

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Volatized Tellurium Interactions with Organics and with Boric Acid

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Cover:

Examples of filters from the experiments in paper I and II. The black filter is coated with tellurium metal, and the white filter is coated with tellurium dioxide TeO_2 .

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Abstract

Tellurium is a volatile fission product with a complex chemistry that must be accounted for in case of a nuclear accident. Specifically, its interactions with organic species and with boric acid, both which are expected in the accident chemistry, are largely unknown. This study aims to explore the interactions between tellurium aerosols and three organic species (acetone, propanol, and methane), and with boric acid at temperatures representing the primary circuit of a pressurized water reactor. Oxidizing (Air), inert (N_2) and reducing (Ar/H_2) atmospheres are all investigated. Analysis involves ICP-MS of liquid samples, XPS of solid filter samples, as well as online measurements, mainly to categorize the aerosols produced during the experiments, as well as FTIR. Furthermore, neutron activation and gamma spectroscopy are used.

The results indicate that tellurium may form small amounts of organic tellurides in these conditions, specifically under inert and reducing conditions. These results are not conclusive, as no analysis detected these species. However, data from both XPS and ICP-MS may imply their formation. In general though, the ICP-MS data imply low volatility of tellurium in these conditions, as the concentration in the liquid trap remains in the μM to nM range.

As for boric acid, no indication was found of direct interaction between boric acid and tellurium. However, XPS analysis does imply that the oxidation of the tellurium is reduced by the presence of boric acid. This was inferred mainly from XPS analysis and may potentially be an important consideration for future studies of tellurium in the context of severe nuclear accidents. Volatility remains at the low μM level, though formation of H_2Te does serve to increase it. This only occurs at high temperature in reducing atmosphere, however.

Keywords: Tellurium, Severe nuclear reactor accident, Volatile fission product, boric acid

List of Publications

- I. Pasi Anna-Elina, Kärkelä Teemu, Börjesson Sandén Fredrik, Tapper Unto, Kajolinna Tuula, Ekberg Christian, "Gas phase interactions between tellurium and organic material in severe accident scenarios", *Annals of Nuclear Energy*, 2024, vol 197
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Contributions: Main author, Part of experimental work, part of data processing and data interpretation

List of Abbreviations

INES Scale - International Nuclear and Radiation Event Scale

Gy - Gray (J/kg)

MOX-fuel - Mixed oxide fuel

PWR - Pressurized water reactor

BWR - Boiling water reactor

DGEBA - Bisphenol A diglycidyl ether

TGA - Thermogravimetric Analysis

MIBK - Methyl isobutyl ketone

INAA - Instrumental neutron activation analysis

MFC - Mass-flow controller

PTFE - Polytetrafluoroethylene

FTIR - Fourier transform infrared (spectroscopy)

ELPI - Electric low-pressure impactor

TEOM - Tapered element oscillating microbalance

ICP-MS - Inductively Coupled Plasma Mass Spectrometry

XPS - X-ray photoelectron spectroscopy

SEM - Scanning electron microscopy

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1. Introduction

Phenomena such as extreme weather and a changed ecosystem are starting to become more and more visible [1]. At the same time, the living standard of all modern countries demand energy for transport and production of wares and services, heating of homes and food, or recreation and cultural expression. Electric cars are becoming more and more common both in Sweden [2] and abroad [3], and heavy industry such as steelworks [4] are looking to turn away from fossil fuels to electricity with a low environmental impact. Furthermore, emergent technology such as hydrogen production through hydrolysis must also be considered [5]. With all these considerations, the demand for electricity is projected, according to some scenarios, to more than double in Sweden by 2050 [5].

Nuclear power is a method for production of electricity which does not contribute appreciably to emissions of greenhouse gases [6] and is not very dependent on the weather. These factors make nuclear power worth considering for the energy mix.

Of course, nuclear power is not without its drawbacks. One of the big challenges for the industry is to gain the trust of the public despite accidents such as those at Chernobyl and Fukushima-Daiichi. Severe nuclear accidents are rare, but when they happen, they pose unique challenges due to the radio-hazardous materials released. Such material is harmful to humans and the environment both in the short and the long term and necessitates expensive cleanup.

Nuclear accidents, therefore, must be studied and prepared for. Such studies can take the form of large research programs spanning years or decades (such as the PHÉBUS FP [7] running 1988-2010), or small-scale experiments exploring a limited facet of accident phenomenology.

This thesis falls in the latter category, focusing on tellurium speciation and behavior in environments and circumstances that may occur during a severe nuclear accident. Tellurium is a volatile fission product, meaning the majority of the inventory is expected to be released in case of a nuclear accident. It has a complex chemistry, belonging to the chalcogen group of the periodic table, just like oxygen and sulfur. Just like its lighter homologues, tellurium interacts with organic molecules to form organic tellurides [8][9], which potentially changes its behavior and source term in an accident. Furthermore, boric acid is a chemical used in certain nuclear reactors to control the reactivity of the reactor core. It can also be used as part of the emergency shutdown sequence. Its presence in an accident scenario, and its potential to interact with and change the tellurium source term, also bears investigating.

This thesis aims to investigate the interaction between tellurium aerosols transported in the gas phase in two chemical circumstances that may occur in a severe nuclear accident, namely its interaction with organic species, and its interaction with boric acid.

2. Background

2.1. Nuclear Accidents and Incidents

Nuclear power plants are large industrial plants involving a myriad of components. Smaller deviations from protocol are bound to happen with some frequency and carry little to no risk of radiation exposure to the public. To communicate consequences of such deviations, a seven-point scale is used, referred to as the INES scale (International Nuclear and radiation Event Scale) [10]. This scale is logarithmic, so in principle the consequences of one rating are one tenth of the consequences of an event of the next rating. The same scale is used not only for nuclear power plants, but for any installation using radioactive material, including hospitals or reprocessing plants, for instance.

It should be stressed that this scale is not used predictively (it does not assess “risk”, meaning consequences of a scenario times the probability of the scenario occurring). It is used to assess deviations in normal operation after they have already occurred and make a statement of the consequences of that deviation. The INES-level assigned is based both on the activity released from the event, the dose committed to people, and the number of people exposed.

The first three levels of the scale are referred to as “incidents” of varying severity. Incidents encompass events such as degradation of radiological safety systems to radiological overexposure without severe consequences. The levels four to seven are referred to as “accidents”. The term “Severe Accident” is not used by the scale but will for the context of this work be taken to mean an event at a level of five or higher, (those being referred to as “Accident with wider consequences”, “Serious Accident” and “Major Accident” respectively).

With regards to an event rating of four, the INES event scale handbook gives the following advice regarding dose releases [10]:

“An event resulting in an environmental release corresponding to a quantity of radioactivity radiologically equivalent to a release to the atmosphere of the order of tens to hundreds of terabecquerels of ^{131}I , or an event resulting in a dispersed release of activity from a radioactive source with an activity greater than 250 times the D_2 value for the isotopes released.”

Where the D_2 -value refers to the activity of a radionuclide source that could cause severe deterministic health effects if uncontrolled and dispersed [11]. Correspondingly for doses to individuals:

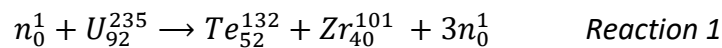
“Level 4 is the minimum level for events that result in: the occurrence of a lethal deterministic effect, or the likely occurrence of a lethal deterministic effect as a result of a whole-body exposure, leading to an absorbed dose of the order of a few Gy.”

For reference, the Three Mile Island accident 1979 is ranked as a level five accident, and the Chernobyl accident and the combined Fukushima-Daiichi accidents are the only events to date to have been classified as level seven.

Related to the INES scale is NEWS “Nuclear Events Web-based System”, which is a website run by the IAEA which lists all nuclear events reported to them at a level two or higher, where the public can access the information of such events.

2.2. Fission products and radioactivity

Nuclear power is a way of generating electricity by using the energy contained in heavy atomic nuclei. The energy is released through splitting the nuclei, which evaporates water to feed a turbine. Today there are two broad categories of nuclear fuel being used; uranium fuel incorporates the fissile nuclide U^{235} , and MOX-fuel (mixed oxide fuel) incorporates mainly Pu^{239} in a matrix of uranium. In a nuclear reactor, both will undergo the same principal operation: upon being struck by a (thermal) neutron, there is a chance for these nuclei to fission. An example with uranium is described in Reaction 1.



