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# Organic Telluride Formation during a Severe Nuclear Accident (ORTEF)

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## Abstract

The potential formation of organic species during a severe nuclear reactor accident is a cause of concern due to the very high volatility of organic compounds. The interactions between organic material and fission products are especially concerning since this could lead to increased source term even in post-accident conditions. One of the fission products potentially forming volatile organic species is tellurium. Although evidence of the formation of organic tellurides in accident conditions was only showed recently, this raises concerns whether organic tellurium species should be considered a significant threat in accident scenarios. The aim of the ORTEF project was to investigate potential interactions between tellurium and organic material in the gas phase. The project was a collaboration between Chalmers University of Technology, VTT Research Center of Finland and University of Oslo. The experimental conditions in ORTEF were chosen to represent those likely present in the containment gas phase where tellurium aerosols would be present simultaneously with organic gas or vapor. The results show that the presence of organic material increases the transport of tellurium in the gas phase. This was observed by measuring higher concentration of tellurium in the trap solution in the presence of organics compared to the reference conditions. Oxidizing atmosphere and the consequent oxidation of elemental tellurium to tellurium oxide inhibited the interactions whereas the increased transport was apparent in inert and reducing atmospheres. Although the speciation analysis was inconclusive, the significant increase in the gaseous fraction is a clear indication of interactions. Whether the increased transport is due to the formation of organic tellurides or other interactions remains for further studies.

## Preface

The ORTEF project was a collaboration between Chalmers University of Technology, VTT Research Center of Finland and University of Oslo. The three organizations have had successful collaborations in the past and ORTEF was no exception. The project focused on studying the interactions between tellurium and organic material in containment gas phase conditions.

The experiments were performed at VTT. The samples obtained from the experiments were analyzed partly at VTT and Chalmers. Samples requiring instrumental neutron activation analysis were sent to Nuclear Research Institute in Rez, Czech Republic for analysis.

The report presents the main results obtained from the ORTEF project. More detailed analysis and discussion will be presented in a scientific publication to be submitted in Spring of 2023.

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## 1. Introduction

During nuclear fission, isotopes of uranium are consumed to generate heat. The fragments of uranium are known as fission products. Some examples include  $^{131}\text{I}$ ,  $^{137}\text{Cs}$ , and  $^{90}\text{Sr}$ . During normal operation, they are mostly retained inside the fuel matrix and will remain there [1] until the fuel is removed from the reactor and eventually sent to its final repository.

However, in the case of a severe nuclear reactor accident, the fission products may be released and pose a radiological hazard to people and the environment [2]. The significance of the fission products depends on factors such as their release behavior from the fuel, speciation and its effect on the retention, and bioaccumulation in the body. In addition, the physical and chemical speciation varies between the elements. Therefore, different fission products will behave in different ways.

Some of the most significant fission products from a nuclear accident standpoint include isotopes of iodine (primarily  $^{131}\text{I}$  with a half-life of 8.02 days), cesium (primarily  $^{137}\text{Cs}$ , with a half-life of 30.08 years) and tellurium (primarily  $^{132}\text{Te}$  with a half-life of 3.2 days). All of these are designated volatile fission products [3], since they can be expected to volatilize and escape the fuel matrix, potentially travelling far from the accident site and cause widespread fallout. Furthermore, the decay of  $^{132}\text{Te}$  will give rise to  $^{132}\text{I}$ , with a half-life of 2.28 hours. As such, tellurium acts as a delayed source of iodine, which is arguably the most significant radio-hazardous element of all as it accumulates in the thyroid gland and gives a prolonged dose of radioactivity, increasing the risk for thyroid cancer [4]. Plenty of research has been conducted with the express goal of understanding how iodine will behave under severe accident conditions. Some of the major experimental programs with this goal includes PHEBUS [5] and VERCORS [3]. These programs have focused on the release and transport behavior of fission product from the core through the reactor coolant system. In addition, several individual experiments have been performed to study the behavior of iodine inside the containment e.g., radiolytic oxidation [6], formation of organic iodides [7] and deposition of different iodine species on containment surfaces [8]. However, experiments on the behavior of other fission products, especially tellurium, in the containment is scarce.

Aside from the volatility, both iodine and tellurium both have a complex chemistry which includes the interaction with organic molecules [9]. In severe accident scenarios, organic material can be present in both gas and aqueous phase originating from e.g., painted surfaces, insulators, or resins [10]. Due to the high dose of radiation present in both phases, the organic species can form radicals which can react quickly with fission products, such as iodine or tellurium to form organic iodides or tellurides. The organic species are of concern due to their extremely high volatility. For example, elemental tellurium has a boiling point at 988 °C whereas one of its organic forms potentially forming in accident scenarios, dimethyl telluride, has a boiling point at 85°C. Therefore, the concern around the volatility and release of organic species are very valid. Some examples of organic compounds capable of forming in accident scenarios include methyl iodide and dimethyl telluride. These compounds alter the chemical and physical properties of the radioactive species. Both organic tellurides and iodides are more volatile than the respective elemental forms. Organic iodides have been determined in severe accident scenarios, and recent studies have shown evidence of the potential formation of tellurides as well [11]. However, the understanding of the formation and behavior of organic tellurides in the context of severe accidents is lacking.

In severe nuclear accident contexts, organic tellurides have only been studied in the aqueous phase. However, as tellurium is a volatile element, and the conditions of an accident can vary, research should also be done to investigate the possibility of organic tellurides forming in the gas phase. This project

aims to investigate the possible interactions between tellurium and organic material in the gas phase. Emphasis has been put on the potential increase in tellurium transport because of such interactions.

## 2. Background

To understand the formation of organic tellurides in a nuclear accident, the nature of the element itself, as well as the sources and types of organic species that can be expected in an accident, must be understood. Organic iodides are also discussed briefly as a comparison.

### 2.1. Tellurium

In the subject of severe nuclear accidents, some isotopes of note that are formed in the fuel include several short-lived isotopes (their half-lives are all less than an hour):  $^{131}\text{Te}$ ,  $^{133\text{m}}\text{Te}$ ,  $^{134}\text{Te}$ , as well as the previously mentioned  $^{132}\text{Te}$  [12]. Together, these make up about 60% of the tellurium activity. With the relatively short half-life, tellurium is a radio hazard chiefly in the earliest stages of the accident. Emitted activities of  $^{132}\text{Te}$  have, in previous severe nuclear accidents, been relatively high. In the Chernobyl and the Fukushima-Daiichi accidents, the release of  $^{132}\text{Te}$  has been estimated to 88 PBq and 1150 PBq respectively. This is less than the releases of  $^{131}\text{I}$  (160 PBq and 1760 PBq, respectively) but much more than  $^{137}\text{Cs}$  (15 PBq and 85 BPq respectively) [13]. The radiological danger of inhaling  $^{132}\text{Te}$  is comparable to that of  $^{131}\text{I}$ , with their dose coefficients being similar ( $2 \times 10^{-9}$  Sv/Bq and  $2.4 \times 10^{-9}$  Sv/Bq, respectively) [13].

In the PHEBUS- FPT1 experiment, nuclear fuel was retrieved from a reactor and subjected to the conditions expected during a core meltdown. This experiment involved 9163 g uranium [5]. Before the experiment was started, the fuel bundle was irradiated as to regain the inventory of short-lived fission products. The fuel bundle was calculated to contain 2.52 g tellurium, and 1.12 g iodine. The fraction of tellurium released during the accident scenario is extremely dependent on the oxidation of the fuel cladding, which in turn is dependent on the temperature of the system. If the cladding remains unoxidized, tellurium is effectively trapped. Once the oxidation breaches a threshold, the tellurium release can become large; 100% release have been measured [14]. Of course, the contents of fission products in the fuel varies significantly with the burnup of the fuel- the higher the burnup, the more fission products are formed in total during the lifetime of the fuel. For high-burnup fuel, at reactor shutdown, the activity of the tellurium is about two thirds of the iodine activity [15].

If tellurium were to be released into the containment following a reactor accident, it can be expected to take on a variety of chemical and physical forms depending on the prevailing conditions of the accident. Tellurium melts at 450°C degrees and boils at 990 °C, both temperatures that can be exceeded inside the reactor during an accident. However, outside the reactor the temperature rapidly falls and may be lower than the melting point. During the PHEBUS FPT1- experiment, the simulated steam generator had a temperature between 700 °C and 150 °C, both well below the boiling point and, for a large segment, also the melting point of tellurium [5]. This, then, indicates that tellurium will likely be released to the containment in the form of aerosols. The physical nature of the aerosols in the primary circuit is expected to be a near lognormal, with an aerodynamic mass median diameter no larger than 2  $\mu\text{m}$  and a geometric standard deviation of 2 [16]. These also form the basis of the aerosols present in the containment, but it is possible that other effects, such as core-concrete interactions alter them at this stage.

