RESEARCH REPORT

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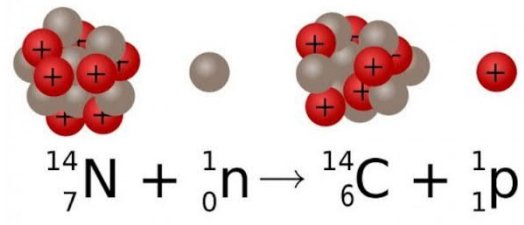

Impurities in LWR fuel and structural materials

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Summary

Nuclear fuel and fuel assembly structural materials contain various impurity elements. Many of these impurity elements are activated in the heavy neutron irradiation in a nuclear reactor and must be considered in the safety analysis of spent nuclear fuel handling, interim storage and final disposal. This report presents a literature survey on impurity elements found in UO_2 fuel, Zry-2, Zry-4, Zr-2.5Nb, Inconel 718, Inconel X-750 and stainless steels SS 304 and SS 304 L. The emphasis is on the fuel and zirconium alloys.

The largest impurity element found in fuel was iron and the second largest thorium. There was wide variation between different literature sources on what elements were reported and on the given concentrations. However, iron was found in all of the studied references and most agreed it to be one of the two largest impurity elements. Altogether 67 different impurity elements were found in the literature sources for the UO_2 fuel.

The largest impurity element in the zirconium alloys was either carbon or silicon. The number of impurity elements mentioned in the different literature sources were 24 (Zry-2), 74 (Zry-4) and 53 (Zr-2.5Nb). Cobalt was the largest impurity element for the inconels and nitrogen for SS 304. Only a few impurity elements were mentioned in the studied sources for these materials.

Maximum nitrogen concentration in the inconels and stainless steel was 1300 ppm. For all of the Zircaloys it was 80 ppm and for the fuel 200 ppm. The maximum concentration for chlorine was 20 ppm in Zry-2 and Zry-4 and 15 ppm in Zr-2.5Nb and the fuel. Chlorine was not mentioned in the references for Inconel and stainless steel. However, chlorine is not likely to be absent in these materials either and a more thorough literature survey concentrating on Inconel and stainless steel would doubtlessly give estimates for it also.

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Contents

1. Introduction

The core of a commercial light water reactor generally comprises a couple of hundred fuel assemblies. Each fuel assembly contains fuel rods and structural materials designed to hold the fuel rods firmly in place. The fuel of commercial light water reactors is composed of uranium oxide UO_2 or in some cases (not in Finland) of mixed oxide MOX. The most common materials in fuel claddings and other structural materials are zirconium alloys mainly because of the low thermal neutron cross-section of zirconium. However, also stainless steels and Inconel can be used and have been used especially in some older designs.

In addition to the intended atomic elements both fuel and structural materials contain various impurity elements. In a nuclear reactor, many of these impurities are activated under the heavy neutron irradiation. This can become problematic when considering the long-term safety analysis of final disposal especially if the activation products are long-lived and highly mobile. For example, according to the POSIVA safety case study TURVA-2012 there are five radionuclides propagating to the biosphere and causing doses to humans in a reference scenario. The nuclides are C-14 (dominant nuclide), I-129, Cl-36, Ag-108m and Mo-93 [1]. C-14 and Cl-36 are mainly produced in the activation of N-14 and Cl-35. The other three nuclides are mainly produced as fission products, but Ag-108m and Mo-93 are also born in the activation of Ag-107 and Mo-92. These nuclides can become important in the long term safety analysis because of their long half lives 5700 y (C-14), 1.6E7 y (I-129), 3.0E5 y (cl-36), 438 y (Ag-108m) and 4000 y (Mo-93).

Short lived activation products must also be considered in radiation protection and overall management of spent fuel assemblies during interim storage and transportation. Such short lived nuclides are e.g. Co-60 ($T_{1/2}$=5.3 y) and Fe-55 ($T_{1/2}$=2.7 y). These nuclides are produced in the activation of Co-59 and Fe-54.

The composition of impurities in fuel and structural materials is not known exactly. The composition can also vary between different fuel assembly batches. Manufacturing techniques and even manufacturing site can influence the exact amounts of impurities. Different literature sources often give different values for impurities. Some sources report considerably more impurity elements and some only a few. The purpose of this report is to gather data on impurities from various sources. Later, this data will be used as a basis for calculations making conservative estimates on the impact of impurities on the spent fuel characteristics.

This report is structured as follows. First in section 2, a brief description of the fuel manufacturing process is given and the main manufacturing companies are listed. In section 3 a short description of some fuel assembly types and structural materials is presented. Section 4 introduces impurities in the materials studied in the report. Maximum values of the impurities found in all the studied literature sources are presented in tables and graphs. Section 5 presents a summary and final conclusions of the study and considers future plans for the research. All the impurity values found in the studied references are presented in tables at the end of the report in Appendix A.

2. Nuclear fuel fabrication

2.1 Manufacturing process

The information in this section on fuel manufacturing is mainly based on reference [2] and the information on zirconium production on reference [3].

The manufacturing process of nuclear fuel includes uranium ore mining and milling, conversion and enrichment and the final fuel fabrication. Uranium mining can be realized by either open

pit mining or by in-situ leaching techniques. In open pit mining the ore is excavated from a large open mine. In in-situ leaching oxygenated groundwater is circulated through porous uranium ore to dissolve the uranium oxide.

Milling is used to extract the uranium from the ore. The milling process involves crushing, grounding and leaching in a sulphuric acid in order to separate the uranium oxide from the waste rock. Finally it is dryed and often heated to produce the uranium oxide (U_3O_8) concentrate or “yellowcake” before shipping it for conversion and enrichment. The yellowcake generally contains more than 80 wt-% uranium.

In order to enrich the fuel it must first be converted into gaseous form. For this purpose, the U_3O_8 is converted into hexafluoride UF_6. It is in solid form in room temperature but sublimes into gas above 57 °C [4]. UF_6 must be handled and transported in water tight conditions since it reacts with water. The enrichment is realized in centrifuges that comprise of thousands of rapidly spinning vertical tubes. Due to the small mass difference between U-235 and U-238, the rapid spinning movement separates the gaseous isotopes.

Fuel fabrication comprises three phases:

1. Production of uranium dioxide from the enriched UF_6.

2. Producing high density accurately shaped ceramic UO_2 pellets.

3. Producing the rigid metal framework of the fuel assembly and loading the fuel pellets into cladded fuel rods, sealing the rods and putting them together into the fuel assembly.

Two types of conversion techniques exist to convert the UF_6 into UO_2, the wet and the dry method. There are two types of wet methods, the ammonium uranyl carbonate procedure (AUC) and the ammonium diuranate procedure (ADU). The procedures are named after their main intermediate compounds [5]. In both wet methods, UF_6 is injected into liquid water forming UO_2F_2 slurry. In the ADU procedure, ammonia (NH_3) is added to the mixture and reacts with UO_2F_2 to produce ammonium diuranate (ADU). In the AUC method the added compound is ammonium carbonate ($(NH_3)_2CO_3$) and the reaction product ammonium uranyl carbonate (AUC). In both procedures pure UO_2 is produced by filtering, drying and heating in a reducing atmosphere. In the dry method, the UF_6 is heated to a vapour and mixed with steam to produce UO_2F_2. Hydrogen H_2 in the steam removes the fluoride and chemically reduces the uranium to a pure microcrystalline UO_2 product. The morphology of the UO_2 powder produced with any of the three ways differs from each other. Therefore, the impurity content of the differently produced powders may also differ depending on the production technique.

Before the fabrication of ceramic pellets, the UO_2 powder may be conditioned by first homogenizing the powder and then adding additives. The additives may comprise e.g. burnable absorbers, lubricants and pore formers as well as U_3O_8. The conditioned powder is pressed into pellets and sintered by heating the pellets in high temperature in a reducing environment to consolidate them. Finally, the pellets are machined to exact dimensions to guarantee uniformity.

The fuel pellets are stacket into a Zircaloy tube, the fuel cladding, and the tube is filled with helium in high pressure. A free space, the plenum, is left between the top pellet and the fuel rod end plug to accommodate for thermal expansion and fission gas release inside the rod. A spring is placed in the plenum to prevent the fuel pellets from moving. The fuel rods are fixed into an assembly structure designed to hold the rods firmly in place. The assembly structures are typically made of steel and zirconium alloys and the fuel cladding consists of a zirconium alloy. The exact composition of the alloy depends on the manufacturer. The goal is to minimize the number of nuclides that absorb neutrons and improve the strength and corrosion resistance of the structures.

