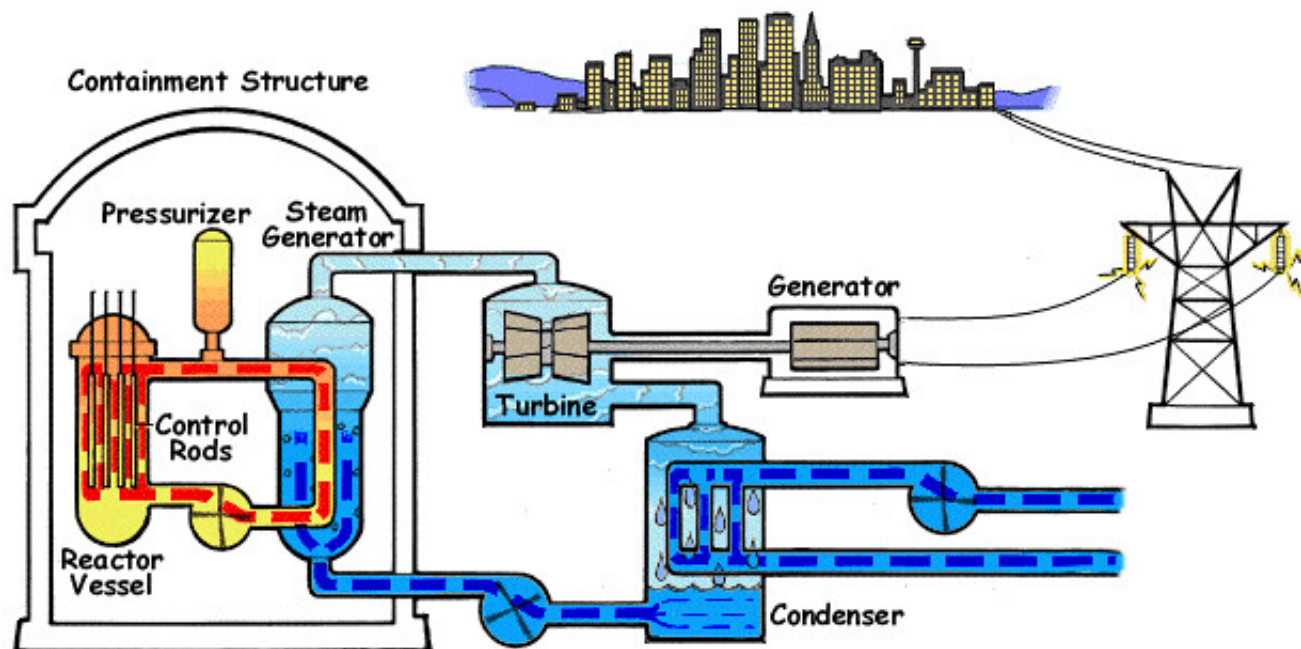




Correlation of Corrosion and Activity Build-up in Structural Components in Relation to the Swedish Nuclear Utilities Requirements

Master's thesis, KBTX12, in Nuclear Chemistry and Materials Chemistry, MPMCN

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CHALMERS UNIVERSITY OF TECHNOLOGY
Gothenburg, Sweden 2020
www.chalmers.se

MASTER'S THESIS, KBTX12, 2020

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This master's thesis project is performed as a collaboration between Ringhals AB nuclear power plant (shortened Ringhals) and the Department of Chemistry and Chemical Engineering, Nuclear Chemistry, Chalmers University of Technology.

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Cover: A schematic figure of a pressurized water reactor connected to a powerline transporting electricity to a city. Available from: <https://www.flickr.com/photos/69383258@N08/47051510834>

Gothenburg, Sweden 2020

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Abstract

In the nuclear industry there are specific requirements which refers to materials that are subjected to radiation and/or primary water environment. One important requirement for materials in nuclear power plants is the amount of cobalt (Co) present in the materials that are in contact with primary water exposed to radiation. The Co present in the materials can be activated to the activation product ^{60}Co by the combined effect corrosion and neutron capture. Since ^{60}Co also causes an activity build-up in the structural components, it is important to see if the strict requirements on the Co level for systems, structures and components that can be exposed to direct radiation are relevant and fulfilled to be within Ringhals framework of nuclear-specific requirements. This was one of the parts the project aimed to investigate. These requirements differs depending on where the material or component is located and how large the surface area is. The subject is also important to investigate from an activity production and radiation safety point of view since a higher radioactive build-up could cause higher doses to personnel. The second aim of the project was therefore to investigate what Co content is reasonable from an "As Low As Reasonably Achievable" (ALARA) and operational perspective.

Literature studies, collecting data and information from reports and documents at Ringhals and estimation calculations of Co levels in the primary circuit were performed. By comparing it to measured chemistry data at Ringhals it was seen that regarding materials and components used inside the reactor pressure vessel (RPV) and its internals, the requirement of 0.05 wt% cobalt is still adequate and should be kept strict. Outside the RPV and its internals, it was seen that the requirement could be less strict if the surface area is $\leq 10 \text{ m}^2$. This could potentially be applied to some larger surface areas as well if the corrosion rate could be kept low. It was also seen that it is of great importance to keep the low cobalt content in the steam generator tubings since most of the Co is released from there.

Keywords: Ringhals, Cobalt, Nickel, Cobalt-58, Cobalt-60, Cobalt requirements in materials, Corrosion, Activity build-up, Pressurized water reactor, PWR, Swedish Nuclear Utilities Requirements, TBM

Acknowledgements

I would firstly like to thank Björn Forssgren and Pål Efsing for being my supervisors during my master's thesis as well as providing me the opportunity to perform it at Ringhals. I would also like to thank both them and my examiner Teodora Retegan Vollmer for taking the time when I have needed help.

Besides my supervisors and my examiner there are many others I would like to give my appreciation to. The following persons are employees at Ringhals. A big thank to Anders Hultin and Fredrik Andreasson for providing me with data about components in the system. Thank you to Fredrik Ehrlén, Madelene Johansson, Torgny Svedberg, Kari Anttila, Angelica Andersson, Kent Sletten and Jimmy Hägg for attending meetings where all have been sharing their thoughts about the project, as well as given me more detailed insight in there respective specialties. A big thank you to Marcus Fredin, Lotta Björklund and Erika Turesson for letting me follow you at the chemistry department and inside the plant to see how the sampling of water and filter samples are performed at Ringhals. I would also like to thank Bernt Bengtsson, Olof Gottfridsson, Bengt Svensson, Erik Börjesson, Gustav Settervik, Åsa Henning, Berit Thoren, Besart Rexhaj, Veine Victorson, Per-Olof Aronsson, Anders Jonsson, Sara Almgren, Christina Andersson and Martina Larsson for helping me with administrative things, forwarding me to other persons and/or for providing me with some reports, thoughts and values of interest. Besides all these persons I would also like to give a big thank you to the personnel at the department GN-UQ, where this master's thesis were performed, for a nice time at Ringhals.

The last persons I would like to thank is Mattias Olsson at Forsmark, Maja Boström at Jernkontoret, Katarina Thyrestam and Carl-Filip Lindahl at Sandvik AB and Nina Cromnier and Andreas von Schmalensee at SSM for providing me with reports of interest and for answering questions I have had. Thank you to all mentioned persons at Ringhals, Forsmark, Jernkontoret, Sandvik AB and SSM for taking the time to help me during my master's thesis.

Hanna Svensson, Sweden, June 2020

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List of Abbreviations and Symbols

In the following section, a compilation of abbreviations and symbols used in the report are listed. All abbreviations are introduced in the text when first being used, and the reader can always come back to this page if the reader needs to see it again without needing to look for it in the text.

Abbreviations and Descriptions

A600 Alloy 600

A690 Alloy 690

AISI American Iron and Steel Institute

ALARA As Low As Reasonably Achievable

ASME American Society of Mechanical Engineers

BWR Boiling Water Reactor

CAD Computer-Aided Design

Crud Chalk River Unidentified Deposit. Crud is often defined as corrosion products deponated on heated primary surfaces. When precipitated on fuel surfaces it becomes activated and could be spread in the primary system causing increased dose rates. Crud could be soluble but a solid of spinel is the most common formed corrosion products.

CS Control System

FCC Face Centered Cubic

FWHM Full Width at Half Maximum

HPGe High Purity Germanium

LWR Light Water Reactor

manSv manSievert, a unit for collective dose

mdm $\text{mg dm}^{-2} \text{ month}^{-1}$, a unit for corrosion

mSv milliSievert, unit for dose

PRZ Pressurizer

PWR Pressurized Water Reactor

R3 Reactor unit 3 at Ringhals

R4 Reactor unit 4 at Ringhals

RC Reactor Coolant

RCC – M Design and Construction Rules for the Mechanical Components of PWR Nuclear Islands
(*French: Règles de Conception et de Construction des Matériels Mécaniques des Îlots Nucléaires PWR*)

RCP Reactor Coolant Pump

RCS Reactor Coolant System

RPV Reactor Pressure Vessel

SAM Surface Activity Measurements

SG Steam Generator

SSM Swedish Radiation Safety Authority (*Swedish: Strålsäkerhetsmyndigheten, SSM*)

TBM Technical Regulations for Mechanical equipment (*Swedish: Tekniska Bestämmelser för Mekaniska anordningar*)

Nuclides (in the form AX or $X-A$, where A is the mass number and X is the element)

^{110m}Ag Silver-110m

^{58}Co Cobalt-58

^{59}Co Cobalt-59

^{60}Co Cobalt-60

^{93}Mo Molybdenum-93

^{58}Ni Nickel-58

^{124}Sb Antimony-124

^{235}U Uranium-235

Denotations

$\bar{\nu}$ Antineutrino

ε Electron capture

$\sigma_{n,p}$ Reaction cross section for the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ reaction

σ_{reaction} Reaction cross section

φ Particle flux

φ_{fast} Fast neutron flux

φ_{thermal} Thermal neutron flux

A_a Activity of nuclide a

A_i Surface area of material i

$C_{j,i}$ Composition of element j in material i

Conc_j Concentration of element j

$\text{Conc}_{j,\text{chem}}$ Concentration of element j , based on measured chemistry data

k_a Production rate of nuclide a

M_j Molar mass of element j

M_k Molar mass of naturally occurring nickel isotope k

$m_{j,\text{chem}}$ Released mass of element j , based on measured chemistry data

$m_{j,i}$ Released mass of element j from material i

$m_{\text{released},i}$ Total released mass of material i by corrosion

n Neutron

$N_{j,i}$ Number of atoms of element j from material i

$N_{t,b,i}$ Number of atoms of nuclide b from material i

$N_{t,b}$ Number of target atoms of nuclide b

p Proton

$r_{corr,i}$ Corrosion rate of material i

$t_{1/2,a}$ Half-life of nuclide a

t_{cool} Cooling time

t_{irr} Irradiation time

t_{oc} Time of one operating cycle at Ringhals

X_b Fractional abundance of nuclide b

X_k Fractional abundance of naturally occurring nickel isotope k

$\dot{m}_{j,i}$ Mass release of element j from material i

$\dot{m}_{released,i}$ Total mass release of material i by corrosion

$\dot{m}_{waterflow}$ Water mass flow

Constants

$\sigma_{n,\gamma}$ Reaction cross section for the $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ reaction 37.2 barn or $3.72 \times 10^{-23}\text{ cm}^2$

$M_{^{58}\text{Ni}}$ Molar mass of ^{58}Ni $57.935347\text{ g mol}^{-1}$

$M_{^{59}\text{Co}}$	Molar mass of ^{59}Co	$58.933198\text{ g mol}^{-1}$
M_{Co}	Molar mass of elementary cobalt	$58.933198\text{ g mol}^{-1}$
M_{Ni}	Molar mass of elementary nickel, weighted with respect to all naturally occurring nickel isotopes	$58.69335111\text{ g mol}^{-1}$
N_A	Avogadro's number	$6,02214129 \times 10^{23}\text{ atoms mol}^{-1}$
$t_{1/2, ^{58}\text{Co}}$	Half-life of ^{58}Co	70.86 days
$t_{1/2, ^{60}\text{Co}}$	Half-life of ^{60}Co	5.2712 years

1

Introduction

In the following section background, purpose and aim, question formulations and delimitations of the master's thesis will be presented.

1.1 Background

Ringhals nuclear power plant, located in Väröbacka on the west coast of Sweden, was until the end of 2019 the largest power plant in Scandinavia. In 2019, Ringhals operated four reactor units - one of them is a so called boiling water reactor (*BWR*) and the other three are pressurized water reactors (*PWR*). The BWR unit is also referred to as reactor unit 1 and the PWRs as reactor unit 2, 3 and 4. In the end of 2019, unit 2 commenced into coast down as a consequence of fuel loading during the final operating cycle in preparation of the final shut down on December 30th in 2019 [1]. Similarly, unit 1 will end its commercial operation in the end of 2020 and be decommissioned to obtain final shut down of that reactor. The final shut down of the two reactor units will therefore result in that Ringhals will operate two PWR reactors after the year of 2020, namely reactor unit 3 and 4 which are referred to as R3 and R4 in this report.

In the nuclear industry there are specific requirements (beside those who applies to the design, manufacture and conformity assessment of stationary pressure equipment) which refers to materials that are subjected to radiation and/or primary water environment. One important requirement for materials in nuclear power plants is the amount of cobalt (Co) present in the materials that are in contact with primary water exposed to radiation. Cobalt can be found in for example valve seats, guide pins and similar items in reactor vessels. It can also be found as an impurity in stainless steel and nickel (*Ni*) base alloys as well as in the reactor pressure vessel manufactured by low alloy steel. The cobalt present in the materials can be activated to the activation product ^{60}Co by the combined effect corrosion and neutron capture [2].

Since ^{58}Co is produced as a result of the activation of nickel, see §2.2, it can be determined that this cobalt isotope comes from corrosion of stainless steel and nickel base material, whereas ^{60}Co mostly comes from corrosion of cobalt-rich materials. Cobalt-rich materials can be for e.g. hardfacing materials used inside as well as outside the reactor vessel such as Stellite [2]. Both isotopes, but mainly ^{60}Co [3, 4], increases the inventory of dose-producing species and may result in increased dose to personnel in connection with maintenance and control work.

Since ^{60}Co also causes an activity build-up in the structural components [3], also known as induced activity, it is important to see if the strict requirements on the cobalt level for systems, structures and components that can be exposed to direct radiation are relevant and fulfilled to be within Ringhals framework of nuclear-specific requirements. This type of activity is more problematic for upcoming changes and/or demolition of the material.

1.2 Purpose and Aim

The purpose of the project is to investigate the correlation of corrosion and irradiation on activity build-up in structural components at Ringhals to see if 1) the framework of nuclear-specific requirements presently used is still adequate when Ringhals will operate PWR technology only, and 2) to investigate what Co content is reasonable from an "As Low As Reasonably Achievable" (*ALARA*) and operational perspective. These two purposes are therefore also the question formulations for the master's thesis. The activation of cobalt in the alloys but also an assessment of corrosion products and their activation and possible dissemination will be studied.

The aim of the master's thesis work is therefore also to provide a basis that can be used when updating requirement specifications.

1.3 Delimitations

Since both reactor units 1 and 2 at Ringhals will be permanently shut down, as previously mentioned, only PWR units will be operated after year 2020. Because of this, one delimitation of the project is to concentrate on the PWR technology.

Another delimitation made is to study only one of the PWR units. This delimitation is made to be able to fulfill the purpose and aim of the project in the amount of time set for the master's thesis, i.e. 20 weeks. The PWR unit chosen to be studied is R3. The study will only be performed on the primary circuit in R3 and not on auxiliary systems since the question formulation only affects the primary circuit with connected service systems such as the pressurizer (*PRZ*), residual heat removal system (*RHR*) and chemical and volume control system (*CVCS*). What type of sample points that are of interest is determined during the literature study in cooperation with the supervisors and employees at the chemistry department at Ringhals.

2

Theory

2.1 Pressurized Water Reactor (PWR) Technology

At Ringhals there are three PWRs, but only two of them are still in use [1], R3 and R4.

A PWR is of a so-called light water reactor (*LWR*) type, which is a reactor using normal water (H_2O) as coolant. [5, 6, 7]. The water also acts as a neutron moderator which means that it reduces the speed of fast neutrons, resulting in that the neutrons instead becomes thermal. At Ringhals, sea water is used as main coolant of the secondary circuit. The PWR reactor unit consists of multiple components, a lot of piping systems and more. The main parts are the reactor pressure vessel (*RPV*), the PRZ, steam generators (*SG*), turbine, electric generator, condenser, valves and different pumps such as the reactor coolant pumps (*RCP*) and the feed water pumps.

2.1.1 The Primary Circuit in a PWR

A PWR circuit can be illustrated as shown in Figure 2.1 [7], with a primary and secondary circuit [6, 7]. The primary circuit in a PWR is a separate system which means that the reactor water never gets in contact with the secondary circuit. As can be observed in Figure 2.1, the primary circuit (shown as dark grey parts in the figure) consists of the RPV, the PRZ, SGs, RCPs (mentioned as pump in Figure 2.1) and the primary loops. The outlet from the RPV is called hot leg, the outlet from the SG is called cross over leg and the outlet from the RCP is called cold leg. All these components are located in the reactor building, also called containment. Even though Figure 2.1 is a very simplified picture of the process system, it is an acceptable simplification for the purpose of this project.

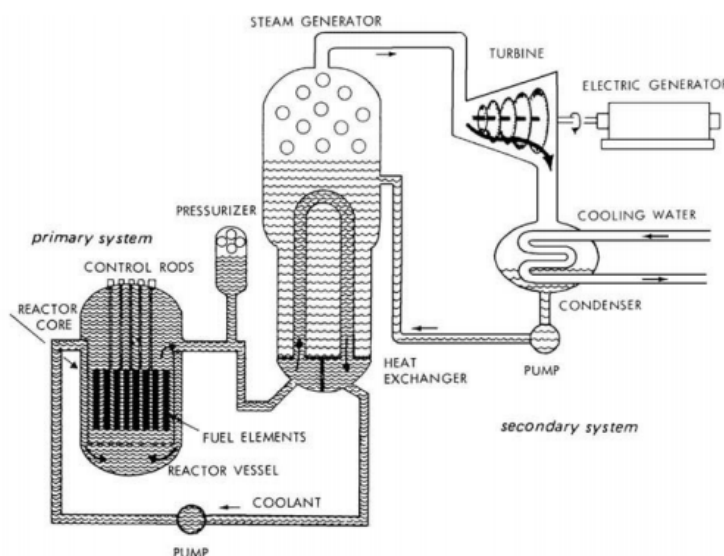


Figure 2.1: The primary and secondary circuit in a PWR [7]. Reproduced with permission.

In Swedish nuclear power plants, the fuel is made out of enriched uranium (U) with approximately 2-5 % of the fissile isotope ^{235}U [5, 6, 7]. The enriched uranium is then transformed to uranium dioxide (UO_2) in powder form via chemical processes, which in turn is made into cylindrical pellets by compaction/pressing. These pellets are then sintered at high temperature to give them high density. After grinding of the final fuel pellets, they are stacked together in zirconium alloy tubes and are then called fuel rods. The zirconium alloy tubes can also be called fuel cladding, and it is the outer layer of the fuel rods that is exposed to the reactor coolant water on one side and the nuclear fuel on the other. When the fuel rods are assembled in bundles, they are called fuel elements and these make up the core of the reactor. In R3, there are 17×17 fuel rods in each bundle. There are also 157 positions for fuel assemblies in each PWR reactor at Ringhals. A normal length of the bundles is 12 feet which is about 4 meters but even longer ones exist.