In principle, the starting nuclei will split into two (in rare cases more) fragments, and free neutrons. These neutrons allow for a continued nuclear reaction if they interact with other fissile nuclei. The fragments from the incident nucleus are now referred to as fission products, which are nuclei of new, lighter elements. These isotopes are nearly always radioactive.

The fission products vary from fission event to fission event. In total, there are several dozens of nuclides that will form from the operation of a nuclear reactor. However, some are more common than others [12]. For fission with thermal neutrons of both ^{235}U and ^{239}Pu , which is what is used in conventional reactors, there are two ranges of fission product masses that tend to form (the probability for formation of these masses is about 5-7% per fission event). The probability per mass is depicted in Figure 1.

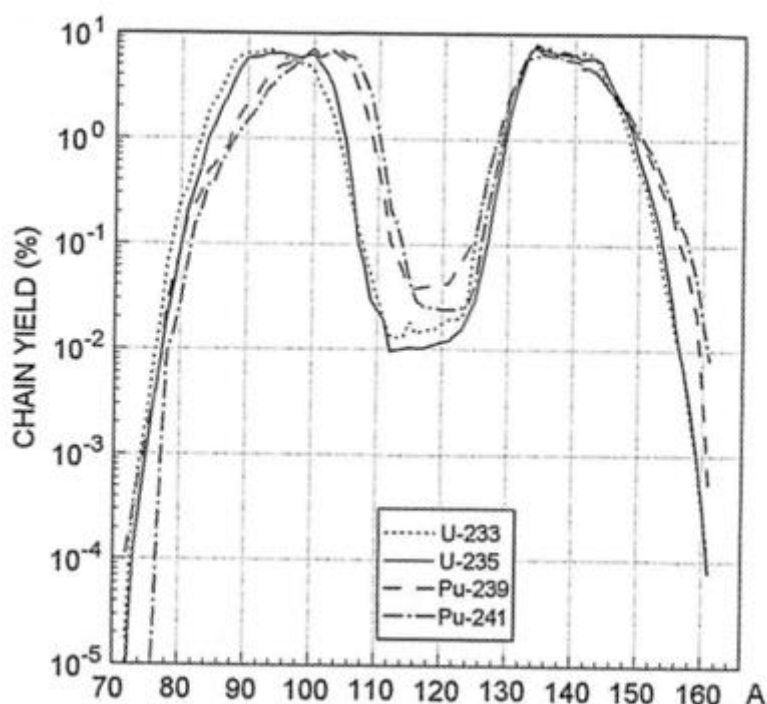


Figure [1]: Graphical depiction of the probability for formation of a certain fission product mass after fission with thermal neutrons of ^{235}U . Notice that fission products of the approximate masses 90-101 u and 132-145 u are comparatively common [12]. Used with permission from the author.

In a severe accident, the danger of radiation exposure to the public derives from the nuclear fuel, and the fission products. The nuclear fuel itself has a high melting- and boiling point and is not considered volatile [13][14]. Fission products, however, can be more mobile. This means that an understanding of the fission products, their chemistry and their assumed interaction between structural material, atmosphere, or each other will all determine the ramifications of the accident and the best ways of mitigating the impact and danger to the public and the environment.

Of the nuclei that will form, a few have been identified as especially problematic due to a combination of high tendency to form in the fuel, a chemical or physical nature that lets them feasibly escape the reactor containment and cause offsite harm, and a long enough half-life to spread beyond the containment. Examples of such nuclides are ^{131}I , ^{137}Cs , and ^{132}Te . Iodine and cesium are both relatively well studied, whereas tellurium is more obscure.

Tellurium has the potential to interact with various species found during a reactor accident. This paper will investigate if and how it interacts with some token organic species (propanol and acetone), and with boric acid.

2.3. The general chemistry of tellurium

Tellurium is a metalloid and element 52 in the periodic table. It is placed in the chalcogen group, alongside oxygen, sulfur, selenium, and polonium. In nature it consists of five stable isotopes (^{120}Te , ^{122}Te , ^{124}Te , ^{125}Te , and ^{126}Te), and another three strictly speaking radioactive ones with very long half-lives (^{123}Te , ^{128}Te and ^{130}Te) [15][16]. In terms of abundance, the three heaviest isotopes together make up almost 85% of the total amount. Tellurium is not

found as a free element in nature but together with for instance gold, silver, bismuth, lead and nickel [15].

In terms of oxidation states, tellurium can adopt five nominal oxidation states (-II, 0, +II, +IV, +VI), and has an electronegativity of 2.1 on the Pauling scale [17]. All these oxidation states may, in principle, become relevant for severe accident research. The +VI oxidation state includes H_2TeO_3 and $\text{Te}(\text{OH})_6$, both of which may form in the sump during an accident [18]. The +IV state occurs in TeO_2 , which is the principal oxide formed by heating tellurium metal in air. The metallic form of tellurium is expected to be rare in the containment, but used for this work and others to represent tellurium. Organic tellurium species are based on the C-Te bond. With the electronegativity of carbon on the Pauling scale being 2.55, it makes organic tellurides strictly speaking belonging to the +II or +IV state (as organic tellurides tends to be di- or tetravalent [16]). The -II state, finally, would be present in species such as Cs_2Te [19]. Potentially H_2Te also exhibits the -II state, though the electronegativity of hydrogen is very similar to that of tellurium, the formal oxidation state is not certain. In keeping with the chalcogen group however, the species is referred to as “hydrogen telluride”, implying the -II state.

Compared to the corresponding bonds for the lighter chalcogens, the bond weakens in strength with the weight of the chalcogen. The C-O bond has a bond dissociation energy of 358 kJ/mol, C-S has the energy 272 kJ/mol, and C-Se has the energy 234 kJ/mol [20]. The tellurium bond energy is about 200 kJ/mol or lower [16], partially explaining its highly limited organic chemistry compared to the lighter chalcogens. Tellurium also does not have any known biological function in humans, whereas oxygen, sulfur, and selenium [21] are essential for our health.

2.4. Tellurium in the reactor core and containment

As a fission product, tellurium will be formed in the nuclear fuel. The chemical form of tellurium outside of the fuel is an extensively studied topic, and the results vary depending on the temperature, presence of oxygen, and on the fuel element cladding.

The fuel is commonly structured as pellets of uranium dioxide stacked inside a fuel pin filled with inert gas (commonly helium). The fuel pins are grouped in assemblies, which in turn make up the reactor. In a typical light water reactor (PWR or BWR) the fuel assemblies will be surrounded by water, which serves to moderate the neutrons in the core.

To ensure a good neutron economy, the fuel pin cladding should be made of a material with a low cross-section as the neutrons should interact with the fuel itself, rather than the cladding. A popular choice is an alloy of zirconium and a few percent of other metals (including among others tin) called Zircaloy. Thus as tellurium is formed in the fuel, interaction with zirconium becomes possible in the fuel-cladding gap forming various of zirconium-tellurium species, either as compounds or into a solid solution [19][23]. This serves to fixate the tellurium into the cladding for as long as it remains intact, preventing further spread of tellurium. The precise species again depends on the particular environment in the fuel cladding gap, especially the tellurium vapor pressure. At normal operation temperatures, the reaction between zirconium and tellurium is thought to be

limited to the cladding surface, rather than the bulk. The zirconium- tellurium system is thought to remain intact up to 2000°C before oxide compounds can start to break it down and liberate the tellurium [19]. As such, the release of tellurium is intimately connected to the absolute temperature and the time of exposure to that temperature [24].

However, some level of corrosion of the zirconium alloy seems to occur even at much lower temperatures (below 700 °C) if oxygen is present in the system, forming some Zr-Te phase. This process seems to be governed by kinetics at those temperatures, rather than thermodynamics [19].

If oxygen or steam comes into contact with the cladding material at temperatures above 877 °C, then the zirconium will be oxidized to zirconium oxide (also producing hydrogen gas) in an exothermal reaction [25]. As the reaction proceeds the temperature increases and pushes the reaction further. The oxide takes precedence over the Zr-Te phase, so if the cladding is breached or (more or less) fully oxidized, the bound tellurium will be released [19].

Tin is used as an alloy element in zirconium cladding. As the oxidation progresses, the tin will be forced away and form an interface between the oxidized and unoxidized phases of the cladding. Tin melts at about 230 °C and has a low vapor pressure, so the tin ought to be in a liquid state at this point. It is then available to react with fission products, including tellurium in which case it would form SnTe [23][26]; a compound which in principle could be stable in these circumstances. Mass spectra suggests it would vaporize as SnTe(g) [19].