Zirconium can not be used as such in the structural materials because of its poor corrosion resistance. Instead, the corrosion resistance is enhanced by alloying. The alloy homogeneity and impurity concentrations of the final products are established in the very first steps of the metal manufacturing.

Nearly all zirconium metal is extracted from zircon sand, Zr-Hf SiO_4, occurring in beach sand all over the world. The zirconium hafnium ratio in zircon is about 50/1. Because of its large thermal neutron cross section hafnium is a very unwanted impurity and is separated from zirconium as effectively as possible during the manufacturing process. Three commercial processes are used to produce zirconium metal; the Kroll process, the Van Arkel process and the electrolytic process. Most manufactures use the Kroll process, sometimes called the sponge zirconium process. According to reference [3] published in 2009, Russian vendors use zirconium metal produced from a mixture of the Van Arkel and electrolytic processes. However, the Russians are also going to shift using the Kroll process.

In the Kroll process, zirconium is extracted from the zircon sand by converting it into $ZrCl_4$ through a carbo-chlorination process. After Zr/Hf separation process, Zr metal is obtained by zirconium reduction by reducing the gaseous $ZrCl_4$ using liquid magnesium at 850 °C in an oxygen-free environment. Possible magnesium remnants are removed from the produced sponge cake by distillation at 1000 °C. The impurity levels of chlorine and magnesium in the sponge are determined in this process step.

The Van Arkel process is capable of producing higher purity zirconium than the Kroll process, but is also considerably more expensive. The Van Arkel process is based on the zirconium iodine reaction producing ZrI_4. Zirconium is then separated from iodine at high temperature on an electrically heated filament.

In the electrolytic process, the concentration of hafnium is first reduced by fractional distillation of the zircon sand. Zirconium is separated from the zircon sand ($ZrSiO_4$) in a separate operation involving reaction with K_2SiF_6 to produce K_2ZrF_6. The electrolytic process is operated in closed gas tight cells at high temperature and inert atmosphere using 10 000 to 20 000 A current. K_2ZrF_6 is used as the electrolyte in KCL and the Zr is deposited on the cathode. Because of external water cooling the cell surfaces are covered with solidified electrolyte to avoid the pick-up of impurities. The impurity concent of zirconium produced by the electrolytic process is lower than in the Kroll process but higher than in the Van Arkel process. The cost of the method is similarly higher than that of the Kroll process but lower than the Van Arkel process.

More information on the manufacturing of fuel and fuel assembly parts can be found e.g. in reference [5] in chapter 5.

2.2 Manufacturers

Presently, the most significant global nuclear fuel vendors are Framatome, GNF (Global Nuclear Fuel), TVEL and Westinghouse [2]. They are also the fuel vendors presently supplying the Finnish power reactors. In addition to these, during 2001-2007 the VVER-440 fuel of the Finnish power company Fortum for Loviisa 1 was provided by BNFL (British Nuclear Fuels Ltd). It was a UK government owned company whose operations included nuclear fuel fabrication and other nuclear power related activities. All of the company's operations were sold by may 2009.

Since 2007, all fuel for the Loviisa power plant has been provided by TVEL who has been the fuel provider for Loviisa since the start of reactor operation in 1977. Also, the fuel for the Finnish power company Fennovoima's reactor Hanhikivi 1 will be provided by TVEL at least for the first 10 years of operation. TVEL manufactures and supplies fuel to commercial VVER reactors in Russia, in 15 European and Asian countries and in 30 research reactors around the world. Additionally TVEL supplies the fuel to all Russian nuclear powered ships [6].

TVO operates the Olkiluoto 1 and 2 reactors with mixed cores including different types of fuel. The fuel is purchased from several vendors including Framatome, GNF and Westinghouse.

GNF is a USA based company which supplies boiling water reactor fuel and fuel-related engineering services. GNF operates in a joint venture with Spain based ENUSA. Most of the fuel components are provided by GNF and mainly manufactured in Wilmington, USA. The components are then shipped to ENUSA for fabrication and final assembly of fuel bundles. TVO and GNF ENUSA have signed a contract for fuel supply (GNF2) to Olkiluoto 1 and 2 between the years 2020-2038 [7]. GNF ENUSA has previously supplied TVO also with GE14 fuel [8].

Framatome (former AREVA) is a designer and supplier of nuclear steam supply system and nuclear equipment including nuclear fuel [9]. Framatome's major share holder (75.5 %) is the power utility EDF largely owned by the French state. Framatome supplies BWR, PWR and research reactor fuel. They are capable of manufacturing conventional uranium oxide fuel, fuel with enriched reprocessed uranium and mixed oxide fuel. TVO's BWR reactors have previously used Framatome's predecessor's AREVA's fuels Atrium 10xM and Atrium 11 [8]. Atrium11 fuel contains the sintering additive chromium at a relatively high concentration [10].

Westinghouse is a versatile company who among many other things provides nuclear power plants and related products such as nuclear fuel [11]. Westinghouse provides fuel assemblies to BWRs, PWRs, AGRs and VVERs. One of the newest additions in the company's supply is the new BWR fuel Triton11. First Triton11 fuel assemblies were delivered to Olkiluoto in February 2019 [12]. Westinghouse has previously provided TVO with SVEA-96 Optima2 and SVEA-96 Optima3 fuel assemblies [8]. Triton11 contains Westinghouse ADOPT pellets, which contain the sintering additives aluminum and chromium at relatively high concentrations [13,14].

3. Fuel assembly types and structural materials

The materials used in fuel assemblies are mostly zirconium alloys due to their low thermal neutron cross section and adequate mechanical, corrosion and high temperature oxidation properties. Other materials such as Inconel and stainless steel (SS 304 L) may be used e.g. in springs and in some components in the bottom and top part of the assembly where the neutron flux is relatively low. The most common zirconium alloys used in light water reactors are [5]

- Zircaloy-2: BWR-cladding and water channels,
- Zircaloy-4: PWR cladding and structural materials, BWR water channels,
- E110 (Zr-1Nb): VVER and RBMK cladding and structural materials,
- M5 (Zr-1Nb): PWR cladding and structural materials,
- Zr-2.5Nb: PHWR and RBMK pressure tubes
- Multicomponent zirconium alloys with additions of iron, niobium and tin such as E635 for VVERs and RBMKs and ZIRLO for PWRs.

Zirconium alloys such as Zry-2 and Zry-4 contain altogether a couple of percents tin, iron, chromium, nickel (Zry-2) and oxygen for higher corrosion resistance. Zr-Nb alloys contain mostly zirconium, niobium and oxygen, but also iron and tin can be added (e.g. Zirlo and E635) [15]. In addition to the intentionally added "impurities" the alloys contain numerous unintentional impurities listed in section 4.

There is a wide variety of reactor and fuel types used around the world in commercial power reactors. Currently in Finland two types of reactors are in operation, VVER-440 in Loviisa and BWR in Olkiluoto. At the time of the writing of this report, the commercial operation of the EPR unit under construction in Olkiluoto is scheduled to start in the spring 2021. Additionally, a VVER-1200 unit is to be built in Pyhäjoki. Some other reactor types currently in operation around the world are the pressurized heavy water reactor CANDU in Canada, the British Advanced Gas cooled Reactor AGR and the graphite moderated Russian design RBMK. Figure 1 presents rough sketches of the VVER, BWR and EPR fuel assemblies including some of their top and bottom structures [16]. The following discussion about the assemblies is restricted mainly to the structural materials.

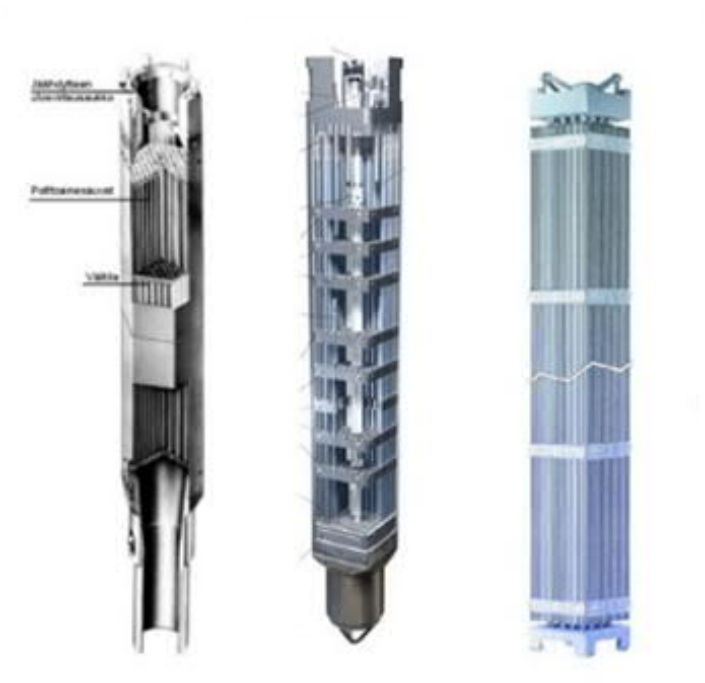

Figure 1. Sketches of VVER, BWR and EPR fuel assemblies. Source: Posiva Oy.

