\ln(2)/T_{1/2})$

t = Time since activation

The source term for activated corrosion products can be defined, as any parent or radioactive nuclide that is outside the fuel cladding, and which may transport through the primary circuit. The selection of significant radionuclides is based on half-life, concentrations, and gamma scan data. The main contributors, based on gamma scan campaigns, include ⁵⁸Co, ⁶⁰Co, ^{110m}Ag, ^{124,5}Sb, ⁵⁹Fe, ⁵⁴Mn, ⁵¹Cr, ⁹⁵Zr and ⁹⁵Nb. Each of these radionuclides is subject to deposition or absorption on ex-core surfaces contributing to the radiation field build-up, or removal on purification media (resins or filters). It should be noted that ⁵¹Cr typically does not present dose rate issues during shutdown due to the low energy decay gamma.

Table 2 captures the dominant radionuclides observed in the industry with the source, activation process, and common sources based on supporting research from multiple references. In general, these specific isotopes dominate the overall source term related to long term dose rates.

There is many other activated corrosion products identified in table-2 for a variety of reasons including half-life and expected concentrations. For example, 55 Fe, 59 Ni and 63 Ni are present in the coolant and impact waste stream classifications. In some cases, these radionuclides become more of a concern if the primary circuits are open for maintenance, due to the -accompanying β emission.

Antimony and silver present unique challenges during shut-down operations. Shutdown chemistry controls may require adjustment and additional dosimetry evaluations may be required. These radionuclides will be discussed in later sections.

Table 2: Origin of the main activation products present in the primary cooling system from structures or corrosion mechanism

Radionuclide	Half Life	Activation Reaction	Major Source
⁵¹ Cr	27.702 days	⁵⁰ Cr (n,γ) ⁵¹ Cr	Stainless steel and nickel based alloy
⁵⁴ Mn	312.1 days	⁵⁴ Fe (n,p) ⁵⁴ Mn	Stainless steel and nickel based alloy
⁵⁵ Fe	2.73 years	54 Fe $(n,\gamma)^{55}$ Fe	Stainless steel and nickel based alloy
⁵⁶ Mn	2.578 hours	⁵⁵ Mn (n,γ) ⁵⁶ Mn	Stainless steel and nickel based alloy
⁵⁸ Co	70.88 days	⁵⁸ Ni (n,p) ⁵⁸ Co	Nickel alloys
⁵⁹ Fe	44.51 days	⁵⁸ Fe (n,y) ⁵⁹ Fe	Stainless steel and nickel based alloy
⁵⁹ Ni	7.46E4 years	⁵⁸ Ni (n,γ) ⁵⁹ Ni	Stainless steel and nickel based alloy
⁶⁰ Co	5.271 years	⁵⁹ Co (n,γ) ⁶⁰ Co	Stellite TM and cobalt bearing components
⁶⁴ Cu	12.701 hours	⁶³ Cu (n,γ) ⁶⁴ Cu	17-4 PH Steel
⁶⁵ Zn	243.8 days	64 Zn $(n,\gamma)^{65}$ Zn	Natural zinc injection
⁹⁵ Nb	34.97 days	⁹⁵ Zr decay	Fuel cladding (Zircaloy, Zirlo™, etc.)
⁹⁵ Zr	64.02 days	94 Zr (n,γ) 95 Zr	Fuel cladding (Zircaloy, Zirlo™, etc.)
⁹⁹ Tc	2.13E5 years	98 Mo (n,γ) 99 Mo \rightarrow 99 Tc	Stainless steel, tramp impurities, and fission

Radionuclide	Half Life	Activation Reaction	Major Source
^{110m} Ag	249.8 days	¹⁰⁹ Ag (n,γ) ^{110m} Ag	Silver-Indium-Cadium Control rod wear, Helicoflex™ seals
¹²² Sb	2.72 days	¹²¹ Sb (n,y) ¹²² Sb	Secondary start-up source
¹²⁴ Sb	60.20 days	¹²³ Sb (n,γ) ¹²⁴ Sb	Secondary start-up source, RCP bearings, impurities
¹²⁵ Sb	2.75 years	¹²⁵ Sn decay ¹²⁴ Sb (n,γ) ¹²⁵ Sb	Fuel cladding impurities and neutron capture by ¹²⁴ Sb
$^{181}\mathrm{Hf}$	42.4 days	180 Hf $(n,\gamma)^{181}$ Hf	Fuel cladding impurities
¹⁸⁷ W	23.9 hours	$^{186}{ m W}$ $({ m n},\!\gamma)$ $^{187}{ m W}$	Stainless steel_carbides, and welding artifacts

3.1.3.1 Corrosion Product Transport and Activation

A series of events must occur before a given radionuclide can reach and incorporate into ex-core oxides. Hussey identified five steps in the process [xx]; each of these steps is a complicated process that should be evaluated in more detail.

- 1) Corrosion product release from out-of-core surfaces,
- 2) Transport to the core and deposition on fuel cladding surfaces,
- 3) Activation of the corrosion product metal,
- Release of the activated corrosion product from the fuel cladding surface and transport from the core.
- 5) Deposition or uptake of the corrosion product on out-of-core surfaces.

An exception to the 5 steps above relates to the release of radionuclides produce by activation of reactor vessel internals, fuel assemblies, and other fuel structure components. In this case, the basic steps are best described as:

- 1) Activation of fuel assembly or structural component metals,
- 2) Release of highly activated corrosion products in the coolant and transport from the core,
- 3) Deposition or uptake of the corrosion products on out-of-core surfaces.

Release of these corrosion products can be affected by processes such as dissolution, spalling, erosion, and corrosion while deposition is driven by diffusion, inertia (the ability to maintain a particle in solution), temperature gradient, surface charge, etc. Plant design, flow, and chemistry play an integral role in the corrosion and corrosion release rate process. Primary coolant chemistry (pH_T, Zn and H₂), as well as local velocity (wall shear forces) and temperature are important factors to consider related to the management of corrosion products.

Equation 4 captures the basic processes activated corrosion products undergo after release from core surfaces. The concern for radiation protection is the uptake terms in Equation 4. These terms (KAC) for stainless steel and SG tubing are dependent on maturity of material (oxides), temperature, porosity of the oxides, and the thermal conditions. Numerous research projects have reviewed and defined the corrosion and corrosion product releases of piping surfaces and alloys in the primary circuit. In order to impact the source term, one must change the corrosion rate of the alloys, change the corrosion product release mechanisms or alter the uptake terms.

Comment [RDCN6]: EPRI Reference

Hussey, D, Impacts of CVCS Cleanup Systems on Activity Release and Dose Rates, June 2007, EPRI Condensate Polishing Workshop, Palm Springs, CA

Disposed by committee - 7/12/2013

Equation 4

$$A_{coolant} = \frac{RR_{\mathit{Fuel}} - W_{\mathit{LD}}C - k_{\mathit{SG}}A_{\mathit{SG}}C - k_{\mathit{SS}}A_{\mathit{SS}}C}{M\lambda}$$

Where:

A = Activity

C = Primary circuit activity concentration, <u>Bq/kg (μCi/kg)</u>

RR = Release rate into the coolant, Bq/s (μ Ci/s)

M = Mass of the primary coolant, kg

 λ = Decay constant, (s⁻¹)

 A_{ss} = Stainless steel area, (em²)

 k_{SG} = Steam generator tubing incorporation rate constant, $(Bq/s)/(Bq/m^2/kg)$

 $(\mu \text{Ci/s})/(\mu \text{Ci em}^2/\underline{\text{kg}})$

 k_{SS} = Stainless steel incorporation rate constant, $\frac{(Bq/s)/(Bq/m^2/kg)}{(\mu Ci/s)/(Bq/m^2/kg)}$

μCi m²/kg)

 W_{LD} = Letdown flow rate, (kg/s) A_{SG} = Steam Generator area, (em²)

The sections below capture some of the more common radionuclides of concern for radiation protection. It is not intended to be an all inclusive, but a review of the major sources of activated corrosion products in more details compared to Table 2.