Tellurium specifically have been shown in the Chernobyl accident to form stable aerosols with Cesium, where the ratio of these two elements remained almost constant across 1,400 km [17]. As such there is evidence that tellurium aerosols can spread far from the accident site and contaminate a large area.

During the Fukushima-Daiichi severe accident, TEPCO (Tokyo Electric Power Company) conducted measurements of air, water (both seawater and water collected from various places in the nuclear power plant), and soil were taken and reported. The earliest press conference mentioning tellurium in the soil is dated to the sixth of April, but the earliest measurement itself is from the twenty-first of March, ten days after the accident began on the eleventh of March. The isotopes  $^{129m}\text{Te}$  and  $^{132}\text{Te}$  are both found, and they exhibit activities of 250 kBq/kg dry soil sample, and 610 kBq/kg dry soil sample, respectively. These samples were taken at a playground area, roughly 500 meters west-north-west of the power plant. For comparison, the same sample contained 580 kBq/kg soil sample of  $^{131}\text{I}$  [18]. Tellurium is also found in the measurements looking for airborne particles.  $^{132}\text{Te}$  was detected in a measurement dating the twenty-second of March, with an activity of  $66.6 \mu\text{Bq}/\text{cm}^3$  [19]. The measurement was taken by the Fukushima Daiichi main gate. For reference,  $^{131}\text{I}$  displayed an activity of  $2.2 \text{mBq}/\text{cm}^3$  in the same measurement.

At room temperature tellurium can remain in its metallic form. However, when heated in air, it will oxidize, primarily forming tellurium dioxide  $\text{TeO}_2$ , whereas the corresponding monoxide  $\text{TeO}$  will be thermodynamically favored at higher temperatures [20]. In reducing hydrogen rich atmosphere, tellurium can form hydrogen telluride,  $\text{H}_2\text{Te}$  [21]. In the presence of steam, there is also the possibility for the formation of tellurium oxyhydroxide,  $\text{TeO}(\text{OH})_2$ , which increases the tellurium transport [22]. Furthermore, tellurium can react with tin to form  $\text{SnTe}$  [21], which is relevant in an accident scenario as tin is a constituent of the nuclear fuel cladding and this present in relatively large amounts. The stability of this compound depends on the oxygen potential. In oxygen-rich conditions, this compound will oxidize and give rise to  $\text{TeO}_2$  or  $\text{TeO}$ . At very high temperatures, tellurium may also exist in the elemental form as a dimer,  $\text{Te}_2$ .

## 2.2. Organic compounds

Organic compounds can be present in the containment in the form of paints, gaskets, or seals, for instance. Some studies have been conducted to determine the exact species released from various paints and primers. Investigating epoxy paints and primers used in the nuclear power plants in Sweden, Ringhals and the shut-down plant at Barsebäck, different organic compounds could be identified, such as xylene, benzyl alcohol and methyl phenols [23].

The organic compounds are not radio hazardous by themselves; however, their presence will affect the chemical interactions possible during an accident. Organic compounds may undergo radiolysis, thus forming highly reactive radical species. Together with tellurium, these may form organic tellurides [11]. This class of compounds have a much-decreased boiling point and chemical behavior compared to other forms of tellurium, making them a risk, especially in post-accident conditions, where re-volatilization from the containment surfaces or liquid phases could occur.

Studies on the dissolution of organic compounds from three paint films into water reveals several organic compounds that can also dissolve into the containment sump. The three films investigated included a polyurethane paint with an epoxy undercoating, a vinyl paint with a zinc primer, and an epoxy paint. Methyl isobutyl ketone (MIBK) is a compound that was found to dissolve from all three paints. Acetone was also found in the epoxy and the vinyl paints, supposedly as an impurity or due to a reaction with the paint surface. Formaldehyde was present in the vinyl and the polyurethane paints. Furthermore, the compounds in the gas phase were likewise investigated, and again MIBK was measured in the gas phase of all three paints. Another such compound which was *m*-xylene. In fact, throughout all three paints, *m*-xylene and MIBK always were the two most abundant compounds detected. In the case of the vinyl and polyurethane paints, MIBK was the most abundant compound

followed by m-xylene, whereas m-xylene was the most abundant compound in the epoxy paint, followed by MIBK. From a kinetic standpoint, MIBK is used to model the release for all paints as it is present in all three coatings. The final concentrations of MIBK in the aqueous phase ranges from 42  $\mu\text{M}$  to 330  $\mu\text{M}$  [24]. The MIBK solubility in water (at 25°C) is about 19g/dm<sup>3</sup> [25].

In a real accident case however, it can be expected that more complex organics will be broken down into simpler compounds due to the high temperature and radiation. As such, the exact composition of organics in a nuclear containment during an accident is difficult to predict. However, simple hydrocarbons such as methane or alcohol like ethanol and propanol are also possible and have been recorded during the degradation of some types of paint [24], even if they are commonly not present in the containment during normal operation. Compounds like these also have low boiling points, meaning they may also be present in the atmosphere rather than only in the sump, which has also been shown experimentally [24].

### 2.3. Previous studies the formation of organic tellurides

The progression of organic compounds undergoing radiolysis and forming radicals which then interacts with tellurium species is one way of forming organic tellurides. However, there are other paths as well.

Organic tellurides can be synthesized by mixing tellurium with an alkali metal (often sodium) in ammonia, and then reacting the resultant Na<sub>2</sub>Te with an organohalide to form the corresponding di-organic telluride [26]. However, this reaction is not possible in a nuclear accident scenario. Another scenario where organic tellurides have been formed is where organic radicals are passed by a tellurium mirror, whereupon the weight of the mirror decreases as the radicals passes by [27]. While such a tellurium mirror is not probable during an accident scenario, the result of this experiment does lend credence to the idea that organic tellurides can form in the presence of organic radicals. There is a strong case for this being possible in a nuclear accident scenario. The evidence of organic tellurides forming from solid tellurium in the presence of organic radicals raises concerns whether this phenomenon could occur inside the containment, either in the gas phase or from tellurium deposits. This work investigates this phenomenon by studying the interactions between tellurium aerosols and organic material in simulated containment conditions. The possible formation of organic tellurides was considered as well as interactions leading to increased transport of tellurium in the gas phase.

### 2.4. Organic Iodides

The most studied, and most problematic fission product from a severe nuclear accident standpoint is iodine. Much like tellurium, it too has a rich chemistry that involves organic compounds, such as methyl iodide. This compound in particular has been studied and found to be possible both in the aqueous phase [28] and in the gaseous phase, where the latter is expected dominate [29]. For the aqueous phase, this is dependent on the form of iodine, the concentration of constituent compounds, in this case the iodine radical (I·) and ·CH<sub>3</sub>, both of which can be formed from radiolysis, or more commonly from interaction with the OH-radical (·OH) formed from the radiolysis of water [28].

For gaseous reactions, again radiolytic reactions will be the main avenue of formation, as thermal reactions are not favored in the expected conditions [29]. However, the presence of oxygen and/or water reduces the yields of methyl iodide, due to radical scavenging, especially by oxygen. In a realistic accident scenario, both of these will be abundant. Again, the reaction principle remains the same as in the aqueous phase, involving radicals of iodine and methane. Furthermore, methyl iodide may decompose by exposure to light, or thermally [30].

The effort put to studying organic iodides as well as the studies done for organic telluride formation were used as references in this work. The aim was to investigate the interactions between tellurium and organic material in the gas phase. The possible formation of organic tellurides and other reactions increasing tellurium transport in containment conditions were considered.