BWR and VVER-440 assemblies are contained inside shroud tubes. BWR shroud tubes typically consist of Zry-2, Zry-4 or in some newer designs ZIRLO [5,13] and VVER-440 tubes can be e.g. Zry-4 or in some early cases Zr-2.5Nb (E125). The control blades in BWRs move outside the shroud tubes. BWR assemblies do not have guide tubes for control rods in the assembly, but can contain water channels of different size and shape depending on the assembly design. The fuel cladding is typically Zry-2.

Fuel pins of the assemblies are generally fixed in place by several spacer grids positioned at different axial levels of the assembly. The spacer material for BWRs can be e.g. Zry-2, Inconel 718 or Inconel X-750. Spacers in PWRs are made of Zry-4, M5, ZIRLO or DUPLEX. Some old VVER-440 had stainless steel grid spacers. Top and bottom spacer for PWRs can be also Inconel 625. [5]

The top and bottom structures include tie plates/nozzles, debris filters and handles that are typically made of stainless steel SS 304 L. These structures and the spacer grid contain also springs that can be made e.g. of Inconel 718. [5]

More information on the exact structures of BWR, PWR and VVER fuel assemblies can be looked for e.g. in reference [5] (chapter 2). Information on a VVER-440 assembly structure and dimensions is available e.g. in reference [17] (Part 2). Exact data on dimensions and materials for a PWR reactor can be found in reference [18]. Information on different fuel assembly designs is also available in Nuclear Engineering International issues in September 2013 and

September 2015. Mass distributions for different structural materials in PWR and BWR fuel assemblies are given in reference [19] (table 4.4).

4. Impurities in fuel and structural materials

This section presents data on impurities in UO_2 fuel and several structural materials. The data has been collected from references [19-31]. Only the maximum values of each impurity from all the literature sources are presented in this section. All impurity values from all used sources are presented in Appendix A. The used literature sources are briefly described in the following list.

- [19] Published 1978. Presents PWR and BWR models for ORIGEN for uranium-oxide and uranium/mixed-oxide fuels. Recommendations for fuel and structural material compositions to be used in ORIGEN are given. The Impurity concentrations in fuel are mostly based on measurement data. For structural materials (Zry-2, Zry-4, Inconel, SS 304), assumed compositions are given.

- [20] Published 1996. Discusses light element radionuclides in used CANDU fuel and their contribution to postclosure safety assessment. Light nuclides include all nuclides between H-Bi (atomic numbers 1-83). The values are based either on measurements or manufacturing specifications. In cases where neither has been available, maximum content has been estimated based on chemical similarity and abundance in the earth's crust. The report gives both maximum values and recommendations for values to be used in ORIGEN-S calculations. Information for UO_2, Zry-4 and Zr-2.5Nb.

- [21] Accessed in the internet 18.12.2019. Technical data sheet of ATI (Allegheny Technologies Incorporated) who manufactures e.g. Zirconium alloys. Maximum impurity contents for Zry-2, Zry-4 and Zr-2.5Nb.

- [22] Published 1975. Presents an ANL study on the long-term management of Zircaloy cladding hulls. The information in the report is mainly based on a literature review, discussions with experts and visits to manufacturing sites and experimental facilities. The paper presents maximum impurity contents in Zry-2, Zry-4 and Inconel 718. Nominal concentrations for Inconel-600 are also presented but is not included in this report.

- [23] Published 2002. A paper published in a book "Characterization and Quality Control of Nuclear Fuels" which is a collection of papers presented in a conference with the same name as the book and held at Hyderabad, India, in 2002. The paper describes a methodology for chemical characterization of nuclear fuel and structural materials at different stages of the manufacturing process. The paper presents specifications e.g. for Zry-2, Zry-4, Zr-2.5Nb and UO2 impurities. It is a little unclear if these are an example of measurement data or maximum impurity levels, but most likely the latter.

- [24] Published 2011. A PNNL collection of material composition data for 372 materials intended to be used in radiation transport simulations. Mostly, the materials interesting for the present work do not include data on impurities. However, for Inconel 718 and stainless steels 304 and 304 L concentrations for some impurity atoms is also included.

- [25] Published 1992. YJT (Nuclear Waste Commission of Finnish Power Companies) report containing elemental compositions for Zry-2, Zry-4 and Inconel X-750.

- [26] Published 2017. Assesment of new zirconium alloys during normal operation, Anticipated Operational Occurrences, postulated accidents and intermediate dry storage. Compositions including some maximum impurity levels for E110, M5, two types of Zr-2.5Nb (E125, CANDU), ZIRLO and a couple of others.

- [27,28] Published 2007, 2005. Benchmark specifications based on the #2670 ISTC project which provides VVER 440 PIE data for eight samples cut out of four fuel pins of the Novovoronezh nuclear power plant fuel assembly. The benchmark provides impurity data for the fuel with 3.6 % enrichment.

- [29] Published 1998. IAEA technical document discussing zirconium alloy corrosion in nuclear reactors. Maximum impurity levels for Zry-2, Zry-4 and two types of Zr-2.5Nb are presented based on the ASTM standard B 353.

- [30] Published 2018. A handbook containing data on various materials including nuclear grade zirconium alloys.

- [31] Accessed in the internet 24.1.2020. Information on the webpages of an American company American Special Metals. The company distributes metal products in North America. Values for impurities are maximum values. Compositions for Inconel 718 and Inconel X-750.

4.1 Impurities in fuel

Maximum values for the impurities in UO_2 fuel are presented in Table I. The impurity values are ppm in uranium and they are based on references [19,20,23,27,28]. The most comprehensive list of impurities in fuel is in reference [20] which presents impurity values recommended to be used in ORIGEN-S calculations. About half of the impurity elements listed in Table I were only found in reference [20]. For those elements that were included in multiple references significant variations between references were observed.

Table I. Maximum impurity content in UO_2 fuel. The impurity values are ppm in uranium.

Element	ppm	Element	ppm	Element	ppm
Ag	25	**Ga**	3	**Pb**	400
Al	400	**Gd**	2.5	**Pt**	1
Ar	0.1	**Ge**	3	**Re**	1
As	3	**H**	1	**Ru**	1
Au	1	**Hf**	10	**S**	20
B	1	**Hg**	1	**Sb**	1
Ba	100	**Ho**	0.3	**Sc**	20
Be	0.1	**In**	2	**Se**	20
Bi	20	**Ir**	1	**Si**	250
Br	5	**K**	20	**Sm**	0.3
C	200	**La**	10	**Sn**	400
Ca	250	**Li**	1	**Ta**	10
Cd	25	**Lu**	0.3	**Tb**	0.3
Ce	10	**Mg**	200	**Th**	500
Cl	15	**Mn**	200	**Te**	1
Co	75	**Mo**	400	**Ti**	20
Cr	400	**N**	200	**Tl**	1
Cu	400	**Na**	400	**Tm**	0.3
Dy	0.3	**Nb**	10	**V**	400
Er	0.3	**Ni**	400	**W**	100
Eu	0.3	**Os**	1	**Yb**	0.3
F	50	**P**	60	**Zn**	400
Fe	600				

Figures 2-4 give a graphical presentation of the maximum impurity concentrations. The impurity elements have been divided in three separate figures for visual reasons. The largest impurity concentrations are presented in Figure 2 and the smallest in Figure 4.