The fission of atomic nuclei occurs in the core of the reactor and since fission results in a release of neutrons, this means that a lot of neutrons exist in the core. The fuel elements, together with control rods, control actuators/CRD, and inlet and outlet for the reactor coolant water are the main components in the RPV. The so-called internal parts of the RPV, whose aim is to regulate the water flow, are a grid support for the core and provide structural strength and coolability and ensure that the fuel assemblies themselves always maintain a geometry that allows this to be met. Other internal parts of the reactor with large surface areas are the baffle assemblies and core barrel. The internal structures are found around the core, and are therefore also exposed to a lot of neutrons. The internal parts are made of stainless steel.

To the RPV of the reactor type Ringhals employs, there are three reactor coolant loops (also called primary loops) connected, each one containing a SG and a RCP [5, 6, 7]. The SG is used as a heat exchanger where the hot reactor water from the hot leg enters in the bottom of the SG. The reactor water then flows through U-formed tubes in the primary side of the SG, where it leaves heat to the condensate formed in the secondary side of the SG. This heat exchange results in the formation of steam which exits in the top of the SG and then continues to the turbine and electric generator where electricity is produced. The reactor water then exits the SG in the bottom to the cross over leg which continues through to the reactor coolant pump and the cold leg. All three loops have a common pressurizer whose function is to adjust the pressure in the primary circuit so that the water is not boiling [6, 7].

2.1.2 Water Chemistry

Ringhals traditionally works at a 12 month operating cycle, resulting in approximately 330 days of operation per year and throughout the operating cycle the water chemistry varies. There are some different parameters which determine the water chemistry in a PWR such as temperature, pH, lithium/boron (Li/B) dosage and hydrogen gas (H_2) dosage [8]. The operating temperature and pressure in the PWR units at Ringhals are around 280-320°C and 154 bar(o), where *o* means over pressure (the Swedish unit: bar(ö)) [3].

In order to regulate the reactivity in the core, boric acid (H_3BO_3) is added. This results in an acidic environment and therefore also a low pH. The pH is regulated by adding lithium hydroxide (LiOH) to the water [3]. There is a continuous discussion about which pH is the most optimal for PWR operation but it is believed that a pH_{300} , that is the pH at 300°C, between 7.1 and 7.4 is the most optimal. The pH is controlled by the cycle burn up and varies throughout the operating cycle due to changed water chemistry, often with a value of around 7.1-7.2 in the beginning. This is due to that the amount of B and Li is around $\leq 1\,800$ ppm and 5 ppm respectively in the beginning of the operation cycle. The

amount of B is then decreased by approximately 4 ppm per 24 hours down to the minimum level of 4 ppm boron. 5 ppm of Li is the maximum amount allowed determined by the fuel suppliers since a higher amount can result in cracks in the fuel. The amount of Li is therefore held constant at 5 ppm in the beginning, until the pH has reached around 7.4. The Li is then decreased with a minimum level of around 0.75 ppm in the end of the operating cycle [8]. The reason for why the pH needs to be kept alkaline is since corrosion of iron (*Fe*) and Ni alloys are the lowest in alkaline conditions. LiOH is therefore added to the water to counteract for the acidic effect caused by the H_3BO_3 [2, 3].

The PWR operates under reducing conditions due to addition of H_2 [2]. This operating condition is used to gain an excess of dissolved hydrogen (*DH*), since an excess of DH can reverse or suppress radiolysis of water where oxygen (O_2), H_2 and hydrogen peroxide (H_2O_2) is formed. By operating under reducing conditions, the corrosion and the risk for so-called stress corrosion cracking (SCC) in the systems, structures and components of the nuclear reactor are also decreased. There is however an optimum regarding the hydrogen content which results in a maximal crack growth rate. The optimum can be seen in a Pourbaix diagram, which is a diagram showing the potential versus the pH and also possible stable phases of an aqueous electrochemical system. For nickel base materials, the optimum is found where the transition between Ni and nickel oxide (*NiO*) is. "Normal operation values of hydrogen content in a PWR is around 40-50 mL/kg hydrogen" [English translation] [3, 8].

2.1.3 Zinc Dosage

One way to lower the ^{60}Co content in oxide layers of components in a reactor, and therefore also the dose rates to personnel during shut down, is to use a method called zinc dosage. The result of the method can be shortly summarized by that zinc changes the properties of the oxides on the fuel and on primary surfaces [3] by blocking free positions to be occupied by ^{60}Co in the spinel oxide structure. This method is not used in the primary circuit at Ringhals because there are not enough positive arguments from an economic and radiation point of view. If the reader wants to know more about zinc dosage, two good references to read are [3] and [9].

2.1.4 Shut Down Release Clean Up Using Hydrogen Peroxide Dosage

To reduce activity and dose rates during refuelling and maintenance work, Ringhals uses H_2O_2 dosage as pretreatment during cold shut down and refueling. To be able to use this method, some preparations are made before shut down to refueling. The changes are made in the primary chemistry, first by adding H_3BO_3 to assure that the system is subcritical in connection with refueling. The second step is to use an ion exchanger to remove Li. These two steps creates a reducing environment to support ^{60}Co in out of core surfaces to easier release into the water and be cleaned. To avoid risks of explosions in an open reactor system, hydrogen is also removed as a third step. Finally at cold shut down, that is a temperature $< 93^\circ\text{C}$, H_2O_2 is also added to assist the fuel crud to dissolve (mainly Ni and ^{58}Co). Ion exchangers are then used to clean the reactor water from released activity [2].

2.2 Production of the Radioactive Cobalt Isotopes ^{60}Co and ^{58}Co

The two cobalt isotopes which are of interest in this project are, as previously mentioned, ^{58}Co and ^{60}Co which are radioactive. Some concepts which are good to know for this section are what isotopes, a nuclide and a reaction cross section means. A particular chemical element always has the same number of protons, but can have different numbers of neutrons. The different variants, protons plus

neutrons, are called the isotopes of that element. One example is deuterium (2H), also known as heavy water, and tritium (3H). Both are of the chemical element hydrogen, but deuterium has one neutron whereas tritium has two neutrons giving them the mass numbers of 2 and 3, respectively. A nuclide has a specific number of nucleons (a nucleon is a proton or neutron). The reaction cross section of a nuclear reaction can be described as a probability that the reaction in question will occur and is given in the unit barn or cm^2 , where 1 barn is equal to 10^{-24} cm^2 . To get a better understanding of these isotopes, the production of the isotopes are described in this section.

^{60}Co can be formed by neutron activation of ^{59}Co , as shown in Reaction (i). Nuclear reactions are often written as in Reaction (i), and means that the cobalt isotope ^{59}Co reacts with a neutron (n) forming ^{60}Co and emits γ -rays. ^{59}Co is non-radioactive and the only naturally occurring and stable cobalt isotope [10], which means that the abundance of ^{59}Co is 100 %. The molar mass of ^{59}Co is $58.933198 \text{ g mol}^{-1}$ [11] and the reaction cross section for Reaction (i), $\sigma_{n,\gamma}$, is 37.2 barn or $3.72 \times 10^{-23} \text{ cm}^2$ [10].



^{58}Co can be formed by neutron activation of the naturally occurring and stable nickel isotope ^{58}Ni [10], as shown in Reaction (ii). Reaction (ii) means that ^{58}Ni reacts with a neutron forming ^{58}Co and a proton (p) is emitted.



^{58}Ni is non-radioactive with a molar mass of $57.935347 \text{ g mol}^{-1}$. ^{58}Ni has an abundance of 68.077 %, and is not the only naturally occurring Ni isotope. This means that the molar mass of naturally occurring Ni in a material must be weighted as shown in Equation (2.1), where M_{Ni} is the weighted molar mass for elementary Ni isotopes in g mol^{-1} , M_k is the molar mass of naturally occurring nickel isotope k in g mol^{-1} and X_k is the fractional abundance of naturally occurring nickel isotope k . The four Ni isotopes which are naturally occurring except ^{58}Ni are ^{60}Ni , ^{61}Ni , ^{62}Ni and ^{64}Ni . Using data for each nuclide in Equation (2.1) [11], this gives a weighted molar mass of $58.69335111 \text{ g mol}^{-1}$.

$$M_{Ni} = \sum_{k=1}^5 (M_k X_k) \quad (2.1)$$

The reaction cross section for Reaction (ii), $\sigma_{n,p}$, is not set. Instead, the reaction cross section can be determined from experimental data in charts showing how the cross section depends on the energy of the neutrons that takes part in the reaction [12].

2.2.1 Neutron Activation/Neutron Capture

The process occurring when ^{59}Co and ^{58}Ni forms the radioactive isotopes ^{60}Co and ^{58}Co respectively is called, as previously mentioned, neutron activation or neutron capture. Since neutrons have no charge they can easily enter atomic nuclei compared to charged particles [7], since repulsion occurs as charged particles nears the nucleus. This means that neutrons existing in the process system are captured by atomic nuclei in e.g. cobalt or nickel, which exist in the materials and as corrosion

products in the system. This results in that the atomic nuclei that have captured a neutron gets heavier due to more nucleons in their nuclei than before. When the nuclei becomes heavier, they also get excited. After excitation, the nuclei often go through a radioactive decay. In Reaction (i), thermal neutrons are most likely involved in the reaction since thermal neutrons are captured through (n, γ) reactions [13].

2.3 Decay of the Radioactive Cobalt Isotopes ^{60}Co and ^{58}Co

Before explaining the decay of the two cobalt isotopes of interest, there are three concepts which are good to know. These are radioactive decay, a mother nuclide and a daughter nuclide. Radioactive decay, or disintegration, is a phenomenon where an unstable nucleus in a radionuclide decays to another nuclide by emitting radiation [7]. A mother nuclide, or also called parent nuclide, is a nuclide before disintegration. The decay product which is formed after disintegration of the mother nuclide is called the daughter nuclide. The time it takes for the amount of mother nuclide to reduce to half of the starting amount is called the half-life ($t_{1/2}$) of the mother nuclide.

There are different ways for a radioactive nuclide to decay, for example by alpha (α) decay or beta (β) decay [7]. β -decay is a general name for three different decay types, such as electron capture (ϵ). This will be further explained in §2.3.1. If the nucleus of the daughter nuclide is left in an excited state after an α - or β -decay, energy will be released in the form of a gamma (γ) emission or by internal conversion.

^{60}Co can decay to ^{60}Ni by a beta decay called negatron (β^-) emission, with gamma lines at 1 173 keV and 1332 keV and with a $t_{1/2}$ of 5.2712 years (a , annual in Reaction (iii)) [10]. The decay of ^{60}Co can be observed in Reaction (iii), $\bar{\nu}$ is an antineutrino.



The decay of ^{58}Co to the iron isotope ^{58}Fe by electron capture can be observed in Reaction (iv), and it has a gamma line at 811 keV and a $t_{1/2}$ of 70.86 days (d in Reaction (iv)) [10].



2.3.1 Beta Decay

β -decay is a collective name for three different types of decay, namely negatron (β^-) emission, positron (β^+) emission and electron capture (ϵ) [7].

As could be seen in Reaction (iii), a β^- -emission takes place when ^{60}Co decays to ^{60}Ni . β^- -emission occurs when a nucleus is unstable due to too many neutrons. This results in that a neutron from the mother nuclide transforms into an antineutrino, a proton and a β^- -particle which is a fast and highly energetic electron. The proton is kept by the daughter nuclide and the β^- -particle and the $\bar{\nu}$ are emitted, which means that a new element is formed. During the β^- -emission, an antineutrino is also released.

Electron capture is a phenomenon which occurs when a nucleus is unstable due to too many protons in comparison to number of neutrons. An electron in an inner orbital is therefore withdrawn from its position into the nucleus, which together with a proton forms a neutron and releases energy in form of γ -rays.

2.3.2 Gamma Emission

When a nucleus goes through an α - or β -decay, a γ -emission can also occur [7]. This is for example seen in Reaction (iii) and (iv), where ^{58}Co and ^{60}Co decay through different types of β -decay with a γ -emission as well. γ -emission, or γ -rays, is an emission of highly energetic photons. This means that there is an excitation energy and this energy can be released through, as in this case, a γ -emission.

2.3.2.1 High Purity Germanium (HPGe) Detector

To be able to measure the γ -emission from different nuclides a high purity germanium (HPGe) detector can be used [7]. A HPGe detector is a detector which is nuclide-specific to γ -radiation due to good resolution on the γ -energies. The peaks in an obtained spectrum from a HPGe detector have a so-called full width at half maximum (FWHM). FWHM is the width at half the amplitude of the peak, see Figure 2.2. A measurement with respect to γ -energies using a HPGe detector yield peaks with a FWHM of around 2 keV, which means that the peaks are quite narrow. This in turn results in that peaks that differ with 1-2 keV can be separated and identified separately in the spectrum.

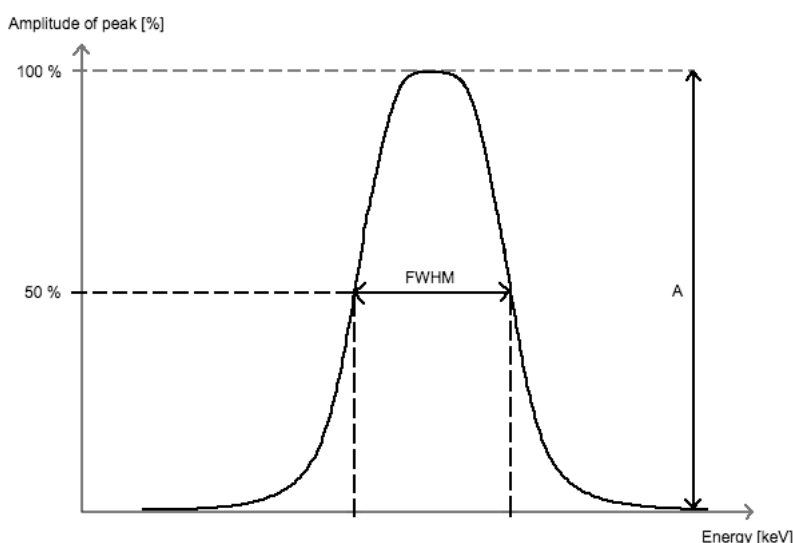


Figure 2.2: A drawing of where to find FWHM of a peak in a γ -spectrum. A denotes the full amplitude of the peak.

2.4 Steel Manufacturing

The structural components of a PWR are mainly manufactured using steels in a variety of grades and the steels can be produced using two different starting points: by using iron ore based material or by using recycled material which is also known as scrap metal. The two principles are described in the following sections. Whether the one or the other method is used in the steel production, both of them start with iron ore as the base [14, 15]. The iron ore, often together with coke and other additives in some forms, is put into a blast furnace which smelts the ore. The next step is what makes it an ore based or scrap metal based steel. "The Swedish production of stainless steel is today based

on scrap metal" [English translation] [16] and low alloy steel is mainly produced via "virgin steel" or iron ore based for e.g. at SSAB in Oxelösund [24].

2.4.1 Primary Steelmaking Step: Iron Ore Based Steel

The first step in this process, which is the making of crude steel, is explained in §2.4. When using the ore based material process for the production of steel, the next step is using a process called basic oxygen furnace (BOF), also called basic oxygen steelmaking (BOS) [14, 15, 17]. The conversion of liquid iron to steel through the BOF process is called the primary steelmaking step.

The molten iron ore is in this process transferred to an oxygen steel converter, see Figure 2.3. In the converter, calcium oxide (CaO , also called lime) and oxygen are blown through the iron [18, 19]. This is to remove potential impurities by oxidizing them, resulting in that the impurities forms a slag which floats on top of the liquid iron. The blowing creates an exothermic reaction and scrap metal is therefore sometimes added to cool down the hot metal. By tilting the oxygen converter to a horizontal position, the slag containing the impurities can then be poured off. The metal that is left can once again be blown through with oxygen and the metal is then removed from the converter.

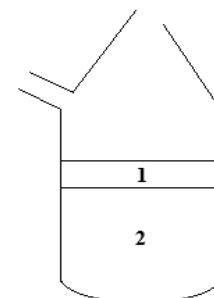


Figure 2.3: A simplified drawing of an oxygen steel converter, where phase 1 is the impurities in the form of a slag and phase 2 is the smelt iron.

2.4.2 Primary Steelmaking Step: Recycled Material Based Steel

The second method to produce steel is to use recycled material, i.e. to reuse scrap metal. This is done by mixing the molten iron from the blast furnace with scrap metal, transferring it to the vessel of an electric arc furnace and then melting it all [14, 15, 18]. The electric arc furnace uses electric arc conducted from graphite electrodes that are lowered down into the scrap metal. By doing this, the scrap metal melts due to that the electric arc converts electrical energy into heat. Temperatures up to 3 500°C can be achieved in the arc and in the steel melt the temperature can be up to 1 800°C. Besides the graphite electrodes, which are positioned on a removable cover, another main component in the electric furnace is the tilting system. The tilting system is used in the end of the process, when the desired chemical composition and steel temperature has been achieved, so that the steel can be poured out from the furnace vessel. When using scrap metal in the production of steel, a very precise composition and good "purity" of the metal is achieved since it is possible to know the composition of the reused steel before using it in the electric arc furnace.

2.4.3 Secondary Steelmaking Step

The next step, which is called the secondary steelmaking step, is used for both the ore based and scrap metal based steel production [14, 18]. In this step, the metal melt is refined and treated to give it the chemical composition needed for the desired steel material. This is done by for example adding alloying elements, removing impurities or homogenizing the melt.

2.5 Metallic Materials Existing in the Primary Circuit

Commonly used materials in the primary circuit of a PWR unit are low alloy steel, austenitic stainless steel and nickel base alloys [3]. Low alloy steel is used in main pressure retaining components and vessels, e.g. the RPV, the SGs and the PRZ. Austenitic stainless steel is used as material for many

internal parts, in the primary loops and in the RPV as weld cladding on the ferritic RPV steel. Nickel base alloys are mostly used as material in the SG tubes [20], but also in internal parts [3]. The cobalt-rich material Stellite also exists, but in small amounts, in the primary circuit.

One alloying element that is used in stainless steel and which is the major component in nickel base alloys is nickel, see §2.5.1 and §2.5.3. When mining nickel or copper (*Cu*), which is also used as alloy element in nickel base materials, cobalt is often mined as a byproduct [21]. This means that besides being the major element in cobalt-rich materials, cobalt can also exist as an impurity in some materials. Sweden's geological survey also says in an article [21] that "according to EU, the amount of recycled cobalt available to manufacturers is zero percent" [English translation], which results in that cobalt is mainly taken from mine production.

2.5.1 Stainless Steel

Stainless steel is a collective name for metallic materials that are iron-based alloys, containing a certain amount of chromium (*Cr*) [3, 16, 22, 23]. Without chromium in the material the iron would corrode, i.e. rust, and alloys must contain at least 10 to 12 weight% (wt%) of chromium (depends on the content of molybdenum (*Mo*)) to be considered as "stainless" and to form the protective properties. An addition of 12 wt% chromium in stainless steel protects the material from corroding since the material then forms a chromium-rich oxide film on the surface, see Figure 2.4. The protective film formed is called "passive film" and it is formed spontaneously when the material is in contact with enough amount of oxygen.

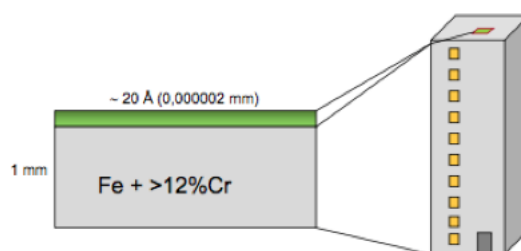


Figure 2.4: The passive film on the surface of stainless steel [16]. Reproduced with permission.

Stainless steel can also contain other elements or alloy additives such as carbon (*C*), nickel, molybdenum, manganese (*Mn*) or nitrogen (*N*) to give it specific properties, e.g. to give the material further possibility to increase the efficiency of the "passive film" and therefore also to increase the protection against corrosion and the targeted mechanical properties [3, 16, 22, 23]. However, what needs to be taken into account when changing the composition of the material is that the material also needs to be for e.g. weldable and possible to manufacture. This means that not all possible combinations of compositions can be used. One of the materials which are used a lot at Ringhals is the austenitic stainless steel [24] since it is great as structural steel in components due to its good weldability as well as good strength properties and processability at low temperatures [3, 25].