As for speciation, there are several stoichiometries possible when tellurium reacts with the cladding, with varying ratio of zirconium and tellurium. Due to the interaction with the cladding, tellurium is retained [27]. This potentially means that that tellurium is never released, if the oxidation of the cladding can be prevented through the accident, or that the release of Tellurium is delayed while the zirconium cladding is being corroded. Also, with the relatively short half-life of several tellurium isotopes, this delay may give some of it the time to decay into iodine instead.

The oxidation level of the cladding must be very high for the tellurium to be released; above 95% if the oxidation happens from one side of the cladding [27]. At oxidation levels less than 90%, the Te-release should be taken as 1/40:th of the release of iodine [27]. The release in that case ought to be in the form of SnTe(g). If zircalloy is not present in the system, then the release may be in the form of CsTe [28].

Other literature [29] states the most likely compounds to form, depending on the atmosphere and relative abundance of hydrogen, oxygen and/or water to be the following:

SnTe if the H_2O/H_2 is about 1.

Te(g), TeO, TeOH_x, SnO₂ if the H_2O/H_2 is above 1.

TeO_yOH_x (oxyhydroxides) if H_2O/O_2 is about 1 (mixed, as the article says)

TeO₂ and SnO₂ in the absence of water.

However, even in oxidizing systems, the release of tellurium is not measurable until about 950 °C [23], to be compared to 777 °C for cesium and 677 °C for iodine. Iodine and cesium do not have the same tendency to be trapped by the fuel cladding as tellurium does.

In tests where the cladding is fully oxidized, the Te release is about the same as the release of cesium and iodine.

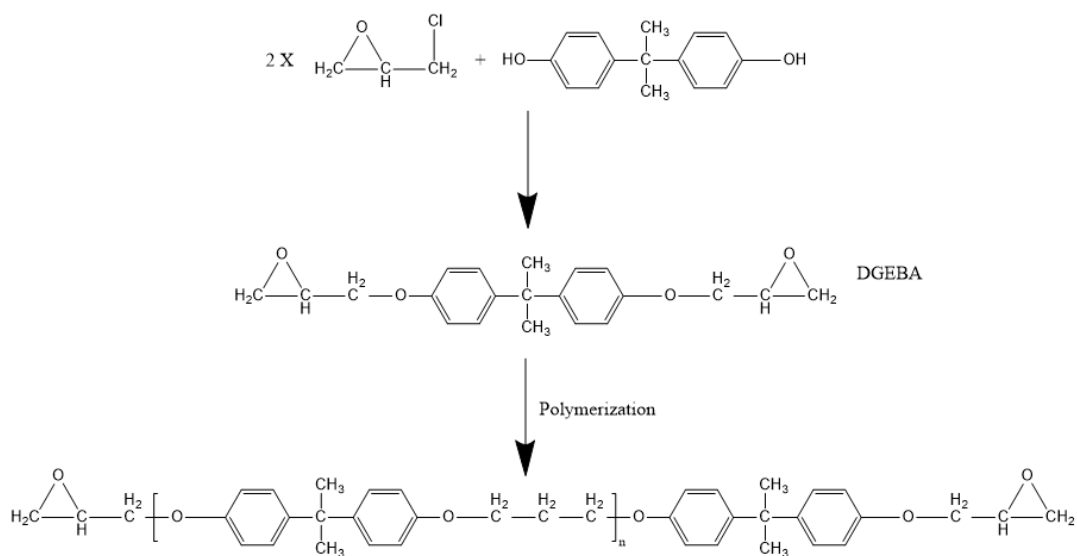
Due to the dependency on the severe accident progression, especially in regard to temperature, oxygen availability and completion of the zirconium oxidation, tellurium can potentially leave the reactor core in various states, all from volatile species such as H_2Te [19], to oxides or potentially the metal [29].

2.5. Organic species in the containment and organic tellurides

Organic species are present in nuclear containment in the form of paints, gaskets, seals, lubricating oils, cables etc. During a severe accident, these can remain intact, or they can be changed due to the accident, for instance through pyrolysis or radiolysis. Radiolysis can also be a direct interaction, where radiation alters the organic molecule, or an indirect reaction where for instance water radiolysis products induces a reaction in the organics [30].

The availability and speciation of organic components in the containment will depend on the organic material available in the containment and the accident progression. The chemical composition of the above listed sources of organics cannot be thought of as homogeneous among different nuclear power plants, meaning that the makeup of the organics during the accident will likely vary from plant to plant and from accident to accident. However, studies have been undertaken with regards to the speciation of epoxy-based paints, a common type of paint in nuclear containments [31], and the interactions between various organics with iodide [32] and with tellurium [27] in the liquid sump, and with tellurium in the liquid sump.

Epoxy based paints are a group of paints that are cured by the polymerization reaction of epoxides. An example of a monomer (diglycidyl ether of bisphenol A, DGEBA) used for this purpose can be seen in Reaction 2.



Reaction 2: Synthesis reaction of DGEBA and principal subsequent polymerization [33].

This class of paint forms a hard, smooth surface that covers the concrete structure of the containment well and can easily be scraped off and replaced in case of contamination. The type of paint used by several Swedish nuclear power plants is “Teknopox Aqua VA paint” [31]. Of course, the behavior of the paint and its chemical inventory is bound to change as the paint ages, further complicating the matter of determining the organic species arising from it [31].

Nevertheless, speciation analysis of the paints reveals that a multitude of different organic products are present in the epoxy paint, however Thermogravimetric (TGA) analysis of the paint indicates that it is stable up to about 150°C, and a severe mass loss only begins around 300 °C [31]. In normal conditions this is much above the ca. 45-55°C expected in the containment, but during an accident the temperature can increase to about 120-150 °C, depending on the reactor type [31].

The containment may be partially submerged during a severe accident. This can happen for instance in a loss-of-coolant-accident where a water pipe bursts and floods the containment, or because of the containment spray system; a sprinkler-like safety measure designed to cool the containment and wash aerosols down into the sump [34]. In general, this system is capable of washing out most of the tellurium aerosols, though the efficiency depends on the chemical content of the spray solution, and the present atmosphere [35]. This makes leaching of organics from the painted surface and into the water followed by volatilization a potential avenue for organics in the containment atmosphere [36]. Studies on the leaching of different paint types indicate that for instance methylisobutyl ketone (MIBK), acetone, and m-xylene, to name a few [36], are all species that can be leached. Organic tellurides have been identified following the irradiation of a simulated water sump containing tellurium dioxide and MIBK (dimethyl telluride) and texanol (diisopropyl telluride) [27].

Interactions between dissolved organic species and tellurium aerosols present in the gas phase, however, have not been studied previously.

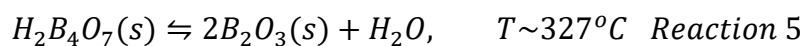
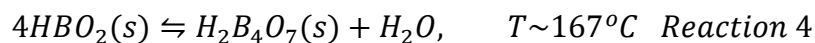
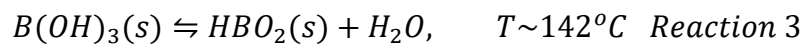
2.6. Boric acid in severe accidents

Orthoboric acid ($B(OH)_3$ henceforth referred to as just “boric acid”) is an important substance in nuclear operations. Naturally occurring boron consists of two isotopes, ^{10}B and ^{11}B , in the ratio roughly 1/5. ^{10}B has a very high cross section for thermal neutrons, about 3840 barn for the alpha-fission reaction [37], which can be compared to 586 barn for the fission of ^{235}U . Therefore, injecting boron into a nuclear reactor will effectively decrease the reactivity.

Boron injection has different purposes depending on the reactor type. In boiling water reactors, boric acid is used as part of the emergency shutdown system. In pressurized water reactors, the most common reactor type in the world [38], boron is also used to control the reactivity of the core during normal operation by increasing or decreasing the neutrons reaching the fuel. Boric acid used for this purpose is known as a “chemical shim”. However, it can also be used for emergency cooling in these types of reactors as well [22].

Boric acid is therefore a chemical that should be expected in a nuclear accident scenario for both reactor types, and its reactions with other species known from this context warrants investigation.

Solid boric acid undergoes a series of dehydration reactions seen in Reactions 3, 4 and 5 as it is heated [39].