3.1.3.2 51 Cr

⁵¹Cr is formed as a result of neutron-gamma activation of ⁵⁰Cr (Equation 5) generated from corrosion of the primary circuit piping. ⁵¹Cr is observed in the primary circuit analysis and typically it is not a major contributor to the overall source term from a dose perspective. ⁵⁰Cr has a natural abundance of about 4%. The low energy gamma can be masked in the Compton continuum during analysis or gamma scans.

Equation 5

$$^{50}_{24}Cr + {}^{1}_{0}\eta \rightarrow ^{51}_{24}Cr + {}^{0}_{0}\gamma(320.1 keV)$$

3.1.3.3 54Mn

⁵⁴Mn is formed as a result of the high energy neutron-proton activation of ⁵⁴Fe (Equation 6). The source ⁵⁴Fe, which is about 6% natural abundance, is primarily from the corrosion of primary circuit piping.

Equation 6

$$^{54}_{26}Fe + {}^{1}_{0}\eta_{f} \rightarrow ^{54}_{25}Mn + {}^{1}_{1}\rho$$

3.1.3.4 58Co

58Co is formed from the neutron-proton activation of 58Ni (Equation 7). On decay, 58Co emits multiple decay energies with the primary energy at ~811 keV. Nickel is a primary constituent in PWR Alloy 600 and 690 steam generator tubing and has in the past been used in some core components (grid straps).

Comment [RDCN7]: SI units will be used

Revised by committee - 7/12/2013 - need to verify on final printing

Equation 7

$$^{58}_{28}Ni + {}^{1}_{0}\eta_{f} \rightarrow ^{58}_{27}Co + {}^{1}_{1}\rho$$

⁵⁸Co is the second largest source to consider for PWRs and can be a significant contributor to shutdown radiation fields. In some cases, ⁵⁸Co may be the largest source for a particular outage because of large releases during end-of-cycle operation (over the last 200 days of operations).

3.1.3.5 ⁵⁹Fe

⁵⁹Fe is formed as a result of a neutron-gamma activation of ⁵⁸Fe (Equation 8). ⁵⁸Fe is approximately 3% natural abundance and is primarily from the corrosion of primary circuit piping surfaces.

Equation 8

$$_{26}^{58}Fe + {}_{0}^{1}\eta \rightarrow _{26}^{59}Fe + {}_{0}^{0}\gamma$$

3.1.3.6 60 Co

⁶⁰Co is formed as a result of neutron-gamma activation of ⁵⁹Co (Equation 9). The decay of ⁶⁰Co presents two problems for plant personnel; the first is the relatively long half-life, and the second is the two high energy gammas emitted on decay.

Equation 9

$$^{59}Co(n,\gamma)^{60}Co \rightarrow ^{60}Ni + e^- + 2\gamma(1.173 and 1.332 MeV)$$

In general, system piping materials have very low cobalt content and minimizing the cobalt content should always be a consideration when replacing components. Low cobalt replacement material should be considered in a plant's cobalt reduction program.

3.1.3.7 110mAg

^{110m}Ag is formed as a result of neutron-gamma activation of ¹⁰⁹Ag (Equation 10). ^{110m}Ag has been observed in large amounts upon plant shutdown after oxygenation, significantly impacting letdown dose rates. The primary source for ^{110m}Ag is believed to be from Ag-In-Cd control rod wear, although in some cases, an additional source could be silver from seal rings or soldering materials.

Equation 10

$$^{109}_{47} Ag + {}^{1}_{0} \eta \rightarrow ^{110 \, m}_{47} Ag + {}^{0}_{0} \gamma$$

3.1.3.8 122Sb, 124Sb, 125Sb

Antimony has two stable isotopes, ¹²¹Sb with an abundance of 57.36% and ¹²³Sb with an abundance of 42.64%. ¹²¹Sb and ¹²³Sb are the source for the activation products ¹²²Sb and ¹²⁴Sb, respectively, through a neutron-gamma reaction (Equation 11 and Equation 12).

¹²⁴Sb is formed as a result of neutron-gamma activation of ¹²³Sb (Equation 12). Secondary start-up sources composed of antimony-beryllium encased with a stainless steel cladding have in the past lead to significant shutdown dose issues on failure. Antimony-impregnated graphite pump seals and bearings have also been sources of radioantimony. Several utilities have replaced these sources on a more aggressive schedule or removed the start-up sources from the plant.

The longest-lived isotope is ¹²⁵Sb with a 2.75 year half-life, which is formed through decay of ¹²⁵Sn and neutron activation of ¹²⁴Sb. ¹²⁵Sb is formed as a result of neutron-gamma activation of ¹²⁴Sn (~5.8% of natural tin). Equation 13 captures the activation step to ¹²⁵Sb, and the subsequent beta decay to ¹²⁵Sb. The source of tin is believed to be from the fuel cladding where tin is a minor constituent of Zircaloy and ZIRLOTM.

Equation 11

$$^{121}_{51}Sb + {}^{1}_{0}\eta \rightarrow {}^{122}_{51}Sb + {}^{0}_{0}\gamma$$

Equation 12

$$^{123}_{51}Sb + {}^{1}_{0}\eta \rightarrow ^{124}_{51}Sb + {}^{0}_{0}\gamma$$

Equation 13

$$^{124}_{50}Sn + ^{1}_{0}\eta \rightarrow ^{125}_{50}Sn \xrightarrow{9.64days} ^{125}_{51}Sb + ^{0}_{-1}\beta^{-} + \upsilon$$

3.1.3.9 Conclusion

Sections 3.1.3.2 through 3.1.3.8 capture only a few of the many activated corrosion products that can impact the overall radiation fields. A comprehensive source-term reduction program evaluating replacement material and the impacts to radiation fields is critical to the long-term success of source-term reduction programs.

3.1.4 Contribution of radionuclides to dose rate

The radiological impact of radionuclides with short radioactive half-lives such as ¹⁶N (7.3 seconds) is extremely high during operations related to containment or drywell entries, but become negligible within a few minutes after shutdown due to the rapid decay. The overall contribution of these short-lived radionuclides to the refuelling outage radiation field is negligible.

As previously noted, fission products represent the largest source of radionuclides within the primary circuit, but have limited impact on radiation fields. However, unless significant fuel defects (number, size, or combination) are present, the fission products are contained within the fuel assemblies and do not contribute to the overall radiation field that the workers are exposed to during operation or maintenance activities. Refuelling operators are shielded from the very high and intense radiation fields from the fuel assembly by the refuelling pool water and distance. Considering the coolant activation products and shielding to refuelling operators, the most important radionuclides with respect to worker dose are the activated corrosion products ⁶⁰Co, ⁵⁸Co, potentially ^{110m}Ag and ¹²⁴Sb, and ⁹⁵Zt and ⁹⁵Zt and ⁹⁵Nb in PHWRs.

In case of specific contamination, ^{110m}Ag and ¹²⁴Sb can strongly contribute to dose rates. These four radionuclides are particularly bothersome during outage work and significant/major contributors to ex-core dose rates. Table 3 provides a summary of the main radionuclides of concern, their fission and activation products and sources.