2.5.2 Austenitic Stainless Steel

In Figure 2.5 [16] a phase diagram of Fe-Cr-Ni alloy at 1 000°C is shown, where the compositions of the different phases can be seen. The austenitic stainless steel is a type of stainless steel containing chromium and nickel additives of more than 18 wt% and 8 wt% respectively, which also is a typical composition [3, 16, 26]. The steel with 18 wt% chromium and 8 wt% nickel is called 18/8-steel.

Austenite is a non-magnetic, high temperature phase of iron with a crystal structure as face centered cubic (FCC). If having nickel additives around 8-10 wt%, the austenite phase becomes stable even down to room temperature [3, 16, 26]. As mentioned in §2.5.1, other additives could be added to the alloy to obtain desired properties. Molybdenum is one element which is often added to austenitic stainless steel to improve the protection against corrosion of the material in chloride environments and reducing acid environments [3].

Since both stainless steels and austenitic stainless steels can have compositions with different percentages of added elements, the materials are assigned names according to different naming systems. For austenitic stainless steels, some of the systems used are the one from the American Iron and Steel Institute (AISI) and the one mostly used in Europe called EN [3, 16]. In AISI, the material is named with a three-digit system e.g. 304 and in EN the same material is named 1.4301. Sweden also has a naming system where the materials get a SS designation, but EN is often used instead. The material called 304 in AISI and 1.4301 in EN is referred to as SS 2333 in the Swedish naming system or as the 18/8-steel mentioned previously.

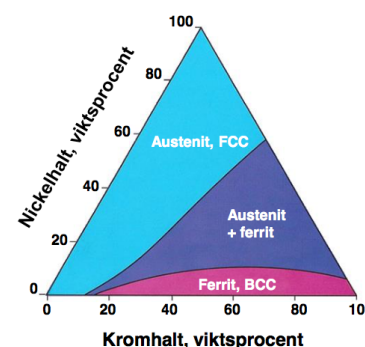


Figure 2.5: Phase diagram of Fe-Cr-Ni alloy at 1000°C [16]. Reproduced with permission.

2.5.3 Nickel Base Alloys

A nickel base alloy is a type of alloy consisting of a large portion of nickel, having an austenitic crystal structure as FCC [3, 27]. To give the material properties such as good strength, good corrosion resistance to reducing acid solutions as well as better resistance to sea water, to oxidizing solutions and under reducing conditions, nickel is often alloyed with elements such as iron, chromium, molybdenum and copper.

Typically used nickel base alloys in nuclear power plants are Alloy 600 (A600) and Alloy 690 (A690). A600 is an alloy with composition 72 wt% nickel, 17 wt% chromium, 9 wt% iron and 0.2 wt% aluminium (Al). Before, Ringhals had SG tubes made of A600 but when they replaced the SGs in 1995 they changed the SG tubes to ones made of A690 [3], all manufactured by Sandvik AB. The chemical composition of A690 for nuclear applications is ≥ 58 wt% nickel, 28-31 wt% chromium, 7-11 wt% iron, ≤ 0.04 wt% carbon, ≤ 0.50 wt% silicon (Si), ≤ 0.50 wt% manganese, ≤ 0.015 wt% sulphur (S), ≤ 0.50 wt% copper and ≤ 0.10 wt% cobalt [28].

2.5.4 Stellite

A material that is a major contributing factor to activated cobalt, in the form of ^{60}Co , if used in nuclear power plants is the cobalt-rich alloy Stellite [3, 4] which contains up to 50-60 wt% cobalt or more. Stellite alloys also contains alloy elements such as iron, nickel, chromium, tungsten (W), carbon, silicon and others. The Stellite alloy which is mostly used in nuclear power plants is Stellite 6, with has a typical alloy element composition of maximum 3 wt% iron, 3 wt% nickel, 56 wt% cobalt, 30 wt% chromium, 5 wt% tungsten, 1.25 wt% carbon.

Stellite is often used as material for the hardfacing of e.g. valves and at wear surfaces such as the clevises and support pins which are part of the support system to guide and connect the reactor internals to the correct position after removal and remounting within the RPV. The function of the clevises is to guide and restrain the reactor internals, thus keeping the core structure intact and coolable [29]. This means that there is a critical importance that the function of the material is of

high reliability [4]. Since the Stellite alloys contain about 50-60 wt% cobalt, using this material near the reactor could result in hotspots of activity on components. This is due to that parts of the material can be released through corrosion, friction or wear. The worn material, also called wear debris, is then transported in the process system to the core of the reactor where it can get activated by neutrons.

Stellite and Stellite 6 are materials with excellent properties such as good resistance against wear, corrosion and oxidation as well as good resistance to high temperature water [3, 4]. Because of this, there are currently no other known alloys available which have equally good tribological properties in the sense that it could replace the Stellite in the components [29]. Tribological properties means properties of surfaces in sliding or rolling contact and includes friction, abrasion and lubrication.

2.6 System of Regulations - PAKT

In Sweden there is a system of regulations and interpretation of the Swedish Regulation (*SSMFS*) called *PAKT* that has been collected and compiled by the Swedish Nuclear Power Companies jointly [30]. *PAKT* is part of the interpretation of *SSMFS* with additional requirements, such as various corrosion phenomena, aging effects and cobalt requirements. The *PAKT* documentation consists of a number of documents describing the technical regulations and specifications regarding use in the Swedish nuclear power plants. These documents are abbreviated *ABM* (General Regulations for Mechanical Equipment), *KBM* (Quality Regulations for Mechanical Equipment), *TBM* (Technical Regulations for Mechanical Equipment), *PBM* (Regulations for In-Service Inspection) and the *PAKT* definitions. *PBM* is divided into *PBM1* (Test Provisions for Mechanical Devices) and *PBM2* (Qualification Scheme for Qualification of OFP Systems in Sweden).

The document which e.g. explains the requirements of cobalt in materials is the *TBM* document [31], and the requirements listed in *TBM* will be further explained in §2.6.1. The *TBM* document consists of requirements for many other elements as well, but since the project focuses on ^{60}Co and its contribution to doses to personnel, only cobalt is considered from the requirements in *TBM* in this master's thesis.

Requirements in the *PAKT* and *TBM* documents apply to the Swedish nuclear power plants. There are therefore other systems of regulations which apply for other countries and parts of the world. The system of regulations which the United States of America and almost the rest of the world uses is called *ASME*. In *ASME*, most regulations are mechanical ones and information and regulations about cobalt content in materials are not defined. *RCC-M* is the French equivalent to *ASME* and is a so-called regulatory code (design code).

2.6.1 Requirements from TBM

The *TBM* document is divided into different chapters and sections, with two of the chapters being "§3 - Material, Design and Manufacturing and Installation" and "§4 - Equipment Specific Regulations" [31].

In the material chapter there is a section describing cobalt and the general requirements of it in materials. In this part of the *TBM* document it is mentioned that materials used in the RPV, in the internal parts of the RPV or for materials with a surface area of $>10 \text{ m}^2$, which is considered as a large area, the cobalt content must not exceed 0.05 wt%. The last mentioned, about large areas, must be true if water that circulates through the reactor could pass by or through the material without passing through an ion exchanger before entering the reactor vessel again.

If the surface area of a material instead is small, that is $\leq 10 \text{ m}^2$, and outside the RPV the requirement

of cobalt content in the material must instead not exceed 0.20 wt%. The requirement of maximum 0.20 wt% cobalt content refers to materials in contact with water which potentially could enter the reactor vessel. If the water passing by or through the material is passing through an ion exchanger before entering the reactor vessel, the requirement must not be achieved.

In the chapter about the equipment in the TBM document, the cobalt requirements for specific equipment is listed. In this section, cobalt requirements exist for materials in heat exchangers and, as previously mentioned, the internal parts of the reactor. For the heat exchangers, the cobalt content must not exceed 0.05 wt% in the parts which are in contact with primary water.

2.7 General Corrosion

As mentioned previously in §2.5 and its subsections, many of the metallic materials of interest have a good resistance against corrosion. Even though the resistance is good, corrosion still occurs since there is a thermodynamic driving force for it to happen. Corrosion is a type of phenomenon where a material reacts chemically or electrochemically when in contact with the medium it is surrounded by [3]. What then happens is that the material strives to transform to its stable oxide form by forming an oxide film on the surface of the material. One type of corrosion is called general corrosion. General corrosion means that the entire surface of the material corrodes when it is in contact with the corrosive medium. This due to that there are anode and cathode surfaces all over the material surface, which in turn results in a relatively even corrosion of the material. A corrosion rate can be expressed in the unit mdm, which is an abbreviation of $\text{mg dm}^{-2} \text{ month}^{-1}$.

2.8 Activity Build-Up

As previously mentioned in §2.1.1, there are excessive amounts of neutrons in the core which are released through fission. These neutrons can irradiate particles or ions existing in system flows and in the structural materials [32], resulting in an activity build-up in the process system [3]. The activated elements can then also accumulate in the process system resulting in hotspots with activity. The activity for nuclide a (A_a) can be calculated using Equation (2.2) [7], where t_{irr} is the irradiation time, t_{cool} is the cooling time and k_a is the production rate of nuclide a . t_{irr} , t_{cool} and $t_{1/2}$ must all be in the same time unit, e.g. seconds. Nuclide a is produced from nuclide b . Nuclide a is either ^{60}Co or ^{58}Co and nuclide b is either ^{59}Co or ^{58}Ni .

$$A_a = k_a \left[1 - \exp(-\ln(2)t_{irr}/t_{1/2,a}) \right] \exp(-\ln(2)t_{cool}/t_{1/2,a}) \quad (2.2)$$

By using Equation (2.3), k_a can be calculated. In Equation (2.3) [7], φ is the particle flux in particles $\text{cm}^{-2} \text{ s}^{-1}$ (in this case the particles are neutrons), $\sigma_{reaction}$ is the reaction cross section in cm^2 and $N_{t,b}$ is the number of target atoms of nuclide b .

$$k_a = \varphi \sigma_{reaction} N_{t,b} \quad (2.3)$$

2.8.1 Activation of Corrosion Products

A structural material can in contact with a corrosive environment corrode, which results in that corrosion products can be released. The corrosion products are then transported with the water into the rest of the system. When entering the core, the corrosion products get irradiated by neutrons,

activating the corrosion products. If it is nickel base alloys or cobalt alloys that corrode, then ^{59}Co and ^{58}Ni would be activated to ^{60}Co and ^{58}Co respectively after irradiation, as previously shown in Reaction (i) and (ii). This process will be referred to as activation of corrosion products.

2.8.2 Induced Activation

The second mentioned type of activation, i.e. irradiation of elements in the materials, will in this project be referred to as induced activation. Induced activation mostly occurs in components near the core of the reactor where most of the neutrons are and is the direct irradiation of structural components in the core zone such as all internal parts and the RPV wall. Induced activity is problematic for upcoming replacements and/or demolition of the material since a higher induced activity in the material means higher costs when the material is to be stored or deposited. The activated material could then be released from the surface by for example corrosion or wear, entering the rest of the system by transportation with the water. In the same way as described in §2.8.1, depending on what material is directly irradiated, ^{60}Co and ^{58}Co can be formed.

2.9 ALARA

ALARA is an abbreviation for "As Low As Reasonably Achievable" and is a concept which can be used in different applications. One of them is to preserve or reduce radiation doses to personnel to a reasonable level. Something to note is that ALARA does not mean that the subject or value of interest, e.g. the dose, needs to be zero if that is not reasonably achievable. The unit for ionizing radiation dose is Sievert (Sv). The Swedish Radiation Safety Authority (SSM) have set limits on how much radiation a person, as a worker or person in public, is allowed to receive as dose. The dose limit for a worker during a single year is maximum 20 mSv and the same limit is true for the lens of the eye [33]. Personnel working at nuclear facilities are usually receiving approximately 2 mSv per year as an average dose. The dose allowed to a person in the public as a consequence of emissions from the nuclear power plants is set to a maximum of 0.1 mSv per year [34].

3

Method

As mentioned early in the report, this master's thesis has been performed in cooperation with Ringhals. Since Ringhals is referred to as a protective object, there are restrictions about what internal information is allowed to be shared to the public. Because of this two versions of the report will be done; one "Chalmers-friendly" and one more specific for Ringhals. A "Chalmers-friendly" report means in this case a report which contains information that is allowed to be shared freely to the public according to the restrictions at Ringhals, and therefore also okay to be published by Chalmers University of Technology. In the "Chalmers-friendly" report, a modified reference system has been used for chapter 4, in the form "Ringhals reference #" where # is a number. This was discussed and approved by the examiner of this master's thesis beforehand.

The major part of this master's thesis has consisted of literature studies and collecting data for interesting sample points and materials at Ringhals, which has been studied during the project. Beside the theoretical part of the report which has been based on literature studies as well as collected data at Ringhals, a practical part has also been performed. The practical part was sampling as well as analyses of the samples, but it was determined early during the master's thesis that performing the actual sampling and analyses of water and filter samples to collect a lot of results would not give that much for the purpose of the project. This was due to that the sampling of interest are performed frequently by personnel at Ringhals which means that a lot of results in the form of collected data already exist. The sampling and analyses of water and filter samples were therefore only studied by following personnel at the chemistry department at R3 and R4 at Ringhals. This was to gain knowledge about how the sampling and analyses are performed but otherwise, the report is based on already gathered data at Ringhals. Another thing which was of interest during the master's thesis was results from the so called surface activity measurements (SAM) which are performed by personnel from the radiology and dosimetry department. The procedures for sampling, measurements and analyses are described in §3.3. All described measurements yield a result showing what γ -emitting radionuclides that are present.

Both the literature studies and the practical part were used as a way to gather information needed to fulfill the purpose and aim of the project.

3.1 Literature Studies and Data Collection

Because of the restrictions from Ringhals, internet, old course literature and course material was used for the theory, see chapter 2, to gain knowledge and information for the report since this type of information is considered as public. Most parts of the theory are also general information which means that internet and similar are better sources to find information at than via internal systems at Ringhals.

Another major part of the master's thesis was to collect data for relevant systems, sample points and materials at Ringhals. This was done by using databases at Ringhals such as their internal document database called *Darwin*, *LabVantage* (where chemistry data is stored) and *Origin* (where data can be gathered and plotted). Some information also existed in paper form in folders, which was also used during the literature studies and data collection. This type of information is not considered public, and it was therefore needed to be shared in a way that is allowed according to the restrictions at Ringhals.

In the part of the project where information and data have been collected, personnel from different departments at Ringhals have also been involved, since all departments and individual personnel have expertise in different areas. This was first done by calling the personnel of interest for a meeting. For the sampling, analyses of samples and trend interpretation, personnel from the chemistry department and the radiology and dosimetry department have been involved. For the ALARA perspective, personnel working with radiation protection have helped during the project. Personnel working at the department where this master's thesis has been performed have helped with data for components, materials and similar. Personnel from different departments have been involved to get a bigger picture of the problem and to gain knowledge from them all. At the first meeting, personnel from the different departments were gathered together to be able to have a discussion where inputs from the involved departments could be given. Later during the project, individual meetings with each department have been booked where more specific topics and delimitations have been discussed. During the project, personnel from other companies such as Forsmark and Sandvik AB have also been involved to answer questions along the way.

3.2 Calculations for Cobalt and Nickel in Primary Circuit

In this section, estimation calculations and calculations for the actual outcome of Co and Ni are presented. These calculations have been performed to estimate mass releases, concentrations and activities caused by Co and Ni from the materials stainless steel, A690 and Stellite to the reactor coolant system (RCS) water, to be able to compare it to measured chemistry data. The compositions of Co and Ni in the materials have been determined from material certificates and reports at Ringhals and will be referred to as *real compositions*. All calculations have also been performed using theoretical compositions of 0.05, 0.10, 0.15, 0.20, 0.25 and 0.30 wt% Co in stainless steel and A690, instead of the real wt% of Co in those materials. These calculations were performed to later estimate how the activity would be affected if having a higher Co content in the materials. Calculations were also made for one conservative and one realistic case since corrosion rates for each case were found in reports from Ringhals.

To calculate the total mass release of material *i* by corrosion, $\dot{m}_{released,i}$, in mg month⁻¹ Equation (3.1) was used. In Equation (3.1), $r_{corr,i}$ is the corrosion rate for material *i* in mdm and A_i is the total surface area of material *i* in dm². Material *i* is either stainless steel, A690 or Stellite.

$$\dot{m}_{released,i} = r_{corr,i}A_i \quad (3.1)$$

By multiplying Equation (3.1) with the operating cycle time, t_{oc} , as shown in Equation (3.2), the total released mass of material *i* by corrosion, $m_{released,i}$, during one operating cycle could be calculated. In Equation (3.1), $\dot{m}_{released,i}$ is in mg and t_{oc} is the time for one operating cycle in months. 30.416667 days was used as an average of days per month when converting t_{oc} from days to months.

$$m_{released,i} = r_{corr,i} A_i t_{oc} \quad \text{Note : } t_{oc} \text{ in [months]} \quad (3.2)$$

$m_{released,i}$ was then multiplied with the compositions of Co and Ni in material i, using Equation (3.3), to estimate how much of each element that is released by corrosion during one operating cycle. In Equation (3.3), $m_{j,i}$ is the released mass of element j from material i in mg and $C_{j,i}$ is the composition of element j in material i in wt%. Element j is either elementary Co or Ni.

$$m_{j,i} = m_{released,i} \frac{C_{j,i}}{100} \quad (3.3)$$

$m_{released,i}$ in Equation (3.3) could also be changed to $\dot{m}_{released,i}$ to determine the mass release of element j from material i, $\dot{m}_{j,i}$, in mg month⁻¹, see Equation (3.4).

$$\dot{m}_{j,i} = \dot{m}_{released,i} \frac{C_{j,i}}{100} \quad (3.4)$$

If $\dot{m}_{j,i}$ is converted to the unit $\mu\text{g s}^{-1}$, the estimated concentrations of Co and Ni in the RCS water could be determined using Equation (3.5). In Equation (3.5), $Conc_j$ is the concentration of element j in ppb (or μg of element j per kg water) and $\dot{m}_{waterflow}$ is the water mass flow in kg water s⁻¹. This was used when the estimated concentrations of Co and Ni in the RCS water were calculated for the real and theoretical composition cases.

$$Conc_j = \frac{\dot{m}_{j,i}}{\dot{m}_{waterflow}} \quad \text{Note : } \dot{m}_{j,i} \text{ in } [\mu\text{g s}^{-1}] \quad (3.5)$$

The number of Co and Ni atoms released during one operating cycle were then calculated using Equation (3.6), where $N_{j,i}$ is the number of atoms of element j released from material i, M_j is the molar mass of element j in g mol⁻¹ and N_A is Avogadro's number. The expression is divided by 1 000 to convert $m_{j,i}$ from milligrams to grams.

$$N_{j,i} = \frac{1}{1000} \frac{m_{j,i}}{M_j} N_A \quad (3.6)$$

Since the abundance of ⁵⁹Co is 100 atom% the calculated released Co atoms during one operating cycle was assumed to be equal to the number of ⁵⁹Co atoms released from the material. Ni on the other hand has more than one naturally occurring isotope which means that the calculated amount of released Ni atoms during one operating cycle was multiplied with its fractional abundance 0.68077 of ⁵⁸Ni to obtain the number of ⁵⁸Ni atoms. The general equation for this calculation is shown in Equation (3.7) where $N_{t,b,i}$ is the number of atoms of nuclide b from material i, also referred to as $N_{t,b}$ in Equation (2.3) in §2.8, and X_b is the fractional abundance of nuclide b.

$$N_{t,b,i} = N_{j,i} X_b \quad (3.7)$$

To calculate the released masses of Co and Ni during one operating cycle based on measured chemistry data, Equation (3.8) was used where $m_{j,chem}$ is the mass of element j in μg , $Conc_{j,chem}$ is the concentration of element j in ppb (this was determined from measured chemistry data) and t_{oc} is the operating cycle time in seconds.

$$m_{j,chem} = Conc_{j,chem} \dot{m}_{waterflow} t_{oc} \quad \text{Note : } t_{oc} \text{ in [s]} \quad (3.8)$$

When $m_{j,chem}$ had been determined, Equations (3.6) and (3.7) could be used, by changing $m_{j,i}$ to $m_{j,chem}$, to determine the number of target atoms for the case based on measured chemistry data.