### 3. Methodology

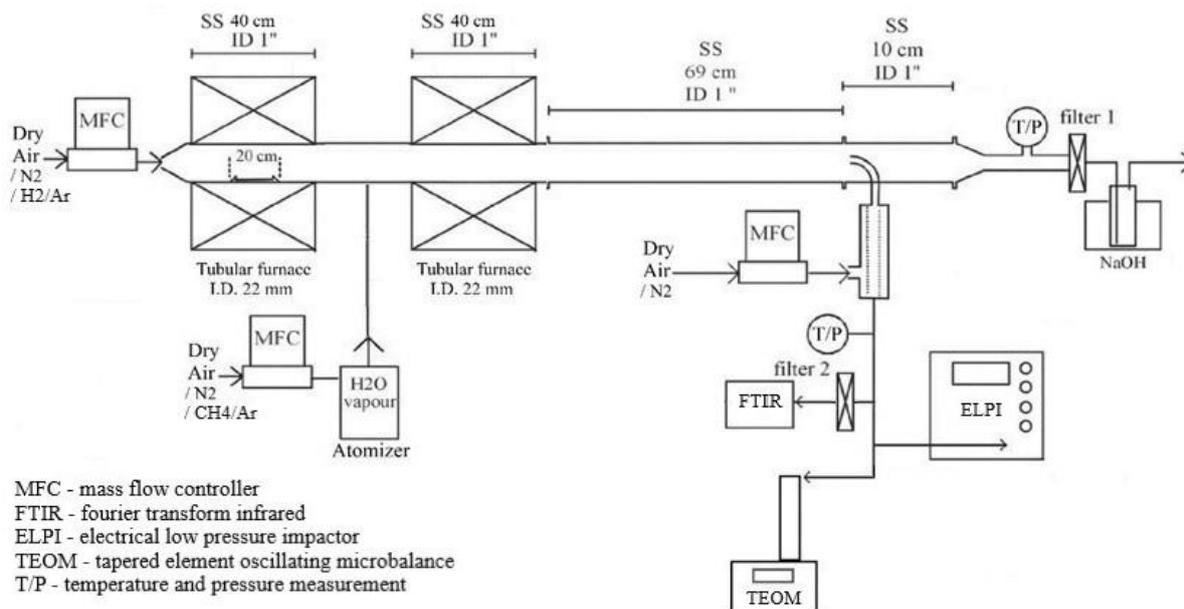
This project was a collaboration between VTT Research Center in Finland, Chalmers University of Technology in Sweden, and the University of Oslo, Norway. The experiments were carried out at VTT, using their aerosol laboratory and the analysis of the results was conducted at VTT, Chalmers, and at the Nuclear Research Institute in Rez, Czech Republic.

#### 3.1. Experimental setup for the tellurium reaction/transport

The experiments were carried out in a tubular flow furnace (Entech/Vecstar, VCTF 4), with a tube made of stainless steel (AISI 316L). One gram (1g) of tellurium metal (Te, Sigma-Aldrich, purity  $\geq 99.997\%$ ) was added to a crucible and placed in the middle of the heating section. During the experiment, this part of the oven was set to  $600^{\circ}\text{C}$ , for the volatilization of the tellurium. At this relatively low temperature compared to the boiling point of tellurium, the volatilization is slow and continuous throughout the experiment.

The furnace tube was connected to a mass-flow control unit (Brooks S5851, Brooks® Instrument) set to  $5\text{ dm}^3/\text{min}$  of the gas mixture used for the experiment. The mixture of Tellurium aerosols and carrier gas then passed a connective junction, where the organic precursor, acetone or 2-propanol and water both entered the system as droplets from an atomizer. This mixture then entered a second furnace with the same type of tube held at  $300^{\circ}\text{C}$ . This is where any chemical reactions took place. Beyond the reaction furnace a part of the stream was diverted through a filter (MilliPore, Mitex™ PTFE, pore size  $5\text{ }\mu\text{m}$ ) to collect the formed aerosols. The filtered stream then passed through  $100\text{ ml } 0.1\text{ M NaOH}$ , as to trap any gaseous species. The rest of the flow was quenched with  $\text{N}_2$  gas and proceeded to be further diverted to several on-line measurements. These were a differential mobility analyzer (DMA), a condensation particle counter (CPC) and finally an electric low-pressure impactor (ELPI). Together, these instruments can count particles in the size range of  $1\text{ nm}$  to  $10\text{ }\mu\text{m}$ . The total mass-concentration of particles in the gas stream was monitored (online) with tapered element oscillating microbalance (TEOM). Fourier-transform infrared spectroscopy (FTIR) was used to determine speciation and concentration of any gaseous reaction products.

Finally, a second identical filter was placed at the end of the analysis line, before the remaining gas flow was sent to the exhaust line. The entire setup can be seen schematically in Figure 1.



**Figure 1:** Schematic representation of the experimental setup at VTT, including the online measurement systems.

In total, eight experiments were conducted, as detailed in the experimental matrix seen in Table 1. The experimental conditions varied the atmospheric composition, and the organic precursor being used. All experiments were performed in singlets.

**Table 1:** Experimental matrix

Number	Precursor	Temperature, C	Atmosphere	Organic precursor
1	Te	540	Air	-
	Te	540	Air	Acetone
	Te	540	Air	1-Propanol
2	Te	540	N <sub>2</sub>	-
	Te	540	N <sub>2</sub>	Acetone
	Te	540	N <sub>2</sub>	1-Propanol
3	Te	540	H <sub>2</sub> /Ar	-
	Te	540	H <sub>2</sub> /Ar	Acetone
	Te	540	H <sub>2</sub> /Ar	1-Propanol
4	Te	540	H <sub>2</sub> /Ar	Methane
4.1	Te	540	N <sub>2</sub>	Methane

## 3.2. Analytical methods

### 3.2.1. Tellurium release

The total tellurium release was determined by weighing the sample crucible before and after the experiment.

### 3.2.2. ICP-MS

The concentration of tellurium in the sodium hydroxide trap solutions was measured with High Resolution Inductively Coupled Plasma Mass Spectrometry (HR ICP-MS, Element 2, ThermoScientific).

The detection limit for tellurium ( $\text{Te}^{126}$ ) was 0.006 ppb. The samples were prepared by diluting with 0.5 M  $\text{HNO}_3$  (Suprapure) to a suitable concentration.

### 3.2.3. ELPI and TEOM

ELPI is a method that can be used to determine the sizes of aerosols. The gas-flow enters the topmost part of the unit and are electrically charged with a corona charger. The particles are then sent through a series of twelve “impactor sieves” with progressively smaller diameters. The concentration of particles on every stage is measured by the impact of the charged particles, and the resulting electrical current is transformed into a mass concentration. The measurement range on the equipment was between 7 nm and 10  $\mu\text{m}$ , with an uncertainty of  $\pm 10\%$ . The ELPI was controlled by the software ELPIVI version 4.0 (Dekati Ltd.). TEOM was used to measure the total mass concentration in the gas stream. Note that neither of these methods, in principle, differentiates between the composition of different aerosols. The only thing these measurements can determine is the mass concentration of particulate matter, not the composition or species of that particulate matter.

### 3.2.4. SEM-EDX

SEM-EDX is a combined method for investigating the morphology, size and composition of the aerosol particles collected on the filters located before the liquid trap (almost immediately after the exit from the reaction furnace) and at the end of the sampling line. The SEM instrument used was a Zeiss Crossbeam 540 operating at 2.0 kV with a probe current of about 100 pA. EDX spectroscopy was performed using 8.0 keV electrons and  $\sim 80 - 100$  pA probe current. Prior to analysis, a piece (1 cm x 1 cm) of each studied filter substrate was attached on aluminum pin stub using double sided adhesive carbon tape. The samples were coated platinum in a sputter coater to improve conductivity of the samples.

### 3.2.5. INAA- Gamma Spectroscopy

Instrumental Neutron Activation Analysis (INAA) is the process wherein a sample is introduced in a neutron field, inducing nuclear reactions. Depending on the sample constituents and the particular neutron field, one can estimate the nuclear reaction products. These products will often be radioactive, meaning they can be analyzed using gamma spectroscopy.

The Neutron Activation and subsequent analysis was performed at the Nuclear Research Institute in Řež, Czech Republic, using their experimental reactor, LVR-15. A small piece of the filters from the furnace lines were cut and saved for future XPS analysis, and the rest of the filters were sent to Řež where they were folded and encapsulated in polyethylene. The polyethylene was folded into aluminium foil and all the samples were stacked inside a pair of cylindrical irradiation capsules, also made of aluminium. The assembly is depicted in Figure 2.



**Figure 2:** Package for neutron irradiation. The filter sample is folded and packaged into the polyethylene, folded into aluminum foil, then stacked into the smaller cylinder. The small cylinder is then loaded into the large one, and the package is sent via a pneumatic delivery system into the reactor.

Alongside the samples, standards, and a blank filter there were, in total, six iron flux monitors spaced out along the capsule. They allow for a measure the neutron flux experienced by different parts of the assembly, as the flux is never completely homogenous. 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. After irradiation, the samples were stored for seven days to allow the shorter-lived fission products to decay.