Table 3: Source terms components - Summary table

nuclei	Fission Product	Activation Products	Actinides or heavy nuclei
--------	-----------------	---------------------	---------------------------

		Fission Products	Activation Products	Actinides or heavy nuclei
Production Methodology		Generated by nuclear fission under neutron flux into fuel rods during operation	All material near reactor core is activated: hard structures ² but mainly corrosion products transported by primary coolant	Constituents of fission (uranium or plutonium) splitting during operation to provide energy
_	Dominant dionuclides	¹³¹ I, ¹³³ Xe, ⁸⁵ Kr, ¹³⁴ Cs, ¹³⁷ Cs	⁶⁰ Co, ⁵⁸ Co, ^{110m} Ag, ¹²⁴ Sb, ⁵⁴ Mn,	²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Am, ²⁴² Cm, ²⁴⁴ Cm
Impact	Operation	Confined in fuel rod cladding, they can be released in case of cladding defects.	Corrosion products are transported into the reactor and auxiliary systems subject to deposition or incorporation onto wall pipe surfaces. 16N is present in the reactor water 41 Ar is present in the ventilation air (mainly for PWRs) or leaked in air (mainly for BWRs)	Confined to the fuel rod cladding, actinides can spread in primary circuit in case of severe cladding defect.
	Maintenance Outage Operations	An activity peak may be observed during plant depressurization activities in case of cladding defects.	These activated corrosion products are the largest contributor to shutdown dose rates. If the recommended procedures are followed, the contribution to worker doses will be minimal.	Generally absent from primary coolant except in case of severe cladding defect occurring during previous cycles or impurities from manufacturing.

In general terms, actinides and fission products do not pose problems regarding dose rates, but can be an issue if the primary system is opened and the plant has a history of fuel cladding failures. As a conclusion, the impact of the activation products (mainly corrosion products) will be the main focus of this report.

3.2 Material Issues

Nuclear steam supply systems (NSSS) are constructed from alloys based on several factors supporting the plant life cycle.. These materials fulfil several requirements including material integrity, wear resistance, satisfactory corrosion behaviour, and low activation in the expected environment. From a radiation protection and dose rate point of view, the most important material issue is the corrosion resistance of the material.-Regardless of NSSS design, alloys corrode when in contact with high temperature water or steam environments. This in turn results in the release of corrosion products, which are then available for deposition and activation. As an example, the PWR fleet is challenged by the high surface areas of the Alloy 600, 690, and 800 tubes used in the steam generators.

² All materials inside the reactor vessel (e.g. internals, fuel cladding and materials of construction) should be considered and understood regarding their source-term impact. Piping penetrations and concrete have many other factors to consider related to long-term maintenance activities and decommissioning activities (i.e. ³⁶Cl)

3.2.1 Pressurized Water Reactors (PWRs)

For PWR's, ⁵⁸Co and ⁶⁰Co are the significant contributors to shutdown radiation fields. Other radionuclides present contributing to radiation fields may include ^{110m}Ag, ¹²²Sb, and ¹²⁴Sb. In general terms, the primary radiation field contributors are ⁵⁸Co and ⁶⁰Co. Table 4 gives an overview of typical PWR materials showing a mixture of stainless steels and alloys.

Table 4: Typical PWR materials of construction

Component(s)	Material	
Primary Circuit -	Reactor Vessel and Piping	
Vessel Cladding	304 SS (Weld Deposited)	
Vessel Internals	304 SS	
Instrument and Control Rod Drive Nozzles	Alloy 600	
Control Rod Drives	304 SS and 410 SS	
RCS Piping	304L SS	
Surge and Spray Piping	316 SS	
	n Generator	
Bottom Head Cladding	304 SS (Weld Deposited)	
Tube Sheet Cladding	Alloy 600 (Weld Deposited)	
Tubes	Alloy 600, 690, or 800	
Divider Plate	410 SS	
The second secon	Pumps	
Casing	316 SS	
Internals	304 SS	
Pr	essurizer	
Cladding	304 SS and / or Alloy 600	
Heaters	304 SS and / or Alloy 600	

3.2.1.1 Steam Generator Material

The primary side of the SG consists of tubes made of Alloy 600, 690, or 800 and represents the largest surface area in the primary circuit, and the principle source for ⁵⁸Ni. The corrosion and release rates of Fe, Ni, Cr, Co and other elements from the SG tubes will have a major effect on the subsequent formation of activated corrosion products. Plants that have replaced Alloy 600 tubes with Alloy 690 should observe a reduction in corrosion and corrosion release rates over time. Laboratory data suggests as much as a factor of three reductions in corrosion may be observed for Alloy 690 tubing compared to Alloy 600 tubing. [6]

3.2.1.1.1 Steam Generator Manufacturing Process

Various manufacturing steps have been considered in an effort to understand the impact on passivation and related to corrosion and corrosion product release. There have been a number of improvements to the manufacturing of Alloy 690 tubes since the initial deployment. The manufacturing processes modified include the sandblasting of tubes with corundum particles has been suppressed, the annealing process is now under H₂ compared to the earlier options of NH₃ or H₂. The carbon content has been reduced and the equivalent carbon content is taken into account for determining the annealing temperature. Pilgering steps and/or drawing have been optimized, cleanings has been introduced, cleaning baths have been modified etc. Accordingly with these manufacturing

process optimizations, the improvements are expected to result in enhanced corrosion product behaviour and subsequent corrosion product release.

The French fleet has conducted extensive research in this area. In French reactors, four main periods can be considered according to the optimized processes. The first period is for SG manufacturing through 1988, the second is the period from 1989-1992, the third is the period from 1993-1995, and the fourth is SG manufacturing after 1995 [7]. In Figure 2, the evolution of ⁵⁸Co peak deposited activity on hot legs over cycles for various French reactors are presented for the 900 MWe series of reactors. Figure 3 is for EDF 900 MWe fleet and shows the evaluation of ⁵⁸Co oxygenation peaks. Both figures represent the evaluation ⁵⁸Co post-steam generator replacement. It should be noted that EDF follows a standardized chemistry program for the various fleets and only minor modifications related to the shutdown procedure have occurred over this period. These modifications should have a minor impact on oxygenation peaks, if any. Based on the EDF data, variations observed in ⁵⁸Co peaks between the different series appear to be mainly due to manufacturing processes, surface condition, and eventual surface preconditioning.

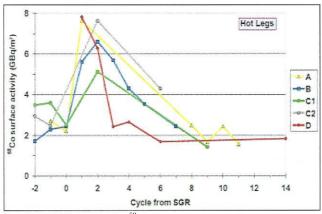


Figure 2: EDF 900 MWe Fleet ⁵⁸Co deposited activity on hot legs over cycles

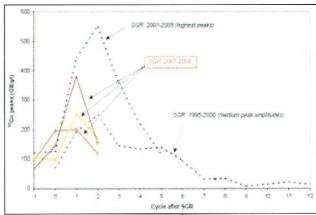


Figure 3: EDF 900 MWe Fleet 58Co oxygenation peaks

Comment [HBO8]: Dates were confirmed.

Reference

"Impact of the Surface State of Steam Generator Tubes on the Release of Corrosion Products in Pressurized Water Reactors", F. Carrette and al, NPC 2006

EPRI, 1018593, Surface Analysis of Pressurized Water Reactor Steam Generator Tubing Specimens: updated review of results obtained from the ID Surfaces of EDF tubes pulled between 1985 to 1990

Agreed with by committee and 2 references – 7/12/2013

The kinetic results and the characterization of the surface state and the oxide layers formed on the surfaces show that corrosion product release is a complex phenomenon. The surface state has a great influence on the release rate but the effects are not easy to summarize and are beyond the scope of this paper.

Figure 4 represents approximately 200 cycles of additional ⁵⁸Co data from across the fleet representing plants injecting zinc and not injecting zinc, higher pH programs to lower pH programs, and plants using speciality resins and enhanced clean-up systems. As shown in the figure, the ⁵⁸ Co peaks approximately 3- 5 cycles post-SGR and trends to lower levels over time. The peaks reach a minimal concentration between 8 and 12 SG effective full power years.

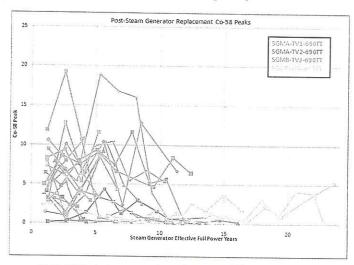


Figure 4 Post-steam generator replacement of 58Co peaks (µCi/g)

3.2.1.2 Primary Circuit and Other Components (Non-SG)

Primary system piping in PWRs (reactor coolant, RHR, and the Chemical and Volume Control System or CVCS) is primarily composed of stainless steels and is exposed to high pressure and temperature environments. The environmental conditions vary from acidic to alkaline conditions under reducing or oxidizing. See appendix-1 table 1 which includes information on primary PWR materials.