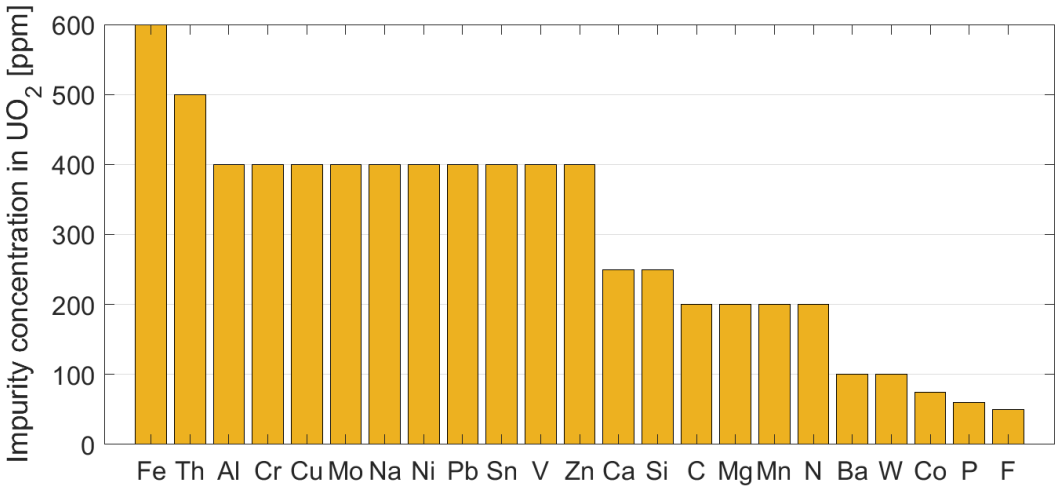

Figure 2. Maximum impurities in the UO_2 fuel for the elements with the largest concentrations.

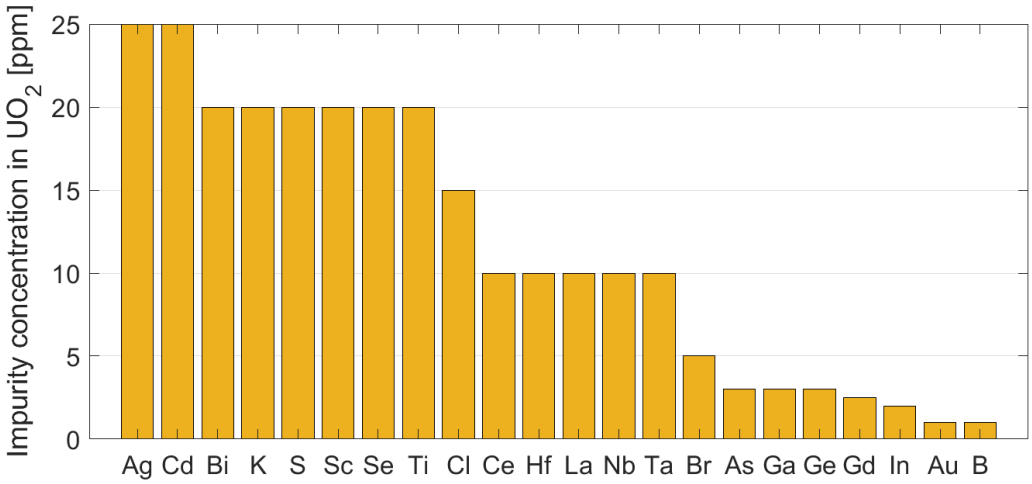

Figure 3. Maximum impurities in the UO_2 fuel for the elements with the second largest concentrations.

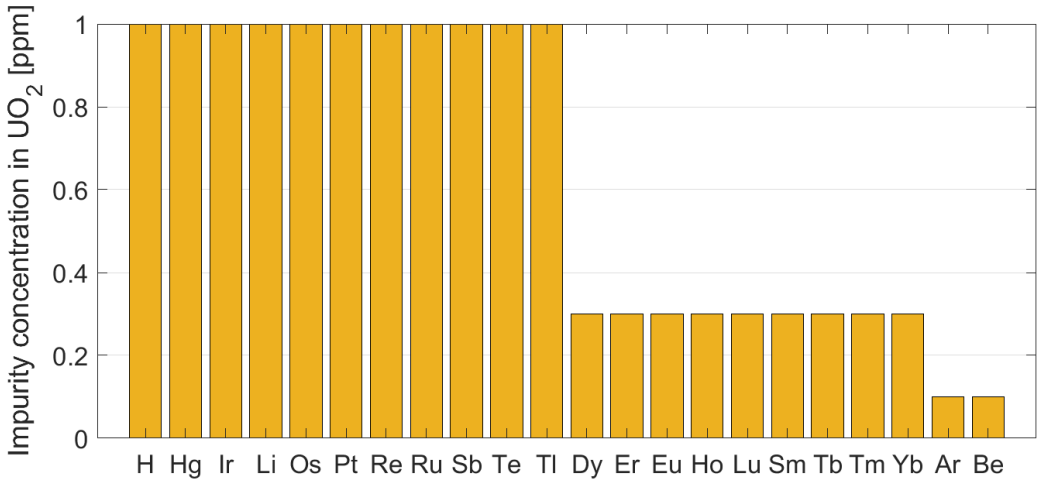

Figure 4. Maximum impurities in the UO_2 fuel for the elements with the smallest concentrations.

The largest value for impurity concentration is for iron whose concentration according to references [27,28] is 600 ppm in uranium. All the references where impurity data for UO_2 was found in this survey included iron [19,20,23,27,28]. All but one reference agreed that iron is the largest or second largest impurity element in UO_2 fuel. However, there was significant variation between the absolute values given for the iron concentration in the different references, the minimum value being 18 ppm [19].

Under neutron irradiation Fe-54 becomes radioactive Fe-55 ($T_{1/2}$ = 2.7 y) through neutron absorption. However, Fe-54 constitutes less than 6 % of natural iron. The dominant nuclide in iron is Fe-56 (>90 %) [32]. Neutron absorption of Fe-56 results in stable Fe-57 which in turn becomes stable Fe-58 through neutron absorption. The activation of Fe-58 through neutron absorption yields a radioactive isotope Fe-59 ($T_{1/2}$ = 44 d) and the decay product is stable Co-59. However Co-59 activates to radioactive Co-60 ($T_{1/2}$ = 5.3 y). In conclusion, the activation of iron might have some significance in the short term handling and radiation protection concerning spent nuclear fuel.

The second largest impurity concentration was thorium which was found in references [20,23], reference [20] having clearly the larger value (500 ppm vs. 10 ppm). Thorium is comprised of Th-232 which through neutron absorption becomes Th-233 ($T_{1/2}$ = 22 min) who in turn undergoes β^- radiation to become Pa-233 ($T_{1/2}$ = 30 d) and finally fissile U-233.

For all nuclides whose maximum concentration was 400 ppm the maximum value was found in reference [23]. In all these cases, the values in the other sources, including the said nuclide, are considerably smaller. However, this is not true for all impurity nuclides found in reference [23].

All elements in Figure 2 except for barium and phosphorus were found in more than one reference and the impurity values between different references varied considerably. Barium and phosphorus were only found in reference [20]. All elements with the smallest concentrations and presented in Figure 4 were only found in reference [20].

Probably the most important activating elements concerning long term safety are nitrogen and chlorine. In neutron irradiation the dominant isotopes N-14 and Cl-35 become C-14 ($T_{1/2}$ = 5700 y) and Cl-36 ($T_{1/2}$ = 3.0E5 y). The values for nitrogen in the studied references were 100 ppm [20,23] and 200 ppm [27,28]. Reference [19] that included impurity data for fuel did not have a value for nitrogen. For chlorine, impurity data was only found in references [20] 5 ppm and [23] 15 ppm. In the other references chlorine was not mentioned.

Other important nuclides concerning long term safety are Ag-108m, I-129 and Mo-93. All these nuclides are formed as fission products. However, Ag-108m and Mo-93 are created also in neutron absorption from Ag-107 and Mo-92, respectively. Natural molybdenum includes several nuclides, but Mo-92 makes up about 15 % of the whole element. Approximately half of silver nuclides are Ag-107 [32]. The values for silver in the studied references were 0.1 ppm [19], 1.0 ppm [20] and 25 ppm [23]. Impurity levels for molybdenum were 8 ppm [20], 10 ppm [19] and 400 ppm [23]. References [27,28] did not include silver or molybdenum.

In addition to the unwanted impurities, some fuel pellets may contain intentional additives whose activation must also be considered. As mentioned in section 2.2 such additives are e.g. chromium in the Atrium11 (BWR) and GAIA (PWR) fuel and chromium and aluminium in the ADOPT pellets of Triton11 fuel (BWR). In the Atrium11 and GAIA fuel, the added Cr_2O_3 concentration is 1600 ppm [10]. At least in tests performed on SVEA-96 Optima2 fuel, the additive concentration in ADOPT pellets was 1000 ppm [14].