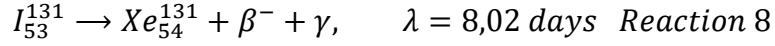
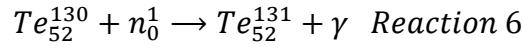


Tellurium and boron are known to interact in certain situations, with one such case being in borotellurite glasses, which can be formed by heating boric acid [40] or B_2O_3 [41] and TeO_2 together. Boron and tellurium can also occur together in organic systems [42], though this has not been seen in accident contexts. However, the chemistry involving tellurium and boric acid is called out specifically by the APRI (accident phenomena of risk importance) final report from 2022, compiled at the behest of the Swedish radiation protection agency [43], as a phenomenon that needs to be studied.

3. Theory

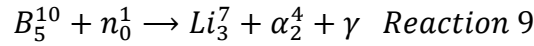
3.1. Instrumental neutron activation analysis

Instrumental neutron activation analysis involves irradiating a sample and analysing the subsequent nuclear decay to get an understanding of the composition and amount of sample irradiated. In the present case, the neutron reaction is described in reactions 6, 7 and 8.



Where ^{130}Te has about a 34% abundance in natural tellurium. ^{131}I has a half-life of about eight days and can be measured with gamma spectrometry. In principle, it is also possible to use the decay of ^{131}Te into ^{131}I , or even the decay of $^{123\text{m}}\text{Te}$ into ^{123}Te . However, ^{131}Te has a short half-life of only 25 min, and $^{123\text{m}}\text{Te}$ has a half-life of 119 days and is produced from ^{122}Te , which only has a 2.55% abundance in natural tellurium. These considerations make the decay of ^{131}I the most practical and achieve the lowest limit of detection and the lowest uncertainties [44].

INAA analysis was only performed on the samples investigating the organic interaction, as it was suspected that the boron on the other filters would interfere with the neutron irradiation, and thus make the measurement unreliable. However, the analysis of boron with neutron activation is possible in principle [45]. Natural boron consists of two isotopes, ^{10}B and ^{11}B , where the prior is much more reactive towards neutrons, according to reaction 9.



However, ^7Li is a stable nucleus, and so it cannot be measured by gamma spectroscopy. Instead, determination of boron with INAA depends on the prompt reaction where either the α - or the γ -ray from the nuclear reaction is measured during the neutron irradiation process [45]. This has not been done for this work but may be relevant for future studies.

4. Experimental

The main experimental work has taken place at VTT Research Center in Espoo, Finland. Analysis has likewise taken place at VTT, as well as at Chalmers University of Technology, Gothenburg, Sweden. All experiments follow the same general pattern in that they include the volatilization of tellurium metal at the temperature of 540°C under a reducing, inert or oxidizing atmosphere. Another compound of interest is then injected into the stream, the temperature is changed depending on the experiment, and the resulting mixture is analyzed.

4.1. Experimental setup

The volatilization of tellurium took place in a tubular furnace (Entech/Vecstar, VCTF 4), with a tube of AISI 316L steel. The tellurium precursor (metallic tellurium, Te, Sigma-Aldrich, purity $\geq 99.997\%$) was placed in an Al_2O_3 crucible which was heated in the center of the heating section of the furnace. The furnace was connected to another, identical tube through a junction where an aqueous solution could be added. The setup is schematically described in Figure [2].

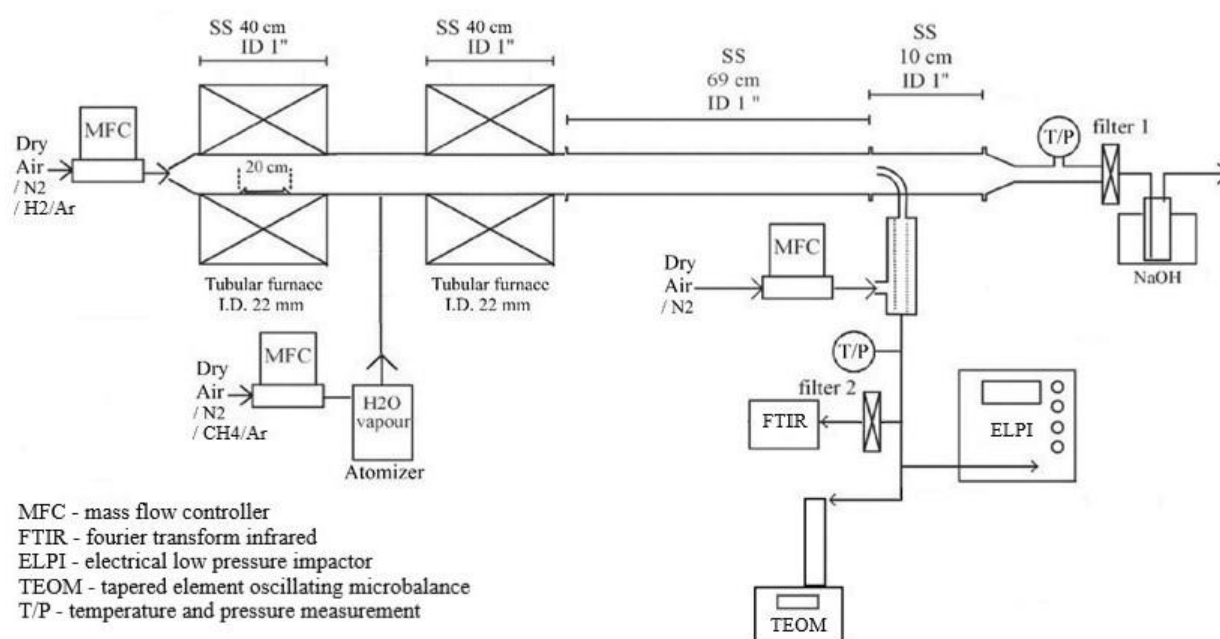


Figure [2]: Schematic representation of the experimental setup used throughout this work.

The flows through the systems were regulated by a pair of mass-flow-controllers (MFC:S). A first MFC (Brooks S5851, Brooks® Instrument) regulated the feed of the carrier gas through the volatilization furnace, and a second, identical one was used to regulate the flow through the atomizer at the middle junction. The carrier gases varied between the experiments. Air, nitrogen, or a mixture of 5% H₂ in 95% Ar were all used to simulate different atmospheres. In total, the gas flow through the reaction furnace was 6 l/min in all experiments. After the reaction furnace, 5 l/min of the gas was directed through a filter (MilliPore, Mitex™ PTFE filter, pore size 5 µm, diameter 42 mm) followed by a liquid trap (100 ml 0.1 M NaOH) before being vented into a fume hood. The last 1 l/min was diverted and quenched with nitrogen gas. This portion of the gas was analyzed online with Fourier-transformed infrared spectrometry (FTIR), before being sent through the electric low-pressure impactor (henceforth referred to as “ELPI”) and the tapered element oscillating microbalance (henceforth referred to as “TEOM”), both of which are used to characterize aerosols.

4.2. Studies on interactions between tellurium and organics

1 g of solid elemental tellurium powder was placed in the volatilization furnace. The furnace was heated to 540°C to volatilize the tellurium, and the actual carrier gas was connected. The reaction furnace was set to 300 °C. In total, three organic species were used. Acetone and propanol solutions were both fed as droplets through the atomizer at the junction. These compounds were chosen to represent species found in epoxy paints, while also being suitably soluble in water.

Methane was introduced along with the carrier gas and chosen since it is a common pyrolysis product. During the experiments using methane, the atomizer was not in use. The experimental matrix is depicted in Table 1.

Table 1: Experimental plan for the influence of organics on volatilized tellurium.

Experiment #	Reaction Temperature [°C]	Atmosphere	Organic precursor
1	300	Air	Acetone
	300	Air	Propanol
2	300	N ₂	Acetone
	300	N ₂	Propanol
3	300	H ₂ /Ar	Acetone
	300	H ₂ /Ar	Propanol
4	300	H ₂ /Ar	Methane
4.1.	300	N ₂	Methane

Notice that methane is not investigated in air atmosphere for safety reasons.

4.3. Studies on interactions between tellurium and boric acid

For each experiment, 5 g of solid elemental tellurium powder was placed in the volatilization furnace and the furnace was heated to 540°C. Each temperature and atmosphere included two experiments, one reference state without boric acid, and then one identical experiment with dissolved boric acid. For the experiments using boric acid an aqueous solution (0.2 M B(OH)₃) was added through the atomizer. For the experiments without boric acid, the atomizer was not in use. The reaction furnace was set to one of two temperatures, 300°C or 650°C. Until the setpoint for both furnaces were reached, the system was flushed with nitrogen, and the exhaust gas was diverted from the sampling position. As the experiment started, the relevant carrier gas was started, and the flow changed to run through the sampling positions. Each experimental condition lasted for 30 min. The experimental matrix is depicted in Table 2.