Gamma spectroscopy was performed using the Canberra detector, relative efficiency 77.8 %, FWHM resolution 1.87 keV, both for the 1332.5 keV gamma-rays of  $^{60}\text{Co}$  with two different counting geometries. Most samples were measured 20 cm above the detector. However, three samples proved highly active and had to be measured 40 cm above the detector, which was the maximum height possible. The samples using the 20 cm geometry were measured for one hour (live time), and most of these samples had a dead time below 30%. The highly active samples that required the 40 cm geometry still had a high dead time, exceeding 40%. They were measured for 2 hours (live-time). Finally, three fresh, unused filters were weighted with a bench scale (capable of measuring down to 0.001 mg) three times each. The mass of tellurium on each filter ( $\mu\text{g}$  tellurium) could be calculated according to equation 1

$$m_{\text{un}} = \frac{N_{\text{un}} m_{\text{st}}}{N_{\text{st}}} \quad (1)$$

Where  $m_{\text{un}}$  is element mass in the sample,  $m_{\text{st}}$  is element mass in standard,  $N_{\text{un}}$  is number of counts corrected for the relative neutron flux and decay time in a sample,  $N_{\text{st}}$  is number of counts corrected for the relative neutron flux and decay time in a standard. Upon dividing with the averaged filter mass, the tellurium “concentration” [mass tellurium per mass filter] is obtained.

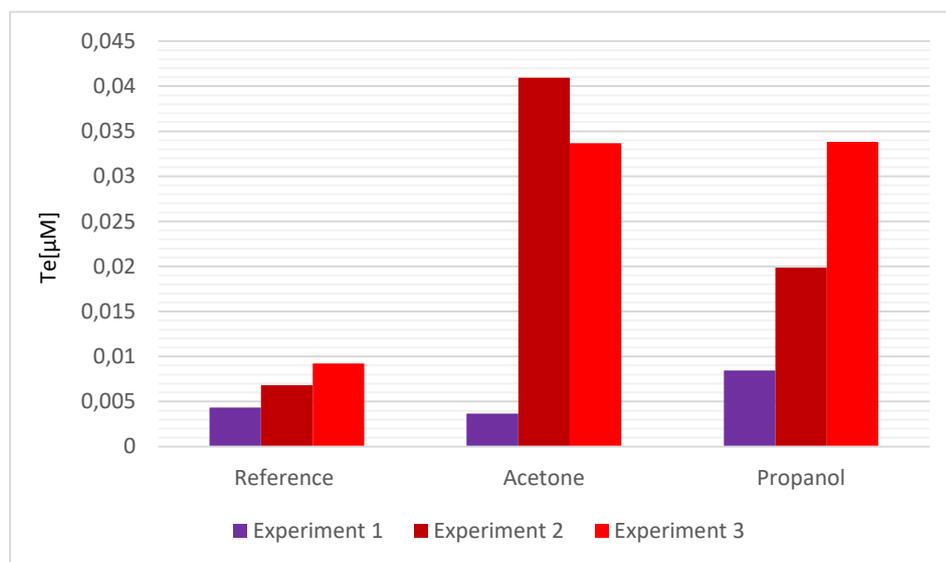
The standards were prepared by dissolving 24.7 mg Te (99.9 % Te. Merck) in 5 mL of warm 3 M  $\text{HNO}_3$  and diluted up to 10 mL in a volumetric flask. From this solution, 50  $\mu\text{L}$  aliquots were deposited with

a gravimetrically calibrated micropipette onto 16-mm diameter disks of chromatographic paper (Whatman 1), then gently dried and packed into the 25-mm diameter polyethylene irradiation capsules. The Te calibrators contained  $124.9 \pm 0.4 \mu\text{g}$  of Te ( $x \pm s$ , N=5)

## 4. Results/Discussion

### 4.1. Tellurium in the gaseous fraction

The results from the ICP measurements of the liquid traps from Experiments 1, 2 and 3, where acetone and propanol were used as the organic precursors, are presented in Figure 3.



**Figure 3:** The relative amounts of tellurium present in the liquid traps, arranged by different organic precursors. “Experiment 1” indicates an experiment conducted in air atmosphere, “Experiment 2” was conducted in inert (nitrogen) atmosphere, and experiment 3 was conducted in reducing atmosphere (hydrogen gas in argon).

The liquid traps were preceded by a filter with the pore size of  $5 \mu\text{m}$ , and that part of the system was significantly cooler than inside the furnace. As such, it is highly unlikely that very many aerosol particles would make it to the liquid traps. This can be inferred with some clarity from the reference experiment. As the reference experiment did not use any organic precursor, and tellurium does not form long-lasting gaseous species at these temperatures, the only likely transport mechanism in the reference experiment is the transport by small aerosols, which also gives only a small amount of tellurium in the trap, no matter the atmosphere. The increase in Experiment 3 (reducing atmosphere) compared to the other two conditions could possibly be related to the formation of  $\text{H}_2\text{Te}$ , which can be formed at elevated temperatures. This species, however, is not stable as the temperatures go down [22].

As for the experiments using an organic precursor, there is a significant increase in the tellurium concentration for both the reducing and inert atmospheres, whereas the oxidizing atmosphere retains a thoroughly low tellurium concentration in the trap. An explanation for the low tellurium concentration in oxidizing conditions may be that the elemental tellurium rapidly oxidizes to tellurium dioxide. Interaction between tellurium dioxide and organic precursors to form organic tellurides is an unprecedented phenomenon.

As for the sharp increase in tellurium concentration measured in the inert and reducing atmospheres in the presence of organic precursor, an implication is that the organic precursors enable the

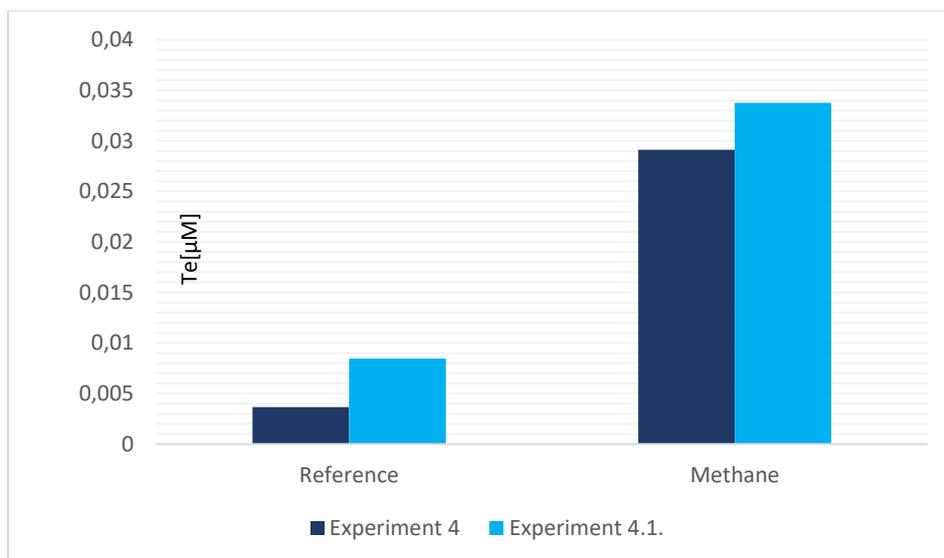
formation of a tellurium species capable of penetrating the filter. A possibility for such a species is some form of organic telluride.

Since only singlets were performed, uncertainty analysis is not possible, but the result nonetheless seems to indicate that under reducing atmosphere, the nature of the organic compound does not seem to matter to a large extent, as evidenced by the concentrations of tellurium being the same in both the experiments conducted. A potential explanation could be that acetone undergoes a reduction to 2-propanol. This species would have a similar behavior as 1-propanol, explaining why the concentration of tellurium is virtually identical for both the organic species under reducing atmosphere. However, reduction of a ketone by solely  $H_2$  is generally not applicable [31]. However, at such high temperatures, and in the presence of tellurium (or even potentially the steel tube, containing trace amounts of nickel), it cannot be dismissed with absolute certainty. Another pathway for formation of organic tellurides in this system could be the pyrolysis of the organic precursors which enables radical reactions. For acetone, however, decomposition does not seem to involve radicals [32], making this idea unlikely since radical reactions is the primary way to form organic tellurides. As for 1-propanol, for the temperatures of 570-621°C, the end products formed by pyrolysis seem to be methane and acetaldehyde and a polymeric material [33]. The proposed reaction mechanism, however, does include several radical species as intermediaries, including  $CH_3-CH_2\cdot$ ,  $\cdot CH_2-OH$  and the hydrogen radical [33].

This process is reported at a higher temperature than was used for this study. However, the ingress of the organic precursor can be expected to be above 500°C, as the organic precursor was introduced into the system just after the furnace used to volatilize tellurium, held at 540°C.

Under inert atmosphere, addition of acetone to the feed results in the highest concentration of tellurium in the liquid trap, about twice as much as the same atmosphere with 1-propanol in the feed. This indicates that while both organic precursors seem to interact with tellurium and increase the transport to the liquid trap, acetone seem to do so faster, or form a compound more stable under these conditions.