Estimated activity calculations were then performed for all cases using Equations (2.2) and (2.3) shown in §2.8. During the calculations, some conservative assumptions have been made. The assumptions are presented in chapter 4 whenever they were made. All calculated masses were also converted to the unit grams to easier compare them later in §5.2.

3.3 Sampling, Sample Preparations and Analyses

In the following section the sampling, sample preparations and analysis procedures are described. The procedures described are referred to the ones which are performed in R3 at Ringhals. The procedures are similar for R4 since R3 and R4 are almost identical, but could differ slightly.

The sampling and analysis of water, filter and gas samples were performed according to the procedures used at Ringhals. Samples were taken from the three SGs, the reactor coolant (RC) and the control system (CS) in R3. The CS-in sampling comes from the cold leg, i.e. the water supplied to the core, see §2.1.1. RC water is coming from the outlet of the core. When being in the areas where the samples are taken, it is necessary to wear a special overall, helmet, safety goggles and protective shoes. If other safety clothing or protection is necessary it is mentioned in the respective procedure.

3.3.1 Analytical Techniques

For measurement of water and filter samples as well as for SAM, a HPGe detector was used. All measurements were performed and analyzed using a software called GammaVision, where the detected γ -energies are shown. From the γ -energies it is possible to determine what γ -emitting radionuclides that are present in the sample.

3.3.2 Water and Filter Samples

In the following section the procedures for sampling, sample preparations and analyses are described for the different sample points. The sample points have been divided into separate sections since the procedures differ.

3.3.2.1 Water from Reactor Coolant

The valve where water samples from RC is taken is inside a glovebox in the plant since the RC water is radioactive. Before taking a sample, the valve was opened to let the water flush for 5 minutes. Then, sample was taken from the valve in a 60 mL plastic bottle. The outside of the bottle was then flushed with regular water to clean it from potential radioactive contamination. After cleaning, the bottle was taken out from the glovebox and was measured to be sure no contamination was stuck on the outside of the bottle.

The bottle containing RC water was then transferred to the lab where the sample preparation was continued. For this step, safety gloves were also used. 50 mL of the sample was filtered through three different filters, placed on top of each other, by using vacuum filtration in a fumehood. The top filter was a particle filter, the middle a cation filter and the bottom an anion filter. By using these three filters, different nuclides will get stuck in each filter.

After filtration, the three filters and the filtrate were divided into four separate plastic jars holding 60 mL each. γ -measurements were then performed on all four samples, separately, in a HPGe detector

for 900 seconds each, i.e. 15 minutes. The results were analyzed using GammaVision.

3.3.2.2 Water from Control System

The sample with water from CS was taken in the same glovebox as where the sample with RC water was taken. The sample was taken from its valve using the same procedure as for RC water, see the first paragraph in §3.3.2.1, but in a plastic bottle holding 125 mL.

The bottle containing water from CS was then transferred to the lab where the sample preparation was continued. 85 mL of the sample was filtered through a filter by using vacuum filtration. The filtrate was transferred to a 90 mL plastic jar and the filter was placed in a 60 mL plastic jar. The samples were left to decay until the next day and were then measured separately in a HPGe detector for 3 000 seconds each, i.e. 50 minutes. The results were analyzed using GammaVision.

3.3.2.3 Water from Steam Generators

The samples with water from the three SGs were taken from valves in a more open system, i.e. not in a glovebox as seen in §3.3.2.1 and §3.3.2.2. This is due to that the water from the SGs should not be radioactive since its tubes belongs to the secondary system and not the primary system where the reactor water flows. Before taking a sample, the valve was opened to let the water flush for 5 minutes. Then, using safety gloves, approximately 1 L of each sample was taken from the valve in separate plastic bottles. The bottles were then flushed with regular water to clean the outside of them, since the SG water contains hydrazine.

The bottles containing water from the SGs were then taken to the lab where each sample was transferred to a special 1 L sample jar which fit in a HPGe detector. The samples were then measured separately in a HPGe detector for 3 000 seconds each, i.e. 50 minutes, and the results were analyzed using GammaVision.

3.3.3 Gas Sample

A gas sample was taken on the RC water from a separate valve in the same glove box as where water sample from RC and CS are taken, see §3.3.2.1 and §3.3.2.2. However, procedure for gas sampling is different compared to water sampling.

The gas sample is taken in a glass equipment called a "bomb", which is pictured as a simplified drawing in Figure 3.1. In the lab, a needle connected to a tube was inserted into one of the septa in a bomb holding a volume of 256 mL. By using a pump connected to the tube, the air was pumped out of the bomb to achieve vacuum. This was to more easily fill the bomb with a sample later during the sampling. The glass vials where the gas sample later were going to be transferred to, were also pumped out of air to create a vacuum in them.

The bomb was thereafter brought to where the sampling takes place. To the valve from where the gas sample was taken, a needle device were attached. The valve was opened to let the water and gas flush through the needle for 10 minutes. When taking the sample, the needle was inserted to the bottom septum of the bomb. The bomb was then filled to half the volume with water and the rest in the bomb was gas.

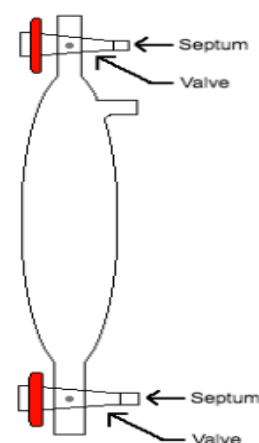


Figure 3.1: A simplified drawing of a bomb which is a glass equipment used in gas sampling.

Since the gas contained different nuclides, also short-lived ones, it was necessary to quickly transfer the bomb back to the lab directly after sampling to later be able to get a correct measurement of all nuclides in the sample. Back in the lab, gas was transferred from the bomb to the emptied glass vials by using a needle device. The volume of water collected in the bomb was thereafter measured and subtracted from the total bomb volume of 256 mL to determine how much gas that was in the bomb. The gas sample was then measured in a HPGe detector for 3 600 seconds, i.e. 1 hour, and the results were analyzed using GammaVision.

3.3.4 Surface Activity Measurements (SAM)

When it is of interest to determine the activity or dose rate of a surface in the nuclear power plant a SAM is performed. Before doing the measurements, a meeting is needed where a walk-through of the procedure is made. Since the work is performed in areas where there are measurable dose rates, a meeting minimizes the dose to personnel since well prepared and organized work results in that the procedure in place takes less time to perform than if not prepared.

At the place where the measurement is taking place, a detector is placed correctly against the surface of interest. The detector is connected to a DigiDart, a multi channel analyzer which contains settings for the measurement and collects the spectra. After having the detector in place, the measurement is started and the detector is left for approximately 10 hours while measuring the surface with respect to γ . The measurement is then stopped and the results are analyzed at the office using a PC with the software GammaVision. The unit of surface activity is Bq m^{-2} where (*Bq*) is the abbreviation of Becquerel. 1 Bq is equal to 1 decay s^{-1} .

3.4 Analysis of Collected Data and Information

By using gathered information from the literature studies together with collected data from measurements, a bigger picture of the Co content could be formed. Since one purpose of the project was to see if the framework of nuclear-specific requirements, which is presently used at Ringhals, is still adequate it was also needed to study the requirements made by the Swedish nuclear utilities to be able to compare it to gathered data at/from Ringhals. The levels of ^{60}Co and ^{58}Co in the systems were then compared with the requirements of Co content in metallic materials to be able to discuss a reasonable level from an ALARA and operational perspective.

4

Results

4.1 System Components of Interest in Primary Circuit

The system components of interest are the SG tubes, the cladding of the RPV, the RPVs internal parts, RCPs, the primary loops and the PRZ. In Table 4.1 the estimated media-affected surface areas of some components in the primary circuit in R3 are shown. The surface areas have been estimated by a design engineer [35] at Ringhals using computer-aided design (CAD), but the areas listed in Table 4.1 are only approximate values to avoid publishing of the exact values. Surface areas of some components have been more difficult to determine, such as the surface area for the surge line and the baffle plates. The most important to note in Table 4.1 is that the surface area of the SG tubes are a very large part of the total surface area in the primary circuit.

Table 4.1: Assumed surface areas (approximate values) of some larger components in R3 [35].

Component	Surface area [m ²]
SG tubes (all three SGs)	20 000
Cladding in RPV	120
RPV internals (with contact surfaces between materials)	1 300
RPV internals (with contact surfaces subtracted)	800
RCPs (all three)	60
Primary loops (all three)	130
Cladding in PRZ	90

4.1.1 Steam Generator Tubes

Since the SG tubes take up a very large part of the surface area in the primary circuit it was of interest to gather more information about the SG tubes, which was done by reaching out to personnel at Sandvik AB. The interview was done with Carl-Filip Lindahl at Sandvik AB.

“It coincides that I was actually in charge of production in the early 90’s when we manufactured the steam generator tubes for Ringhals new steam generators.” [English translation] [36]

In the following section the answer to the questions asked will be presented as quotes to get the right picture of the given answers. The first question asked was if Sandvik AB uses iron ore based steel production or recycled material steel production. The answer given was as below.

“We normally use very small quantities of directly ore based pre-material in our meltshop. On average, our uploads consist of 40 % recycled material ... from our own production, 40 % purchased recycled material and 20 % raw material in the form of Ni in various forms,

ferro-chrome etc...” [English translation] [36]

From this answer, the supervisors of this project filled in with the answer as below.

“Just as answered, we specify a requirement specification to the supplier where requirements for alloy content/mechanical properties/corrosion tests etc. must be fulfilled. You can see these in part in TBM and KBM, but for critical components such as the SG tubes, there may be even more stringent requirements, which can only be seen in the project’s specification.” [English translation] [29]

The second question asked was if Sandvik AB uses any manufacturing regulations when producing of stainless steel and nickel base alloys for determination of what amount of a particular element that is allowed in the material depending on the area of use of the material.

“...the simplest answer is that it is our customers who define what our steels are allowed to contain in the form of main elements and levels of impurities. They do this in their specification, where they either explicitly print it, or by referring to norms or standards. In addition to our customers’ requirements, we have for each alloy we manufacture our own internal requirements at impurities levels. The reason for them is that in some cases they affect the manufacturability or the properties of the finished products and that we want as much freedom as possible for our own use of the recycled material generated during our manufacture. Since the recycled material is such a large part of our uploads, it is really important for us to have a good internal system to keep recycled materials of different kinds apart. Otherwise, levels of impurities will soon increase in general.” [English translation] [36]

What type of analysis methods do Sandvik AB use to determine the composition of elements in stainless steels and nickel base alloys that they manufactures was the third question that was asked. The answer given was as below.

“The main method of analysis is X-ray spectrometry. Some lighter elements are determined by wet chemical methods. We have a fairly large and well-reputed accredited chemlab where we run the analyzes.” [English translation] [36]

The fourth question asked to Sandvik AB was how the SG tubes, in material A690, are manufactured.

“...let me give you an extremely short version [otherwise very long answer]. Melting + casting. Rolling of ingot to round bar. Peeling, grinding and testing against surface and center imperfections. Cutting to billet lengths for extrusion, drilling, hot extrusion. Straightening, acid pickling, grinding on the outside, polishing inside. Cold step rolling for final dimension, cleaning, heat treatment, straightening, external grinding, ultrasound and eddy current testing. Finished heat treatment in vacuum, bending, renewed finished heat treatment, testing and packing.” [English translation] [36]

The next question was if Sandvik AB treat the material A690, to the SG tubes, to decrease corrosion of the material in for example PWR environment or if these treatments in this case is performed by the customer? The answer given was as below.

“The material is in itself corrosion protected by its chemical composition and by the finished heat treatment it is given. After all, it is so with stainless steel and nickel alloys that contain chromium that spontaneously a nanometer-thick, chemically stable chromium oxide layer is formed on the material surface when exposed to air or oxygen. The layer is so thin that it is not visible, and ordinary handling also makes very light

holes in the layer very easily. But the chromium oxide layer is also self-repairing when you make holes in it if only the environment is oxidizing. One can talk about self-passivation. No treatments are really needed to be done. There are customers who request an extra passivation in the oxidizing acid bath, but even such [a layer], possibly slightly thicker passive layer will be damaged in normal handling and then you still have to rely on the damaged areas to self-passivate.” [English translation] [36]

The last question asked to Sandvik AB was if they are performing any corrosion tests on their materials before they are sold/delivered to the customer. For this question the answer given was as below.

“We carry out the corrosion tests that our customers request in their specifications. I do not remember what the specification for steam generator tubes looked like in the Ringhals case. There is a large number of common and less common corrosion tests, most of them try to somehow mimic the environment in which the tubes are intended to be used. We rarely deliver anything that has not undergone some kind of corrosion test. One of the most common corrosion tests done on stainless steel and nickel alloys is the grain boundary corrosion test. Such a test is intended to confirm that the tube in the delivery state is properly heat treated and will not be attacked by the special form of corrosion called Intergranular Attack (*IGA*).” [English translation] [36]

4.2 Mapping of Cobalt in Materials in Primary Circuit

The materials containing cobalt and nickel have been mapped and in the following sections they are divided into stainless steel materials, nickel-base alloys and Stellite. The estimated surface areas of the respective material have been found in a report from 2017 at Ringhals, and might not be the exact true value but is an acceptable estimation for the study.

4.2.1 Stainless Steel

Stainless steel is widely used throughout the power plant, but it has been seen in reports at Ringhals that it is used as material in for e.g. the RPV, the RPV internals and in the PRZ [37]. The total surface area of stainless steel in the primary circuit in R3, which is not exposed to induced activation, is estimated to 3 000 m² [38].

4.2.2 Nickel-Base Alloys

Nickel-base alloys are mainly used in the SG tubes and take up a very large area of the total surface area in the primary circuit [24]. Before, Ringhals used SG tubes made of A600 which was mentioned previously in section §2.5.3. In 1995 the SGs in R3 were replaced with new ones and the SG tubes were changed to ones made of A690, a material containing a lower wt% of nickel and a higher wt% of chromium than A600. It has also been seen from a report that the higher amount of Cr in A690, compared to in A600, possibly results in that the chromium(III) oxide (Cr_2O_3 , an oxide layer on the material) forms faster which in turn results in a reduced release of Ni and Fe [39]. Despite a lower content of both Ni and Co in A690 compared to A600, the increase in surface area of the new SGs gave rise to similar or even higher source term, which is how much is coming from the material [40].

The total surface area of A690 in the primary circuit in R3, which is not exposed to induced activation, is estimated to 19 318.5 m² [38].

4.2.3 Stellite

It has been seen from reports at Ringhals that Stellite 6 (will be referred to as Stellite) is used, and the material is located in the parts called lower radial support clevis insert, radial support key, upper core plate guide pin and upper core plate insert [37]. Since Stellite is used, a certain activation will occur whether it is placed in or outside the core region.

The total surface area of Stellite in the primary circuit in R3, which is not exposed to induced activation, is estimated to 10 m² [38]. The surface area is as mentioned only estimated but it is known that reactor unit 2 at Ringhals has had the largest sources of Stellite and R4 has had the lowest ones based on measured data. In the same report where the estimated surface area of Stellite in R3 was found, it was also found that the estimated surface area of Stellite in R4 was 1 m².

4.2.4 Summary of the Surface Areas of the Materials

In Figure 4.1, a summary of the assumed total surface areas of stainless steel, A690 and Stellite are shown in the form of a bar and pie chart. The pie chart is attached to get an overview of the surface areas as percentages. When comparing the surface areas of the materials stainless steel, A690 and Stellite, A690 takes up 87 % of the total surface area as can be seen in Figure 4.1. Since there are other materials used in components, especially in the internal structure, in the primary circuit which are not taken into account in this report the material A690 probably takes up about less than 87 % of the total surface area in the primary circuit in R3. An assumption made was that all mentioned materials and their surface areas described in §4.2.1, §4.2.2 and §4.2.3 are placed outside of the core and therefore not exposed to direct irradiation of neutrons [38].

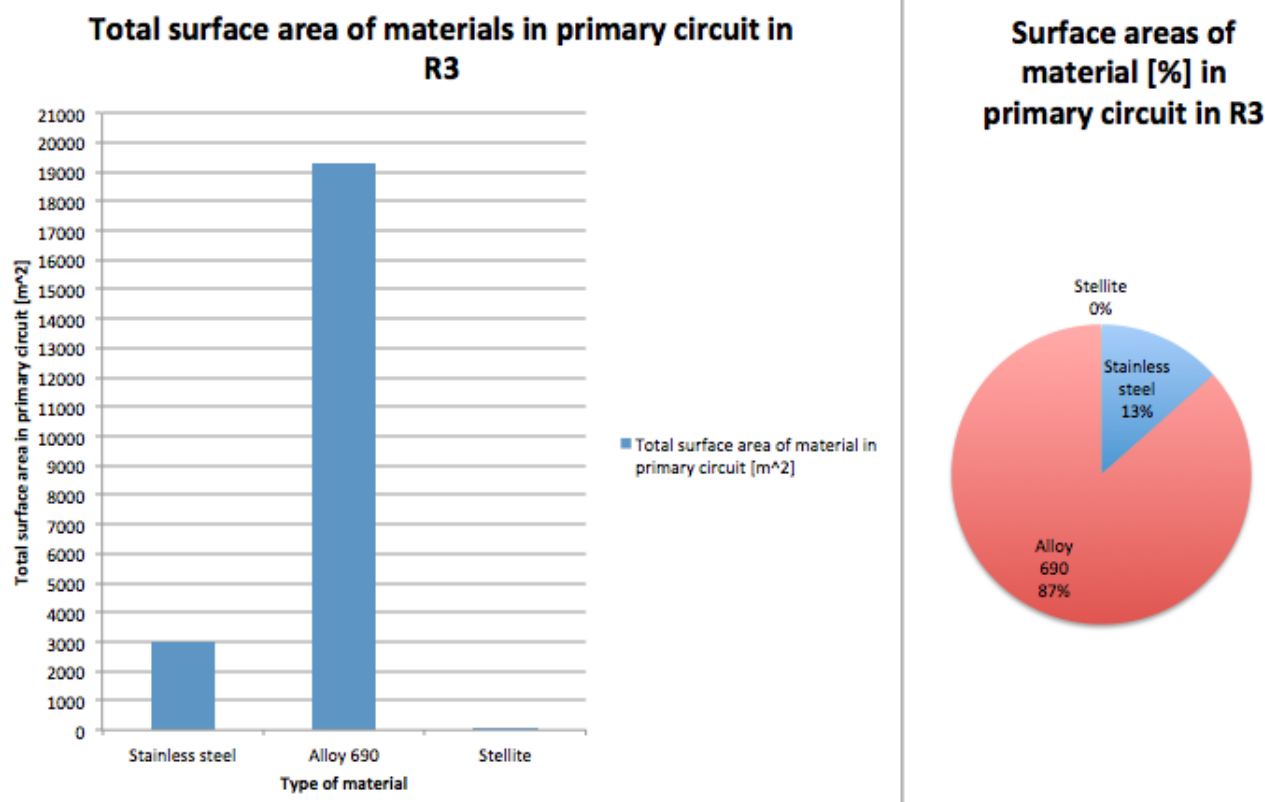


Figure 4.1: A summary of the total surface areas of each material; stainless steel, A690 and Stellite.