3.2.1.3 Other Systems or Components

3.2.1.3.1 Cobalt Sources

Early plant designs applied a high cobalt alloy material supporting long-term wear resistance and component or equipment reliability. These components contained Stellite® for the hard facing surfaces and other reactor internals (BWR Control Blade roller bearings, etc) where identified as the primary source of ⁵⁹Co which undergoes neutron capture and activates to ⁶⁰Co. These activated species migrate to ex-core surfaces and can deposit through a several mechanisms on piping surfaces, thereby contributing to the overall radiation fields workers are exposed to during operations and maintenance activities.

Utilities have developed various processes to address and minimize the amount of cobalt material placed in-service. These efforts continue to shown benefit and have reduced the overall source term due to component replacement. It has been demonstrated that numerous cobalt-free alloys with appropriate characteristics are available to replace the cobalt-based alloys previously used as hard facing materials. Cobalt-free or reduced cobalt materials have undergone extensive testing programs and are now being introduced and used with increasing frequency in numerous applications throughout the industry with acceptable results. Also, a structural alloy with good wear resistance such as 400 grade stainless steel may be used in some applications where a cobalt-base hard facing alloy previously had been used. Controlling the cobalt impurity level in structural alloys used in replacement components is discussed in the EPRI Radiation Field Control Manual TR-1003390

Revision 1 of the EPRI Cobalt Reduction Sourcebook identified several tables and the composition of several commonly applied materials in the PWR fleet [_]. See Appendix-1, table 2 and 3 for example of hard faced cobalt materials composition and examples of hard faced nickel material composition.

Each station should have cobalt reduction guidance documents developed to address cobalt source term reduction efforts and processes. The EPRI *Cobalt Reduction Sourcebook* provides example flowcharts that utilities can apply in the identification of appropriate action plans.

3.2.1.3.2 Fuel Support Material

Early fuel assemblies in the fleet used stainless steel fuel cladding with InconelTM grids. These grids were not only high in nickel content, but high in residual cobalt content. The resulting impact was high coolant radionuclide concentrations and transport to ex-core surfaces, which resulted in higher plant dose rates. The fuel vendors replaced this material with a zirconium-based material, resulting in a significant reduction in ⁵⁸Co and ⁶⁰Co levels. In general terms, western-style PWRs have replaced all of the high nickel and cobalt content fuel assembly materials of construction with low-cobalt, zirconium-based alloys. Table 5 list example materials of fuel assembly construction.

Table 5: Example Modern PWR Fuel Assembly Materials of Construction

Component	Example Materials of Construction		
Fuel cladding material	Example: M5 ^{TM3} or Zirlo TM		
Spacer grids	8 x grids with M5 straps and Inconel™ springs		
Upper end fittings	AISI 304L with Inconel TM springs + spring screw and AISI 308 lock pins		
Lower end fitting	AISI 304L with AISI 660 anti-debris device and AISI 304 pins		
Guide tubes	Example: M5 TM or Zirlo TM		
Grid spacer spring strip	Inconel™ springs		

Western style PWRs require boron for reactivity control in the primary coolant, but rod control cluster assemblies (RCCA) ensure sufficient negative reactivity to ensure the reactor shutdown margin is maintained. The neutron absorbing material is hafnium, or a silver (80%), indium (15%), and cadmium (5%) alloy. ¹⁰⁹Ag is produced from RCCA rod wear and is the source for ^{110m}Ag. It should be noted that silver may also originate from seal rings. Based on EDF data, the amount of metallic silver contamination is estimated to be from 1 to 10 grams and contributes significantly to the dose rate in the shutdown cooling and clean-up systems.

Comment [E9]:

Reference: Cobalt Reduction Sourcebook. EPRI. Palo Alto, CA: 2010. 1021103.

³ Inconel is the registered trademark of Special Metals Corporation referring to group of austenitic nickel-chromium-based superalloy.

 110m Ag contamination can significantly impact dose rates in the shutdown cooling system, clean-up system heat exchangers, and downstream components to the clean-up system due to precipitation.

3.2.2 Water-Water Energetic Reactors (VVERs)

VVER type reactors represent a separate group of PWRs. Within Europe, two main types of reactors are operated; VVER-440 and VVER-1000. There are several significant differences between VVERs and PWRs that significantly influence the dose rate build-up processes, as described further and in chapter 3.3.2.

3.2.2.1 Steam generator materials

All of the primary circuit of a VVER-440 is made of stabilized austenitic stainless steel 08CH18N10T (AISI 321); for VVER-1000 units, the situation is very similar except that the SG collectors are made of perlitic 10GN2MFA (10NiMo8 5) steel. The VVER SG materials of construction contain significantly less nickel compared to their western-style PWR counterparts. This results in much lower ⁵⁸Co concentrations and simpler shutdown chemistry.

The steam generators are of horizontal design and this fact has special importance, especially in the decontamination processes. For VVER-440 type units, the steam generators are employed for RHR operation during shutdown/refuelling.

3.2.2.2 Cobalt inventory

VVER reactors are typically very low cobalt plants; with the exception of the Loviisa plant, no Stellite® components are used in the primary and auxiliary systems. The cobalt content of the SG tubes material is typically less than 50 ppm, so there little ⁶⁰Co produced. At NPP Loviisa, presence of Stellite® components lead to significant growth of dose rates at the cold legs of the primary loop in the 1980s and full system decontamination was performed at Loviisa Unit 2. Program for gradual replacement of Stellite materials was implemented.

3.2.2.3 Surface preconditioning

There are no specific methods applied for surface preconditioning during manufacturing processes, only HFT passivation is performed during plant commissioning.

3.2.2.4 Fuel support material

VVER fuel metallic structures are made of Russian E110 alloy (Zr+1%Nb) with very low impurity content. The fuel assembly head and bottom nozzle are constructed from AISI 321 steel; in the 1990s the spacer grids were made of AISI 321 steel and later replaced by E110. This replacement brought a corresponding reduction of radiation fields due to the removal of one important Co source.

VVER units typically operate with a 12 month fuel cycles with moderate fuel duty. On-going duty increases within power uprating and fuel burn-up extension projects are being introduced at many plants, extended cycle lengths are now also considered by some operators.

3.2.3 Pressurized Heavy Water Reactors (PHWRs)

Comment [RDCN10]: To David.

Is it possible to prepare a comparison table in appendix-1

D Perkins – Need to get together with VVER contact to build this comparison table. Due by September 1st, if possible.

While the NSSS of PHWRs resembles that of a PWR, the core design is significantly different (nuclear fuel contained in pressure tubes, use of natural U as the fuel, separate moderator system) which results in significant differences in activity transport. Activity transport in the CANDU primary heat transport system (HTS) involves the release; activation and deposition of corrosion products present as particulate (including colloids) or dissolved species [Guzonas 2004, 2006, 2010]. In addition to corrosion, wear of various system components, such as pump seals, valve hard facings, bearings, and material released by the movement of fuel along pressure tube surfaces during refuelling, can release particulate material into the coolant. These wear products can be major contributors to activity transport in CANDU reactors. Once released into the coolant, the transport of both inactive parent and active corrosion or wear products can potentially involve many phases (e.g., particles or dissolved species in the coolant, deposits on surfaces) and processes (e.g., dissolution, precipitation, adsorption) before activated species are finally deposited on out-core surfaces.

Two limiting cases with respect to surface area and impurity concentration are important in the CANDU HTS:

- 1. high surface area materials with trace concentrations of an impurity e.g., steam generator tubes and feeder pipes with ppm concentrations of Co.
- 2. low surface area materials with high concentrations of an impurity—e.g., Stellite valve hard facings or fuelling machine load balls in which Co is a major constituent of the alloy.