Table 2: Experimental plan to study the influence of boric acid on tellurium speciation and transport.

Experiment designation	Reaction Temperature [°C]	Atmosphere	Injection
Ref_Ox_LT	300	Oxidizing (Air)	-
Exp_Ox_LT	300	Oxidizing (Air)	B(OH) ₃ (aq)
Ref_Ox_HT	650	Oxidizing (Air)	-
Exp_Ox_HT	650	Oxidizing (Air)	B(OH) ₃ (aq)
Ref_In_LT	300	Inert (N ₂)	-
Exp_In_LT	300	Inert (N ₂)	B(OH) ₃ (aq)
Ref_In_HT	650	Inert (N ₂)	-
Exp_In_HT	650	Inert (N ₂)	B(OH) ₃ (aq)
Ref_Rd_LT	300	Reducing (H ₂ /Ar)	-
Exp_Rd_LT	300	Reducing (H ₂ /Ar)	B(OH) ₃ (aq)
Ref_Rd_HT	650	Reducing (H ₂ /Ar)	-
Exp_Rd_HT	650	Reducing (H ₂ /Ar)	B(OH) ₃ (aq)

4.4. Analytical Techniques

The following techniques were used to analyze the experiments. Inductively Coupled Plasma Mass Spectrometry (henceforth referred to as “ICP-MS”) was performed on the liquid trap solutions, Instrumental Neutron Activation Analysis (henceforth referred to as “INAA”), X-ray photoelectron spectroscopy (henceforth referred to as “XPS”) and scanning electron microscopy henceforth referred to as “SEM”) were all performed at the filters, and TEOM/ELPI data were collected online during the experiment. FTIR data was likewise collected, but did not reveal much of interest, and those results will not be discussed further.

4.5. ICP-MS

Inductively Coupled Plasma Mass Spectrometry (Element 2, ThermoScientific) was used to measure the concentration of the tellurium and other relevant species in the liquid samples following each experiment. The samples were diluted with 0.5 M nitric acid (Suprapure) to a fitting concentration. The limit of detection for the instrument was 0.006 ppb for tellurium (¹²⁶Te). An internal standard of 1 ppb rhodium was used.

4.6. XPS

Following the experiments, the filter was cut into pieces. Some of these were analyzed with XPS (X-ray photoelectron spectroscopy, PHI5000 VersaProbe III- Scanning XPS Microprobe). The X-ray source was a monochromatic AlK α -source with an energy of 1486.6 eV. The beam diameter was 100 μ m, 25W, 15 kV. The survey scan was performed across the energies 0-1100 eV, with a step-size of 1 eV. The detailed scan had a step-size of 0.05 eV for the C s1 signal (used as a reference signal), and 0.1 eV for the other signals. The detailed scan measurements were aligned to the carbon signal at 284.6 eV before analysis, and the system was aligned with the signals 83.96 eV from Au, 368.21 eV from Ag, and 932.62 eV

from Cu. The actual measurement time typically did not exceed ten minutes per individual measurement.

4.7. SEM

The filters were analyzed with SEM (Scanning Electron Microscopy). The instrument was Zeiss Crossbeam 540 operating at 2.0 kV with a probe current of about 100 pA.

4.8. TEOM and ELPI

TEOM (Tapered Element Oscillating Microbalance) and ELPI (Electrical Low-pressure Impactor) are two analytical on-line methods for determining information about the aerosols present in the gas stream during the experiments, and how they change. TEOM measures the total mass concentration of aerosols in the gas stream, and ELPI measures mass distribution and particle number.

4.9. INAA

Instrumental neutron activation analysis (INAA) involves irradiating a material and then measuring the radiation. The irradiation and subsequent analysis took place in Prague at the Nuclear Physics Institute CAS (UJF), using their LVR-15 research reactor. The irradiation lasted for 2.5 hours, and the neutron fluence rate was $3 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$, $8 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$, and $8 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ for thermal, epithermal, and fast neutrons, respectively. Subsequent analysis was performed in a HPGe detector (Genie 2000 Canberra). Two sample geometries were used, 20 cm from the detector for most samples, and 45 cm from the detectors for a few especially active samples. All samples were measured for 1 hour (live-time) for the 20 cm geometry, and 2 hours (live time) for the 45 cm geometry.

A pair of calibrating samples were also prepared and irradiated alongside the normal samples. These were prepared by dissolving 24.7 mg Te metal (99.9 %, Merck) in 5 ml hot HNO_3 and diluting the solution to 10 ml. 50 μm of the solution was deposited onto 16 mm chromatographic paper disks (Whatman 1), dried, and irradiated in the same way as the rest of the samples. The calibrators contained $124.9 \pm 0.4 \mu\text{g}$ of Te and are used to calculate the amount of tellurium in the rest of the samples.

5. Results and Discussion

This report is based on two papers employing similar experimental and analytical methods. As such, the result of both papers will be presented together with each section concerning a certain method, to allow for easy comparison between the two experiments and discussions about the differences or similarities where appropriate.

5.1. Analysis of the liquid trap solutions

The tellurium contents for the liquid trap in the experiment involving organic species is presented in Table 3, and the contents found in the experiment involving boric acid is found in Table 4.

Table 3: Content of tellurium in the liquid trap after experiments involving organic species.

Experiment Designation	Organic species	Tellurium content [$\mu\text{mol}/\text{dm}^3$ trap solution]
1	None (reference)	0.004
1	Acetone	0.004
1	Propanol	0.008
2	None (reference)	0.007
2	Acetone	0.041
2	Propanol	0.020
3	None (reference)	0.009
3	Acetone	0.034
3	Propanol	0.034
4	None (reference)	0.004
4	Methane	0.029
4.1	None (reference)	0.008
4.1	Methane	0.034

These concentrations are all very low, implying that no, or very little, volatile species are formed. In general, the processes including organics do display a higher content of tellurium in the liquid trap, but as the concentrations are always low, and these experiments are performed in singles, no certain conclusions can be drawn from this fact.

Table 4: Content of tellurium in the liquid trap after the experiments involving boric acid.

Experiment Designation	Tellurium content [$\mu\text{mol}/\text{dm}^3$ trap solution]	Boron Content [mmol/ dm^3 trap solution]
Ref_In_LT	2.35	0.17
Exp_In_LT	2.35	1.57
Ref_In_HT	0	0.24
Exp_In_HT	0.78	6.15
Ref_Rd_LT	0.78	0.59
Exp_Rd_LT	0.78	0.75
Ref_Rd_HT	41.5	1.11
Exp_Rd_HT	7.05	3.27
Ref_Ox_LT	0.78	0.13
Exp_Ox_LT	0.78	2.16
Ref_Ox_HT	0.78	0.23
Exp_Ox_HT	0.78	10.37

The tellurium content is somewhat stable across most experiments and in general there is no, or very little, difference between the reference experiments and the experiments involving boric acid. Since there were plenty of precursor left in the crucible after all experiments were done, the ICP-MS measurement does not indicate direct interaction between boric acid and tellurium. There are two outliers in this experiment: namely the two experiments in high temperature and under reducing atmosphere, which display much higher tellurium content than all other experiments. Presumably this is due to the formation of hydrogen telluride, H_2Te . This gas is the tellurium analogue of water and is reported to form at elevated temperatures from hydrogen and tellurium [46], making its presence in these conditions possible. Being a gaseous species also would allow it to pass through the filter and into the liquid trap.

As for the boron, it should be noted that the liquid trap was made from borosilicate glass, and that alkaline solutions, such as the trap solution, does etch such glass. Some contamination of boron can therefore be expected. The presence of boron in the reference experiments, as seen in Table 4 is attributed to this.

The amount of boron in the liquid trap is about three orders of magnitude higher than for tellurium in all cases. Furthermore, the boron added to the gas stream is able to penetrate the filter reliably in both inert and oxidizing atmosphere, as the boron contents of the trap increase an order of magnitude for all such experiments. As for the reducing system, there is an increase in boron in the liquid trap compared to the references, but the difference is much smaller than in the previous conditions. This may indicate an interaction between boric acid (or its derivatives) and hydrogen that forms an aerosol species.

Comparing the tellurium contents of the two experiments can only be done tentatively, as the amount of precursor was different in the experiments, with the boric acid experiments starting with five times as much precursor as the experiments with organics.

In general, the amount of tellurium in the liquid traps is much lower after the experiments with organic species than after the studies with boric acid. This includes the boric acid experiments conducted at 300 °C, which is the same temperature used for the experiments with organics.