4.3 Environment in System

The water chemistry in a PWR reactor unit was previously presented in §2.1.2, and in Table 4.2 the used parameters for the water chemistry in R3 are summarized.

Table 4.2: Typical water chemistry in R3 [8].

pH at the beginning of an operating cycle	7.1-7.2
pH during and at the end of an operating cycle	7.4
B at the beginning of an operating cycle to overcome 0 % reactor power	$\leq 1\,800$ ppm
Minimum level of B at the end of an operating cycle	4 ppm
Li at the beginning of an operating cycle	5 ppm
Minimum level of Li at the end of an operating cycle	0.75 ppm
Hydrogen content	40-50 mL/kg hydrogen

It has also been seen that the total mass flow of water in the RCS through the SGs, the RCPs and the reactor is $12\,860\text{ kg s}^{-1}$ which in volume flow corresponds to approximately $5.7\text{ m}^3\text{ s}^{-1}\text{ loop}^{-1}$ [41].

4.4 Estimation Calculations of Cobalt and Nickel in Primary Circuit

In the following section, results from performed estimation calculations will be presented. The calculations have been made to determine the releases of elementary Co and Ni in the primary circuit, as well what activity this would correspond to if activated when entering the core. A conservative assumption made was that all material that corrodes is transported by the water to the core where all of it is activated by neutrons. More calculated data are found in more detail in appendix A. The calculations have been performed using both compositions for A690 found in material certificates [42] and for stainless steel and Stellite in a report [38] from Ringhals (real compositions) and for theoretical compositions to see how an increase of Co content in the material effect the results.

4.4.1 Estimation Calculations with Real Compositions

From a report at Ringhals, the assumed corrosion rates for stainless steel, A690 and Stellite in the PWR environment in R3 was found to be the ones shown in Table 4.3. Different corrosion rates have been assumed depending on if it is a conservative case or a realistic case.

Table 4.3: Assumed corrosion rates of stainless steel, A690 and Stellite in the PWR environment in R3 [38].

Material	Corrosion rate [mdm]	
	Conservative case	Realistic case
Stainless steel	50	10
A690	5	1
Stellite	200	60

In Table 4.4 the real compositions of Co and Ni in the materials used in the calculations are shown. Something to note is that the Co content in both the stainless steel and A690 are below 0.05 wt% which is the lower limit listed in the TBM document, see §2.6.1.

Table 4.4: Real compositions of Co and Ni in stainless steel, A690 and Stellite in the primary circuit in R3 [38, 42].

Material	Co [wt%]	Ni [wt%]
Stainless steel	0.03	10
A690	0.013	59.5
Stellite	62.8	0.4

By using Equation (3.1) and (3.3) shown previously in §3.2, the released masses of Co and Ni from stainless steel, A690 and Stellite during one operating cycle (i.e. 330 days) were estimated using both the conservative and the realistic case of the corrosion rates for each material. The estimated released masses are presented in Table 4.5. For these calculations, the real compositions shown previously in Table 4.4 were used. In Table 4.5, a summation of released masses of Co and Ni from stainless steel, A690 and Stellite during one operating cycle is also presented, which is referred to as total.

Table 4.5: Estimated released masses of elementary Co and Ni from stainless steel, A690 and Stellite in the RCS water during one operating cycle (330 days) in primary circuit in R3.

Material	Released mass, conservative case		Released mass, realistic case	
	Co [g]	Ni [g]	Co [g]	Ni [g]
Stainless steel	48.82	16 273.97	9.76	3 254.79
A690	13.20	62 324.95	2.64	12 464.99
Stellite	1 362.67	8.68	408.80	2.60
Total	1 424.70	78 607.60	421.21	15 722.39

The number of ^{59}Co and ^{58}Ni atoms were calculated using Equation (3.6) and are shown in Table 4.6.

Table 4.6: Estimated number of ^{59}Co and ^{58}Ni atoms in primary circuit in R3 during one operating cycle (330 days), which are assumed to be the number of target atoms of each nuclide when entering the core.

Material	Number of atoms, conservative case		Number of atoms, realistic case	
	^{59}Co [atoms]	^{58}Ni [atoms]	^{59}Co [atoms]	^{58}Ni [atoms]
Stainless steel	4.99×10^{23}	1.14×10^{26}	9.98×10^{22}	2.27×10^{25}
A690	1.35×10^{23}	4.35×10^{26}	2.70×10^{22}	8.71×10^{25}
Stellite	1.39×10^{25}	6.06×10^{22}	4.18×10^{24}	1.82×10^{22}
Total	1.46×10^{25}	5.49×10^{26}	4.30×10^{24}	1.10×10^{26}

The estimated number of atoms for each material and in total, shown in Table 4.6, was assumed to be the number of target atoms which are irradiated by neutrons when entering the core. As mentioned in §2.2 the cross section of the reaction $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ is known to be 37.2 barn, that is $3.72 \times 10^{-23} \text{ cm}^2$. The neutron fluxes in the core and the cross section for the reaction $^{58}\text{Ni}(n,p)^{58}\text{Co}$ were at first unknown but after discussion with a core physicist at Ringhals [43], these were estimated to be as shown in Table 4.7.

Table 4.7: Estimated values used in the calculations [43].

Cross section for the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ reaction, $\sigma_{n,p}$	0.6 barn (that is $6 \times 10^{-25} \text{ cm}^2$)
Thermal neutron flux, ϕ_{thermal}	$4 \times 10^{14} \text{ neutrons cm}^{-2} \text{ s}^{-1}$
Fast neutron flux, ϕ_{fast}	$1 \times 10^{15} \text{ neutrons cm}^{-2} \text{ s}^{-1}$
Irradiation time, t_{irr}	330 days*
Cooling time, t_{cool}	0 days**

* Reasonable conservative assumption: irradiation time of about 330 days, corresponding to deposition at the beginning of the cycle and release at the end of the cycle. The fuel is washed between each cycle, so material should not be deposited from cycle to cycle.

** Reasonable conservative assumption: set to zero, to count on maximum activity.

As mentioned previously in §2.2.1, thermal neutrons are most likely involved in the reaction $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$. ϕ_{thermal} were therefore used for the calculations regarding ^{60}Co . When looking at experimental data for cross sections of the reaction $^{58}\text{Ni}(n,p)^{58}\text{Co}$, the energies are much higher than for thermal neutrons. Together with the core physicist at Ringhals [7, 43], an assumption was made that it is likely that fast neutrons are involved in the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ reaction. ϕ_{fast} were therefore used in the calculations regarding ^{58}Co .

Using the assumed values observed in Table 4.7, the activities of the produced ^{60}Co and ^{58}Co was calculated as described in §3.2. The activities for both the conservative and realistic case are calculated with the very conservative assumption that all the corroded material is irradiated in the core at the same time. The estimated activities in Bq from ^{60}Co and ^{58}Co as well as the total activities as the sum of the activities from each material are presented in Table 4.8.

Table 4.8: Estimated activities from ^{60}Co and ^{58}Co in primary circuit in R3 during one operating cycle (330 days).

Material	Activity, conservative case		Activity, realistic case	
	^{60}Co [Bq]	^{58}Co [Bq]	^{60}Co [Bq]	^{58}Co [Bq]
Stainless steel	1.02×10^{10}	2.55×10^{12}	2.04×10^9	5.10×10^{11}
A690	2.76×10^9	9.76×10^{12}	5.53×10^8	1.95×10^{12}
Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
Total	2.98×10^{11}	1.23×10^{13}	8.81×10^{10}	2.46×10^{12}

The concentrations of elementary Co and Ni in ppb in the RCS water were estimated, as described in §3.2, to be the ones which are shown in Table 4.9 during one operating cycle. 1 ppb is equal to $1 \mu\text{g}$ Co per kg water. A total water mass flow in RCS of $12\,860 \text{ kg s}^{-1}$, as described in §4.3, was used [41].

Table 4.9: Estimated concentrations of elementary Co and Ni in the RCS water in primary circuit in R3, calculated for one operating cycle (i.e. 330 days), using the real compositions of the materials.

Material	Concentration, conservative case		Concentration, realistic case	
	Co [ppb]	Ni [ppb]	Co [ppb]	Ni [ppb]
Stainless steel	1.33×10^{-4}	4.44×10^{-2}	2.66×10^{-3}	8.88×10^{-4}
A690	3.60×10^{-5}	1.17×10^{-1}	7.20×10^{-6}	3.40×10^{-2}
Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
Total	3.89×10^{-3}	2.14×10^{-1}	1.15×10^{-3}	4.29×10^{-2}

4.4.2 Estimation Calculations with Theoretical Compositions of Cobalt

As mentioned in chapter 3, Co and Ni compositions as shown in Table 4.10 were also used to give a comparison on how the activity changes with higher content of Co in the materials. The Co and Ni compositions in Stellite was not varied since Stellite is an exception not following the restrictions of 0.05 wt% Co. Since the requirement is for Co, the amount of Ni and therefore also the activity of ^{58}Co will not be varied in the calculations either.

When combining Equations (2.2), (2.3), (3.1), (3.3), (3.6) and (3.7) for the calculations regarding ^{60}Co , it was seen that the activity caused by increasing Co content in a material is linear (for that particular material) if making the conservative assumption that ϕ_{thermal} , $\sigma_{n,\gamma}$, t_{irr} and t_{cool} are the same for all cases, see full derivation in appendix B.

Table 4.10: Compositions of Co and Ni in stainless steel, A690 and Stellite used in estimation calculations of the activity in primary circuit in R3.

Material	Co [wt%]	Ni [wt%]
Stainless steel	0.05	10
	0.10	10
	0.15	10
	0.20	10
	0.25	10
	0.30	10
A690	0.05	59.5
	0.10	59.5
	0.15	59.5
	0.20	59.5
	0.25	59.5
	0.30	59.5
Stellite	62.8	0.4

The linearity of the activity increase was also shown as numbers, by calculating the estimated activity for the materials using the compositions shown in Table 4.10. After calculations, released masses of Co and Ni from stainless steel, A690 and Stellite during one operating cycle (i.e. 330 days) were obtained as presented in Table 4.11. The calculations were performed using both the conservative and the realistic case of the corrosion rates for each material previously shown in Table 4.3.

Table 4.11: Estimated released masses of elementary Co and Ni during one operating cycle (330 days) in the primary circuit in R3 using the different compositions of Co, shown previously in Table 4.10, in the materials.

Composition Co wt%	Material	Conservative case		Realistic case	
		Co [g]	Ni [g]	Co [g]	Ni [g]
0.05	Stainless steel	81.37	16 273.97	16.27	3 254.79
0.05	A690	52.40	62 324.95	10.48	12 464.99
62.8	Stellite	1 362.67	8.68	408.80	2.60
	Total	1 496.44	78.61	435.56	15 722.39
0.10	Stainless steel	162.74	16 273.97	32.55	3 254.79
0.10	A690	104.80	62 324.95	20.96	12 464.99
62.8	Stellite	1 362.67	8.68	408.80	2.60
	Total	1 630.21	78 607.60	462.31	15 722.39
0.15	Stainless steel	244.11	16 273.97	48.82	3 254.79
0.15	A690	157.19	62 324.95	31.44	12 464.99
62.8	Stellite	1 362.67	8.68	408.80	2.60
	Total	1 763.98	78 607.60	489.06	15 722.39
0.20	Stainless steel	325.48	16 273.97	65.10	3 254.79
0.20	A690	209.59	62 324.95	41.92	12 464.99
62.8	Stellite	1 362.67	8.68	408.80	2.60
	Total	1 897.75	78 607.60	515.82	15 722.39
0.25	Stainless steel	406.85	16 273.97	81.37	3 254.79
0.25	A690	261.99	62 324.95	52.40	12 464.99
62.8	Stellite	1 362.67	8.68	408.80	2.60
	Total	2 031.51	78 607.60	524.57	15 722.39
0.30	Stainless steel	488.22	16 273.97	97.64	3 254.79
0.30	A690	314.39	62 324.95	62.87	12 464.99
62.8	Stellite	1 362.67	8.68	408.80	2.60
	Total	2 165.28	78 607.60	569.32	15 722.39

The estimated activities obtained after calculations with each of the cases of Co composition in stainless steel and A690 are shown in Table 4.12.

Table 4.12: Estimated activities from ^{60}Co and ^{58}Co during one operating cycle (330 days) in primary circuit in R3 using the different composition of Co, shown previously in Table 4.10, in the materials.

Composition Co wt%	Material	Conservative case		Realistic case	
		^{60}Co [Bq]	^{58}Co [Bq]	^{60}Co [Bq]	^{58}Co [Bq]
0.05	Stainless steel	1.70×10^{10}	2.55×10^{12}	3.40×10^9	5.10×10^{11}
0.05	A690	1.09×10^{10}	9.76×10^{12}	2.19×10^9	1.95×10^{12}
62.8	Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
	Total	3.13×10^{11}	1.23×10^{13}	9.11×10^{10}	2.46×10^{12}
0.10	Stainless steel	3.40×10^{10}	2.55×10^{12}	6.81×10^9	5.10×10^{11}
0.10	A690	2.19×10^{10}	9.76×10^{12}	4.39×10^9	1.95×10^{12}
62.8	Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
	Total	3.41×10^{11}	1.23×10^{13}	9.67×10^{10}	2.46×10^{12}
0.15	Stainless steel	5.11×10^{10}	2.55×10^{12}	1.02×10^{10}	5.10×10^{11}
0.15	A690	3.29×10^{10}	9.76×10^{12}	6.55×10^9	1.95×10^{12}
62.8	Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
	Total	3.69×10^{11}	1.23×10^{13}	1.02×10^{11}	2.46×10^{12}
0.20	Stainless steel	6.81×10^{10}	2.55×10^{12}	1.36×10^{10}	5.10×10^{11}
0.20	A690	4.39×10^{10}	9.76×10^{12}	8.77×10^9	1.95×10^{12}
62.8	Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
	Total	3.97×10^{11}	1.23×10^{13}	1.08×10^{11}	2.46×10^{12}
0.25	Stainless steel	8.51×10^{10}	2.55×10^{12}	1.70×10^{10}	5.10×10^{11}
0.25	A690	5.48×10^{10}	9.76×10^{12}	1.10×10^{10}	1.95×10^{12}
62.8	Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
	Total	4.25×10^{11}	1.23×10^{13}	1.14×10^{11}	2.46×10^{12}
0.30	Stainless steel	1.02×10^{11}	2.55×10^{12}	2.04×10^{10}	5.10×10^{11}
0.30	A690	6.58×10^{10}	9.76×10^{12}	1.32×10^{10}	1.95×10^{12}
62.8	Stellite	2.85×10^{11}	1.36×10^9	8.55×10^{10}	4.08×10^8
	Total	4.53×10^{11}	1.23×10^{13}	1.19×10^{11}	2.46×10^{12}

The estimated concentrations of elementary Co and Ni in ppb in the RCS water were estimated, as described in §3.2, to be the ones which are shown in Table 4.13 during one operating cycle when using the different Co compositions shown previously in Table 4.10. A total water mass flow in RCS of $12\,860\text{ kg s}^{-1}$, as described in §4.3, was used [41].

Table 4.13: Estimated concentrations of elementary Co and Ni in the RCS water in the primary circuit in R3, calculated for one operating cycle (i.e. 330 days), using the different composition of Co, shown previously in Table 4.10, in the materials.

Composition Co wt%	Material	Conservative case		Realistic case	
		Co [ppb]	Ni [ppb]	Co [ppb]	Ni [ppb]
0.05	Stainless steel	2.22×10^{-4}	4.44×10^{-2}	4.44×10^{-5}	8.88×10^{-3}
0.05	A690	1.43×10^{-4}	1.17×10^{-1}	2.86×10^{-5}	3.40×10^{-2}
62.8	Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
	Total	4.08×10^{-3}	2.14×10^{-1}	1.19×10^{-3}	4.29×10^{-2}
0.10	Stainless steel	4.44×10^{-4}	4.44×10^{-2}	8.88×10^{-5}	8.88×10^{-3}
0.10	A690	2.86×10^{-4}	1.17×10^{-1}	5.72×10^{-5}	3.40×10^{-2}
62.8	Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
	Total	4.45×10^{-3}	2.14×10^{-1}	1.26×10^{-3}	4.29×10^{-2}
0.15	Stainless steel	6.66×10^{-4}	4.44×10^{-2}	1.33×10^{-4}	8.88×10^{-3}
0.15	A690	4.29×10^{-4}	1.17×10^{-1}	8.57×10^{-5}	3.40×10^{-2}
62.8	Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
	Total	4.81×10^{-3}	2.14×10^{-1}	1.33×10^{-3}	4.29×10^{-2}
0.20	Stainless steel	8.88×10^{-4}	4.44×10^{-2}	1.78×10^{-4}	8.88×10^{-3}
0.20	A690	5.72×10^{-4}	1.17×10^{-1}	1.14×10^{-4}	3.40×10^{-2}
62.8	Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
	Total	5.18×10^{-3}	2.14×10^{-1}	1.41×10^{-3}	4.29×10^{-2}
0.25	Stainless steel	1.11×10^{-3}	4.44×10^{-2}	2.22×10^{-4}	8.88×10^{-3}
0.25	A690	7.15×10^{-4}	1.17×10^{-1}	1.43×10^{-4}	3.40×10^{-2}
62.8	Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
	Total	5.54×10^{-3}	2.14×10^{-1}	1.48×10^{-3}	4.29×10^{-2}
0.30	Stainless steel	1.33×10^{-3}	4.44×10^{-2}	2.66×10^{-4}	8.88×10^{-3}
0.30	A690	8.57×10^{-4}	1.17×10^{-1}	1.71×10^{-4}	3.40×10^{-2}
62.8	Stellite	3.72×10^{-3}	2.37×10^{-5}	1.11×10^{-3}	7.10×10^{-6}
	Total	5.91×10^{-3}	2.14×10^{-1}	1.55×10^{-3}	4.29×10^{-2}

4.4.3 Comparison - Real versus Theoretical Compositions

The estimated values of released masses (see Tables 4.5 and 4.11), activities (see Tables 4.8 and 4.12) and concentrations (see Tables 4.9 and 4.13) are summarized as bar charts in appendix C to be able to compare them more easily.

4.5 Actual Outcome of Cobalt and Nickel in Primary Circuit

To be able to determine the actual outcome of activated cobalt in the primary circuit, collected data from sample points of interest in the power plant have been studied. Since the conservative assumption that all material that corrodes is transported to the core where it is irradiated, it is of interest to see how much Co and Ni there actually is in the primary circuit.

4.5.1 Water and Filter Samples

In charts from the chemistry department [8], as shut-down transient normalized to full power year, the determined or measured activities of ^{60}Co and ^{58}Co have been found. The activity of ^{60}Co have

from the year 2000 to 2019 varied between approximately 7×10^{10} Bq and 5×10^{11} Bq. The activity of ^{58}Co has from the year 2000 to 2019 varied between approximately 1×10^{11} Bq and 5×10^{13} Bq. From 2016 and forward, the activities seem to have been more stable with an activity of around 3×10^{11} Bq for ^{60}Co and just below 1×10^{13} Bq for ^{58}Co .