Furthermore, two experiments were conducted using methane as the organic precursor, studying its influence on the system in both reducing and inert atmosphere. The results from these experiments can be seen in Figure 4.



**Figure 4:** The relative amounts of tellurium present in the liquid traps after methane was used as the organic precursor. “Experiment 4” indicates an experiment conducted in a reducing atmosphere (hydrogen gas in argon) and “Experiment 4.1” was conducted in an inert (nitrogen) atmosphere.

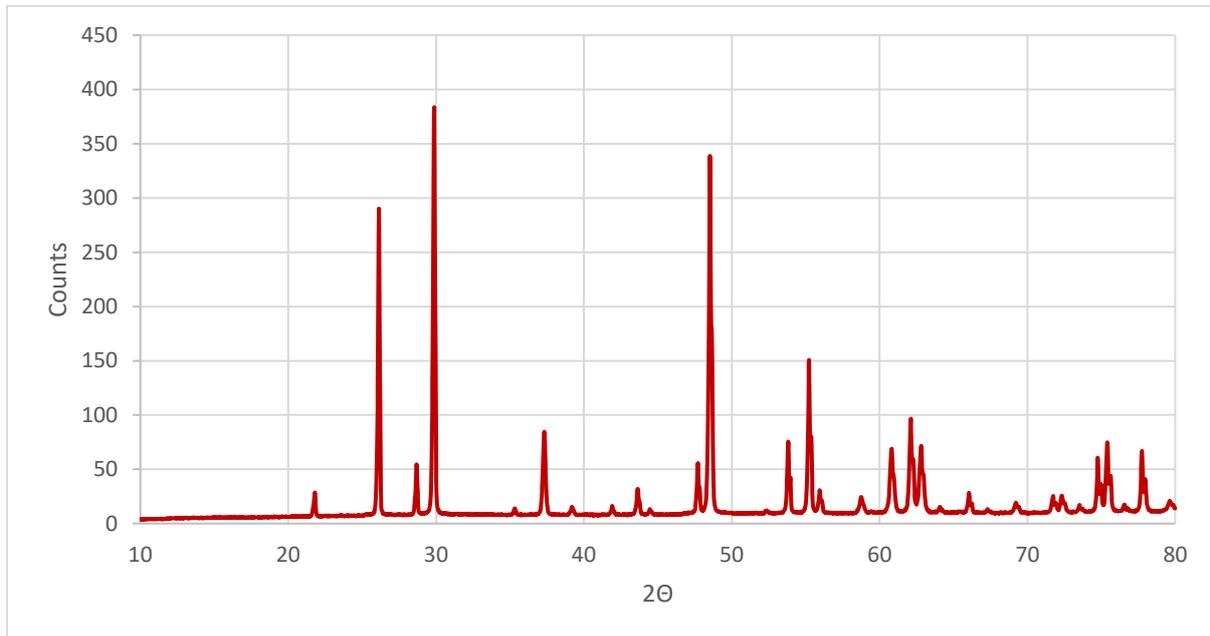
Similar to the experiments with liquid organic precursors, the addition of methane to the feed also marks a significant increase in tellurium concentration in the liquid trap. Again, as no speciation was possible during the experiment, nor has the contents of the liquid traps been studied with a species-sensitive technique. It is thus not possible to say what species, if any, are formed that explains this behavior. However, a gas-phase species would be able to penetrate the filter and end up in a liquid trap, as could very small aerosols. Most likely the reason for the tellurium content in the reference experiment is due to tiny aerosols penetrating the filter and staying airborne long enough to flow through the piping and ending up in the liquid trap.

Pyrolysis of  $\text{CH}_4$  results in solid carbon and  $\text{H}_2$ , and there are some instances where liquid and vapor tellurium have been used to catalyze the pyrolysis [34]. This experiment was, however, conducted at a higher temperature of 1100-1250 K where methane was bubbled through a column of liquid tellurium. With a residence time of 12 s, a 30% pyrolysis rate was achieved. In the present case, both the tellurium and the methane will be diluted by the carrier gases, the temperatures are much lower, and the residence time at the elevated temperature is also less than 12 s. However, the pyrolysis forming carbon and  $\text{H}_2$  might explain the similarity between experiment 4 and 4.1. Experiment 4 uses 5%  $\text{H}_2$  in argon, and experiment 4.1 uses only  $\text{N}_2$ . If the pyrolysis forms  $\text{H}_2$ , then these conditions become very similar, which is reflected in the similar concentrations of tellurium in the liquid traps.

## 4.2. Precursor Analysis

The tellurium precursors left in the crucible were analyzed with XRD. In Experiment 1, where air was used as the carrier gas, there was enough precursor left for a reliable analysis. The precursor was found to have oxidized from metallic tellurium to tellurium dioxide during the experiment. The diffractogram for the precursor from Experiment 1 is presented in Figure 5. No signals were observed for elemental tellurium, only  $\text{TeO}_2$ .  $\text{TeO}_2$  has a significantly higher melting and boiling point than metallic tellurium, at 1245 °C. This may explain why there was more precursor material left after Experiment 1 than after the experiments performed in inert or reducing atmosphere. This oxidation and subsequent retardation of the precursor in the crucible may also explain the gaseous results where no

significant increase of tellurium was observed in the liquid traps for the experiments conducted in air atmosphere.



**Figure 5:** Diffractogram for the precursor from Experiment 1 (air atmosphere). All signals attributed to tellurium dioxide.

### 4.3. Filter weights

Every experiment made use of two filters. These were weighted before and after each experiment in order to determine the mass of tellurium collected on the filter. The weight change of the filters was relatively consistent across all experiment. In general, the filter increased their weight by 0.01-0.015 g after each experiment. This was measured with an analytical scale, and so there are likely significant uncertainty to these numbers.

The total weight of the precursor in each experiment was 1 g of tellurium, that means only a small fraction of the material reaches the filter, and by the Figures 3 and 4, only a minute amount of tellurium can penetrate the filter. This means then that most of the tellurium ends up as aerosols in the various measuring techniques running parallel to the liquid trap or, most likely, that it deposits in the piping as the temperature goes down.

There is some evidence of the latter happening, as during some of these experiments, needle-like crystals could be seen in the connection between the first furnace and the second, about where the organic tellurides were fed. These crystals can be seen in Figure 6.

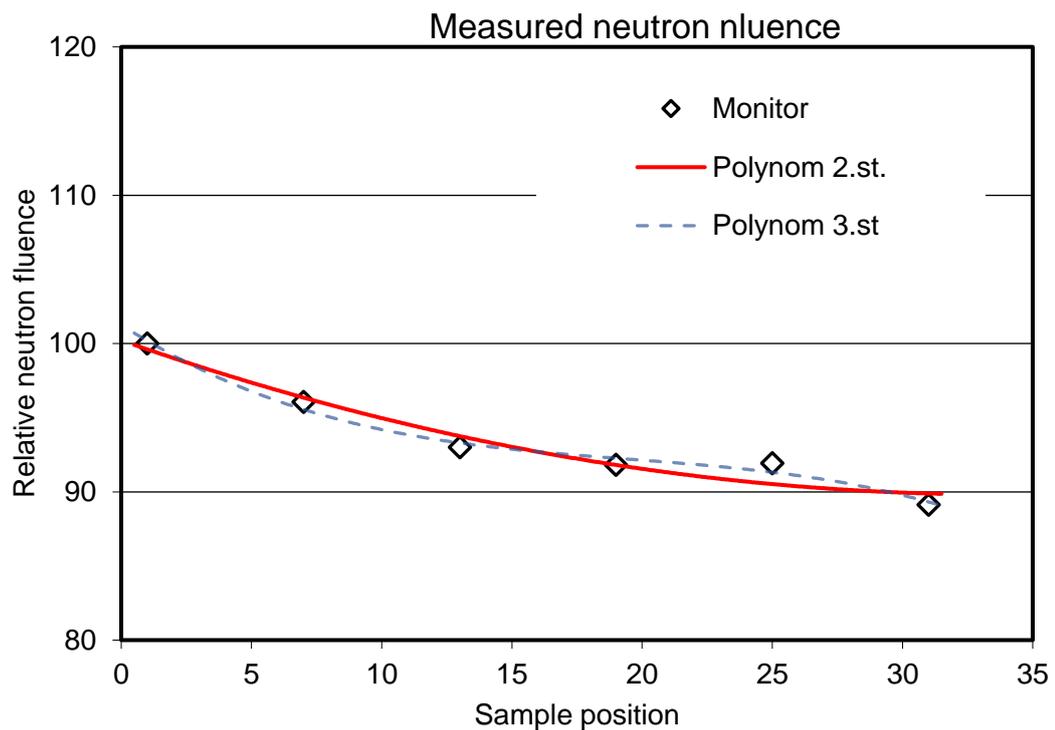


**Figure 6:** View of the crystals formed by the tellurium inside the junction connecting the two furnaces.

#### 4.4. Neutron Activation Analysis

In total, twenty-two samples were measured, alongside six iron monitors, two standards and one blank filter, giving a total of 31 measurements for the INAA analysis.