4.2 Impurities in structural materials

This section presents impurity data for some structural materials found in nuclear fuel assemblies. The materials included in the study are Zircaloy-2, Zircaloy-4, Zircaloy-2.5Nb, Inconel 718, Inconel X-750 and stainless steel 304 (L). Nominal compositions of these materials are given in Table II. The table includes the composition for SS 304 L. Stainless steel SS 304 is otherwise similar, but the allowed carbon concentration is somewhat higher 0.08 wt-% when for SS 304 L it is 0.03 %. Also the nickel concentration in SS 304 L may be (but not necessarily is) a little higher than in SS 304.

Table II. Nominal compositions of the studied structural elements in wt-%. TBS stands for "to be specified with the customer".

Element	Zry-2	Zry-4	Zr-2.5Nb	Inconel 718	Inconel X-750	SS 304 L
Al				0.2-0.8	0.4-1.0	
C						0.015
Cr	0.05-0.15	0.07-0.13		17-21	14-17	18.2
Fe	0.07-0.2	0.18-0.24	0.03-0.15	Balance	5.0-9.0	Balance
Mn						1.6
Mo				2.8-3.3		
Nb			2.4-2.8	4.75-5.5	0.7-1.2	
Ni	0.03-0.08			50-55	min 70	8.5
O	0.095-0.14 (TBS)	0.09-0.145 (TBS)	0.09-0.13 (TBS)			
Si						0.5
Sn	1.5-1.7	1.5-1.7	0.005-0.01			
Ti				0.65-1.15	2.25-2.75	
Zr	Balance	Balance	Balance			
Fe+Cr+Ni	0.18-0.38					
Fe+Cr		0.28-0.37				

Table III presents maximum values of Zircaloy alloy impurities found in all of the studied references [19,20,21,22,23,25,26,29,30]. For Zircaloy-2.5Nb, some of the references specify an ASTM designation for the alloy, either R60901 or R60904. Some of the references do not specify any designation. Table III does not make a distinction between these designations since for some references it is unclear what designation has been utilized. References [29] and [30] contain information for both designations. For some elements the impurity data presented in these two references for the two ASTM designations differs from each other.

However, only in the case of carbon, nitrogen and nickel the maximum value in Table III is based solely on the information found in [29] or [30] and having differing values for the two designations. These values are highlighted with red colour in Table III.

Table III. Maximum impurity concentrations [ppm] in Zircaloy alloys.

Element	Zry-2	Zry-4	Zr-2.5Nb	Element	Zry-2	Zry-4	Zr-2.5Nb
Ag		10	120	**Mn**	50	50	50
Al	75	75	75	**Mo**	50	50	50
Ar		0.1		**N**	80	80	80
As		1	0.1	**Na**	20	2	18
Au		0.1	0.4	**Nb**	100	100	
B	0.5	0.6	0.5	**Nd**		0.1	
Ba		0.1	0.02	**Ni**		80	80
Be		100	0.004	**Os**		1	
Bi		1	0.9	**P**		60	240
Br		1	0.1	**Pb**	130	130	130
C	270	4200 (300)	270	**Pd**		10	
Ca	30	30	25	**Pr**		0.1	
Cd	0.5	0.7	0.5	**Pt**		1	0.3
Ce		0.1	0.006	**Rb**		1	0.3
Cl	20	20	15	**Re**		1	
Co	20	20	20	**Rh**		5	
Cr			200	**Ru**		5	
Cs		1	0.1	**S**	35	35	16
Cu	50	50	50	**Sb**		10	13
Dy		0.1		**Sc**		10	1
Er		0.1		**Se**		1	0.2
Eu		0.1		**Si**	200	320	120
F		6		**Sm**		0.1	
Ga		5	1	**Sr**		1	0.6
Gd		0.6		**Ta**	200	200	200
Ge		1	0.04	**Tb**		0.1	
H	25	25	25	**Th**		10	1
Hf	200	200	100	**Te**		1	0.3
Hg		0.1	0.07	**Ti**	50	50	50
Ho		0.1		**Tl**		0.1	
I		1	0.08	**Tm**		0.1	
In		1	11	**U**	3.5	10	3.5
Ir		1		**V**	50	50	50
K		1	0.2	**W**	100	100	100
La		0.1	0.006	**Y**		1	0.4
Li		0.1	0.008	**Yb**		0.1	
Lu		0.1		**Zn**		1	0.3
Mg	50	50	20				

The most comprehensive list for Zircaloy-4 and Zircaloy-2.5Nb is in reference [20]. About half of the impurities listed in Table III for Zircaloy-2.5Nb are only found in reference [20] and as many as two thirds for Zircaloy-4. The reference does not contain information on Zircaloy-2 and it does not specify ASTM designations.

In case of Zircaloy-4 in reference [20], the impurity values include impurities due to CANLUB, which is a graphite coating used in some CANDU fuel assemblies. This explains the rather

large value for carbon impurities in Zircaloy-4 (4200 ppm). CANLUB coating is purely a CANDU feature and therefore one could assume that the impurity concentration of carbon in LWR reactors is lower. The largest value for carbon impurities in Zircaloy-4 without CANLUB coating is 300 ppm and it is presented inside brackets for Zircaloy-4 in Table III.

The maximum impurity concentrations in Zircaloy alloys are presented also in Figures 5 (Zircaloy-2), 6-8 (Zircaloy-4) and 9-10 (Zircaloy-2.5Nb). Figures 6-8 and 9-10 are divided according to the impurity concentration similarly as the figures presenting impurities in UO_2 fuel. The carbon concentration in Zircaloy-4 in Figure 6 is that without CANLUB coating.

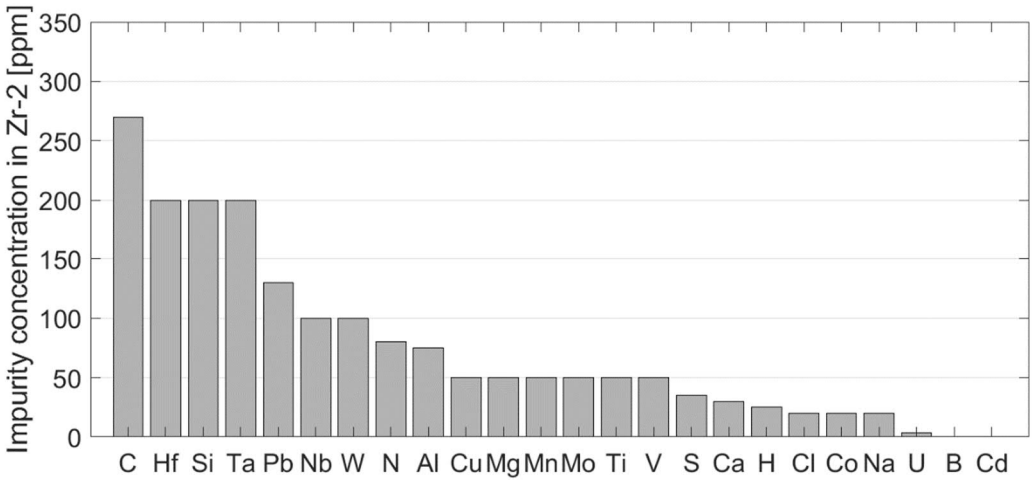

Figure 5. Maximum impurity concentration in Zircaloy-2 [ppm].

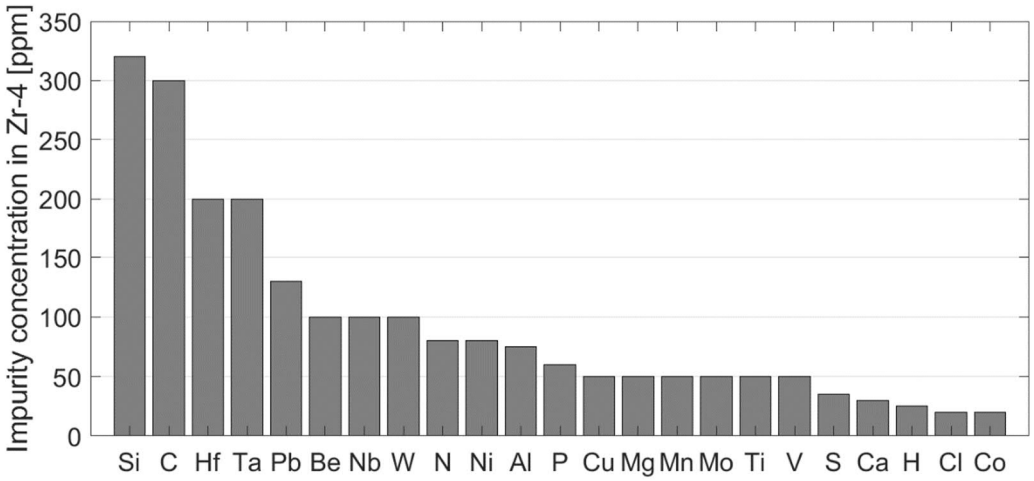

Figure 6. Maximum impurity concentrations in Zircaloy-4 for the elements with the largest concentrations.