The chemistry department also provides charts where the results from so-called integrated sampling are plotted [8]. Integrated sampling is the measurements they are performing on water and filter samples during operation, see previously in §3.3 for how the sampling is performed. From these charts it has been possible to see the measured amounts of elementary Co and Ni in ppb in RC and in CS (which have been cleaned up by the ion exchanger during one operating cycle). The charts showed measured values from the third quarter of year 2009 to the first quarter of 2019. From this chart it could be seen that the measured concentration of elementary Co has been around 0.001 ppb during these 10 years. It was also seen in the respective chart for Ni that the concentration of elementary Ni during these 10 years has been around 0.02 ppb. By using an estimated water mass flow of 7 kg s^{-1} through the ion exchanger [8], the masses of elementary Co and Ni were calculated, using Equation (3.8), to be the ones shown in Table 4.14. Assuming that the concentrations of Co and Ni measured in the CS ion exchanger inlet are representative to the concentrations in cold leg water (i.e. inlet water to the core), the total amount of Co and Ni in the RCS water exposed to the core could be determined using Equation (3.8) and a water mass flow in RCS of $12\,860 \text{ kg s}^{-1}$ [41]. The calculated released masses are shown in Table 4.14.

Table 4.14: Calculated released masses of Co and Ni in the CS ion exchanger inlet and in the RCS water during one operating cycle, with the assumption that the concentrations of Co and Ni are 0.001 ppb and 0.02 ppb respectively.

Element	Released masses during one operating cycle [g]	
	In ion exchanger	In RCS water
Co	0.2	367
Ni	4	7 333

The calculated masses resulted in the activities in Bq shown in Table 4.15, using the calculation procedure as described in §3.2.

Table 4.15: Calculated activities of ^{60}Co and ^{58}Co in the CS ion exchanger inlet and in the RCS water during one operating cycle, with the assumption that the concentrations of Co and Ni are 0.001 ppb and 0.02 ppb respectively.

Nuclide	Activities during one operating cycle [Bq]	
	In ion exchanger	In RCS water
^{60}Co	4.18×10^7	7.67×10^{10}
^{58}Co	6.25×10^8	1.15×10^{12}

4.5.2 Surface Activity Measurements (SAM)

In CS before the ion exchanger (CS ion exchanger inlet), it has been seen in charts that the surface activity of ^{58}Co has had a peak during shut-down of R3 around year 1998-1999 [44]. The surface activity for ^{60}Co has during down regulation been stable through the years. The corresponding surface activities during operation showed a peak in 1998 for ^{58}Co while ^{60}Co has been stable. From the charts with results from SAM it was also seen that from around year 1998/1999, the surface activity of ^{58}Co in CS before the ion exchanger in R3 started to decrease (both during operation and down regulation). The surface activity has thereafter been stable from around 2006 until now, at a slightly lower surface

activity than before the peak.

In CS after the ion exchanger, the surface activity follows the same trend as in CS before the ion exchanger as was just described, but with a lower surface activity for ^{58}Co and ^{60}Co since the water has passed through the ion exchanger [44]. The surface activity in CS after the ion exchanger has decreased from year 1998 to become stable from 2004 until now. The surface activity from ^{60}Co has been quite stable.

Also for charts showing the surface activity in the SGs, the same trend has been observed e.g. for SG 3 (i.e. the SG in loop 3) [44]. The surface activity for ^{58}Co has had peaks around year 1998 for the hot side (that is the inlet to the SG) and between 1999 and 2003 for the cold side (that is the outlet from the SG). The surface activities have then decreased. When studying charts showing the surface activities for other components such as cross over leg and hot leg for loop 3, these also follow the same trend with a peak for ^{58}Co which then decreases to become stable.

The trend that was seen in all charts [44], the increase of ^{58}Co which then decreased and became stable, is due to the change of the SG tubes in R3 in 1995, as previously described in §2.5.3. When replacing the SGs in R3 in 1995, the initial increase of oxide release resulted in a higher amount of Ni (and therefore also ^{58}Co) in the primary circuit. These initial deposited oxides have then decayed since the half-life of ^{58}Co is only 70.86 days, as described in §2.3, resulting in a lower amount of Ni and ^{58}Co .

4.6 ALARA Perspective

Every year, SSM writes an annual report where for example the highest calculated dose to a person in the public as a result of emissions from the nuclear power plants and the collective dose to personnel at nuclear power plants are presented. The dose and collective dose are expressed in mSv per year and manSievert (*manSv*) respectively. As mentioned in §2.9, the allowed dose to a person in the public is 0.1 mSv per year.

From the annual report by SSM from 2019 it has been reported that the highest calculated dose to a person in the public, as a consequence of emissions from the nuclear power plants, was approximately 0.0005 mSv in 2006 from Ringhals, see Figure 4.2 [34]. This dose has then decreased to be around 0.0003 mSv, from the year of 2009 and until now. Compared to the nuclear power plants Oskarshamn and Forsmark, two power plants which are also located in Sweden, the highest calculated dose to a person in the public from those have been around 0.0001 to 0.0002 mSv. For all the mentioned power plants, the doses are well under the allowed limit of 0.1 mSv per year.

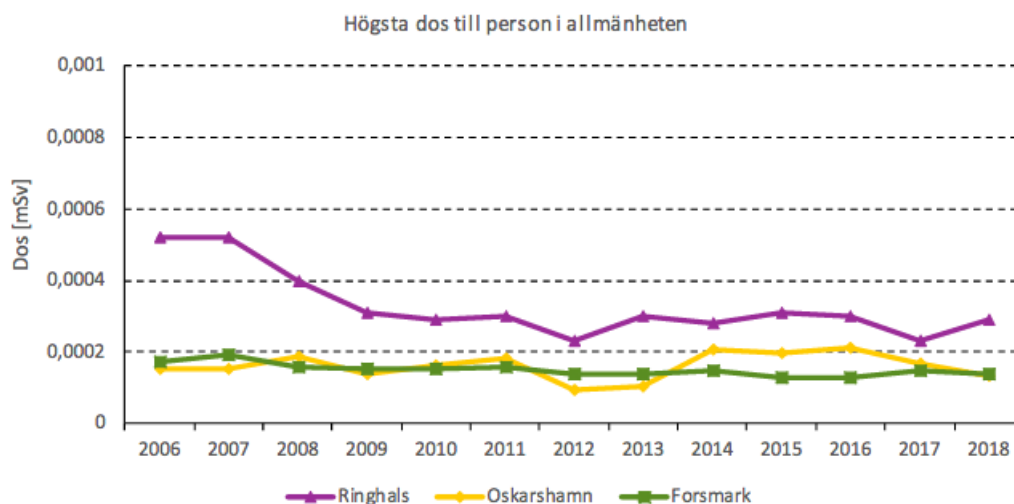


Figure 4.2: The highest calculated dose to a person in the public, as a consequence of emissions from the nuclear power plants Ringhals, Oskarshamn and Forsmark [34]. Reproduced with permission.

The collective dose to personnel at the nuclear power plants Ringhals, Oskarshamn and Forsmark was in 2006 around 3, 4 and 2 manSv respectively, see Figure 4.3. The collective dose has then varied until 2017 where it decrease to around 1 manSv for each nuclear power plant. SSM have expressed in their annual report from 2019 that their "overall assessment is that nuclear power plants handle radiation protection issues in a good way and that the radiation doses are at a reasonable level" [34].

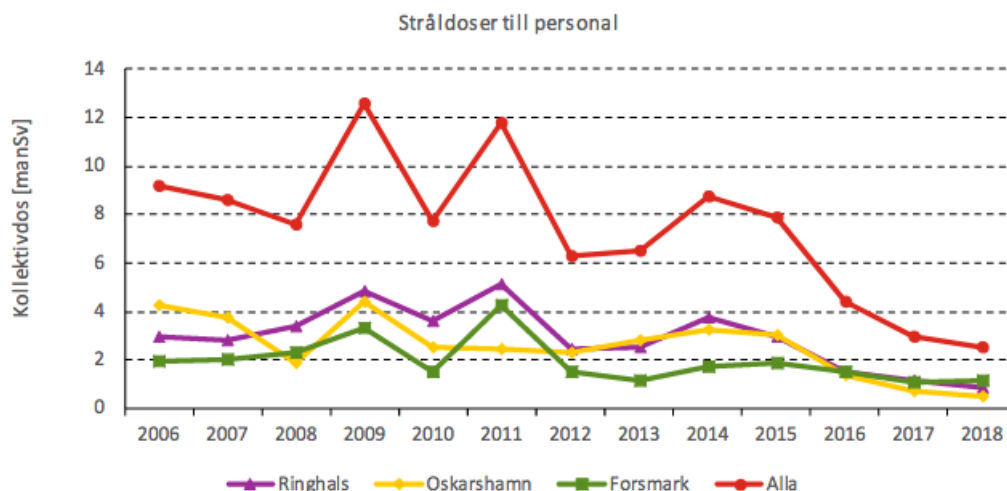


Figure 4.3: Collective dose to personnel at the nuclear power plants Ringhals, Oskarshamn and Forsmark, expressed in manSv [34]. The red line is the sum of all three. Reproduced with permission.

There is also something which is called the alpha value, defined as "the constant that specifies the monetary value which for radiation protection purposes is assigned to the collective dose unit 1 manSv" [English translation] [45], which is the amount of money a company is willing to invest to reduce the collective radiation dose by 1 manSv. In 2007 the alpha value at Swedish nuclear power plants was 4.5 MSEK per manSv, which is relatively low compared to alpha values used at nuclear power plants in other countries. However, it has been seen that an increase of the alpha value is reasoned. The alpha value has therefore been increased to around 10 MSEK per manSv but it is still discussed what is a reasonable alpha value for Sweden.

When speaking with radiation protection personnel at Ringhals [46], it has been understood that

Ringhals have been working a lot with decreasing the dose to personnel as much as possible. They mean that the *R* in ALARA is not used anymore and that the power plants now are working more towards "*As Low As Achievable*", which in other terms would be the same as working towards zero dose to personnel. Discussions are therefore made if the *R* in ALARA should be reinstated to instead work towards reasonable doses. SSM have also begun to renew their regulatory collection which, if finished in time, will be ready in the end of 2020 [47]. In an article about it, it is said as below.

“Characteristic of the new regulations that are now being developed is what SSM calls co-regulation of reactor safety, radiation protection and physical protection. Lars Skånberg describes it as a clarification of the importance of regulating the common goal of protecting people and the environment against the harmful effects of radiation. The principle means that instead of regulating pipe reactor safety, radiation protection and physical protection in separate regulations, SSM intends to produce regulations for construction, analysis and accounting, operation and decommissioning, which each includes all aspects of radiation safety - that is, reactor safety, radiation protection. and physical protection.” [English translation] [47]

Another fact which has been taken up during meetings with for example radiation protection personnel [46] is that ^{60}Co and ^{58}Co are not the most problematic nuclides. Antimony-124 (^{124}Sb), silver-110m (^{110m}Ag) and ^{93}Mo are examples of nuclides which are also problematic [46, 48]. ^{124}Sb and ^{110m}Ag for example "cause some local and occasional problems, such as control rod leakages or wearing/erosion of bearing surfaces" [48]. These two nuclides therefore cause radiation build-up that is more local and related to shut down chemistry. ^{93}Mo is a nuclide which at first is manageable but after many years it gets dominant which means that this also needs to be handled as waste [46]. Reactor unit 1 and 2 at Ringhals are the ones which have resulted in large amount of ^{60}Co and ^{58}Co .

It is therefore of great importance to know what you install in the system to understand how it needs to be handled later. In summary it is important to think about the ALARA concept during operation, waste during operation and waste after decommissioning. This might be more clearly described in the renewed regulatory guidance by SSM.

5

Discussion

Some information has been presented in the results but will not be discussed in this section. The information has been compiled in this report in the case that the project will be continued in the future.

5.1 Assumptions in the Estimation Calculations

As mentioned in §4.4 many assumptions were made, some more conservative than others. The first conservative assumption was that all the corrosion products are transported by the reactor coolant to the core where all of them are activated by neutrons. This is not completely true since some of the products may be deposited on surfaces or get stuck in cracks and bumps on the surfaces, resulting in that not all corrosion products reach the core. The estimated corrosion rates for each material could also have been overestimated. When speaking to personnel at Ringhals [40], the corrosion rate of A690 was claimed to be $\ll 0.1$ mdm in R3 rather than 1 mdm as used for the calculations of the realistic case, see Table 4.3. Therefore the actual corrosion rate, for at least A690, and in turn the amount of Co and Ni from the material in R3 are probably lower than estimated in this project. The calculations were also made for one operating cycle, i.e. 330 days.

The corrosion of materials result in that some material corrodes with a particular rate, for e.g. as mg of the material per month. This means that smaller amounts of material will continuously enter the core, instead of that all the material that has corroded during one operating cycle enters the core at the same time. This means that the activities calculated are the total activities calculated for one operating cycle. One could use Equation (3.1) to determine how much mass that is released per month. By using Equation (3.1) and then using $m_{released,i}$ instead of $m_{released,i}$ in the activity calculations, one would get an answer in how much activity is built up per a month instead of during one operating cycle if one could estimate what the irradiation time would be during a month.

As described in §4.4.2 and appendix B, there might be a linearity which could be used when estimating the activity increase caused by increased Co content in the materials. Even if not all material that corrodes during one operating cycle enters the core at the same time, the same principle could be used for a smaller mass of the material. In the calculations, the total surface areas of each of the materials stainless steel, A690 and Stellite that are not exposed to induced activity are used. Since most of A690 is located in the SG tubes, the estimated surface area of A690 might be a good assumption when determining the released masses etc. from the material and therefore also how the calculated values would change with increasing Co content. For stainless steel and Stellite, these materials are used in a variety of components which have different surface areas. As mentioned in §2.6.1, different requirements are applied depending on how large the surface area is and where the material is located. If one would like to know what the change in mass released, activity etc. would be if changing a particular component, it is therefore needed to determine the surface area

and requirement of that particular component to be able to determine the increase in percentages.

The cross section for the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ reaction, the neutron fluxes as well as the irradiation time and cooling time shown previously in Table 4.7 were also assumed. Neutron fluxes and neutron energies differs depending on where in the core the material is irradiated. Since the cross section of the (n,p) reaction is dependent on the neutron energy, as mentioned in §2.2, the cross section of the reaction is dependent on where in the core it occurs. The assumptions which resulted in the values in Table 4.7 were made in cooperation with a core physicist [43] and were based on that the significant contributions to the total activation likely come from materials deposited directly on the fuel rods which are then released and deposited elsewhere. This is due to that, on the one hand, the deposit itself will be greatest there as a result of the heat generation, and on the other, it will be exposed to most radiation. A reasonable conservative assumption was therefore that the irradiation time is about 330 days, corresponding to deposition at the beginning of the operating cycle and release at the end of the cycle. The cooling time was set to zero to estimate the maximum activity. The fuel is washed between each cycle, so it was also assumed that there would be no material deposited from cycle to cycle.

5.2 Estimated Versus Actual Outcome of Cobalt and Nickel

When comparing the calculated masses and concentrations of elementary Co and Ni in the primary circuit for the cases real compositions and the ones measured in the chemistry department, see Tables 4.5, 4.9 and 4.14 in §4.4.1 and §4.5, the values differ a bit. The calculated total masses released of Co and Ni from the real composition case are shown in Table 5.1 together with the respective masses estimated in the RCS water based on chemistry measurements, that is a composed and modified table of Tables 4.5 and 4.14. The same is done for the concentrations of Co and Ni in Table 5.2 and for the activities of ^{60}Co and ^{58}Co in Table 5.3. All values are for the RCS water and they are calculated for one operating cycle. The upper values in Tables 5.1 and 5.2 are based on measured data from the chemistry department and the lower values are the ones estimated in calculations using the real compositions of the materials stainless steel, A690 and Stellite in the primary circuit.

Table 5.1: Estimated mass releases of elementary Co and Ni from stainless steel, A690 and Stellite in the RCS water during one operating cycle (330 days) in primary circuit in R3. The top half of the table is based on calculations from measured chemistry data and the bottom half is based on calculations from real compositions.

Element		Mass released during one operating cycle [g] In RCS water (water mass flow: 12 860 kg s ⁻¹ [41])		
Co		367		
Ni		7 333		
Material	Mass released, conservative case		Mass released, realistic case	
	Co [g]	Ni [g]	Co [g]	Ni [g]
Total*	1 424.70	78 607.60	421.21	15 722.39

* Total means the sum of stainless steel, A690 and Stellite.

Table 5.2: Estimated concentrations of elementary Co and Ni in the CS water in primary circuit in R3, calculated for one operating cycle (i.e. 330 days), using the real compositions of the materials. The top half of the table is based on measured chemistry data and the bottom half is based on calculations from real compositions.

Element		Concentration [ppb] In RCS water (water mass flow: 12 860 kg s ⁻¹ [41])		
Co		1×10^{-3}		
Ni		2×10^{-2}		
Material	Concentration, conservative case		Concentration, realistic case	
	Co [ppb]	Ni [ppb]	Co [ppb]	Ni [ppb]
Total*	3.89×10^{-3}	2.14×10^{-1}	1.15×10^{-3}	4.29×10^{-2}

* Total means the sum of stainless steel, A690 and Stellite.

Table 5.3: Estimated activities of ⁶⁰Co and ⁵⁸Co in the RCS water in primary circuit in R3, calculated for one operating cycle (i.e. 330 days), using the real compositions of the materials. The top half of the table is based on calculations from measured chemistry data and the bottom half is based on calculations from real compositions.

Nuclide		Activity [Bq] In RCS water (water mass flow: 12 860 kg s ⁻¹)		
⁶⁰ Co		7.67×10^{10}		
⁵⁸ Co		1.15×10^{12}		
Material	Activity, conservative case		Activity, realistic case	
	⁶⁰ Co [Bq]	⁵⁸ Co [Bq]	⁶⁰ Co [Bq]	⁵⁸ Co [Bq]
Total	2.98×10^{11}	1.23×10^{13}	8.81×10^{10}	2.46×10^{12}

* Total means the sum of stainless steel, A690 and Stellite.

As can be seen in Table 5.1, the estimated released masses using calculation from the real compositions are higher than the ones calculated from measured chemistry data. The same goes for the concentrations in Table 5.2 and activities in Table 5.3. However, What can be stated is that the "realistic case" is closer to the ones from measured chemistry data than what the "conservative case" is which could indicate that the values used in the calculations for the realistic case are more like the actual values. The fact that the conservative and realistic cases are higher than the actual values could be due to that some material gets stuck in cracks and bumps on the surfaces but most likely it is because of the conservative assumptions regarding surface areas and corrosion rates. With these different things in mind, it could be determined that the estimation calculations based on the real compositions in the materials could be seen as a worst-case-scenario since that is what the maximum of released masses, activities etc. would be. If one can estimate the surface activity caused by the materials, this could also be compared with the SAM results presented in §4.5.2, to estimate what the surface activity would be in different parts of the primary circuit. The calculated activities can also be compared to the ones measured as shut down transient normalized to full power year, see measured activities in the first paragraph in §4.5.1. These activities are in the same range as the ones calculated and shown in Table 5.3.

From charts it was also seen that Co is measured in the RC water, that is the water which comes from the outlet of the core. This means that the assumption made, when determining the reaction cross section for the (n,p) reaction as well as neutron fluxes as described in §4.4.1, that all Co and Ni is deposited on the surfaces such as the fuel is not a realistic assumption. When doing the estimated calculation over the whole core, the assumed reaction cross section and neutron fluxes might still be good. If the conservative assumption that all Co and Ni is deposited on the fuel without being

released again is used, the RC water flow could be used to calculate the source term. If so, one could use the difference between CS and RC sampling to calculate how much is deposited on the fuel. The reason why the masses and concentrations of Co and Ni were determined for the CS water was due to that the RC sampling is not entirely representative of what really is in RC due to long sampling lines and large temperature differences.

5.3 Manufacturing and Cost Issue

From the interview with Carl-Filip Lindahl from Sandvik AB [36], it was stated that their production of steel is mostly based on recycled material which in turn means that they are able to have good knowledge about the composition of their materials as well as achieving a good "purity" of their materials. It seems like there is no problem for Sandvik AB to produce materials containing of 0.05 wt% Co if needed, which otherwise could be more difficult.