The relative neutron fluence distribution across the whole assembly of 31 samples is presented in Figure 7.



**Figure 7:** Neutron fluence related to sample position across the capsule. The diamonds indicate the iron monitors distributed evenly across the capsule. The calculation is based on the gamma energies 1291keV and 1099 keV for the nuclide  $Fe^{59}$ .

The drop in neutron flux is significant across the capsule, as the bottommost part of the capsule only experiences roughly 90% of the flux compared to the very top of the capsule. The reduction in flux can roughly be described with a second degree- polynomial, as is seen with the red line.

The mass  $^{131}\text{I}$  per filter calculated from the neutron activation analysis is presented in Table 2. Measurement of this radionuclide makes for a sensitive method of determining Te via INAA [35]. Since every experimental condition had two filters, only the ones closest to the furnace outlet, the standards, and the blank filter are presented here.

**Table 2:** Results of the neutron activation analysis. The concentration is calculated based on the number of counts for the  $^{131}\text{I}$  gamma-line at 364.5 keV.

Experiment name	Sample [precursor, atmosphere]	Measurement Geometry [Cm]	Conc. $^{131}\text{I}$ [ $\mu\text{g/g}$ filter]	Combined uncertainty [ $\mu\text{g/g}$ filter; 1 std. dev.]	Rel. Com. Uncertainty [%]
-	Blank Filter	20	4.62	0.135	0.73%
-	Standard-1	20	126	0.906	0.72%
-	Standard-2	20	124	0.905	0.73%
1	Reference, Air	20	23571	138.669	0.59%
1	Acetone, Air	20	77	1.035	1.35%
1	Propanol, Air	20	80	0.739	0.93%
2	Reference, $\text{N}_2$	45	181000	1294.439	0.71%
2	Acetone, $\text{N}_2$	20	2213	13.139	0.59%
2	Propanol, $\text{N}_2$	20	170	1.249	0.74%
3	Reference, $\text{H}_2/\text{Ar}$	45	124000	882.481	0.71%
3	Acetone, $\text{H}_2/\text{Ar}$	20	10849	63.825	0.59%
3	Propanol, $\text{H}_2/\text{Ar}$	20	5898	34.702	0.59%
4	$\text{CH}_4$ , $\text{H}_2/\text{Ar}$	45	127000	903.908	0.71%
4.1	$\text{CH}_4$ , $\text{N}_2$	20	3025	17.958	0.59%

Clearly, there is a significant variance to the iodine, and by extension, the tellurium concentration on the different filters, with the highest concentrations being more than a thousand times more concentrated than the lowest.

In general, the reference cases always have a high concentration of  $^{131}\text{I}$ , throughout displaying the highest concentration of iodine on the filter for their respective conditions. The reference cases often having a high  $^{131}\text{I}$  concentration is congruent with the ICP-MS analysis, where the reference always had the lowest concentration of tellurium in the liquid trap. This is logical if one considers that the filter is located before the trap in the experimental setup. If the filters catch a lot of tellurium, that means less can end up in the liquid trap.

Experiment 1, using the air atmosphere, gave overall low  $^{131}\text{I}$  concentrations, presumably because tellurium oxidizes to  $\text{TeO}_2$ , which has a much-reduced mobility and thus does not make it to the filters.

Of the organic precursors, propanol seems to give a low concentration of  $^{131}\text{I}$ . In all but the experiment conducted in air, it shows a significantly lower concentration of  $^{131}\text{I}$ , and in for that experiment both organic precursors gave a very low and very similar concentration. A tentative explanation is that the pyrolysis of propanol does form species which can react further with tellurium and give volatile compounds. However, this explanation is not entirely satisfactory, as it is not confirmed by the liquid trap measurements. In inert conditions, the acetone gives significantly higher concentration of tellurium in the liquid traps than propanol, and for reducing conditions the liquid trap concentrations are practically the same.

The uncertainty of the measurements seems relatively stable with only one experiment having higher than 1% uncertainty. The background and mathematical detail behind the uncertainty calculation for an INAA procedure is a complex matter that must, in some sense, be determined for the individual lab doing the measurement. In principle, there are three overarching sources of uncertainty present: uncertainties related to the sample preparation, uncertainties related to the irradiation process, and uncertainties related to the gamma spectroscopy measurement [36]. The details and magnitude of these uncertainties, as well as the detailed calculations performed for the uncertainty calculation have been published elsewhere and are not elaborated upon here [36]. The relative uncertainty is taken to be 0.5% for all these calculations.

The individual spectrum for each sample has been analyzed, and several nuclides have been found to be common in most of them. The most frequently observed nuclides, their half-lives, associated nuclear reaction and the most important gamma-ray energies are presented in table 3.

**Table 3:** Nuclides frequently seen in the samples.  $^{131}\text{I}$  and the tellurium isotopes are natural constituents from a sample containing natural tellurium, whereas  $^{134}\text{Cs}$  and  $^{192}\text{Ir}$  would be contaminations.

Nuclide	Half-life	Reaction for formation	Selected gamma lines [keV]
$^{131}\text{I}$	8.03 d	$^{130}\text{Te} (n,\gamma) ^{131}\text{Te} \rightarrow ^{131}\text{I}$	364.48, 636.97
$^{131\text{m}}\text{Te}$	1.25 d	$^{130}\text{Te} (n,\gamma) ^{131\text{m}}\text{Te}$	334.27, 773.68, 852.24, 1206.59
$^{129\text{m}}\text{Te}$	33.6 d	$^{128}\text{Te} (n,\gamma) ^{129\text{m}}\text{Te}$	556.56, 695.84
$^{121}\text{Te}$	16.8 d	$^{120}\text{Te} (n,\gamma) ^{121}\text{Te}$	507.59, 573.13
$^{134}\text{Cs}$	2.062 a	$^{133}\text{Cs} (n,\gamma) ^{134}\text{Cs}$	569.31, 604.71, 795.87
$^{192}\text{Ir}$	73.83 d	$^{191}\text{Ir} (n,\gamma) ^{192}\text{Ir}$	295.96, 308.45, 316.51, 468.07
$^{194}\text{Ir}$	19.15 h	$^{193}\text{Ir} (n,\gamma) ^{194}\text{Ir}$	328.46

All samples contained  $^{131}\text{I}$ , which is to be expected since it is the nuclide used to calculate the mass of tellurium on the filter, and the filters were coated with natural tellurium, making the formation of  $^{131}\text{I}$  likely. It was the only nuclide to be present in every measured filter. The three tellurium isotopes are likewise expected, as they are formed directly from the irradiation of natural tellurium, and their half-lives are long enough that they have not yet decayed seven days after the irradiation, when the measurement is performed.

The cesium and iridium contents are unexpected. Both of the measured isotopes are formed from nuclides found in natural cesium and iridium, however the presence of either iridium or cesium is surprising as neither for them have been used at any point during this project, nor have any of the other analytical methods revealed their presence.

Cesium is a well-known volatile fission product and previous studies at VTT has involved it, in the form CsI. Its inclusion in these samples could be a contaminant from the steel lining and/or furnaces used, or potentially the batch of tellurium powder used. However, this theory cannot be verified. Cesium only has one stable isotope ( $^{133}\text{Cs}$ ), so seeing only  $^{134}\text{Cs}$  is sensible.

Iridium is not an important fission product, so there is no obvious reason for it to appear as a contaminant. Natural iridium consists of two isotopes:  $^{191}\text{Ir}$  and  $^{193}\text{Ir}$  partitioned in a ratio of 36.3% and 62.7%, respectively. In the measured samples,  $^{192}\text{Ir}$  is the dominant isotope due to the relatively short

half-life of  $^{194}\text{Ir}$  of 19.28 hours, meaning that about 8.7 half-lives have passed between the irradiation and the measurement. However,  $^{194}\text{Ir}$  is faintly observed.

#### 4.5. Filter appearances and SEM

Their appearance of the filters changes significantly after each experiment. They can be divided in four groups with four different colors: black, white, gray, and light brown. Examples of the colors can be seen in the Figure 8.