The use of a relatively high surface area of carbon steel piping (inlet and outlet feeder pipes)

In addition, magnetite deposition in the SGs enables them to act as 'full-flow' purification ion exchangers, removing a significant fraction of both activated species (e.g., ⁶⁰Co) and the inactive parent (e.g., ⁵⁹Co). By removing nickel [Burrill and Guzonas] it also minimizes the deposition of nickel phases in the core and the production of ⁵⁸Co.

3.2.3.1 Cobalt inventory

The CANDU design is unique in its ability to refuel on-line, using a pair of fuelling machines able to move across the reactor face and attach to the opposite ends of the specific fuel channel to be refuelled, while the reactor is operating. The main components of the fuelling machine ram are 4 ball screws that contain Stellite Star-J load balls (36 wt.% cobalt). Measurements of the reduction in the ball diameters suggest that wear and/or corrosion of these balls can release a significant mass (on the order of grams) of ⁵⁹Co into the fuelling machine circuit. These wear products are mixed into the fuelling machine water and are either removed by the purification filters in the fuelling system or injected into the HTS during fuelling [Guzonas, 2006].

The fuelling machine purification circuit filters are expected to remove wear particles, the effectiveness depending on both the particle size and the pore size of the filters. Recent data [Gauthier and Guzonas] suggests that some of the Co released from the load balls is present as a charged species (dissolved or colloidal) that can only be removed by ion exchange resins. Therefore, both the fuelling machine purification system filters and ion exchange resins must be effective in removing ⁵⁹Co from the coolant serving the fueling machines.

3.2.4 Boiling Water Reactors (BWRs)

Comment [HBO11]: David Miller with Rick Doty

3.2.5 Material and Technology options

Section 3.2.5 provides a general overview of options related to materials preconditioning and improvements that may be applied to aid in the minimization of corrosion products released and subsequently activated.

3.2.5.1 Electropolishing

Electropolishing (EP) is the electrochemical removal of microscopic irregularities from metal surfaces. The process involves the controlled anodic dissolution of metallic surfaces using an electrolyte and a cathode suitably shaped to accommodate the geometry of the component. This process has been applied to BWR and PWR primary systems including replacement piping RWCU spools, steam generator manway seals, and steam generator channel heads. The application of this technology has demonstrated a great reduction in activity uptake and reduced dose rates.

3.2.5.2 Stabilized Chromium Process (SCrP)

SCrP was developed and patented by the Electric Power Research Institute (EPRI) and works by the application of thin films of electroplated chromium followed by preoxidation in moist air. The process has been shown to significantly reduce activity pickup when applied to the surfaces of replacement components [2, 3]. The surface conditioning method has been applied to reactor water cleanup piping (RWCU) and Jet Pumps in BWRs and steam generator manway covers in PWRs for example reducing activity uptake on those components and reducing dose rates fields in those areas of the plant.

3.2.5.3 Technology - Surface preconditioning

Corrosion product release is a complex phenomenon. The surface state has a great influence on the release rate, but it is not easy to describe and to quantify the impact of the surface parameters. In the case of SG tubing, preconditioning oxidation can be potentially performed by two processes; factory preconditioning and onsite conditioning.

The first step is to perform a surface treatment at the manufacturing facility as part of the original manufacturing process. The second step is through preconditioning after installation. In general terms, this surface preconditioning corresponds to a preconditioning phase during the plant start-up (similar to hot functional tests). Regarding this second step, R&D experience indicates that pre-oxidation under basic and reducing conditions at high temperature is the most effective environment during hot functional tests. However, EDF has on-going studies in this area.

It has been demonstrated that in pure primary water, the release phenomenon was controlled by the rate of formation and growth of the oxide scale, in particular the inner oxide film enriched in chromium. The outer oxide layer, formed mainly by thermochemical and diffusion mechanisms, is generally made of nickel oxide (NiO) or a phase with the spinel structure (such as nickel ferrite $NiFe_2O_4$).

Comment [HBO12]: References

"Cleaning the primary circuit after the first high temperature oxidation of steam generator tubing: why and how to do it", M. Bachet and al, NPC 2010

"SG tubes Pre-oxidation and Cleaning – Effect on Release and Oxides Formation", S. Leclercq, M. Bachet (EDF) – France, NPC 2012, Paris

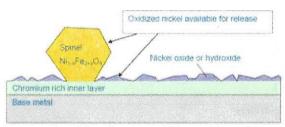


Figure 5: Schematic representation of the oxide grown in high temperature primary water on tubes made of alloy 690TT

Even if significant differences in the oxidation behaviour of alloy 690TT tubes are observed, it seems to be a common feature that the oxidation rates are strongly reduced after passivation, which is a shorter timescale than the observed reduction in ⁵⁸Co oxygenation peaks. In the early cycles of operation, a significant fraction of the nickel would be released during the first cycles of operation.

EDF research has shown that oxidation occurs rapidly on initial exposure to water and higher temperatures. This provides an opportunity to remove a large inventory of nickel before reactor operations, thereby minimizing the formation of activated ⁵⁸Co. The first step is to dissolve the nickel from the outer oxide without damage to the protective inner oxide, and then remove it from water with the help of purification systems like the Chemical and Volume Control System (CVCS). To do this within a relatively short time, it is required that nickel concentrations in the mg.kg⁻¹ range or higher are attained. By examining the available data on the solubility and dissolution kinetics of the possible nickel containing solid phases, EDF has found that the best compromise within chemical specifications is a temperature around 170 °C, an acidic pH and a hydrogen concentration between 10 and 30 cc.kg⁻¹ range or higher. This kind of pre-oxidation and cleaning procedure was rolled out by EDF on a French NPP in 2011 but did not seem to be as efficient as expected.

3.2.5.4 Component Preconditioning

Preconditioning of the surfaces of replacement components can significantly reduce recontamination rates, as well as reduce the cobalt release rate. The nature of primary component surfaces affects the ability of the passive oxides that form on them to incorporate the activated corrosion products, primarily ⁶⁰Co, ⁵⁸Co, and ⁶³Zn, that are responsible for occupational radiation exposure. Surface roughness, surface chemistry, and even surface residual stresses play a role in determining the amount of activity pickup. It was recognized early that electropolishing might lower activity pickup simply by reducing the total surface area in contact with the primary coolant. Another approach to reducing the build-up of radioactivity is to effectively film or coat components that contact the primary coolant. Such coatings could serve two main functions: [2] they form a diffusion barrier against the outward migration of cobalt that is present as an impurity in reactor construction materials, which is desired because the release of cobalt to the coolant is the first step leading to its activation, and [2] coatings may render the surface less susceptible to the incorporation of radioisotopes following their formation in the reactor core. Replacement of primary system components affords utilities an opportunity to specify a surface treatment that is designed to lower the incorporation of activated corrosion products.

Comprehensive programs to develop effective preconditioning techniques have been successful, and the two most widely used surface modification techniques now used in nuclear power plants are electropolishing (EP) and a chromium coating and passivation technique that is designated the Stabilized Chromium Process (SCrP).

Comment [HBO13]: Reference

"Optimized high temperature oxidation and cleaning at Bugey 3", G. Ranchoux and al., NPC 2012, Paris

Agreed - 7/12/2013

3.2.5.5 Other Preconditioning Methods

Recently a new method has been developed by Sumitomo Metals for the formation of a chromium rich surface oxide layer on Alloy 690 steam generator tubes. In laboratory testing this material has demonstrated reduced release of nickel when exposed to simulated PWR environments for up to 1000 hrs. The coating has also been applied to Feedwater Heater Tubes in the Higashidori BWR[3].

3.2.5.6 Technology - Preventive filtration with specific devices

The simplified diagram below illustrates the principles of the preventive filtration methods, which are proposed (Figure 6). It consists of the filtration of all the effluents, which could transport hot spots outside the reactor building.