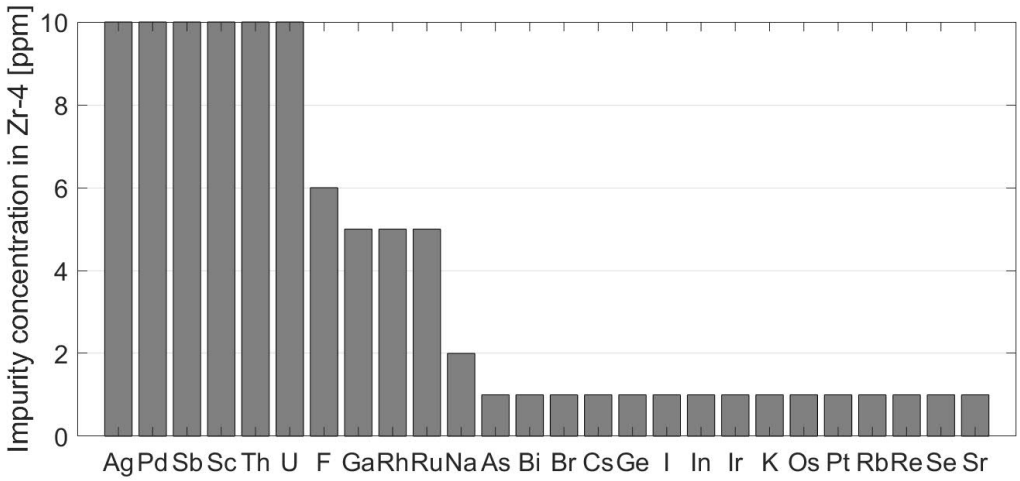

Figure 7. Maximum impurity concentrations in Zircaloy-4 for the elements with the second largest concentrations.

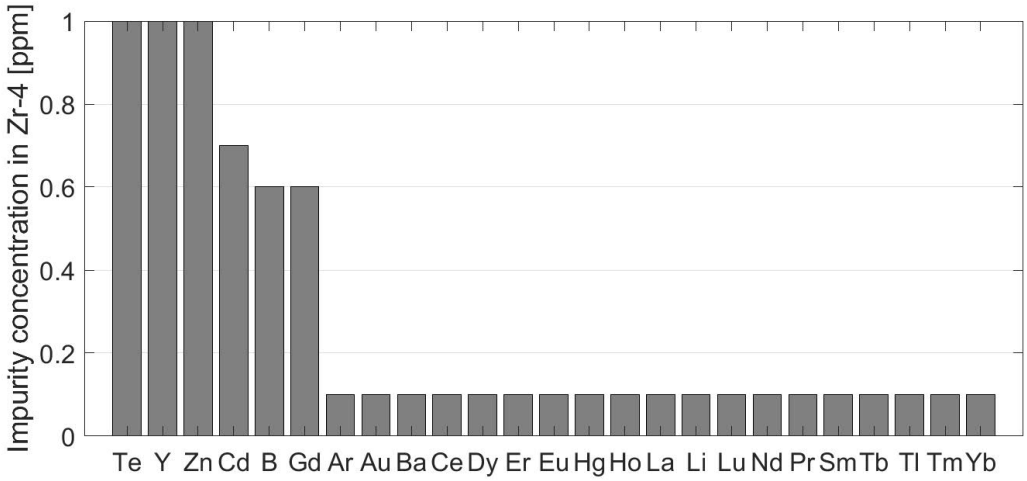

Figure 8. Maximum impurity concentrations in Zircaloy-4 for the elements with the smallest concentrations.

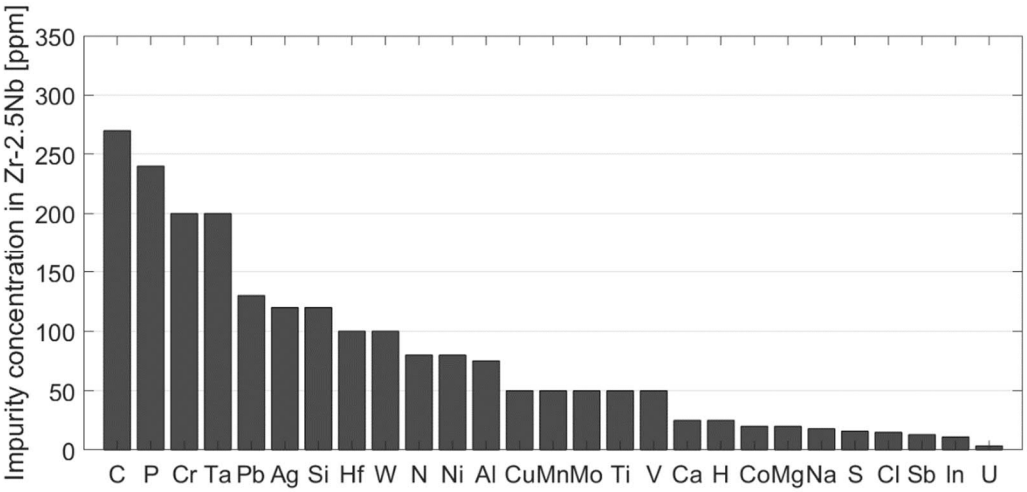

Figure 9. Maximum impurity concentrations in Zircaloy-2.5Nb for the elements with the largest concentrations.

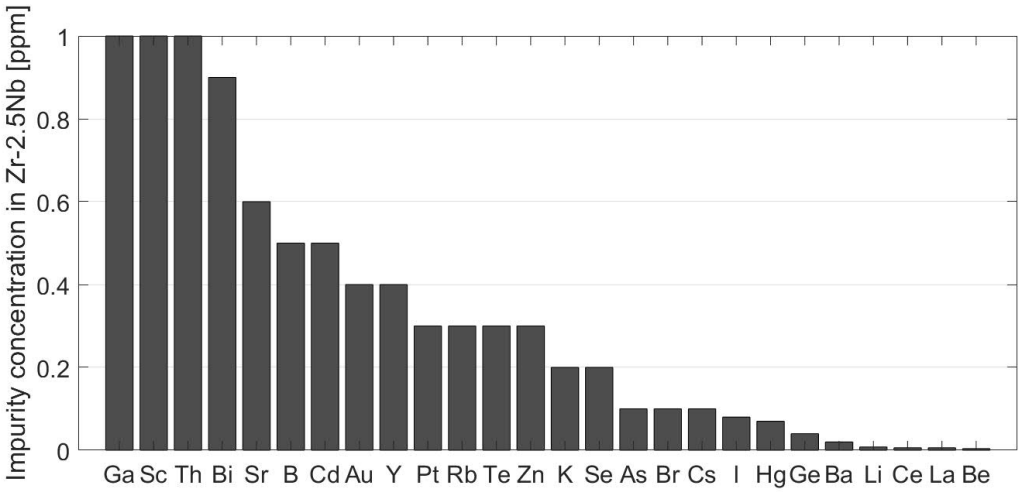

Figure 10. Maximum impurity concentrations in Zircaloy-2.5Nb for the elements with the smallest concentrations.

The maximum impurity elements in Zircaloy-2 and Zircaloy-4 in Figures 5 and 6 contain mostly the same atoms with similar concentrations. Zircaloy-4 contains also beryllium which is according to reference [20] used in brazing Zircaloy-4 spacers and bearing pads and is not found in the other references. The similarity of the maximum impurity elements in these two alloys is natural due to the similarity of the alloys.

The four impurity elements with the largest concentrations in Zircaloy-2 and Zircaloy-4 are carbon, hafnium, silicon and tantalum. Carbon and hafnium are found in all the studied references, the concentrations ranging from 120 ppm to 270 ppm in Zircaloy-2 and to 300 ppm (4200 ppm) in Zircaloy-4. Hafnium concentrations ranged from 60 ppm (Zircaloy-4) or 70 ppm (Zircaloy-2) to 200 ppm. Silicon concentrations are found in all but one reference with values between 20-200 ppm in Zircaloy-2 and 35-320 ppm in Zircaloy-4. Tantalum in Zircaloy-2 is mentioned only in reference [23]. In Zircaloy-4, tantalum is found in references [20] and [23] both giving the same value.