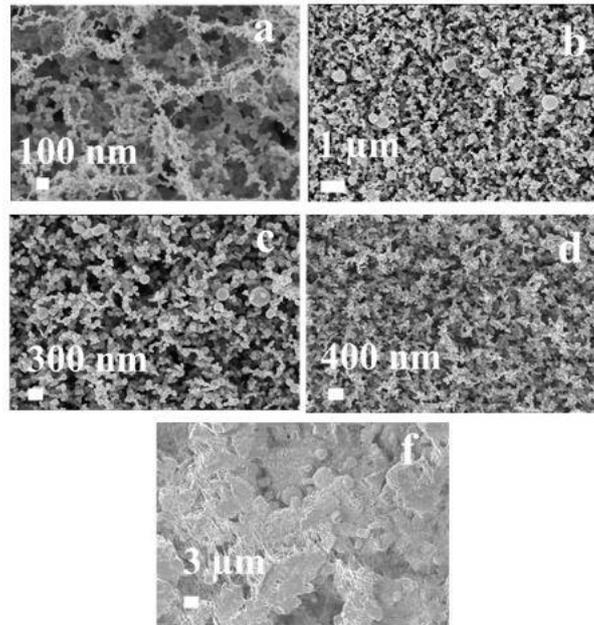
**Figure 8:** The three principal types of filter deposition from the different experiments.

The white filters nearly all came from Experiment 1, using air as the atmosphere and acetone as the precursor. The white color can thus be inferred to be  $\text{TeO}_2$ . It has a white coloration and tendency to be formed when tellurium is heated in an oxygenated atmosphere. There is also one white filter from Experiment 2 (the secondary filter), which is odd, since no oxygen should be present, aside from the small amount added with the organic precursor. That amount is should not be enough to oxidize all the tellurium, and furthermore, no such thing is seen with any of the other filters in Experiment 2.

That same experiment is also responsible for the one brown filter. One possibility is that the brown color occurs due to a mix of black metallic tellurium, and white tellurium oxide. However, this still does not explain the apparent oxygen in the system. The most likely scenario seems to be a leak in the system, though this cannot be verified.

The black filters were the most common, and were seen in Experiments 2, 3, 4 and 4.1. These experiments all took place in reducing or inert atmosphere, meaning the black color is from metallic tellurium. The gray filters were seen only a few times. All of them were secondary filters, placed a significant distance from the furnace. The gray color is thought to be due to a lesser coverage of the filter surface of the tellurium, however, this will be investigated with XPS (X-ray photoelectron spectroscopy).

The filter surfaces were investigated using SEM-EDX, and a typical image from the different filters is presented in Figure 9.

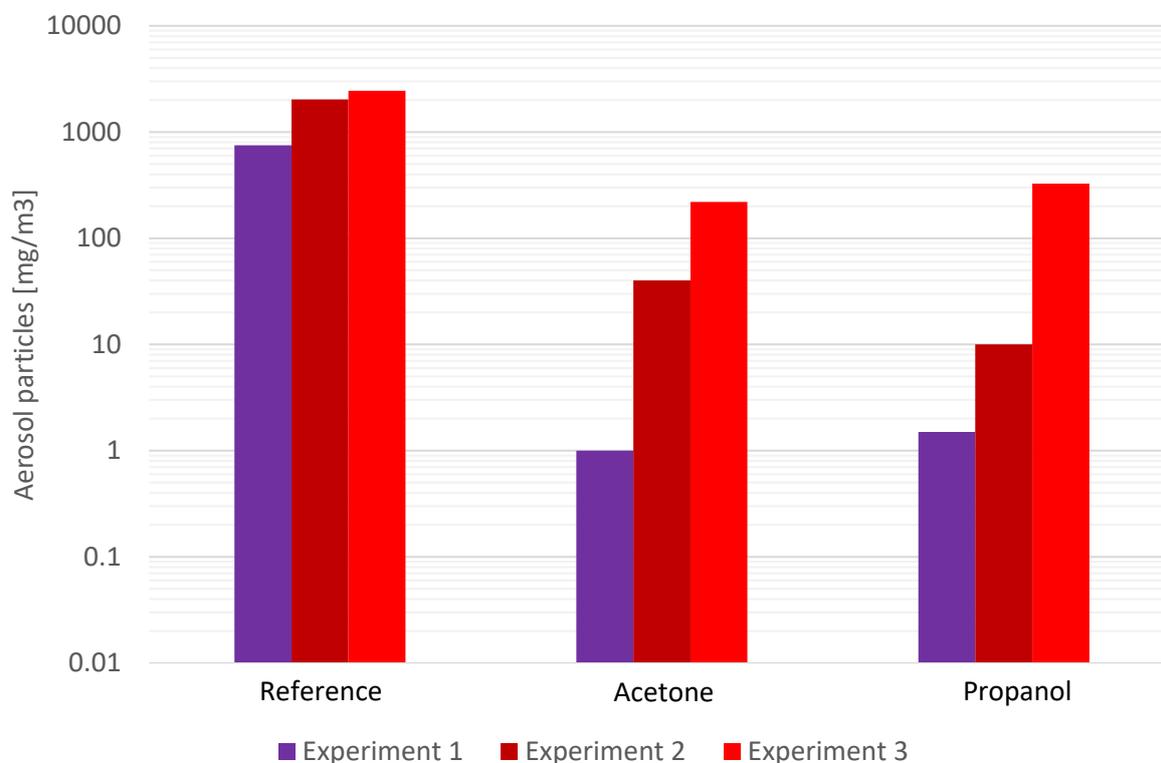


**Figure 9:** SEM images of the particles observed in each experiment. a: Experiment 1(filter 1), b: Experiment 2 (filter 2), c: Experiment 3 (filter 1), d: Experiment 4 (filter1). In f is shown filter material surface without particle collection. Filter 1 and filter 2 indicate sampling location of the sample as given in fig. 1. Scale bars are shown in each image.

The filters a-d all differ from filter f, which is the unused filter, displaying varying levels of particulate matter on the surfaces. As for the nature of the particles, they seem rather similar across the four investigated filters, consisting of spherical, particulate matter: potentially aerosols of tellurium. However, since the scale is different in all the micrographs, there is some uncertainty involved.

#### 4.6. Mass concentrations in the carrier gas

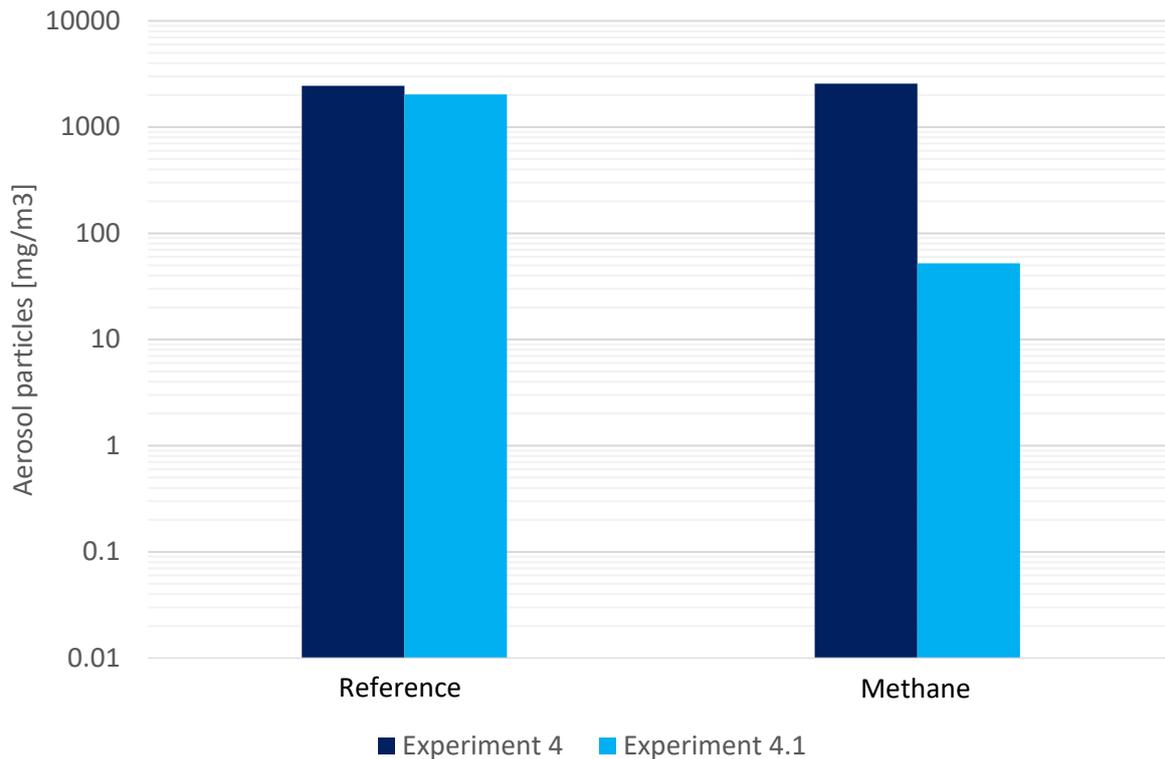
The mass concentration was measured online during the experiments with TEOM, and the results for the first three experiments are presented in Figure 10.