The drains of pools and the primary circuit are important routes for the development of hot spots and the transfer of material that will settle in low flow areas, contributing to the build-up of hot spots. The installation of fine filters or an initial barrier is proposed for the drain orifices of each pool. The drain lines of the primary cooling circuit represent the second transport mechanism of hot spot migration. This process enables both draining processes to be treated with the same device.

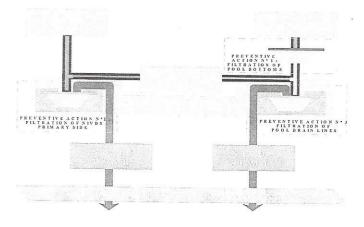


Figure 6: Preventive filtration methods

These modifications are a strategic decision related to the spread of hot spot contamination outside of the reactor building. However, it is a complicated design modification process with regulatory requirements.

3.2.6 Materials Overview Preventive Strategies

Cobalt reduction programs are essential to the long term source term management. Section 3.2.1 provides some information related to key discussions. The understanding and application of technologies can significantly impact source term over several cycles. Some key aspects of a good program include:

- Identification of the overall cobalt source term
- Establishment of a Cobalt Reduction Program
 - Including the limited usage of cobalt based components

 Optimization processes to remove existing materials and understanding methodologies for clean-up following maintenance activities

Surveillance programs are designed to alert the site as early as possible to the presence of hot spots (mapping) in order to take the appropriate measures to prevent their propagation and/or to eradicate them. During unit operation, most hot spots will remain fixed to the fuel. Others may fall, by gravity, to the bottom of the pool or the low points of the primary coolant system or be trapped in the special devices. The most common locations are as follows:

- Thermal sleeves of nozzles of the boiler,
- Valves of the primary cooling system, etc.

3.3 Overview of available chemical methods

3.3.1 Purification / Clean-up System Basics

Each NSSS design has clean-up systems that were originally designed to maintain coolant impurity concentrations within specifications and fission product activity levels supporting off-site dose calculations during accident conditions. Each of the PWRs has basically the same design for clean-up systems. The names for the systems or components vary slightly across the fleet, but in general the systems support five basic functions:

- Maintain the programmed water level in the pressurizer that in turn maintains the required water inventory in the RCS;
- Provide operators with the ability to fill and drain the RCS or during outage and after maintenance conditions pressure-testing of the RCS;
- 3. Provide the flow to the RCS during safety injection conditions;
- 4. Control RCS chemistry including activities per design basis documentation, the chemical neutron absorber (boron) and makeup impurities to the RCS;
- 5. Maintain seal water injection to the reactor coolant pumps.

In the case of the Westinghouse designs, there are three sections or branches of the system: charging, seal water and letdown. During normal operations the plant maintains a continuous flow or feed to the RCS via the charging segment of the system. This flow path is typically charging water back into the RCS and seal injection systems. The primary source of water or at least early in the cycle is recycled coolant from letdown that has passed through a makeup tank covered with hydrogen gas and from seal leak-off. Related to seal injection, a significant amount of this water is routed back to the charging section and the balance is add to the RCS combined with charging flow matched to letdown flow to maintain pressurizer level.

Given the demands placed on plant personnel to reduce the overall source-term, plant personnel and research institutes have expended significant efforts to optimize these systems. Resin vendors continue to improve resin performance in efforts to increase the removal efficiencies of different species.

In order to understand the limitations of clean-up systems, a basic background on clean-up calculations is required. The effect of clean-up flow to system volume and efficiency are variables that can provide insight into the impact of changes related to resins, flow and volumes. Equation 12 provides the basis for the discussions in Section 3.3.1.

Equation 12

$$\begin{split} N(t) &= e^C e^{-\lambda t} = N_o e^{-\lambda t} \\ N_f &= N_t e^{-\lambda t} \\ OR \ More \ Commonly : \\ A_f &= A_t e^{-\lambda t} \end{split}$$

Where:

 $A_{(f)}$ = Final Activity concentration $A_{(i)}$ = Initial Activity concentration λ = Decay constant $(ln(2)/t_{1/2})$, s⁻¹ t = Time delta

3.3.1.1 Clean-up System Impacts

Reactors are designed with numerous clean-up systems, and in many cases, these systems are NSSS-specific designs. As previously mentioned, the original design specifications for these systems focused on maintaining the fission product inventory within specifications to not exceed off-site dose limitations and to minimize impurities in the reactor coolant system. In many cases, the clean-up flow represents less than 5% of the total flow during operation or shutdown conditions, which in turn can significantly limit the ability to optimize clean-up operations.

There are two key areas to consider related to clean-up systems. The first is the impact of clean-up systems during operations and second is the impact during outages for -refuelling or maintenance. The impact during refuelling operations is critical and can significantly impact worker dose, while the impact during operations and limitations of the system related to the overall impact on source-term requires much more detailed evaluations including the effects of materials and fuel design.

During refuelling operations, a simple analogy is that the system has a fixed volume to consider based on the rapid releases observed during shutdown and cool down or in the case of the BWR fleet, the ⁶⁰Co release observed during the refuelling pool flood-up. Both present different demands on the clean-up system performance.

The equations that determine the clean-up system performance impact include system mass, system let-down and clean-up efficiencies. System mass and let-down are easily defined and recorded on many plant computer applications. Clean-up efficiency is simply determined based on the decontamination factor (DF) and defined below in Equation 13. As shown in Equation 13, there can be different DFs for different radioactive species depending on the resin and filtration removal capabilities. This requires an understanding of the efficiencies related to purification system operations.

Equation 13

$$DF = \frac{Activity_{Influent}}{Activity_{Effluent}}$$

$$And \ Efficiency \ is:$$

$$Eff (\%) = \frac{DF - 1}{DF}$$

Equation 13 can then be used in Equation 14 to calculate the purification constant considering the impact of clean-up flow, mass and efficiency.

Equation 14

$$\beta = \frac{\left(\left(Ltdn_{gpm} * \rho_{ltdn} \right) \right)}{\left(\left(\left(V * \rho_{RCS} \right) * 60 \frac{\sec}{\min} \right) \right)} x \frac{DF - 1}{DF}$$

Where:

In order to evaluate the overall effect, an effective half-life must be determined considering both the decay constant and purification half-life (Equation 15).

Equation 15

$$T_{Eff} = \frac{\ln(2)}{\left(\lambda + \beta\right)}$$

Where:

$$\lambda$$
 = Isotopic Decay Constant, s⁻¹
 β = Purification Constant, s⁻¹

Equation 16 now can be used to obtain the effective half-life. As expected, and with the exception of short-lived radioisotopes, the isotopic decay term has little effect on the overall removal rate for power plants during shutdown activities.

Equation 16

$$A_{f} = A_{i}e^{-\lambda i}$$
Where:
$$\lambda = \frac{\ln(2)}{T_{Effective}}$$

Figure 7 simply compares the different effective half-lives using the equations above assuming 100% impurity removal by the resin and filters. As expected, with the exception of short-lived isotopes, the higher purification flows improves the purification half-life. This discussion is expanded in the following sections.

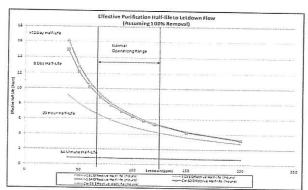


Figure 7: Half-life Impact on Purification Half-life

This basic principle provides an understanding of the factors impacting system clean-up. It should be noted that in many cases, vendors will attempt to focus on chemistry metrics in the evaluation of resin and/or filter performance, but with very low concentrations the same metric may not apply.

3.3.1.2 Clean-up System Resins and Filters

Using the equations described above, one can calculate the effects and/or limitations of clean-up systems on corrosion products and other impurities. Figure 7 assumed 100 % efficiency and Section 3.3.1.2 expands the discussion related to various improvements over time. It should be noted that ⁵⁸Co is used only as an example, and the efficiencies for specific radionuclides should be considered.