The four impurity elements with the largest concentrations in Zircaloy-2.5Nb are carbon, phosphorus, chromium and tantalum. Carbon is found in several references with values between 125-270 ppm. Concentrations of phosphorus vary between 10-240 ppm, the largest being a recommendation to be used in ORIGEN-S in reference [20], the second largest value being 20 ppm [21]. For chromium, two values were found 100 ppm and 200 ppm. Tantalum was mentioned in two references with 100 and 200 ppm concentrations.

The maximum nitrogen content in all three Zircaloy alloys according to the studied references is 80 ppm. All studied references except for reference [30] included nitrogen as an impurity in Zircaloy-2 and Zircaloy-4. The values ranged from 39 to 80 ppm, 80 ppm being the most popular. For Zircaloy-2.5Nb two values for nitrogen concentration were found 65 ppm [21,23,29] and 80 ppm [29].

The maximum chlorine content is 20 ppm in Zircaloy-2 and Zircaloy-4 and 15 ppm in Zircaloy-2.5Nb. For Zircaloy-2, only two references mentioned chlorine both giving the value 20 ppm [23,25]. For Zircaloy-4, the values ranged from 5 to 20 ppm [23]. In Zircaloy-2.5Nb, chlorine concentration ranged from 0.5 to 15 ppm [20].

Table IV presents the maximum impurity concentrations found in the studied references in Inconel alloys 718 and X-750 and stainless steels SS 304 and SS 304 L. Information on Inconel 718 was found in references [19,22,24,31] and the maximum values were found in references [19,22,31]. Information on Inconel X-750 is based on data in references [19,25,31]. Data on stainless steels was found in references [19,24].

Table IV. Maximum impurity concentrations in Inconel alloys and stainless steel [ppm].

Element	Inconel 718	Inconel X-750	SS 304	SS 304 L
B	60			
C	1000	800		
Co	10000	10000	800	
Cu	7500	5000		
Mn	5000	10000		
N	1300	1300	1300	
P	150		450	230
S	300	100	300	150
Si	7500	5000		
Ta		1000		

The maximum impurity concentrations in the Inconel alloys and stainless steel are presented also in Figure 11. The vertical axis is in logarithmic scale because of the very large deviation on the impurity concentrations of the different elements.

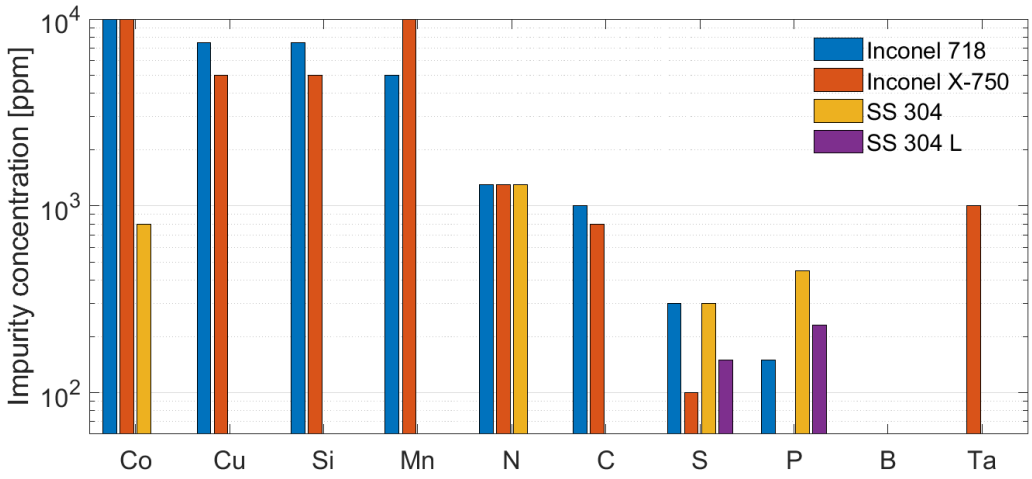

Figure 11. Maximum impurity concentrations in the Inconel alloys and stainless steel [ppm]. The vertical axis is in logarithmic scale.

The largest impurity concentrations in the Inconel alloys are cobalt, copper, silicon and manganese. Cobalt is important in the short term radiation safety examination because of its activation to Co-60 ($T_{1/2}$ = 5.3 y). Cobalt concentration in the Inconels varied largely depending on the referenced source. The range of values for cobalt was 4694-10000 ppm in Inconel 718 and 300-10000 ppm in Inconel X-750.

Nitrogen in the Inconels and stainless steel 304 was included only in reference [19]. Chlorine was not mentioned in any of the studied references for the Inconels or stainless steel.

5. Summary and conclusions

Impurity concentrations in UO_2 fuel and fuel assembly structural materials has been studied from several literature sources. The structural materials studied were Zircaloy-2, Zircaloy-4, Zircaloy-2.5Nb, Inconel 718, Inconel X-750 and stainless steel SS 304 and SS 304 L the emphasis being on the Zircaloy alloys. The amount of impurity elements mentioned and the absolute concentrations of these elements in the various literature sources varied greatly.

The largest impurity element in UO_2 fuel is iron. Quite a good agreement between the references existed on iron being among the two largest impurity elements. Nitrogen concentration in the fuel in the different references was either 100 ppm or 200 ppm. Chlorine was mentioned only in two references with values 5 ppm and 15 ppm.

The four largest impurity elements in Zircaloy-2 and Zircaloy-4 were carbon, silicon, hafnium and tantalum. For Zircaloy-2.5Nb these were carbon, phosphorus, chromium and tantalum. The maximum nitrogen concentration found in the references for all of the Zircaloy alloys was 80 ppm. The maximum chlorine concentration for Zry-2 and Zry-4 was 20 ppm and for Zr-2.5Nb it was 15 ppm.

Much less information was found for the studied Inconel alloys and stainless steels mostly because the emphasis of the study was in the other materials. The maximum impurity element in the Inconels was cobalt with 10000 ppm. The maximum impurity concentration for stainless steel SS 304 was nitrogen. Maximum nitrogen concentration in SS 304 and the Inconels was 1300 ppm. Chlorine was not mentioned as an impurity element in any of these references, but is not likely to be absent in reality.

This study will continue with a computational investigation of the effects of the impurities and their activation on the spent nuclear fuel characteristics. Generally interesting characteristics are e.g. decay heat, activity, photon source, fissile nuclides and the mobile and long-lived nuclides. The most significant impact of impurity activation is expected on the inventory of the long-lived and mobile radionuclides such as e.g. C-14 and Cl-36 that are mainly produced as activation products. However, the study will include also the other interesting characteristics and a wide set of impurity elements based on the results of this study.

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Appendix A

Table A 1 presents impurity concentrations in UO_2 fuel in all the studied references. All but reference [19] agree that iron is the largest or second largest impurity element in fuel. There is wide disagreement between the references for the second largest element. Rather large values are given for iron, carbon and nitrogen in all or most of the references. The concentration of boron and gadolinium is similarly very small according to all or almost all of the references. Otherwise quite a lot of differences exist between the values reported in the various references or only one or two references are mentioning the said element.

Table A 1. Impurity concentrations in UO_2 fuel in all the studied references. The four largest values in all references are highlighted with different colours in the order of red, orange, green, purple, where red indicates the largest and purple the fourth largest value. "Max" means the maximum concentration of the element from all the references and "Max ref" gives the reference number that presents the largest value for the impurity element. Reference [20] presents values recommended to be used in ORIGEN-S calculations (*) and maximum values given by the manufacturer (**).