A noteworthy fact is that both experimental plans do involve reference experiments. In principle, the low-temperature reference experiments in the boric acid project with the references of the organic project are nearly identical, using the same setup and the same conditions aside from the amount of precursor. The fact that they display very different results indicate a strong dependence on the precursor amount.

5.2. XPS analysis

Relevant signals for XPS measurements of various tellurium species are listed in Table 5.

Table 5: Relevant XPS signals for different tellurium and boron compounds observed in this work.

Tellurium species	Associated XPS signal [eV]	Orbital
Te	573,0-573,5 [47], 572,9 [48], 572.7-573.3 [49]	3d _{5/2}
Organic Tellurides*	574.0-575.3* [50], 573.8-574.2** [49]	3d _{5/2}
TeO ₂	576.0-576,2 [47], 575,8 [51] 575.5-576.0 [49]	3d _{5/2}
TeO ₃	576.8 [48] 576.5-576.9 [49]	3d _{5/2}
TeO ₃ *(3H ₂ O)/Te(OH) ₆	576.9-577.3 [49]	3d _{5/2}
B(OH) ₃	192.9-193.5 [49]	1s
B ₂ O ₃	192-193.5 [49]	1s

* Refers to various Te²⁻-based compounds, see further [50]

** Refers to Ph₂Te specifically.

The XPS analysis of the tellurium in the presence of organics is presented in Table 6.

Table 6: XPS signals for the various conditions of the experiments involving tellurium in the presence of organics. All these experiments took place at 300 °C.

Atmosphere	Added compound	Measured Tellurium 3d5/2 signals [eV]
N ₂	None (reference)	573.4 576.5
H ₂ /Ar	None (reference)	573.1 576.2
Air	None (reference)	575.2 577.6 579.4
N ₂	H ₂ O, Acetone	572.9 575.0 576.7
H ₂ /Ar	H ₂ O, Acetone	-
Air	H ₂ O, Acetone	-
N ₂	H ₂ O, Propanol	572.5 574.6 576.4
H ₂ /Ar	H ₂ O, Propanol	573.1 576.2
Air	H ₂ O, Propanol	572.7 574.9 576.8
N ₂	H ₂ O, Methane	573.2 575.1 576.6
H ₂ /Ar	H ₂ O, Methane	573.3 576.3

The results of the XPS can roughly be divided into three groups: signals of energies around 573 eV, energies around 576 eV, and finally energies around 575 eV. There are also some signals of 577-580 in the case of the air-reference sample.

All of these can be assigned different tellurium compounds with varying levels of certainty, based on the values from Table 5. The signals around 573 eV occur in nearly every sample and is ascribed to metallic Te. As this is the precursor its presence is logical and expected, especially for the samples not exposed to oxygen. The signals around the energy 576-577 eV can be ascribed to some oxide, either TeO₃, or potentially TeO₂.

The major question posed by the XPS results is the reason for the signal at 574-575 eV, as there are two possibilities. It can be TeO₂, or it can be organic tellurides, as they both display energy signals in this range, though organic species tend to have a lower range (ca 574-

575.5 eV) than TeO_2 (575.5-576.2 eV) according to the literature. However, in the reference measurement performed in air a signal is measured at 575.2 eV. With no organics in the system, this signal is attributed to TeO_2 , and making the minimum signal for TeO_2 575.2 eV. However, no such signal is seen in the references for the reducing or inert conditions.

Upon adding organics to the system, signals of the range 574.6-575.1 are seen. These are lower than the literature values for TeO_2 , and lower than the “confirmed” signal of TeO_2 from the reference. As such, there is a case for the formation of organic tellurides being displayed on the filters, as the signals from the XPS matches the literature values of organic tellurides, while seemingly avoiding the one credible alternative in TeO_2 . This is especially true in the case of the reducing and inert atmospheres, as no compound with the signal around 574-575 are seen in the reference cases for those compounds.

What speaks for the formation of TeO_2 is the fact that, compared to the reference cases, all the experiments were also done in the presence of water vapour, which could act as a supply of oxygen. However, this does not explain the fact that according to literature, the signals indicating TeO_2 should be slightly higher than what is measured in the experiments performed. Furthermore, some level of oxidation is inevitable when the sample is exposed to open air, especially at the topmost surface of the sample, which is the portion of the sample probed by XPS.

The XPS signals for the experiments involving boric acid are listed in Table 7.

Table 7: Measured signals of tellurium and boron in experiments investigating the impact of boric acid on tellurium speciation.

Atmosphere	Temperature [°C]	Added compound	Measured tellurium 3d5/2 signals [eV]	Signal ratio [% of total signal]	Measured Boron 1S signals [eV]
N ₂	300	None (reference)	573.0 576.3	60 40	-
N ₂	650	None (reference)	573.0 576.4	56 44	-
H ₂ /Ar	300	None (reference)	573.0 576.4	62 38	-
H ₂ /Ar	650	None (reference)	573.1 576.4	61 39	-
Air	300	None (reference)	573.4 576.2 578.0	8 48 44	-
Air	650	None (reference)	576.8	100	-
N ₂	300	H ₂ O, B(OH) ₃	573.3 576.7	68 32	193.2
N ₂	650	H ₂ O, B(OH) ₃	573.3 576.5	77 22	193.0
H ₂ /Ar	300	H ₂ O, B(OH) ₃	573.1 576.5	66 34	192.9
H ₂ /Ar	650	H ₂ O, B(OH) ₃	573.1 576.5	75 25	193.3
Air	300	H ₂ O, B(OH) ₃	573.3 576.3 578.6	9 55 36	193.3 195.4
Air	650	H ₂ O, B(OH) ₃	576.7	100	193.5

The signals themselves are relatively uninteresting by themselves as all samples display almost the same species already identified; the signal at 573 eV corresponds to metallic tellurium, and the signal at 576 eV to TeO₃, or TeO₂. The samples from oxidative environments at 300 °C also show signals as high as 578 eV. The signal is tentatively assumed to be due to TeO₃, as it is a compound that could form in these conditions and has a high-energy signal, though the measured signal is still higher than the values found in the literature.

These samples were measured with XPS within a week of the experiments. Oxidation of samples in storage was confirmed by re-measuring a pair of samples from the high temperature reducing conditions after one week of room exposure to ambient conditions.

There was a noticeable increase of oxidation, but no new species seems to have formed. An example of this is seen in Figure [3], where the signal at 576 eV had increased over the course of fourteen days.