Since the requirements in the Swedish system of regulations coupled with the Swedish utilities Interpretation and additional requirements are more strict than in the rest of the world, it is a bit more difficult when Swedish nuclear power plants need to use suppliers of materials from other parts of the world. That in turn makes it more difficult from an economic point of view since it would be more costly to hold down the Co content and also more difficult to find suppliers who produce materials with that low Co content. If one would only look at the cost aspect, it is easy to find materials for a good price but since the requirements also have to be fulfilled, the matter in question gets more complex. When speaking to personnel at Ringhals who are working with these type of questions, e.g. what the requirements should be and similar, it seems like all agree that the requirement of 0.05 wt% Co in the RPV and the RPVs internals should still be strict and that the current requirement is relevant. This is due to that all material exposed to induced activity later must be taken care of during maintenance, replacements or repository of the material. This will result in a higher dose to personnel and higher costs if the material is more active.

As has been shown in §4.4.1 and §4.4.2, Ni is the element which in largest amount is released to the primary circuit. This also indicates that replacement of smaller components or surfaces to something containing of a slightly higher Co content than now, would probably not make too much of a difference. This shows that the requirement of 0.20 wt% for components outside the radiation field with a surface area of $\leq 10 \text{ m}^2$ could be changed in the future. This could potentially be applied to some larger surface areas as well if the corrosion rate is kept low. If all components were to be replaced by materials with higher Co content, then the difference would probably be a bit larger due to that there will be a larger amount of Co in the primary system after a while. If one would like to replace all components of e.g. stainless steel, to ones with a higher Co content, this report could be used as a basis to see how it would change.

The Co content in the SG tubes are of more importance, than in smaller components, due to the large surface area of A690. This means that if the Co content were to be increased in the SG tubes, this would result in a larger difference in the activity build-up in the system. Since the SG tubes are manufactured by Sandvik AB, which is a Swedish company, it was possible to have lower Co contents in the material than if the SG tubes were manufactured by another, foreign company. As was seen in Table 4.4 in §4.4.1, the average composition of Ni and Co in the material A690 used in the SG tubes are 59.5 and 0.013 respectively. 0.013 wt% Co is well below the requirement of 0.05 wt% Co.

5.4 ALARA Perspective

As was mentioned in §4.6, Sweden has a lower alpha value than many other countries. This could be because the dose to personnel is not as much of a problem as it maybe is in the rest of the world. Sweden is very keen on keeping workplaces safe and if more money was needed to keep the personnel doses down even more, money would probably be inserted to the nuclear power plants to obtain this. This could however be discussed further since it also is a political question. SSM also checks how the nuclear power plants operate, how much doses personnel take, how much emissions there are etc. which mean that if the doses to personnel would increase much, they will take action and make up action plans for the particular power plant with the problem.

Since the personnel doses also have been decreased as much as they have, so that the doses are well under reasonable levels according to radiation protection personnel [46], this might also be an argument for that higher dose rates and possibly an increase in personnel doses because of a slightly higher Co content in the materials might be accepted. Even though it is easy to put a price on a material, it is more difficult to put a price on the dose to personnel when deciding if the requirement should be changed to allow a higher Co content in the materials. The use of the alpha value could potentially be a good way of doing this if deciding to go through with a change of the requirements. Depending on what the renewed regulatory collection by SSM will look like, this might also be a factor to consider in the future.

5.5 Future Works

Since this project has been performed during 20 weeks, there are more interesting things which could be studied to complement the project. Since it was seen that the Co content in stainless steel materials might not be the largest problem, but instead the Ni which is released from the SG tubes, an interesting thing to study more is how the chemistry control could be optimized to keep the doses low. Changing the chemistry parameters or water chemistry can result in that the stability of the oxides could change. Other ways of reducing the activation and release of crud from the fuel could also be studied.

Since there are other nuclides than ^{60}Co and ^{58}Co that are problematic, the study could be extended to also look at other nuclides of interest. Beside radioactive Co, isotopes of Ag and especially Sb are also about to be regulated in the same way, since it is of importance to reduce these. It could therefore be interesting to study from where ^{93}Mo , ^{110m}Ag and ^{124}Sb originate and in what amounts, and what could be done to reduce them.

A third thing which could be performed is to not only look at R3, but also reactor unit 1 (BWR) to better see the difference of the result when comparing a PWR unit to a BWR unit at Ringhals. The study can also be performed for R4.

It could also be interesting to see if it is possible to study and measure old material samples from e.g. hotspots (i.e. a hotspot of activity). This is to see if there are particular locations in the power plant where activity more easily accumulates, resulting in a higher surface activity in that spot. For this, SAM could be used as a tool to also measure the surface areas where the old material once was located.

A fifth interesting thing would be to see if it is possible to create a model in e.g. the calculation software Matlab which is often used at Chalmers University of Technology. This program has been used in previous chemical engineering courses to set up equations for flows and compositions in a

system. However, this case might be too complex to be able to simulate the problem studied in this master's thesis.

A last thing which could be examined is to see if some of the conservative assumptions made in this project could be made more realistic. This could be done by looking even closer at design drawings, material certificates and corrosion rates of specific components and materials, instead of looking at the total surface area of a material in the primary circuit in R3, to see what the change would be if changing a particular component. This could then be linked to in what way and where in the power plant personnel receive the largest dose, by studying dose reports from the dosimetry department at Ringhals.

6

Conclusion

From this master's thesis it has been seen that an increase in Co content would probably not make a large difference in the power plant from an activity production and radiation safety point of view. Since most of the released Co and Ni comes from the SG tubes and the Stellite it is important to keep the Co content low in the SG tubes. Currently, there are no other known alloys available which have equally good tribological properties as Stellite in the sense that it could replace the Stellite in the components. This means that the activity build-up caused by Stellite will not change if changing the requirements of Co content. It might be more relevant to study how to optimize the chemistry control and how to clean the systems efficiently if increasing the Co content in the materials.

Because of this, the framework of nuclear-specific requirements presently used will probably not be adequate when Ringhals will operate PWR technology only. From the study it has been seen that the requirement of Co content in materials inside the RPV and its internals should be kept strict at 0.05 wt%. Co should be continually strict. Outside the RPV and its internals, it has been seen that the requirement could be less strict if the surface area is $\leq 10 \text{ m}^2$. This could potentially be applied to larger surface areas as well. It has also been stated that the strict requirement of 0.05 wt% Co in the SG tubes, which has also been achieved with margins since the Swedish company Sandvik AB has manufactured them, is of great relevance and should be unchanged. An idea is that the requirements could be a bit more flexible depending on what it applies to. For example in small tube parts in the PRZ, the Co content could maybe be 0.30 wt% instead of 0.20 wt%. Since SSM is renewing their regulatory collection, it is also of great interest to see what is written in it when they publish it.

It has been a bit difficult to determine a reasonable level from an ALARA perspective. The Swedish power plants such as Ringhals are working more towards "As Low as Achievable", that is working towards zero dose to personnel. Therefore, an interest to reintroduce the R - *Reasonable* in the ALARA concept exists among radiation protection personnel. This might be an argument for that it would be acceptable if the dose rates increases slightly as a consequence of increased Co content in some components.

Since the Co requirements have been discussed for a very long time, this project has also been a way to lift the question and to provide a basis that can be used when updating requirement specifications in the future.

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A

Appendix: Estimation Calculations

The estimation calculations were performed according to the procedure described in §3.2. In this appendix, all input data and data that has been calculated are presented. The figures in this section are not described more than with their figure texts.

A.1 General Data

R3						
Material	<i>Stainless steel</i>					
Density		7,93	Mg/m ³			
Composition assumed in Ringhals report	Al	0,00002	or	0,002	wt%	
	Si	0,006	or	0,6	wt%	
	Ti	0,0001	or	0,01	wt%	
	V	0,00001	or	0,001	wt%	
	Cr	0,185	or	18,5	wt%	
	Mn	0,013	or	1,3	wt%	
	Fe	0,690809	or	69,0809	wt%	
	Co	0,0003	or	0,03	wt%	
	Ni	0,1	or	10	wt%	
	Cu	0,001	or	0,1	wt%	
	As	0,0001	or	0,01	wt%	
	Nb	0,0001	or	0,01	wt%	
	Mo	0,002	or	0,2	wt%	
	Sn	0,0001	or	0,01	wt%	
	Sb	0,00001	or	0,001	wt%	
	Ta	0,0001	or	0,01	wt%	
	W	0,0001	or	0,01	wt%	
Corrosion rates	Conservative case	50	mdm	=	mg/(dm ² *month)	
	Realistic case	10	mdm	=	mg/(dm ² *month)	

Figure A.1: Assumed composition of stainless steel in primary circuit in R3 [38].

Material	Stellite					
Density		8,69	Mg/m ³			
Composition assumed in Ringhals report	Al			or		
	Si		0,011	or	1,1 wt%	
	Ti			or		
	V			or		
	Cr		0,28	or	28 wt%	
	Mn		0,005	or	0,5 wt%	
	Fe		0,011	or	1,1 wt%	
	Co		0,628	or	62,8 wt%	
	Ni		0,004	or	0,4 wt%	
	Cu		0,001	or	0,1 wt%	
	As			or		
	Nb			or		
	Mo			or		
	Sn			or		
	Sb			or		
	Ta			or		
	W		0,05	or	5 wt%	
Corrosion rates	Conservative case	200	mdm	=		mg/(dm ² *month)
	Realistic case	60	mdm	=		mg/(dm ² *month)

Figure A.2: Assumed composition of Stellite in primary circuit in R3 [38].

The composition of Co and Ni in A690 has previously been described in Table 4.4.

Data for R3 and primary side		
Stainless steel	3000	m ²
	300000	dm ²
A690	19318,5	m ²
	1931850	dm ²
Internals	500	m ²
	50000	dm ²
Stellite	10	m ²
	1000	dm ²
Operating cycle (days during a year)	330	days

Figure A.3: Total surface areas of the materials stainless steel, A690 and Stellite in primary circuit in R3 [38].

A.2 Estimation Calculations with Real Compositions

ppb = ug Co / kg water			Mass flows [ug element/s]		
			Material	Conservative case	Realistic case
Water flow through CS ion exchanger	7	kg/s	A690		
Total mass flow of coolant water	12860	kg/s	Co	0,463114726	0,092622945 ug Co/s
Mass flow of coolant water per loop	4286,666667	kg/s	Ni	2185,919884	437,1839769 ug Ni/s
Volume flow of water through each loop	5,7	m ³ /s	Total	3675,513699	735,1027397 ug/s
Density of water	758	kg/m ³	Stainless steel		
			Co	1,712328767	0,342465753 ug Co/s
			Ni	570,7762557	114,1552511 ug Ni/s
			Total	5707,762557	1141,552511 ug/s
Operating cycle	330	days	Stellite		
	7920	h	Co	47,79299848	14,33789954 ug Co/s
	28512000	s	Ni	0,304414003	0,091324201 ug Ni/s
			Total	76,10350076	22,83105023 ug/s
			Total		
			Co	49,96844197	14,77298824 ug Co/s
			Ni	2757,000554	551,4305522 ug Ni/s
			Total	9459,379756	1899,486301 ug/s
			Concentrations [ppb]		
			Material	Conservative case	Realistic case
			A690		
			Co	3,6012E-05	7,20241E-06 ppb Co
			Ni	0,169978218	0,033995644 ppb Ni
			Total	0,285809774	0,057161955 ppb
			Stainless steel		
			Co	0,000133152	2,66303E-05 ppb Co
			Ni	0,044383846	0,008876769 ppb Ni
			Total	0,443838457	0,088767691 ppb
			Stellite		
			Co	0,003716407	0,001114922 ppb Co
			Ni	2,36714E-05	7,10142E-06 ppb Ni
			Total	0,005917846	0,001775354 ppb
			Total		
			Co	0,003885571	0,001148755 ppb Co
			Ni	0,214385735	0,042879514 ppb Ni
			Total	0,735566077	0,147705 ppb

Figure A.7: Estimated concentrations of elementary Co and Ni in CS in primary circuit in R3.

A.3 Estimation Calculations with Theoretical Cobalt Contents

All calculations for the compositions 0.05, 0.10, 0.15, 0.20, 0.25 and 0.30 wt% Co in the materials stainless steel and A690 are not shown in this section. Instead, the calculations using 0.05 wt% Co in stainless steel and A690 are shown as one example. If one would like to know the values for the other compositions, the values for 0.05 wt% Co can be multiplied with a factor as described later in appendix B. The values of Stellite and the calculations which belongs to the Ni or ⁵⁸Co calculations will not change with increasing Co content (since Stellite was assumed to have the same composition as the real composition, due to that the very high Co content in Stellite is an exception not following the requirements).

Calculated with composition of Co: 0,05; 0,10; 0,15; 0,20; 0,25; 0,30 wt% (for A690 and Stainless steel)				0,05 wt% Co			
The calculated total amount mass released is during one operating cycle (~330 days).							
$m_{\text{released}} [\text{mg}] = r_{\text{corr}} [\text{mdm}] \cdot A_{\text{material}} [\text{dm}^2] \cdot t_{\text{cycle}} [\text{months}]$							
Release rate $[\text{kg/month}] = r_{\text{corr}} [\text{mdm}] \cdot A_{\text{material}} [\text{dm}^2]$							

[illegible]

Figure A.9: Estimated number of target Co and Ni atoms from the materials in primary circuit in R3 using 0.05 wt% as theoretical Co content in stainless steel and A690.

0,05 wt% Co		Mass flows (ug element/s)	
Material		Conservative case	Realistic case
A690	Co	1,837756849	0,36755137 ug Co/s
	Ni	2185,919884	437,1839769 ug Ni/s
	Total	3675,513699	735,1027397 ug/s
Stainless steel	Co	2,853881279	0,570776256 ug Co/s
	Ni	570,7762557	114,1552511 ug Ni/s
	Total	5707,762557	1141,552511 ug/s
Stellite	Co	47,79299848	14,33789954 ug Co/s
	Ni	0,304414003	0,091324201 ug Ni/s
	Total	76,10350076	22,83105023 ug/s
Total	Co	52,48463661	15,27622717 ug Co/s
	Ni	2757,000554	551,4305522 ug Ni/s
	Total	9459,379756	1899,486301 ug/s
		Concentrations [ppb]	
Material		Conservative case	Realistic case
A690	Co	0,000142905	2,8581E-05 ppb Co
	Ni	0,169978218	0,033995644 ppb Ni
	Total	0,285809774	0,057161955 ppb
Stainless steel	Co	0,000221919	4,43838E-05 ppb Co
	Ni	0,044383846	0,008876769 ppb Ni
	Total	0,443838457	0,088767691 ppb
Stellite	Co	0,003716407	0,001114922 ppb Co
	Ni	2,36714E-05	7,10142E-06 ppb Ni
	Total	0,005917846	0,001775354 ppb
Total	Co	0,004081231	0,001187887 ppb Co
	Ni	0,214385735	0,042879514 ppb Ni
	Total	0,735566077	0,147705 ppb

Figure A.10: Estimated concentrations of elementary Co and Ni in CS in primary circuit in R3 using 0.05 wt% as theoretical Co content in stainless steel and A690.

A.4 Estimation Calculations of Activities

Summary of calculated activities						
Compositions from	Material	Conservative case		Realistic case		Unit
		Co-60	Co-58	Co-60	Co-58	
Material certificates + Ringhals report	A690	2,7627E+09	9,7587E+12	5,5254E+08	1,9517E+12	Bq
	Stainless steel	1,0215E+10	2,5481E+12	2,0430E+09	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	2,9809E+11	1,2308E+13	8,8128E+10	2,4618E+12	Bq
0,05 wt% Co	A690	1,0963E+10	9,7587E+12	2,1926E+09	1,9517E+12	Bq
	Stainless steel	1,7025E+10	2,5481E+12	3,4049E+09	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	3,1310E+11	1,2308E+13	9,1130E+10	2,4618E+12	Bq
0,10 wt% Co	A690	2,1926E+10	9,7587E+12	4,3852E+09	1,9517E+12	Bq
	Stainless steel	3,4049E+10	2,5481E+12	6,8099E+09	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	3,4108E+11	1,2308E+13	9,6727E+10	2,4618E+12	Bq
0,15 wt% Co	A690	3,2889E+10	9,7587E+12	6,5778E+09	1,9517E+12	Bq
	Stainless steel	5,1074E+10	2,5481E+12	1,0215E+10	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	3,6907E+11	1,2308E+13	1,0232E+11	2,4618E+12	Bq
0,20 wt% Co	A690	4,3852E+10	9,7587E+12	8,7705E+09	1,9517E+12	Bq
	Stainless steel	6,8099E+10	2,5481E+12	1,3620E+10	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	3,9706E+11	1,2308E+13	1,0792E+11	2,4618E+12	Bq
0,25 wt% Co	A690	5,4815E+10	9,7587E+12	1,0963E+10	1,9517E+12	Bq
	Stainless steel	8,5124E+10	2,5481E+12	1,7025E+10	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	4,2505E+11	1,2308E+13	1,1352E+11	2,4618E+12	Bq
0,30 wt% Co	A690	6,5778E+10	9,7587E+12	1,3156E+10	1,9517E+12	Bq
	Stainless steel	1,0215E+11	2,5481E+12	2,0430E+10	5,0963E+11	Bq
	Stellite	2,8511E+11	1,3590E+09	8,5532E+10	4,0770E+08	Bq
	Total	4,5303E+11	1,2308E+13	1,1912E+11	2,4618E+12	Bq

Figure A.11: Summary of estimated activities for both the real and the theoretical compositions of the materials.

A.5 Calculations Based on Data from Chemistry Data

ppb = ug of element / kg water		
Water flow through CS ion exchanger	7	kg/s
Total mass flow of coolant water	12860	kg/s
Mass flow of coolant water per loop	4286,666667	kg/s
Volume flow of water through each loop	5,7	m ³ /s
Density of water	758	kg/m ³
Operating cycle	330	days
	7920	h
	28512000	s
Co in RC and CS from measurements before ion exchange	0,001	ppb = ug Co/kg water
Co in RC and CS from measurements before ion exchange	0,02	ppb = ug Ni/kg water
In ion exchanger:		
Mass of Co	0,007	ug Co/s
	199584	ug Co/operating cycle
	0,199584	g Co/operating cycle
Mass of Ni	0,14	ug Ni/s
	3991680	ug Ni/operating cycle
	3,99168	g Ni/operating cycle

Figure A.12: Data gathered from charts and reports at Ringhals, including the chemistry department. Calculations of how much Co in grams that are measured in the ion exchanger per operating cycle.

Activity calculations (based on mass from ion exchanger, water mass flow of 7 kg/s):			
Total			
m_Co(tot)	0,199584	g	
M_Co	58,933198	g/mol	Molar mass for Co-59
n_Co(tot)	0,003386614	mol	
N_Co(tot)	2,03947E+21	Co atoms	
Abundance Co-59	100	atom%	
	1		
N_Co-59	2,03947E+21	Co-59 atoms	
Total			
m_Ni(tot)	3,99168	g	
M_Ni	58,69335111	g/mol	Molar mass, weighted for the naturally abundant Ni-isotopes: Ni-58, Ni-60, Ni-61, Ni-62, Ni-64
n_Ni(tot)	0,068009066	mol	
N_Ni(tot)	4,0956E+22	Ni atoms	
Abundance Ni-58	68,077	atom%	
	0,68077		
N_Ni-58	2,78816E+22	Ni-58 atoms	
Total			
Nt_Co-59	2,03947E+21	Co atoms	
Nt_Ni-58	2,78816E+22	Ni atoms	
Total			
k_Co-60	3,03E+13	1/s	
k_Co-58	1,67E+13	1/s	
Total			
N_Co-60	1,00146E+16	atoms	
N_Co-58	5,52046E+15	atoms	
Total			
A_Co-60	41758278,48	Bq	
	41,75827848	MBq	
A_Co-58	625008335,3	Bq	
	625,0083353	MBq	

Figure A.13: Activity calculations (in the ion exchanger) based on calculated data from reports and charts from the chemistry department. The activities are based on the mass calculated from measured chemistry data, using the assumption that it is 0.001 ppb Co and 0.02 ppb Ni in the water as measured in the ion exchanger.

ppb = ug of element / kg water		
Water flow through CS ion exchanger	7	kg/s
Total mass flow of coolant water	12860	kg/s
Mass flow of coolant water per loop	4286,666667	kg/s
Volume flow of water through each loop	5,7	m ³ /s
Density of water	758	kg/m ³
Operating cycle	330	days
	7920	h
	28512000	s
Co in RC and CS from measurements before ion exchange	0,001	ppb = ug Co/kg water
Co in RC and CS from measurements before ion exchange	0,02	ppb = ug Ni/kg water
In CS:		
Mass of Co	12,86	ug Co/s
	366664320	ug Co/operating cycle
	366,66432	g Co/operating cycle
Mass of Ni	257,2	ug Ni/s
	7333286400	ug Ni/operating cycle
	7333,2864	g Ni/operating cycle

Figure A.14: Data gathered from charts and reports at Ringhals, including the chemistry department. Calculations of how much Co in grams that are measured in the CS water per operating cycle, using the assumption that it is 0.001 ppb Co and 0.02 ppb Ni in the water as measured in the ion exchanger.

Activity calculations (based on mass calculated in CS, water mass flow of 12 860 kg/s):			
Total			
m_Co(tot)	366,66432	g	
M_Co	58,933198	g/mol	Molar mass for Co-59
n_Co(tot)	6,221693925	mol	
N_Co(tot)	3,74679E+24	Co atoms	
Abundance Co-59	100	atom%	
	1		
N_Co-59	3,74679E+24	Co-59 atoms	
Total			
m_Ni(tot)	7333,2864	g	
M_Ni	58,69335111	g/mol	Molar mass, weighted for the naturally abundant Ni- Isotopes: Ni-58, Ni-60, Ni-61, Ni-62, Ni-64
n_Ni(tot)	124,9423701	mol	
N_Ni(tot)	7,52421E+25	Ni atoms	
Abundance Ni-58	68,077	atom%	
	0,68077		
N_Ni-58	5,12225E+25	Ni-58 atoms	
Total			
Nt_Co-59	3,74679E+24	Co atoms	
Nt_Ni-58	5,12225E+25	Ni atoms	
Total			
k_Co-60	5,58E+16	1/s	
k_Co-58	3,07E+16	1/s	
N_Co-60	1,83982E+19	atoms	
N_Co-58	1,01419E+19	atoms	
A_Co-60	76715923031	Bq	
	0,076715923	TBq	
A_Co-58	1,14823E+12	Bq	
	1,148229599	TBq	

Figure A.15: Activity calculations (in the ion exchanger) based on calculated data from reports and charts from the chemistry department. The activities are based on the mass which is assumed to be in the CS water using the assumption that it is 0.001 ppb Co and 0.02 ppb Ni in the water as measured in the ion exchanger.

B

Appendix: Derivation of Estimated Activity Increase

In this appendix, the derivation of the estimated activity increase obtained if having a higher Co content in a material is shown. All denotations are previously described in §2.8, §3.2 and in the list of abbreviations and symbols in the beginning of the document. The reader is therefore referred to these sections if an update on what they mean is needed.

Equations (2.2), (2.3), (3.1), (3.3), (3.6) and (3.7) are collected in this appendix as Equation (B.1) to (B.6) where the nuclides of interest have been inserted in the equations.

$$m_{released,i} = r_{corr,i} A_i t_{oc} \quad (B.1)$$

$$m_{Co,i} = m_{released,i} \frac{C_{Co,i}}{100} \quad (B.2)$$

$$N_{Co,i} = \frac{m_{Co,i}}{M_{59Co}} N_A \quad (B.3)$$

$$N_{t,59Co,i} = N_{Co,i} X_{59Co} \quad (B.4)$$

$$k_{60Co} = \phi \sigma_{n,\gamma} N_{t,59Co} \quad (B.5)$$

$$A_{60Co} = k_{60Co} \left[1 - \exp(-\ln(2)t_{irr}/t_{1/2,60Co}) \right] \exp(-\ln(2)t_{cool}/t_{1/2,60Co}) \quad (B.6)$$

Since all expressions in the equations consists of only factors, all equations can be combined to one by inserting Equation (B.1) into Equation (B.2), then Equation (B.3) into Equation (B.4) and so forth. This yields an expression for the activity of ^{60}Co as shown in Equation (B.7).

$$A_{60Co} = \phi \sigma_{n,\gamma} \frac{r_{corr,i} A_i t_{oc} \frac{C_{Co,i}}{100}}{M_{59Co}} N_A X_{59Co} \left[1 - \exp(-\ln(2)t_{irr}/t_{1/2,60Co}) \right] \exp(-\ln(2)t_{cool}/t_{1/2,60Co}) \quad (B.7)$$

Equation (B.7) can be rewritten as Equation (B.8), since $C_{Co,i}$ can be moved around due to that all are factors, as previously mentioned. By putting brackets around the expression and by moving $C_{Co,i}$ out from the brackets, it is easier to see that all inside of the brackets are or are assumed to be constant.

$$A_{60Co} = C_{Co,i} \left[\varphi \sigma_{n,\gamma} \frac{r_{corr,i} A_i t_{oc} \frac{1}{100}}{M_{59Co}} N_A X_{59Co} \left[1 - \exp \left(- \ln(2) t_{irr} / t_{1/2, 60Co} \right) \right] \exp \left(- \ln(2) t_{cool} / t_{1/2, 60Co} \right) \right] \quad (B.8)$$

Since all inside the large brackets are or are assumed to be constant, this is further abbreviated as B . This gives a shorter expression as shown in Equation (B.9), where B is a constant.

$$A_{60Co} = C_{Co,i} B \quad (B.9)$$

If assuming that $C_{Co,i} = 0.0005$, that is 0.05 wt%, this results in Equation (B.10).

$$A_{60Co,1} = 0.0005 B \quad (B.10)$$

What is of interest now is what happens to the activity if $C_{Co,i}$ is increased to 0.001, that is 0.10 wt%. To go from 0.0005 to 0.01 the expression in Equation (B.10) needs to be multiplied by 2 which means that the starting activity yielded from a 0.05 wt% Co content will also increase by a factor 2. To go up to 0.015, Equation (B.10) can be multiplied by a factor 3, yielding in an increase of the activity by a factor 3 compared to the initial value and so forth up to in this case 0.30 wt% Co content.

C

Appendix: Bar Charts of Estimated Values

In this appendix, the estimated values of masses released, activities and concentrations are shown. Since all charts takes up much space, they are all first described in this page to then be seen as figures on the following pages, one by one.

The estimated released masses of elementary Co in grams from each material are shown as a bar chart in Figure C.1, which also is rotated 90° to be able to see the data better. The values are taken from Tables 4.5 and 4.11. The released masses from Stellite as well as the released masses of Ni in each of the materials are not shown since these masses do not change in the calculations and is therefore not of interest to compare. The released mass of Co from Stellite is however included in the total mass released in Figure C.1 since it contributes to the increased total mass released.

The estimated activities of ^{60}Co in Becquerel shown in Table 4.8 and 4.12 are shown as a bar chart in Figure C.2, which also is rotated 90° to be able to see the data better. The activities from ^{58}Co and all activities formed by mass released from Stellite are not shown since their activities do not change in the calculations and is therefore not of interest to compare. The activity of ^{60}Co from Stellite is however included in the total activity in Figure C.2 since it contributes to the increased total activity.

The estimated concentrations of elementary Co in ppb shown in Table 4.9 and 4.13 are shown as a bar chart in Figure C.3, which also is rotated 90° to be able to see the data better. The concentrations of Ni from each material as well as Co formed by mass released from Stellite are not shown since the concentration do not change in the calculations and is therefore not of interest to compare. The concentration of Co from Stellite is however included in the total concentration in Figure C.3 since it contributes to the increased total concentration.

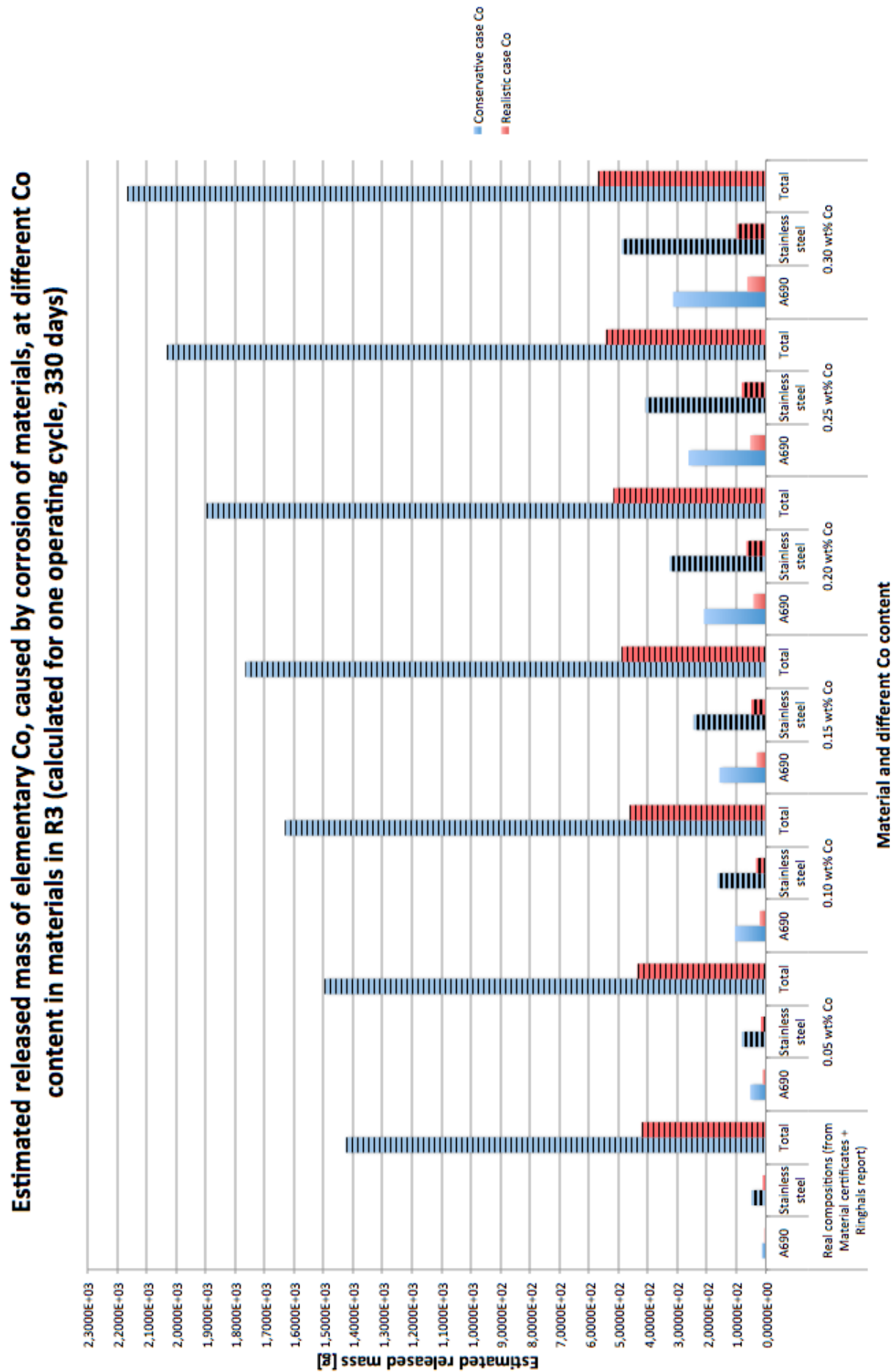


Figure C.1: Estimated released masses of elementary Co using both the real Co contents and different theoretical Co contents in the materials.

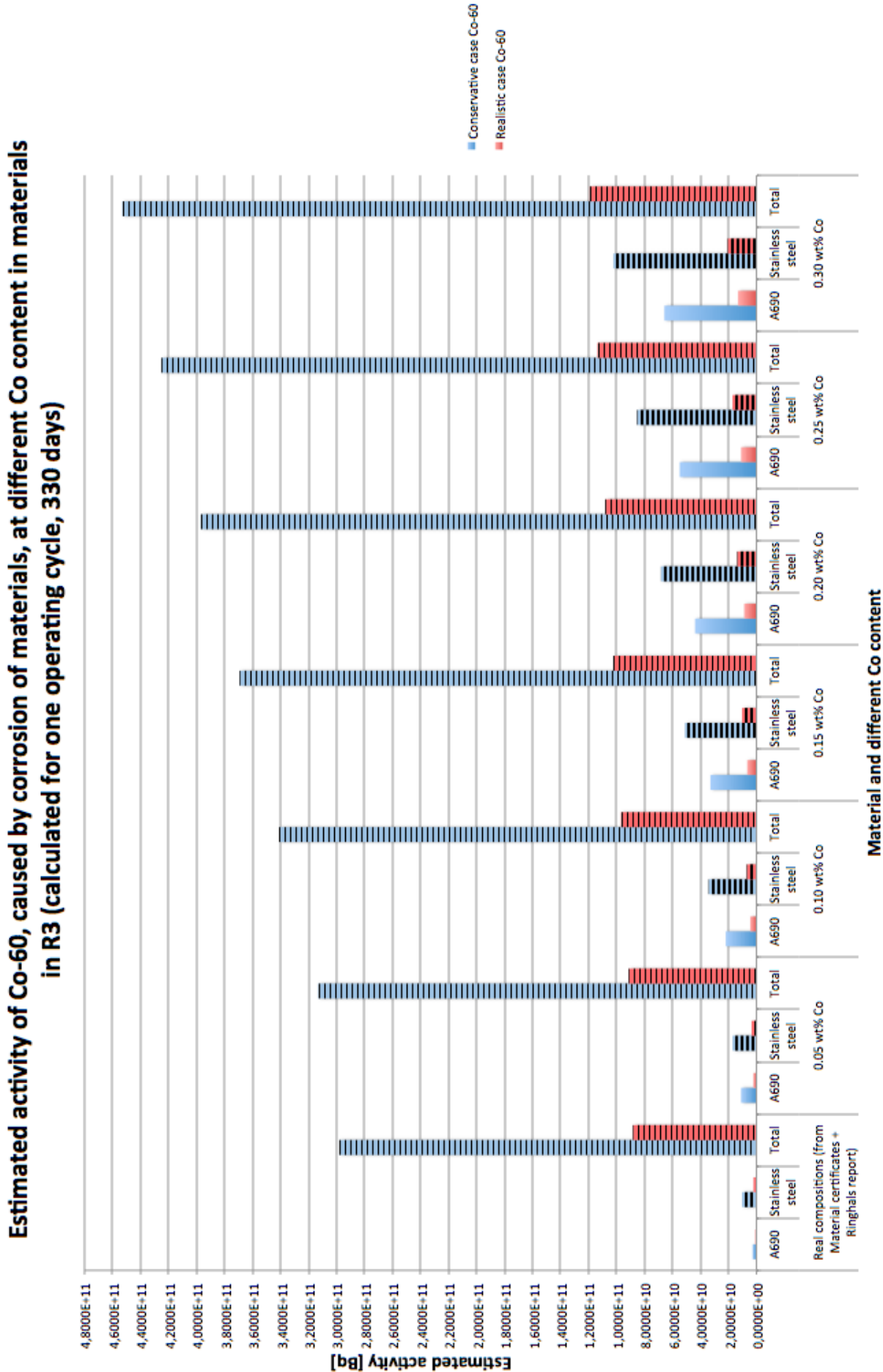


Figure C.2: Estimated activities of ⁶⁰Co using both the real Co contents and different theoretical Co contents in the materials.

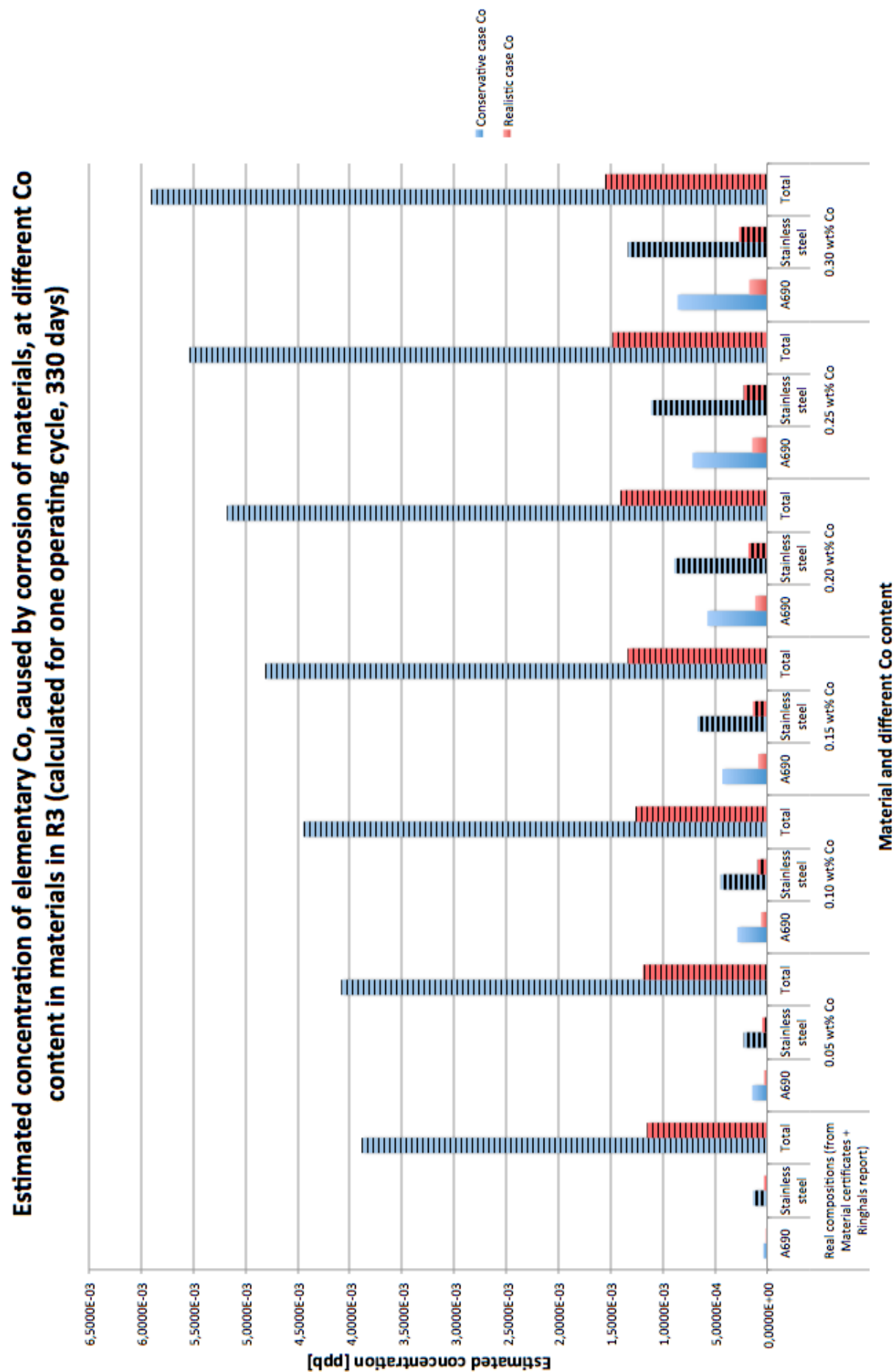


Figure C.3: Estimated concentrations of elementary Co using both the real Co contents and different theoretical Co contents in the materials.

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