**Figure 10:** The impact of different organic precursors on the total mass concentration in the carrier gas, compared to a reference case without any organic species present. “Experiment 1” indicates an experiment conducted in air atmosphere, “Experiment 2” was conducted in inert (nitrogen) atmosphere, and experiment 3 was conducted in reducing atmosphere (5% hydrogen gas in argon).

From a mass perspective, it appears that the organic precursors indeed do have an impact on the aerosols, as their combined mass seems to decrease, no matter the atmospheric conditions, if there is an organic species present. This is especially clear in the case of an oxidizing atmosphere (experiment 1 in Figure 10), where the mass concentration decreases with a factor of almost one thousand upon addition of an organic component. The other two atmospheres also displayed a significant decrease in aerosol concentration, with the inert conditions giving 20-100 times less mass concentration upon addition of an organic component compared to the reference case, and the reducing atmosphere giving about ten times decrease.

Similar measurements were conducted for the experiments involving methane, using inert and reducing conditions. The results are graphically presented in Figure 11.



**Figure 11:** The impact of methane on the total mass concentration in the gas phase, compared to a reference case without methane. “Experiment 4” indicates an experiment conducted in reducing atmosphere (5% hydrogen gas in argon), and experiment 4.1 was conducted in inert (nitrogen) atmosphere.

The reducing atmosphere does not seem to have any impact on the aerosol mass concentration, given that the mass concentration remains virtually unchanged from the reference case. The inert atmosphere, however, do show a decrease in aerosol concentration, to the tune of some 20-40 times reduction. In both these measurements, the total volume of carrier gas used was comparable, meaning that the absolute mass of transported aerosols will look similar to the concentrations presented in the Figures 10 and 11.

TEOM measures the total mass of the aerosols in the gas stream, rather than their amount. For all of experiment 1 and the experiment using propanol in experiment 2, the filters indicate that the aerosols were mostly  $\text{TeO}_2$ , which weighs about 25% more than metallic tellurium. This means that the number of aerosols implied by “Experiment 1” columns in Figure 10 is about 20% higher than indicated.

The reason for the decrease in mass concentration upon the addition of organics is uncertain. Part of the explanation may be that the addition of organic precursors and water further dilutes the gas thus reducing the concentration slightly. Since the exact amount of gas for individual measurements could not be measured with this setup, the magnitude of this issue is uncertain. Also, since water was added together with the organic precursor, condensation of water droplets on the inside of the tubing as the system cools may also serve to trap tellurium particles and decrease the mass concentration when organic precursors are added to the system.

A similar experiment is described in [37], using the same equipment at the same site for the measurement of tellurium aerosols. The authors clearly see that the mass concentration of Tellurium decreases with time during the TEOM measurement. Especially for the experiments conducted in oxidizing atmosphere, the drop in tellurium concentration is clear. This means that the average

tellurium mass concentration heavily depends on the measurement time; especially for the experiments involving oxidizing conditions.

Overall, the mass concentration measurement clearly points to the decrease of a mass concentration upon the addition of organics, which could be the result of organic telluride formation. However, this is contradicted by the ICP-measurements of the liquid traps. If organic tellurides were formed to a large extent, then the concentration in the liquid traps ought to be high when the mass concentrations were low, which is not the case. For instance, there is a decrease in mass concentration for experiment 1 (purple in Figure 10), but the corresponding ICP measurements show a thoroughly low amount of tellurium in the liquid trap. Similarly, for experiment 2 the lowest mass concentration does not correspond to the highest ICP concentration.

There appears to be some other mechanism, aside from organic telluride formation, responsible for the decrease of the mass concentration upon the addition of organics, such as the dilution of the gas, or the retention of tellurium due to the condensed steam.

## 5. Conclusions

Tellurium is known to be a volatile fission product in nuclear installations, and its potential for forming organic tellurides with an even greater volatility has been speculated. However, the question of organic tellurides in the context of nuclear technology is still debated, though there is some evidence pointing towards their formation [11]. In this study, the presence of organic tellurides forming in the gas-phase without the influence of radiation cannot be confirmed with certainty, but the presence of organics in the gas phase alongside the tellurium can increase the volatility of the element. The ICP-MS measurements of the liquid traps show an increased concentration of tellurium reaching the traps in the presence of organic precursors for both the reducing and the inert atmosphere. However, in oxidizing atmosphere the organic precursors do not increase the concentration, presumably due to the rapid oxidation of the tellurium into  $\text{TeO}_2$ , which requires higher temperatures to volatilize.

The TEOM-ELPI measurements also support the idea of oxidation of tellurium taking place in an oxidizing atmosphere, as the mass concentration of tellurium in the gas phase is significantly lower for the oxidizing conditions than for reducing and/or inert conditions. However, in general, the mass concentrations decrease upon addition of organic precursors to the system. This could be due to increased deposition of tellurium aerosols in the system after the addition of steam with the organic precursors, or partial oxidation decreasing the release.

The INAA measurements reveal, as expected that the main constituents of the filter samples were tellurium, as well as possible contaminants of cesium and iridium. However, these contaminants likely did not impact the results to any large degree as neither of those elements reacts to any large degree with organic species or tellurium under these conditions.

The mass of tellurium determined on the filters, based on the gamma spectroscopy varies strongly across the 22 measurements conducted. The reference cases always show the highest tellurium content of the respective experiment group. This seems to indicate that the addition of organics prevents tellurium from reaching the filter. There are several possible explanations for this. One can imagine that it is because of the formation of organic tellurides, which should be able to penetrate the filters. This would also explain the increase in tellurium concentration seen in the liquid trap upon the addition of organics. Another possibility that must be considered is that the addition of water compared to the reference case also reduces the amount of tellurium that reaches the filter by condensing and trapping the aerosols on the tube interior. This is potentially supported by the TEOM results. However, it does not account for the increase of tellurium in the liquid trap. Furthermore, in

the experimental setup, the liquid trap, and the filter before it was located closer to the reaction furnace than the TEOM- measurement was, meaning that the issue of water condensation would have had a much bigger impact on the amount of tellurium reaching the TEOM measurement than on the amount of tellurium reaching the filters.

## 6. Future Work

The ORTEF project has explored the interaction between tellurium and several organic precursors in a variety of different atmospheres, and several results must be explored in greater depth in order to fully understand the results presented here. A first step would be to attempt online analysis of the organics formed, as to determine their exact species.

Further analysis of the filters will be preformed using XPS analysis. It is our hope that this will reveal the chemical speciation present on the filters, and especially if the speciation changes upon the addition of organics to the system.

Furthermore, one can further develop the system to make it more representative of a severe accident, for instance by including other fission products or investigate the interaction between volatized tellurium and organic precursors in the presence of radiation.

## 7. Acknowledgements

The authors want to acknowledge Professor Jan Kučera from the Nuclear Research Institute in Czech Republic for the neutron activation analysis. Thank you also to NKS-R for supporting this research.

## 8. Disclaimer

The views expressed in this document remain the responsibility of the author(s) and do not necessarily reflect those of NKS. Neither NKS nor any other organization or body supporting NKS activities can be held responsible for the material presented in this report.

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Title	Organic Telluride Formation during a Severe Nuclear Accident (ORTEF)
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**Abstract**

The potential formation of organic species during a severe nuclear reactor accident is a cause of concern due to the very high volatility of organic compounds. The interactions between organic material and fission products are especially concerning since this could lead to an increased source term in post-accident conditions. One of the fission products potentially forming volatile organic species is tellurium. Although evidence of the formation of organic tellurides in accident conditions was only showed recently, this raises concerns whether organic tellurium species should be considered a significant threat in accident scenarios. The aim of the ORTEF project was to investigate potential interactions between tellurium and organic material in the gas phase. The experimental conditions in ORTEF were chosen to represent those likely present in the containment gas phase where tellurium aerosols would be present simultaneously with volatile organics. The results show that the presence of organics increases the transport of tellurium in the gas phase. This was observed by measuring higher concentration of tellurium in the trap solution in the presence of organics compared to the reference conditions in both reducing and inert conditions. Oxidizing atmosphere and the consequent oxidation of elemental tellurium to tellurium oxide inhibited the interactions. Although the speciation analysis was inconclusive, the significant increase in the gaseous fraction is a clear indication of interactions. Whether the increased transport is due to the formation of organic tellurides or other interactions remains for further studies.

**Key words** severe accident, source term, fission product, tellurium