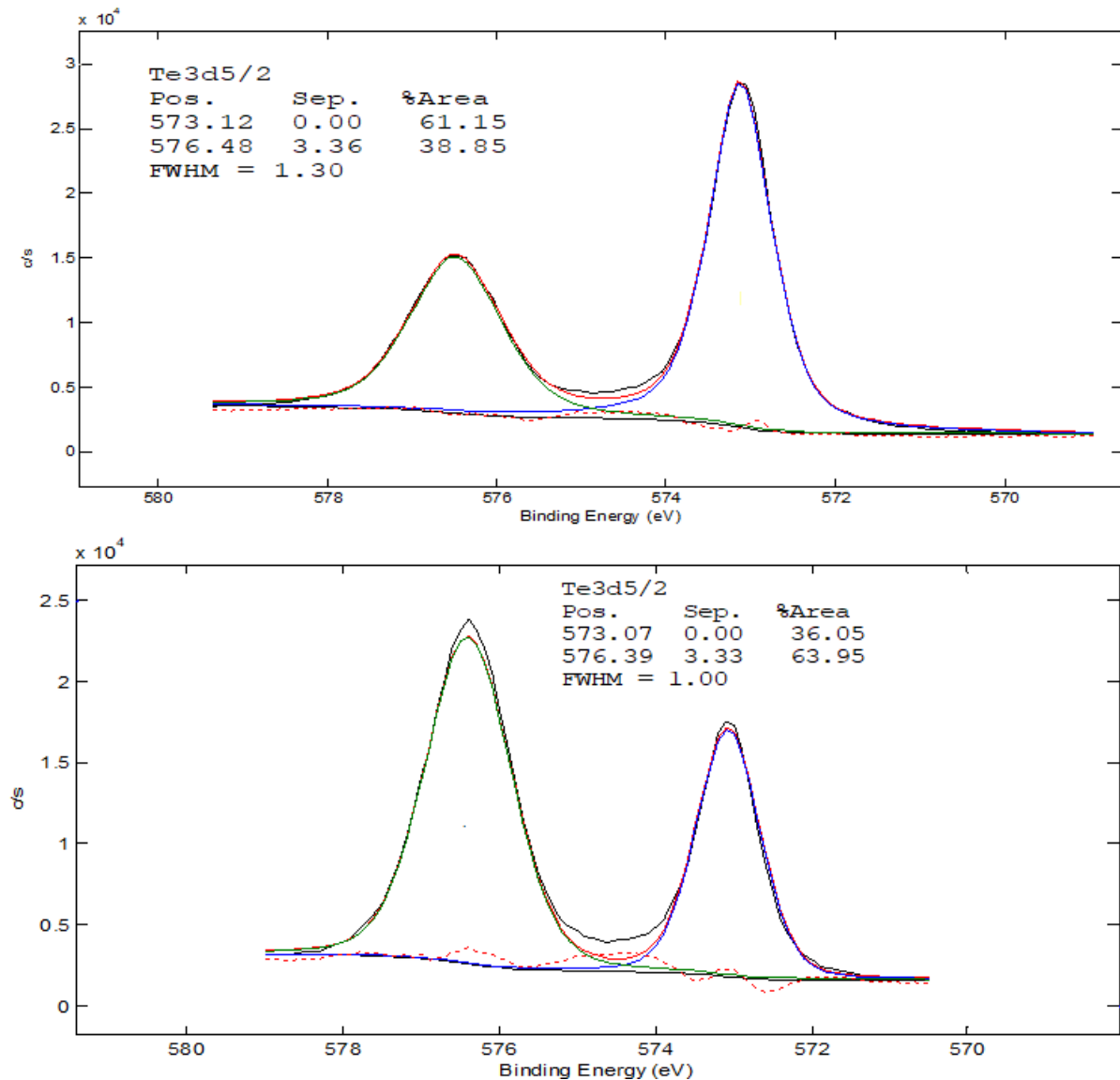


Figure [3]: XPS spectra of the filters exposed to tellurium in reducing atmospheres at 650 °C. The two spectra were collected from the same filter, two weeks apart with the leftmost spectrum being the first measurement, and the rightmost spectrum being collected fourteen days later. The unit on the y-axis is counts per second.

Comparing the signal ratios between the references and the experiments, the first thing of note is that the oxidating environments display a very little change, and that the samples are nearly completely oxidized in all cases.

In the case of inert and reducing conditions, the oxidation level typically decrease slightly in the experiments (the ratio of the signals is shifted towards metallic tellurium). This pattern is consistent across all of these samples and would have some implications from a nuclear safety perspective as metallic tellurium is more volatile than the oxidized form.

Finally, the boron displays two signals, most commonly around 193 eV and once at 195.4 eV. In general, the boron displayed very low signal counts and the associated spectra have a poor signal-to-noise ratio. The higher of those signals does not fit any expected compound, though it does match reasonably well with the signal for $\text{B}(\text{FH})_3$. In principle it would be possible for boron to interact with the PTFE filter and form boron-fluoride bonds, though this explanation is tentative. The second signal at 193 could correspond to either $\text{B}(\text{OH})_3$ or B_2O_3 . These compounds have overlapping signals (as they both contain the same bond: B-O) and differentiating one from the other with certainty is not possible using only XPS.

The two filters exposed to high temperature and reducing conditions were measured twice: once together with the rest of the samples, and then once again thirteen days later. During those thirteen days, the samples were exposed to ambient conditions. As XPS can determine the elemental composition of the surface under investigation, this was used to get an idea in how the oxidation of the tellurium surfaces changes over time. The results of these measurements are seen in Table 8. From this experiment, it appears that the oxidation behaviour changes due to the boric acid, or potentially the boric acid in combination with the reducing atmosphere. There is virtually no difference in oxidation on the filter treated with boric acid, whereas the reference has oxidized considerably.

Table 8: Change in surface composition after thirteen days of ambient temperature for the filters collected from reducing conditions and high temperature. The filter exposed to boric acid has hardly changed, whereas the reference has oxidized further.

Experiment	First measurement			Second measurement		
	Te [%]	O [%]	B[%]	Te [%]	O [%]	B[%]
Ref_Rd_HT	50.5	49.5	-	70.9	29.1	-
Exp_Rd_HT	54.2	34.4	11.3	54.0	33.0	13.0

This result lends some credence to the idea that boric acid does reduce oxidation of tellurium.

Comparing the results from both experimental series lends further credence to the idea that the signals around 575 presented in Table 6 does correspond to the formation of organic tellurides, rather than TeO_2 , as the signal at 575 eV is never seen in the second experimental series, despite this series also including water. It is then unlikely that water is responsible for forming TeO_2 in the first experimental series.

5.3. INAA-analysis

During the gamma-spectroscopy measurements, a few activated samples displayed very high activity, leading to an unacceptable dead-time in the system. To compensate, the distance between sample and detector had to be increased. However, this made measurements of the calibrators more difficult, and the dead-time was still high (up to 60% for the most active sample).

However, the fact that these filters display such a high activity indicates that, for every atmosphere, the system without organics displays considerably more tellurium on the filters

than any system with organics. As previously mentioned, the same experiments consistently show the least tellurium in the liquid traps.

The results of the neutron activation, when accounting for the blank measurement and normalizing against the signal of the calibrators, can be found in Table 9.

Table 9: Results for the neutron activation, after the counts from the blank filter were subtracted. Mass calculation is based on the amount of Te added to the calibrator; see further section “Analytical techniques, subsection INAA”.

Experiment #	Sample [precursor]	Measurement Geometry [cm]	CPS [of ^{131}I , 364.39 KeV]	CPS normalized against calibrator	Mass of Te [mg]
-	Calibrator	20	16.19	1	0.125
-	Calibrator	45	3.32	1	0.125
1	Reference	20	3416.04	211.00	26
	Acetone	20	10.42	0.64	0.08
	Propanol	20	10.70	0.66	0.08
2	Reference	45	5099.43	1535.97	191.84
	Acetone	20	148.23	9.16	1.14
	Propanol	20	21.68	1.34	0.17
3	Reference	45	3506.51	1056.18	131.92
	Acetone	20	1389.92	85.85	10.72
	Propanol	20	744.22	45.97	5.74
4	Methane	45	3528.77	1062.88	132.75
4.1	Methane	20	374.81	23.15	2.89

5.4. Particle transport

The particle mass-concentration was investigated for both sets of experiments. The relevant data is presented in Figure [4].

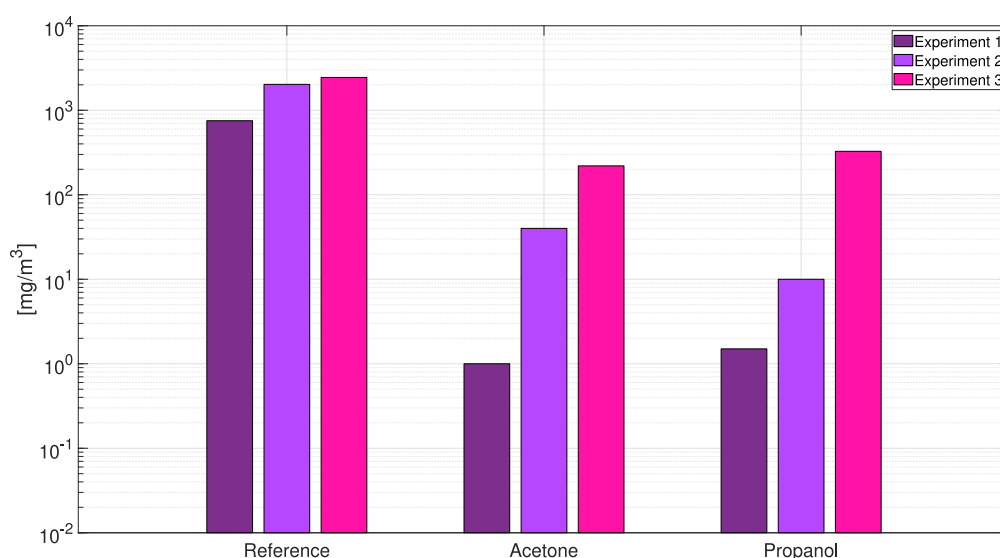


Figure [4]: Mass-concentration data for the addition of acetone and propanol to the volatilized tellurium. As was suggested by INAA, the reference cases produced significantly more aerosols than the systems involving organics.

The highest mass concentration was always found among the reference cases, and indeed the mass concentration of particles was somewhat similar in every reference case, though the oxidizing case was relatively low. This trend continues is consistent with the rest of the experiments.