3.3.1.2.1 Clean-up Flow Path

In general, the clean-up flow path is as follows. Hot water from the primary circuit is passed through one or more heat exchangers to the resin and filters. Piping size and length is designed to allow for the decay of ¹⁶N or other short-lived radionuclides. The water ultimately reaches the clean-up demineralizers and filters. In some NSSS designs, filters are placed in front of the demineralizers and after the demineralizers. The filters allow for removal of particulates depending on the filter rating. The outlet filter was originally designed to remove resin fines, not for corrosion product management. In many cases, plants use sub-micron filters on the demineralizer outlet to aid in corrosion product management. The resins are designed to remove ionic impurities and combined with the filters optimize clean-up system performance. The purified coolant is then returned to the primary circuit.

3.3.1.2.2 Clean-up Resins

Resin vendors continuously try to improve resin removal efficiency. Resins may be designed to remove specific radionuclides or to have improved removal of all impurities. The application of macro-porous resins and other speciality resins has been identified in improvements in overall source-term reduction strategies. The challenge for plant personnel is to identify all the factors related to source-term management and the impact of these strategies on the overall source-term.

Figure 8 shows the effect of increasing resin efficiency on purification half-life. As shown, increasing the resin efficiency from 75% to \sim 95% has a significant effect on the effective purification half-life while increasing efficiencies from \sim 95% to 100 has little effect.

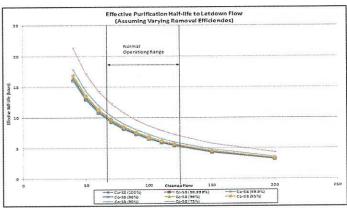


Figure 8: Resin Efficiency Impact on Purification Half-life

3.3.1.2.3 Primary Circuit Filter Application

Primary circuit filteration is typically accomplished through upstream and downstream filters in the cleanup systems located around the demineralizers. The application of the upstream filter is designed to remove particulate material before the demineralizer, while the downstream filters are designed more to minimize the potential impact of resin fines entering the primary circuit and the degradation effects of resin decomposition. These upstream filters can accumulate significant amounts of activated corrosion products compared to the filters downstream of the demineralizer and as a result may be significantly higher in dose rates from each location. The filters are typically rated from 0.05 to 40 microns with the size selected on operating experience and plant specific experiences. In general terms, the upstream filters are between 1 to 40 microns, while the down stream filters are 1 micron or less, but is very plant specific.

3.3.1.3 Clean-up System Operations (Refueling and Operations)

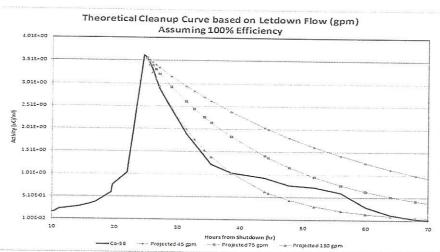
This section is only intended to be an overview and a more detailed discussion is beyond the scope of this report.

3.3.1.3.1 Shutdown Operations

The nuclear power industry continues to review and reduce refuelling outage durations. This reduction in time has placed increased demands for plant personnel to clean-up released corrosion products in minimal time, thereby allowing workers to perform refuelling operations in as low as achievable radiation fields. This reduction in time requires personnel to optimize clean-up systems and coordinated efforts by chemistry, radiation protection, and operation personnel.

There are two key factors to consider related to clean-up systems; clean-up flow and resin efficiency improvements.

Figure 9 plots the clean-up times based on clean-up flow improvements only. As expected from Equation 16, clean-up time is significantly improved for the same volume by increasing the clean-up flow. Figure 10 shows the improvement with optimized flow and increasing resin efficiency from 70 % to 100 %. As expected, this impact is less significant than flow optimization.



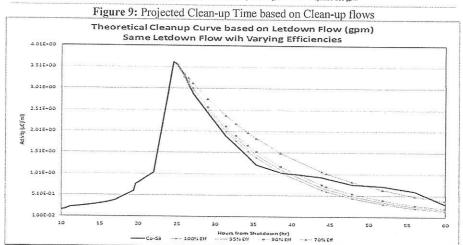


Figure 10: Projected Clean-up Time based on Efficiency Improvements

Based on Equation 16, the key driver to minimize the time for shutdown clean-up activities is to optimize the clean-up flow and/or volume. It should be noted that improvements in clean-up efficiency are always beneficial, especially related to downstream components (i.e., seals, clean-up system piping).

3.3.1.3.2 Normal Operations

As previously stated, the alloys of construction used in the fleet are subject to varying degrees of corrosion and wear, depending on the material composition, chemistry, and thermal - hydraulic conditions. Deposition occurs by precipitation, adsorption, or particle deposition and depends on the nature of the metal oxide developed during operation. Corrosion product deposition on fuel surfaces is much faster than clean-up system removal ($t_{1/2\text{fuel}} < <<< t_{1/2\text{purification}}$).

Table 6 lists data on the typical coolant concentrations of species important for activity transport in western-style PWRs according to the EPRI sponsored Chemistry Monitoring and Assessment program. The typical concentrations are based on equilibrium conditions with nominal clean-up flow. For a normal PWR reactor with clean-up flow maximized, in a given hour only $\sim 12\%$ of the coolant is passed through the clean-up system.

A simple calculation shows that under equilibrium conditions and with optimized clean-up flow and a resin efficiency of 100%, there is still a sufficient number of atoms/gram in the primary coolant for oxide uptake. Improved resin can potentially maintain a lower number of atoms/gram in the coolant, but still sufficient atoms are available for uptake.

Table 6: Typical Coolant Concentrations			
Species	Typical Concentration, ppb	Typical Concentrations, μCi/g	Atoms/gram
Fe	3		3.24E+13
Nu	0.1		1.026E+12
Cr	< 0.01		<1.16E+10
Zn	10		9.13E+13
Co	0.04		4.4E+11
⁵⁸ Co		1.00E-03	3.27E+8
⁶⁰ Co		2.00E-05	1.77E+8
⁵⁴ Mn		8.00E-05	1.15E+8
⁵⁹ Fe		1.00E-05	2.06E+6
51Cr		5 00F-04	6.38E+7

3.3.1.4 Clean-up Conclusion

Resins continue to evolve and improve, which in-turn can result in lower coolant concentrations, but are limited due to system design. Improvements in resin efficiencies during shutdown activities from <95% can significantly impact clean-up durations. Reducing the amount of time to reach clean-up goals during shut down operations allows workers to enter into the containment buildings with potentially lower radiation fields and minimizing the impact on outage schedules. In this case, optimized resins can impact radiation fields allowing workers to enter.

In looking at a simple refuelling outage resin, clean-up flow and volume reduction, Figure 11 provides the improvement that can be observed.

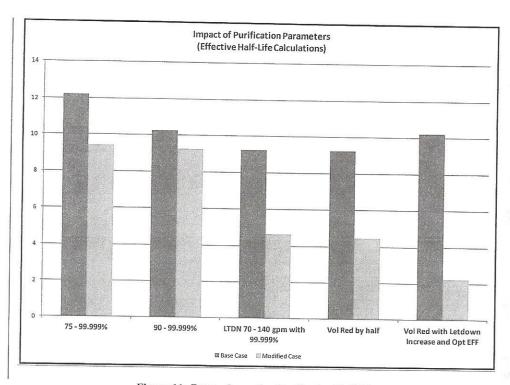


Figure 11: Factors Impacting Purification Half-life

Therefore the overall impact is dependent on the condition or operation and should be understood by plant staff.

3.3.2 Pressurised Water Reactors (PWRs)

The plant chemist tool box is limited to primary circuit pH adjustment, hydrogen controls, and zinc injection. Section 3.3.2 covers the western style PWR chemistry options.

3.3.2.1 pH_T Control

The main corrosion issue related to the primary circuit materials is driven by pH, hydrogen and temperature. As shown by equation 4, even if the concentration of these corrosion products is very low, reactor control and radiation dose rates may be strongly influenced by the deposition of corrosion products on fuel cladding, activation of these corrosion species to radiation source terms, and deposition on out-of-core surfaces.