Ref: Element	[20]* ppm	[20]** ppm	[19] ppm	[23] ppm	[27,28] ppm	**Max** ppm	**Max ref**
Fe	250	250	18	400	600	600	27,28
Th	500	500		10		500	20
Al	100	100		400		400	23
Pb	100		1	400		400	23
Ni	60	60	24	400	150	400	23
Cr	50	50	4	400	100	400	23
Cu	20	20	1	400	50	400	23
Na	20			400		400	23
V	20		3	400		400	23
Zn	20		40.3	400		400	23
Mo	8	8	10	400		400	23
Sn	1		4	400		400	23
Si	120	120			250	250	27,28
Ca	100	100	2	250		250	23
C	200	200	89.4	100	100	200	20
N	100	100		100	200	200	20, 23
Mg	60	60		200		200	23
Mn	20	20	1.7	200	20	200	23
Ba	100					100	20
W	10		2	100		100	23
Co	20		1	75		75	23
P	60					60	20
F	30	30		25	50	50	27,28
Ag	1	1	0.1	25		25	23
Cd	0.4	0.4	25	1		25	19
Bi	20		0.4			20	20
K	20					20	20
S	20	20				20	20
Sc	20					20	20
Se	20					20	20
Ti	20		1			20	20
Cl	5	5		15		15	23
Ce	10					10	20
Hf	10					10	20
La	10					10	20

Nb	10					10	20
Ta	10					10	20
Br	5					5	20
As	3					3	20
Ga	3					3	20
Ge	3					3	20
Gd	0.3	0.2	2.5	1		2.5	19
In	1		2			2	19
Au	1					1	20
H	1					1	20
Hg	1					1	20
Ir	1					1	20
Li	1		1			1	19,20
Os	1					1	20
Pt	1					1	20
Re	1					1	20
Ru	1					1	20
Sb	1					1	20
Te	1					1	20
Tl	1					1	20
B	0.6	0.6	1	1	0.4	1	19,23
Dy	0.3	0.3				0.3	20
Er	0.3					0.3	20
Eu	0.3					0.3	20
Ho	0.3					0.3	20
Lu	0.3					0.3	20
Sm	0.3					0.3	20
Tb	0.3					0.3	20
Tm	0.3					0.3	20
Yb	0.3					0.3	20
Ar	0.1					0.1	20
Be	0.1					0.1	20

Table A 2 presents impurity elements in Zry-2 found in all the studied references. There is a clear consensus between all references that the largest impurity element is carbon. For the next largest elements there is somewhat more disagreement. However, hafnium, silicon and wolfram are ranked high by nearly all the references. Hafnium is an important impurity element in the neutronics point of view because of its large thermal neutron cross section. Boron, cadmium and uranium is found in similar small quantities in most of the references except for references [25,30].

Table A 2. Impurity concentrations in Zry-2 in all the studied references. The four largest values in all references are highlighted with different colours in the order of red, orange, green, purple, where red indicates the largest and purple the fourth largest value. "Max" means the maximum concentration of the element from all the references and "Max ref" gives the reference number that presents the largest value for the impurity element.

Ref: Element	[19] ppm	[21] ppm	[22] ppm	[23] ppm	[25] ppm	[29] ppm	[30] ppm	**Max** ppm	**Max ref**
C	120	270	270	270	120	270	270	270	21,22, 23,29,30
Hf	78	100	200	200	100	100	100	200	22, 23
Si		120	200	20	50	120	120	200	22
Ta				200				200	23
Pb				130				130	23
Nb				100				100	23
W	20	100	100	50	50	100	100	100	21, 22,29,30
N	80	80	80	65	40	80		80	19,21,22,29
Al	24	75	75	75	50	75	75	75	21,22, 23,29,30
Cu	20	50	50	50	50	50	50	50	21,22, 23,29
Mg		20	50	20	10	20		50	22
Mn	20			50	30	50	50	50	23,29,30
Mo		50		50	5	50	50	50	2, 23, 29,30
Ti	20	50	50	50	50	50	50	50	21,22, 23,7, 29,30
V	20			50				50	23
S	35							35	19
Ca				30				30	23
H	13	25	25	25		25		25	21,22, 23,29
Cl				20	20			20	23,25
Co	10	20	20	20	2	20		20	21,22, 23,29
Na					20			20	25
U	0.2	3.5	3.5	3.5		3.5		3.5	21,22, 23,29
B	0.33	0.5	0.5	0.5		0.5		0.5	21,22, 23,29
Cd	0.25	0.5	0.5	0.5		0.5		0.5	21,22, 23,29

Table A 3 presents impurity concentrations in Zry-4 in all the studied references. The last column presenting the references of the maximum impurity values has been omitted for visual reasons. Many of the impurity nuclides are presented only in reference [20]. Only reference [25] never gives the largest impurity value for any of the presented elements. Carbon is consistently the largest impurity element in all the references. Most of the references agree silicon and hafnium to be among the four largest impurity elements. Only reference [19] does not mention silicon. Relatively good agreement between the references is found on the value of nitrogen, most of them given the value 80 ppm. Boron, cadmium and uranium is found in similar small quantities in all of the references except for [25,30].

Table A 3. Impurity concentrations in Zry-4 in all the studied references. The four largest values in all references are highlighted with different colours in the order of red, orange, green, purple, where red indicates the largest and purple the fourth largest value. "Max" means the maximum concentration of the element from all the references. Reference [20] presents values recommended to be used in ORIGEN-S calculations (*) and maximum values given by the manufacturer (**). Reference [22] presents two sets of impurity values designated as "nominal" (+) or "typical" (++).

Ref: Element	[20]* ppm	[20]** ppm	[19] ppm	[21] ppm	[22]+ ppm	[22]++ ppm	[23] ppm	[25] ppm	[29] ppm	[30] ppm	**MAX** ppm
C	4200	300	120	270	270	120	270	130	270	270	4200
Si	320	300		120	120	35	120	70	120	120	320
Hf	100	100	78	100	200	78	200	60	100	100	200
Ta	200	200					200				200
Pb	130	130					130				130
Be	100										100
Nb	100	100					100				100
W	100	100	20	100	100	20	50	25	100	100	100
N	80	80	80	80	80	39	65	45	80		80
Ni	70	70	20	70	40	20	70	35		80	80
Al	75	75	24	75	15	24	75	45	75	75	75
P	60	60									60
Cu	50	50	20	50	50	20	50	20	50	50	50
Mg	20	20	20	20	50	20	20		20		50
Mn	50	50					50	25	50	50	50
Mo	50	50	0	50			50		50	50	50
Ti	50	50	20	50	50	20	50	25	50	50	50
V	50	50	20			20	50				50
S	20		35								35
Ca	2						30				30
H	25	25	13	25	25	13	25		25		25
Cl	6	5					20	5			20
Co	20	20	10	20	20	10	20	2	20		20
Ag	10										10
Pd	10										10
Sb	10										10
Sc	10										10
Th	10										10
U	10	3.5	0.2	3.5	3.5	0.2	3.5		3.5		10
F	6	5									6
Ga	5										5
Rh	5										5
Ru	5										5
Na	2										2
As	1										1
Bi	1										1
Br	1										1
Cs	1										1
Ge	1										1
I	1										1
In	1										1
Ir	1										1
K	1										1
Os	1										1
Pt	1										1

Rb	1										1
Re	1										1
Se	1										1
Sr	1										1
Te	1										1
Y	1										1
Zn	1										1
Cd	0.7	0.5	0.25	0.5	0.5	0.25	0.5		0.5		0.7
B	0.6	0.5	0.33	0.5	0.5	0.33	0.5		0.5		0.6
Gd	0.6										0.6
Ar	0.1										0.1
Au	0.1										0.1
Ba	0.1										0.1
Ce	0.1										0.1
Dy	0.1										0.1
Er	0.1										0.1
Eu	0.1										0.1
Hg	0.1										0.1
Ho	0.1										0.1
La	0.1										0.1
Li	0.1										0.1
Lu	0.1										0.1
Nd	0.1										0.1
Pr	0.1										0.1
Sm	0.1										0.1
Tb	0.1										0.1
Tl	0.1										0.1
Tm	0.1										0.1
Yb	0.1										0.1

Table A 4 presents impurity concentrations in Zr-2.5Nb in all the studied references. The last column presenting the references of the maximum impurity values has been omitted for visual reasons. Most references agree carbon to be the dominant impurity element or at least one of the top three. Reference [20] does not contain a value for carbon or many of the nuclides ranked high by the other references such as e.g. chromium, silicon, hafnium and wolfram. However, it includes many other low concentration impurities not mentioned in the other references. This may be due to what has been chosen or has been possible to measure in reference [20].

Table A 4. Impurity concentrations in Zr-2.5Nb in all the studied references. The four largest values in all references are highlighted with different colours in the order of red, orange, green, purple, where red indicates the largest and purple the fourth largest value. "Max" means the maximum concentration of the element from all the references. Reference [20] presents average (ave) and maximum (max) values based on measurements. ASTM designations are presented in the cases it was given in the reference.

Ref: **ASTM:** **Elem.**	[20] ave ppm	[20] max ppm	[21] R60904 ppm	[23] ppm	[26] ppm	[29] R60901 ppm	[29] R60904 ppm	[30] R 60901 ppm	[30] R 60904 ppm	**Max** ppm
C			270	125	<270	270	150	270	150	270
P	10	240	20	10						240
Cr			100	200		200	100			200
Ta				200			100			200