In general, there is not a lot of difference between the different organic species, as they look rather similar. The difference appears to be mainly due to the different atmospheres.

The TEOM-results for experiment 4 and 4.1 are presented in Figure [5]. The reference cases give a similar result to the previous experiments, with a mass concentration of about 1 g/m³. However, the systems involving methane in inert gas (experiment 4) do not exhibit a decrease of the particle mass compared to the reference, whereas the same experiment in reducing conditions do. In fact, of all the experiments using organics and being run in reducing conditions, this experiment appears to have the lowest particle mass concentration.

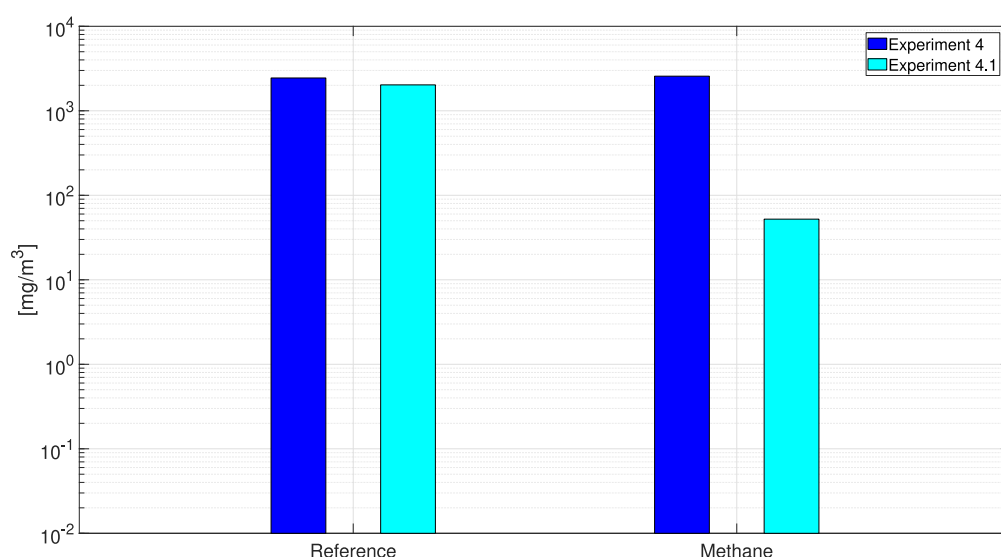


Figure [5]: Mass-concentration data for the addition of methane to the volatilized tellurium.

It is possible that a reason for the decrease in particle mass-concentration upon addition of organics has to do with the acetone and propanol being dissolved in water, which was also injected. Potentially, the water causes the tellurium particles to deposit in the tubing at a higher degree than in the reference case, leading to a decrease in the particle mass concentration. This would also explain why the methane experiments in the inert atmosphere do not see a decrease in aerosols, as the methane is not added as a water matrix.

It does not explain the mass concentration reduction in the case of methane in reducing conditions, however. This system also did not involve water.

Compared to the references, both the systems including methane did notice an increase in tellurium content in the liquid trap (see Table 3), indicating that some volatile species may form under both conditions. However, the difference between the liquid trap contents for the inert and reducing systems is extremely small and does not explain the difference in mass concentrations between the systems.

The relative mass concentrations determined by TEOM match well with the particle masses on the filters determined by INAA, as the high mass concentrations correspond to a high mass on the filters, and vice versa, as could be expected.

Similar data was collected for the boric acid investigation, in the case of inert and reducing atmospheres, as presented in Figure [6]. The processes in oxidizing conditions, however, did not yield any clear data, and so those conditions have been omitted.

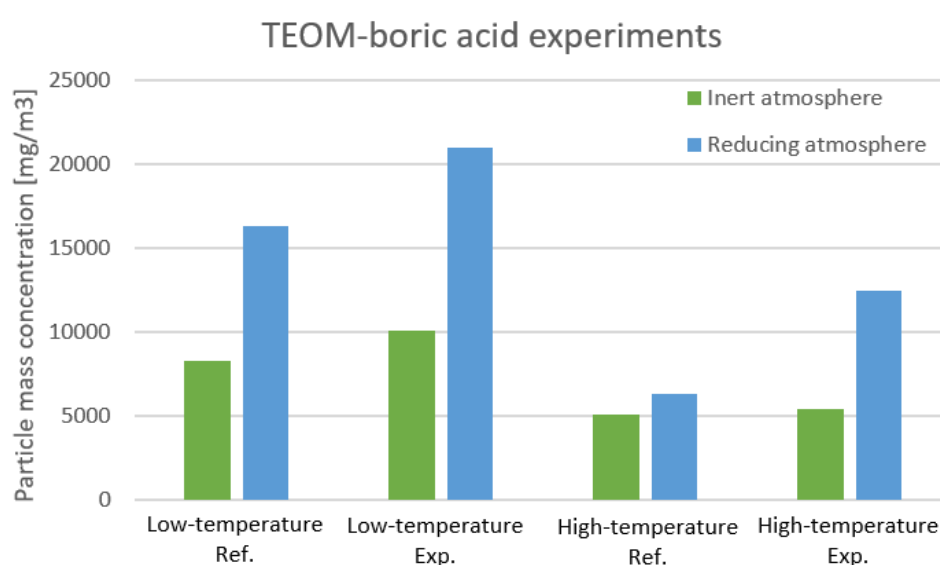


Figure [6]: Mass concentration of the boric acid investigation. The conditions designated “Ref.” indicates reference states; that is, without boric acid. “Exp”, accordingly, refers to an experiment involving boric acid.

In these experiments the reducing atmospheres consistently gives a higher mass concentration than the inert series, and the processes involving boric acid also gives higher concentrations than their corresponding reference cases without acid.

The reason for higher mass concentration in the inert case is uncertain, but it does line up with what was seen in the experiment with organics, where the reducing atmosphere again yielded higher mass concentrations than the inert case. This may be due to oxidation of the tellurium happening easier inert conditions than reducing conditions, but of course there should be very little oxygen available for the formation of TeO_2 in either case. This hypothesis also does not line up with XPS data, which does not imply a large difference in oxidation level between these conditions in the reference or the experimental cases.

Comparing the overall values between the study of organics and the study of boric acid, the values in the boric acid study tend to be higher. Presumably this is due to a higher amount of precursors in this experimental series (5 g tellurium compared to 1 g).

6. Conclusions

There is still no definite proof of interaction between organic species and volatilized tellurium in the gas phase, but there are results implying the formation. The amount of tellurium in the liquid traps is very low, but it does increase when organics are added to the system both in the reducing and inert conditions. There is no, or very little, increase in the case of oxidizing conditions, presumably due to the formation of TeO_2 . The XPS measurements can also be interpreted to indicate the formation of organic tellurides. Finally, the TEOM data implies a lower number of aerosols in every case involving organics compared to the reference cases.

The study of the impact of boric acid on tellurium speciation implies that the boric acid does not directly interact with tellurium to form Te-B bonds. This is inferred from the liquid trap analysis, which does not indicate an increase of volatile compounds, and the XPS which does not reveal any solid particles with that type of bond. The high-temperature process in reducing conditions does result in a large increase of tellurium in the liquid trap though, presumably due to the formation of TeH_2 .

However, boric acid does seem to influence tellurium in that the level of oxidation on the tellurium is decreased when boric acid is added to the system. This too was seen with XPS. A pair of samples (one treated with boric acid and the corresponding reference) were re-measured after thirteen days, and while oxidation had noticeably increased in the reference case, the second sample was virtually unchanged.

7. Future work

Another paper relating to the interactions between boric acid and tellurium is currently in the works. This work builds on the studies of boric acid by also including iodine and cesium iodine in an attempt to clarify whether their presence alters the tellurium speciation, or the interaction between tellurium and boric acid.

The question of organic tellurides being formed in the gas-phase from tellurium aerosols has not been conclusively solved yet and warrants further investigation, as does the question if the formation of organic tellurides can compete with organic iodides. It is not impossible for both to form under similar conditions, but the experiment that determined organic tellurides [27] did not include any iodine. As such, it is possible that either tellurides or iodides takes precedence and makes the other class irrelevant. This needs to be investigated. Furthermore, organic species including both tellurium and iodine may need to be considered.

Furthermore, there is the question of tellurium release from the fuel. The process is, in principle, relatively well understood but questions about certain interactions and processes

still warrant more detailed investigation. One such process is the interaction/ release of tellurium from epsilon-particles- that is, particles of fission product noble metals that form distinct inclusions in the fuel.

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9. References

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