The Analysis Report on 1999-2001 Field Experience with Elevated, Constant pH [5] provides detailed explanations on various pH ranges to optimize nickel release, deposition on fuel, activity transport, deposition, and potential dose reductions.

Chemistry departments have limited options related to primary circuit pH programs due to fuel concerns and other corrosion related issues. It is expected that in the normal range of operation, primary coolant pH_T (7.0 – 7.4) has minimal impact on corrosion and release rates of associated plant materials. Table 7 is reproduced from Reference ________to show the potential improvement in corrosion

Comment [HBO14]: EPRI primary water guidelines

EPRI, 1014986, Pressurized Water Reactor Primary Water Chemistry Guidelines, Volume 1, Revision 6 product release for various pH programs relative to $pH_T = 6.9$. There is approximately a 4% reduction in corrosion rate by increasing pH_T from 6.9 to 7.1, and by increasing pH an additional 0.1 units would potentially further reduce the release rate by ~1%. Based on current plant performance, there is not an immediate plan to change the primary pH control program in some utilities and peak cycle lithium is ~3.5 ppm with the 7.2 pH_T regime.

Table 7: Relative Corrosion Rates versus primary pH_T

pH_T	Alloy 600	Stainless Steel	
6.5	1.154	1.158	
6.9	1.000	1.000	
7.1	0.962	0.962	
7.2	0.949	0.948	
7.4	0.930	0.929	

It is well known that a pH lower than 6.9 will induce higher risks of contamination of out-of-core surfaces and of axial offset anomalies. Axial offset anomaly (AOA), or later referred to as Crud Induced Power Shift (CIPS), has been observed in PWR cores with sub-cooled nucleate boiling and sufficient circulating corrosion products. Deposition predominantly takes place on the upper portion of the highest powered fuel assemblies. This effect may cause local core power depression through accumulation (hideout) of borates in corrosion product layer on the fuel rod cladding surface. Many plants have experienced AOA, either mild or severe for one or more fuel cycles. However, other plants that have operated with aggressive thermal conditions have been free of the effect. The most severe occurrences of AOA have been observed at the Callaway PWR.

Through the B/Li coordination, which sets the lithium concentration according to the boric acid concentration, optimum pH_T is defined worldwide between 7.2 and 7.4 depending on the alloys used in the primary system. To ensure the core reactivity control in the PWR, the concentration of boric acid is defined according to the neutron calculations and decreases from the beginning of cycle (BOC) to the end of cycle (EOC). Primary pH_T is mainly defined by the concentration of lithium and boron in the primary water. Nevertheless, there are many factors to optimize primary pH_T control. Some alternatives are listed below.

- fuel management and the cycle length,
- increase of lithium concentration at the BOC,
- use of neutron poisons in some fuel rods to decrease the boron concentration at the BOC,
- use of ¹⁰B enriched boric acid to get the same reactivity control with lower boric acid concentrations.

Worldwide, there are many B/Li control programs in use today. In some cases, limits are based on technical specifications and others may be based on fuel vendor limitations. Examples of primary pH control programs include "modified", "elevated Li", or "constant elevated".

[Li] and pH 300°C 7 and 7,2 versus boron

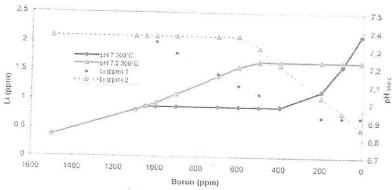


Figure 12: B/Li Ratio and pH

As an example, in 2010 EDF has approximately six plants follow the so-called "modified" B/Li coordination. For all the other EDF reactors, the technical specification is currently at 2.2 ppm maximum lithium concentration ("standard" as defined by EDF) (Figure 12).

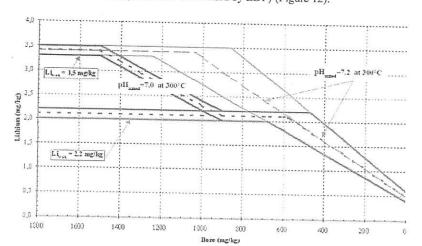


Figure 13: Diagram of the three used or foreseen B/Li coordinations in EDF PWRs: Classic coordination (pink) modified coordination (blue) and elevated lithium coordination (red)

EDF performed many studies in order to test "elevated lithium" control before a possible implementation across the fleet. The main objective of this modification was to raise the maximal lithium concentration from 2.2 ppm to 3.5 ppm to reach as quickly as possible a target pH $_{\rm T}$ of 7.2 (Figure 13). Different issues concerning the impacts of increasing the lithium concentration have been taken into account: dose rate and radiation fields, mitigation of axial offset anomalies, and material degradation (cracking mitigation). Concerning the last point, the influence on the main alloys used for

Comment [HBO15]: Provided from "EDF strategy for the primary coolant pH", J.L. Bretelle, NPC 2004

Agree - 7/12/2013

Comment [HB016]: Provided from "Corrosion products behavior and source term reduction: Guidelines and feedback for EDF PWRs, concerning the B/Li coordinations and steam generators replacement", S. Taumier and al, NPC 2010

Agreed by committee - 7/12/2013

the primary system components have been considered.

3.3.2.1.1 Overview of pH_T program

Plant chemists continue to optimize primary pH programs considering a wide range of issues. These issues include fuel vendor concerns, fuel management, material corrosion, and any impact on support systems. It is up to plant specific evaluations to determine the optimal pH_T programs.

3.3.2.2 Zinc injection

Zinc injection is primarily considered as part of an overall dose reduction strategy, although some consider zinc as part of the primary water stress corrosion cracking (PWSCC) mitigation plan. The application of zinc has been successfully performed at approximately 80 PWRs worldwide since the mid-1990s representing 30% of the global PWR fleet.

EDF has developed a strategy for taking advantage of zinc injection. The target zinc concentration has been fixed considering the benefits expected for material corrosion, source term reduction and radiation fields. Safety analysis, Chemical Specifications, Operational Guides have been elaborated to facilitate the NPP actions permitting the zinc injection continuation at Bugey 2 and Bugey 4. Regarding radiation field reduction, zinc injection has not clearly shown positive results but no contra-indication has been highlighted neither. Nevertheless, the main interest in zinc injection is its multiple benefits and the implementation at 14 more EDF units from 2010 to 2012 has been decided not only for dose rates reduction but also for PWSCC and AOA mitigation. Zinc injection should be considered as a strategy with benefits in the short, medium and long term.

3.3.2.2.2 Conclusion

NPP operational experience and laboratory results show that zinc injection application seems to provide positive effects in all of these domains without inducing adverse impacts. EDF has developed a strategy to implement zinc injection in its fleet.

Nevertheless, even if the popularity of zinc injection is due to its fast impact on surface contamination, the main interest of zinc injection is its multiple benefits not only for dose rates reduction but also for PWSCC and AOA mitigation. The zinc injection should be considered as a strategy with benefits in short, medium and long term.

3.3.2.3 Shut down and start-up operations

During the shutdown, physico-chemical conditions can significantly vary: pressure and temperature drop, hydrogen content decreases, boron and lithium concentrations changes, hydrogen peroxide is injected, the primary fluid switches from reducing to oxidizing conditions. These changes of the primary water conditions result in the dissolution of deposits on fuel assemblies. Because of the oxygenation, corrosion product activity concentrations significantly increase in the primary circuit, especially the ⁵⁸Co volume activity. The level and form of ⁵⁸Co activity at forced oxygenation drives the clean-up time required to reach the activity threshold for which the RCS breaking can occur, thus impacting the outage duration. In order to minimize and decrease this activity, various parameters can be taken into account, such as the surface state of the tubes.

The objectives in terms of chemistry/radiochemistry will depend on shutdown and restart scenarios considered and must ensure a balance between various constraints: