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Main Outcomes of the TETRA Project on Tellurium Chemistry in a Severe Accident

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Abstract

A joint study between VTT Technical Research Centre of Finland Ltd, Chalmers University of Technology, and the University of Oslo in the NKS-R TETRA activity (2019) was carried out to investigate the tellurium transport in the primary circuit system and the effect of the different spray solutions used in the containment spray system for the tellurium removal efficiency. Tellurium is considered to be one of the volatile fission products released in a severe nuclear power plant accident. The experimental work was carried using two different experimental setups at VTT. One setup utilized two furnaces; one used to volatilize (540 °C) the precursor (Te-metallic) and another one used as the reaction zone (1227 °C). This setup was used to observe the transport behavior of tellurium under the different investigated atmospheres (oxidizing and inert), both dry and humid. For the emergency spray solution investigation (water or two different alkaline borate solution), another setup with one furnace and spray chamber was used. The furnace was used to volatilize (840 °C and 540 °C) the two different precursors used $(TeO_2 \text{ and } Te-metallic)$ and the volatilized species were transported to the spray chamber by the carrier gas establishing the experimental conditions (oxidizing and inert), both dry and humid. Both the transport experiments and the spray experiments also investigated the effect of CsI on Te behavior under humid conditions (oxidizing and inert).

The transport experiments indicated the highest transport of the released Te through the experimental setup under oxidizing conditions compared to inert conditions. However, the highest release fraction of Te occurred under inert conditions. Increasing the humidity content of the atmosphere resulted in an increase in the fraction of tellurium transported under oxidizing and a decrease under inert conditions. The addition of CsI reduced the amount of tellurium transported to the filter and also the released amount of tellurium under oxidizing conditions. Moreover, CsI increased the tellurium fraction on the filter in humid inert conditions in comparison to the humid conditions without CsI.

The containment spray system experiments showed that the tellurium bearing aerosols were generally efficiently removed under both oxidizing and inert conditions. Although, a decrease in the removal was observed when metallic Te precursor was used instead of TeO_2 . The airborne CsI increased the removal efficiency of tellurium. Furthermore, the addition of chemicals to the spray solution did have a noticeable effect when TeO_2 was used as a precursor and less apparent when metallic Te was used.

Key words

Tellurium, Cesium iodide, Containment spray system, Aerosol, Primary circuit, Severe Accident, Source Term

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Final Report from the NKS-R TETRA

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1. Preface

The NKS-R TETRA project (project year: 2019) was performed in a collaboration involving Chalmers University of Technology (Sweden), VTT Technical Research Centre of Finland Ltd (Finland), and University of Oslo (Norway). The objective in the TETRA project was to study the tellurium chemistry in a severe nuclear power plant accident. The experiments were focused on the tellurium transport in a primary circuit and on the tellurium retention by the containment spray system.

The experiments on the tellurium transport in the primary circuit and the tellurium retention by the containment spray system were performed at VTT. The further analysis of the collected tellurium samples was performed at VTT and Chalmers. The instrumental neutron activation analysis (INAA) was intended to be carried out using the research reactor at Halden, Norway. Unfortunately, this became impossible during the project. Hence, an alternative reactor in the Czech Republic was used instead; specifically, the experimental reactor LVR-15 at Řež (Research Centre Řež, Ltd., Czech Republic). This alternative was made possible by the Jon Petter Omtvedt (UiO, Norway).

The main outcomes of the TETRA project are summarized in this report. The detailed results are presented in two journal publications to be submitted for journal review in June 2020. As an additional task outside the scope of the TETRA project, the publication about the tellurium retention by spray includes a simulation using a MELCOR code to enable a comparison between experiments and modeling. Furthermore, as an additional task, a third publication about the capability of the INAA method for the analysis of tellurium samples has also been prepared and submitted for journal review in March 2020.

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1. Introduction

Using nuclear power plants (NPP) for the production of electricity through nuclear fission reaction results in the build-up of several fission products (FP) in the fuel during the NPP operation. One of these FPs is tellurium, which has four main isotopes that constitute roughly 60% [Alonso & González, 1991] of the total tellurium in the fuel. These are ¹³¹Te (25 min [Magill et al., 2015]), ¹³² Te(76.3 h [Magill et al., 2015]), ^{133m} Te(55.4 min [Magill et al., 2015]), and ¹³⁴ Te (41.8 min [Magill et al., 2015]), all of which decay to different radioactive isotopes of iodine [Magill et al., 2015]. Iodine is especially problematic due to its radiotoxicity related to accumulation in the thyroid [Yosida et al., 2014]. Due to the relatively long half-lives of tellurium isotopes and the fact that many of these decay to iodine, tellurium release is a considerable risk and therefore further research and knowledge is needed.

After being released from the reactor core the FPs, including tellurium, are transported to the containment through the reactor coolant system (RCS). In the RCS, the FPs released from the core are subjected to the surrounding prevailing conditions (e.g., oxidizing, inert, reducing) and significantly lower temperatures than in the core. The change of conditions enables the FPs to condense and form aerosols of different composition and size. Most FPs are transported as part of fission product aerosols of different composition [Laurie et al., 2013]. Two exceptions are iodine and ruthenium, which can both exist in gaseous form under certain (e.g. oxidizing) conditions [Shegal, 2011; Kärkelä et al., 2017]. Once the FPs have reached the containment, they are exposed to various removal processes. One of the important safety features is the containment spray system (CSS), which removes both aerosols and gaseous species from the containment atmosphere and flushes them down to the sump. Although tellurium source term has been relatively well studied in terms of its release behavior from the core, the reactions and behavior of tellurium species following the core release still remain unclear. This project, and especially the transport experiments, aims to fill in the gaps in the knowledge related to tellurium behavior beyond the initial core release, and thus give valuable information for tellurium source term estimations. In addition, the effectiveness of the containment spray system for the airborne tellurium aerosol removal has been investigated.

Research on the source term of tellurium has been carried out previously, both in bench-scale work [Potter & Rand, 1983; McFarlane, 1996a] and in larger, integral FP release programs [Sangiorgi et al., 2015; Jones et al., 2015; Pontillon et al., 2010]. Two important programs investigating FP release and transport behavior, including tellurium, are VERCORS and PHÉBUS. One of the main outcomes from these programs in terms of the release behavior of FPs has been how to classify the FPs based on their volatility. From these programs, the tellurium has been classified as one of the volatile FPs, alongside cesium and iodine. In the VERCORS program [Pontillon et al., 2010; Pontillon & Ducros 2010a, 2010b], the release of tellurium from the core was considerable in almost all of the experiments. Only VERCORS tests 1 and 2 had very low releases, whereas tests 3 to 5 had significantly higher tellurium releases from the core. The difference in the release fractions of tellurium was suggested to be due to the time spent at maximum temperature. VERCORS test 1 had the shortest time at maximal temperature and the smallest tellurium release, while VERCORS test 2 had a slightly longer time and consequently somewhat higher tellurium release. Higher temperatures also increased the level of oxidation of the zirconia cladding. This has been found to be important for tellurium, as its release is delayed due to its retention in the zirconia cladding by interactions with zirconium and tin [Boer & Cordfunke, 1995]. However, once the cladding becomes sufficiently oxidized, tellurium is released completely, which happened in the VERCORS test 6, RTs-test, and HT-test series. This trend continued in the later VERCORS experiments, where the oxidation of the cladding was complete and there was almost always total release of tellurium. Furthermore, it was observed that the atmosphere had a significant effect on the tellurium speciation entering the thermal gradient tube (TGT) used in the different VERCORS HT tests. Concurrently, the distribution of cesium was found to coincide with that of tellurium in the VERCORS HT tests [Pontillon and Ducros, 2010a], thus indicating a potential correlation between cesium and tellurium.

In addition to the release and transport behavior investigated in the VERCORS FP program, the PHÉBUS FP program also included work regarding the behavior of FPs in the containment. In the PHÉBUS program, tellurium isotopes were found to be transported and entered the containment as a part of FP aerosols. Tellurium represented around 1-4% of the overall mass of the aerosols [Laurie et al., 2013]. The percentage varied due to different experimental conditions. Moreover, the behavior and distribution of tellurium were also monitored in the containment and it was found that tellurium-bearing species were removed from the containment atmosphere by both natural aerosol processes and engineered systems (e.g., washing phase). Tellurium species were found to deposit and condense on the inner surfaces of the vessel. Furthermore, at the end of the experiment, most of the tellurium was found in the sump as non-soluble species.

Both VERCORS and PHÉBUS experimental programs provide valuable information about the behavior of tellurium during the release and transport into the containment, as well as behavior once subjected to the removal processes. However, details about the exact speciation of tellurium formed under various conditions and the efficiency of the spray system require further attention.

2. Background

Several tellurium species can be present in the RCS during a nuclear accident. What actual species are present will depend on e.g., the type of accident, condition, and timing of the accident. Especially, the type of condition will have a great impact. The focus of this work has been on two of the relevant conditions, which are oxidizing and inert conditions. Moreover, a certain part of the chemistry involving the two other fission products cesium and iodine are also of interest.

After entering the containment, the species are subjected to various physical and chemical processes, either natural or engineered, which determine the rate of removal of the FPs. The main natural processes removing aerosols from the containment atmosphere (airborne particles ranging mainly up to 20 μ m in diameter [Kim et al., 2015]) include gravitational settling, include gravitational settling, diffusiophoresis, and deposition on the walls [Powers et al., 1996]. Engineered systems consist of e.g., containment spray system (CSS), steam suppression pool, and fan coolers [Sehgal, 2011]. The speciation of the FPs, the size and the mass of the potential aerosols, attraction to different surfaces, solubility in water, and reactivity towards the chemically engineered systems are parameters that affect the behavior and distribution of different fission products in the containment.

2.1. Tellurium

Under oxidizing conditions, tellurium can form several different oxides and affect the formation of other tellurium species (e.g., tin telluride). The main oxides of tellurium that can be formed are TeO and TeO₂. Initially, the dioxide will be the main species at lower temperatures. As the temperature increases to approximately 833 °C, thermodynamic studies [Garisto, 1982] have shown that the formation of the monoxide begins. According to the literature, two reactions are possible for metallic tellurium directly to form the two different oxides, according to equation 1

$$Te + 0.5 \cdot O_2 \rightarrow TeO [BG - 1] (1)$$

and by combustion in air [Dutton & Cooper, 1966] as exemplified by equation 2

$$Te + O_2 \rightarrow TeO_2$$
 (2).

However, the presence of humidity also enables another pathway for the formation of the monoxide [Garisto, 1982], through the tellurium dimer, as shown in equation 3.

$$Te_2 + H_20 \Leftrightarrow TeO + Te + H_2$$
 (3).

Furthermore, a new compound may also form in the presence of steam; $TeO(OH)_2$ [Garisto, 1982; Malinauskas et al., 1970]. A possible way that this may form is according to equation 4, which the thermodynamic calculations performed by Makinauskas [Malinauskas et al., 1970] are based on.

$$TeO_2(s) + H_2O(g) \Leftrightarrow TeO(OH)_2(g)$$
 (4)

A specific difference for tellurium is the reaction with the cladding. Consequently, the cladding will trap the tellurium, and for tellurium releases to occur the cladding needs to be oxidized. At the point at which tellurium release occurs, the tellurium can be released as tin telluride [Boer & Cordfunke 1995, 1997]. This new species is stable and can be part of the mix of tellurium species leaving the core. However, if oxygen potential is high (higher than $H_2O/H_2 = 100/1$), the tin telluride may oxidize to tin dioxide and metallic tellurium [Boer & Cordfunke 1995; Bettini & Richter, 1979]. The elemental tellurium would then be able to further oxidize and/or take part in other reactions.

Under very high temperatures (2300 °C, based on the VERCORS [Pontillon & Ducros, 2010a]) tellurium may be released from the core under inert conditions. This would change the main speciation of tellurium and result in the formation of metallic tellurium and the corresponding dimer (i.e., Te₂). Of these two, Te₂ would be the main species at lower temperatures (around 330 °C). However, as the temperature increases the elemental tellurium would become the dominating species [Garisto, 1982]. Furthermore, if steam is present it is possible that equation 3 could occur. Hence, some oxidation may occur. Still, as indicated by McFarlane [McFarlane, 1996] the predominating species, even in the presence of steam, would be metallic tellurium and its dimer.

Besides the already mentioned species, two more important species exist for tellurium source term considerations. One of these is tin telluride and the other is a species between cesium and tellurium. The occurrence of tin telluride, as already mentioned, is dependent on the prevailing oxygen potential. However, the formation of cesium telluride is more complex and several compounds between these are possible. Experimental information in the literature of such speciation is limited. However, it has been shown that it is possible that dicesium telluride can exist at higher temperatures. Moreover, not only dicesium telluride but also several other species are possible with different ratios of cesium to tellurium [Portman et al., 1989; McFarlane and Leblanc, 1996].

2.2. Containment Spray System

The inventory and distribution of fission products in the containment at any given time depends on the rate at which FPs enter the containment compared to the rate of removal [Soffer et al., 1995]. The removal happens via natural or engineered processes. One of the main engineered mitigation systems is the aforementioned CSS. It has been designed to help maintain the containment building integrity by decreasing the pressure inside the containment, as well as to mitigate the release of radionuclides by removing the FP-containing particles from the containment atmosphere into the sump. The removal efficiency of the CSS relates to the size of the aerosols in the containment atmosphere. The CSS has been found to be the most efficient at removing particles larger than 1 μ m or smaller than 0.1 μ m [Sehgal, 2011], but the removal efficiency remains low for the accumulation mode particles (0.1 to 1 μ m). For the larger particles, the most effective removal happens through sweep-out and interception due to the droplets as the aerosols follow the streamlines of the flow. The smaller particles are removed by diffusion to the droplet surface [Sehgal, 2011]. Gaseous species (e.g., organic iodides) can be removed by adjusting the chemistry of the spray solution to efficiently react with the gaseous compounds and convert them to a non-volatile form.

The chemical composition of the spray is primarily designed to mitigate iodine releases. Moreover, the main components of the CSS solution are a base (e.g., sodium hydroxide, potassium hydroxide or trisodium phosphate), boric acid, and possibly an additive (e.g., sodium thiosulphate, hydrazine). The spray solution pH is generally kept alkaline to shift the disproportionation of volatile iodine to a more non-volatile iodate/iodide side [Neeb, 2011]. Similarly, the use of additives, usually reducing agents, reduces the amount of volatile iodine species (especially organic iodides) by decomposing and trapping the gaseous species inside the droplets [Parsly, 1971; Joyce, 1971]. Finally, boric acid is used to maintain the subcriticality of the reactor core as well as to buffer the pH with the base [Neeb, 2011].

Most of the experimental work in terms of spray effectivity has been focused on iodine removal, e.g., [Parsly, 1971; Joyce, 1971; Hilliord et al., 1971; Postma et al., 1975]. Therefore, the data about the effectiveness towards tellurium species is yet unclear. Some modeling work regarding the spray efficiency, which takes into account tellurium, has been done [Dehjourlan et al., 2016]. The data suggests that the CSS is efficient in removing tellurium. However, the

chemistry and complexity of the speciation are limited in modeling codes and thus the results possess a high uncertainty when it comes to the speciation of tellurium under different conditions. Therefore, the spray effectiveness results need to be considered with caution. Parameters of most importance considering spray effectiveness are e.g., spray droplet size and particle size distribution. The TETRA experiments aim to provide more detailed information about the effectiveness of the spray system in removing tellurium species that have formed under different conditions. Furthermore, the results obtained from these TETRA experiments give information about the removal of species including cesium and iodine in addition to tellurium.

3. Experimental procedure and matrix

The experiments on the tellurium transport in RCS and removal by the CSS were performed using two different experimental setups - see the descriptions in Chapter 4.1. An overview of all the experiments performed is given in Tables 1 and 2. Tellurium was exposed to a maximum temperature of 1230 °C (metallic tellurium precursor) in the transport experiments, whereas the airborne tellurium species were generated at 540 °C (metallic tellurium precursor) and 880°C (tellurium dioxide precursor) in the spray experiments. In the experiments, the composition of the gaseous atmosphere was varied from inert to oxidizing, and the humidity content was also varied. In some experiments, additional cesium iodide aerosol was fed together with the tellurium precursor.

Table 1. Experimental matrix for the transport experiments and the parameters of the atmosphere used in each experiment. The temperature refers to the maximum temperature the airborne tellurium precursor and additives were exposed to in the experiments.

Experiment#	Precursor	Temperature	Atmosphere	Humidity	CsI
		[°C]			
T1	Те	1230	Air	No	No
T2	Te	1230	Air	Yes	No
T3	Te	1230	Air	Yes	Yes
T4	Те	1230	Nitrogen	No	No
T5	Те	1230	Nitrogen	Yes	No
T6	Te	1230	Nitrogen	Yes	Yes

Table 2. Experimental matrix for the spray experiments - the spray solutions are described in Chapter 4.1.1.

Experiment#	Precursor	Temperature	Atmosphere	Humidity	CsI
		[°C]			
1	TeO ₂	880	Air	No	No
2	TeO ₂	880	Air	Yes	No
3	TeO ₂	880	Air	Yes	Yes
4	Те	540	Air	No	No
5	Те	540	Air	Yes	No
6	Те	540	Air	Yes	Yes
7	Те	540	Nitrogen	No	No
8	Те	540	Nitrogen	Yes	No
9	Те	540	Nitrogen	Yes	Yes

3.1. Experimental facilities

3.1.1. Tellurium transport experiments

Based on the previous NKS-R collaboration between VTT and Chalmers (NKS/ATR-1 and NKS/ATR-2 projects), the already existing facility at VTT for the study of fission product transport was used for these TETRA project experiments. The schematics of the "VTT fission product transport" facility can be seen in Figure 1.



Figure 1. Schematics of the experimental VTT facility for the tellurium transport studies, extracted from [Kärkelä et al., 2017].

The experiments were carried out using a tubular flow furnace (Entech, ETF20/18-II-L), inside a tube made from high purity alumina. An additional small furnace (not shown in Figure 1) was located just before the entrance of the tubular furnace in Figure 1. The same alumina tube extended through both furnaces. Metallic tellurium (ca. 1 g) was added to a ceramic crucible placed in the small furnace (Entech/Vecstar, VCTF 3). The small furnace was heated to 540 $^{\circ}$ C, as then the volatilization of tellurium (melting point 500 $^{\circ}$ C) took place moderately. The tubular furnace was heated to 1230 °C to simulate the primary circuit conditions. During the heat-up phase of both furnaces a nitrogen flow of 0.5 l/min was directed through the furnaces. A flow rate of 5 l/min (air or nitrogen) through the system was initiated when the temperature set-points of the furnaces were achieved. The flow rate is a sum of the carrier gas flow rates of tellurium (generation by the small furnace), steam (generation by atomizer), and CsI (generation by atomizer) in various experiments. The mixture of gas flows and precursors entered the (high temperature) tubular furnace, in which the reactions between the precursors took place. Downstream of the tubular-furnace, a stainless steel (SS; AISI 316L) tube was positioned and the cooling of the flow to containment temperatures occurred. The aerosol reaction products originating from the interaction between tellurium, the atmosphere, other precursors, and/or additives were filtered out at a location of 106 cm from the outlet of the furnace (at 30 °C). Beyond the filter, water/NaOH traps were positioned to capture gaseous species.

3.1.2. Tellurium retention by containment spray experiments

The schematics of the "VTT spray chamber" facility is shown in Figure 2. The facility is a containment model with a containment spray system (CSS) on top. A tubular furnace (Entech/Vecstar, VCTF 3) was used to vaporize the tellurium precursor in an alumina crucible inside a stainless-steel tube (AISI 316L). In the spray experiments, two precursors were used: tellurium dioxide (TeO₂) and metallic tellurium (Te). The mass of the precursor was adjusted to represent 1 g of tellurium. For metallic tellurium, the mass was 1 g and for TeO₂ 1.26 g (the latter of these was slightly higher to maintain equal molar amounts of Te). The furnace was heated to 540 °C for Te and to 880 °C for TeO₂. A flow rate of 5 l/min (air or nitrogen) through the furnace was initiated at the beginning of the furnace heat-up. The flow rate is a sum of the carrier gas flow rates of tellurium compound (generated by the furnace), steam (generation by

atomizer before the furnace - not shown in Figure 2) and cesium iodide (generation by atomizer before the furnace - not shown in Figure 2) in various experiments. The mixture of gas flows and precursors reacted inside the furnace. Downstream of the tubular furnace a stainless steel (SS; AISI 316L) tube was positioned and the cooling of the flow to ca. 20 °C occurred. The aerosol reaction products originating from the interaction between tellurium, the atmosphere, other precursors, and/or additives were directed to the vertical spray chamber through a connection in the lower section of the chamber. The aerosols were subjected to the spray droplets (ca. 10 µm). The spray solutions (at 20 °C) chosen for these experiments were either pure water (Milli-Q water) or were based on an alkaline borate solution containing 0.23 M boric acid, H₃BO₃, and 0.15 M sodium hydroxide, NaOH, to adjust the pH to 9.5. To test the effect of the additives, 0.064 M of Na₂S₂O₃ was added to the alkaline borate solution. The compositions of the spray solutions were chosen to represent the general conditions used in severe accident management. The aerosol flow transported out of the spray chamber was diluted and dried with hot gas flow (air or nitrogen at 100 °C). Aerosols were filtered out after the hot dilution (at a location with a temperature of 30 °C). Beyond the filter, water/NaOH traps were positioned to capture gaseous species. Each experiment started by collecting a reference sample where all of the tellurium species transported through the system without the spray were collected on the filter and trap. The reference samples were used to calculate the removal efficiency of the sprays. Each spray duration was 20 minutes and sump, filter, liquid trap, and scanning electron microscopy (SEM) samples were collected for analyses.



Figure 2. Schematics of the experimental VTT facility for the studies on the tellurium retention by containment spray system.

3.2. Analysis methods

3.2.1. INAA – Instrumental Neutron Activation Analysis

Each experimental run had all used filters and the first sodium hydroxide liquid trap analyzed by INAA. The aim was primarily to determine the amount of tellurium. However, in the cases when CsI was part of the experiments the amounts of cesium and iodine were also determined. Determining these three elements required different approaches timewise; short-term irradiation (tellurium), medium-term (tellurium), and long-term (tellurium, cesium, and iodine). Two reasons exist for the three different irradiation times: (1) to be able to determine all elements and (2) to determine the efficiency of measuring tellurium according to the three different modes in INAA.

Prior to analysis, the samples had to be slightly processed before the analysis. The filters ("Filter 1" in Figure 1) were folded and pressed prior to analysis, except a small part that was removed for other analyses. From the liquid samples, 100 μ L was pipetted and added to a cellulose chromatography paper (Whatman 1) and then dried. In addition to the samples, calibrators were also made consisting of dissolved tellurium (99.9% Merck) in HNO₃. Aliquots (50 μ L) were taken and prepared similarly to the liquid samples. The amount of tellurium in these calibrators was 103 ± 0.5 μ g. Blank filters were also investigated to determine possible interfering peaks. The irradiation times for the samples were 1 min or 2.5 hours, depending on what nuclides were to be measured. Furthermore, the difference in time required the use of different transport containers (polyethylene or aluminum) and cooling medium (air or water).

A gamma-spectrometer (Canberra Genie 2000) with three options for choice of coaxial high purity germanium detector (HPGe-detector) was used. Depending on if short-term (Princeton Gamma-Tech), medium-term (Ortec or Canberra), or long-term (Ortec or Canberra) analysis were performed, different HPGe-detectors were used with different counting geometries. The relative efficiency and resolution (⁶⁰Co-photons) for the short-term radionuclides were 20.3% and 1.75 keV at 1332.5 keV, respectively. For the medium/long-term radionuclides the relative efficiency and the resolution (⁶⁰Co-photons) were either 52.9% at 1.76 keV at 1332.5 keV (Ortec) or 77.8% at 1.87 keV at 1332.5 keV (Canberra), respectively. The gamma lines [Magill et al., 2015] for determining the different elements were for cesium 606 keV (¹³⁴Cs-decay), for iodine 443 keV (¹²⁸I-decay), and for tellurium 149.8 keV (¹³¹Te-decay).

3.2.2. ICP-MS – Inductive Coupled Plasma Mass Spectrometry

At Chalmers, the analysis using Inductive Coupled Plasma Mass Spectrometry (ICP-MS) was performed to analyze cesium and tellurium content of the liquid samples as an alternative to INAA. Aliquots from the sodium hydroxide liquid traps were taken and diluted using 0.5 M nitric acid (Suprapure®, Sigma Aldrich). Rhodium (Elemental Standard Foratomic Spectroscopy 1000 ± 5 mg/ml, 20° C, Spectrascan) was used as an internal standard. The samples were analyzed with ICP-MS (Thermo ScientificTM iCap Q) and the data was evaluated using QtegraTM Intelligent Scientific data solution (v.2.21465.44).

3.2.3. Gas-phase analysis and sampling of aerosols

In the tellurium transport experiments (Figure 1), at 74 cm from the furnace outlet, aerosol gasphase sampling took place. The sample was diluted and quenched to ca. 30 °C with a porous tube diluter in order to minimize losses during the dilution. The number size distribution of particles was online measured with a combination of differential mobility analyzer (DMA) and condensation particle counter (CPC) for a particle range of 2 nm to 1 μ m. At the same time, the mass concentration of aerosol particles was online analyzed with Tapered Element Oscillating Microbalance (TEOM). In the spray experiments (Figure 2), downstream of the hot dilution step, an electrical low-pressure impactor (ELPI) was used to online measure the particle number size distribution in a particle range from 7 nm to 10 μ m. In both transport and spray experiments, the formed particles were also collected on a carbon/copper grid directly from the gas phase (just before the filter location) with an aspiration sampler. This was done in order to analyze the particles by SEM afterwards. Furthermore, in the transport experiments, a sample of particles in the gas flow was also collected on an additional analysis filter ("Filter 2" in Figure 1) through the online measurement line of the facility.

3.2.4. SEM - Scanning Electron Microscopy analysis

Samples of particles in the gas phase were collected with an aspiration sampler on a copper/carbon grid. The size and morphology of the collected particles on the copper/carbon grid substrates were analyzed using scanning electron microscopy (SEM, Zeiss Crossbeam 540). SEM was operated at 2 - 3 kV and the probe current was 100 - 200 pA during imaging.

4. Results and Discussion

4.1. Tellurium transport experiments

Filters ("Filter 1" in Figure 1), sodium hydroxide liquid traps, and online data were acquired from the transport experiment part of the TETRA project. In this report, an overview of the most important results is presented. For detailed analyses of all the results, more information can be found in the different articles from the TETRA project presenting the transport experiments (to be submitted in June 2020, planned journal: Progress in Nuclear Energy). and the INAA analysis (submitted 2020 to Microchemical Journal).

4.1.1. Mass Difference

The weight change¹ that occurred for the tellurium precursor in an alumina crucible in each experiment was determined by weighing using a tabletop balance. For the two first oxidizing conditions (Experiments T1 and T2) the weight changes were roughly 56 mg (5.6% of total amount precursor) for the dry and 44 mg (4.4% of the original precursor amount) for the humid. Moreover, the addition of cesium iodine (Experiment T3) resulted in a release of 25 mg (2.5% of the total amount tellurium) of the precursor. Of these released amounts of tellurium, the mass fractions reaching the filter 1 (in Figure 1) during each experiment was 90% (Experiment T1) for the dry experiment and 91% (Experiment T2) for the humid experiment, and when cesium iodide was added the fraction was 80% (Experiment T3). The mass change of the dry inert (Experiment T4), humid inert (Experiment T5), and humid inert with added cesium iodide (Experiment T6) were similar at 280 mg (28.3% of total amount tellurium), 300 mg (29.6% of total amount tellurium), and 275 mg (27.5% of total amount tellurium), respectively. Of these amounts released, the fraction of tellurium reaching the filter 1 (in Figure 1) was 22% for the dry inert conditions (Experiment T4), 12% for the humid inert condition (Experiment T5), and the addition of cesium iodide (Experiment T6) resulted in 19% of the released tellurium reaching filter 1. An overview of all values from the experiments can be found in Table 3.

Observing the precursor in the alumina crucible from all experiments, a slight color change (black to gray) occurred with the precursor of the three oxidizing experiments (Experiments T1-T3) and no color change occurred with the precursor of the three inert experiments (Experiments T4-T6). In the former, the color change can be explained by oxidation. If this mainly occurred at the surface, it would mean that the bulk part of the precursor is still mainly tellurium and the effect of oxidation on the precursor mass increase will be negligible. However, the depth of the oxidized layer was not analyzed. Observing the mass change of what remained in the crucible from the oxidizing conditions, only the addition of CsI notably changed the amount of precursor leaving. For the inert conditions, the release fraction of tellurium was roughly the same for all experiments. However, the amounts reaching filter 1 were different, indicating that something did occur at high temperature. From these values, humidity under inert conditions reduced the amount of tellurium. The mass increase observed for the experiment involving CsI (Experiment T6) can be explained by the CsI itself depositing on the filter, thus increasing the weight.

¹For the Experiments T1-T3, the oxidation is assumed to be negligible.

Table 3. Released mass of tellurium in the alumina crucible and the mass of tellurium transported to the filter (filter 1 in Figure 1). Measured by a tabletop balance. Values in parenthesis; First row: the released fraction of the precursor; Second row: the fraction on the filter of the total release, with the assumption that the oxidation of the precursor was negligible.

Experiment	T1	T2	T3	T4	T5	T6
Released [mg]	56	44	25.0	283	296	275
	(5.6%)	(4.4%)	(2.5%)	(28.3%)	(29.6%)	(27.5%)
Total transported,	50	40	20	62.5	35	52.5
filter 1 [mg]	(90%)	(91%)	(80%)	(22%)	(12%)	(19%)

4.1.2. INAA

The data obtained using the INAA method for both oxidizing and inert conditions showed the content of tellurium on the filters (filter 1 in Figure 1) from all experiments, and cesium and iodine in relevant experiments (Experiments T3 and T6). However, no measurable amounts of the three elements were detected in the sodium hydroxide liquid traps from any of the experiments (Experiment T1-T6). Observing the data for the filters, the fraction of tellurium transported of the total released (when utilizing the release data from Table 3) was determined in all oxidizing experiments (Experiment T1-T3). The percentage for each was 58% (Experiment T1), 63% (Experiment T2), and 50% (Experiment T3) for the dry, humid, and humid with the addition of CsI oxidizing condition, respectively. For the inert conditions, the tellurium fraction reaching the filters was considerably lower compared to the oxidizing conditions. For the dry (Experiment T4), humid (Experiment T5), and humid with added CsI (Experiment T6) the fraction of tellurium reaching the filters of the total released was 16%, 13%, and 15%, respectively. An overview of the outcome can be observed in Figure 3.



Figure 3: The estimated fraction of the tellurium reaching the filter of the total released amount from the precursor. The release data used are from the tabletop measurements presented in Table 3.

The INAA results for the oxidizing condition showed that the amount of tellurium reaching the filter increased when humidity (Experiment T2) was added and then decreased when cesium

iodide was also included (Experiment T3). The initial increase observed could be explained by the existing literature, which suggests that water vapor increases the volatility of TeO₂. However, the decrease observed when CsI was added is not as readily explained. Still, one explanation could be that the presence of CsI prevents/decreases the water vapor from interacting with the TeO₂ directly by the CsI interacting with the water vapor itself or interacting with the TeO₂. Alternatively, it may be that the increased number of particles due to adding CsI increased the size of the aerosols and thereby increasing the deposition prior to the filter. Observing the INAA data for the three experiments under inert conditions (Experiments T4-T6), the tellurium fractions reaching the filters were similar. Potentially, a slight decrease could be distinguished during the humid inert experiment (Experiment T5). This would then be explained by the oxidation due to the humidity via the formation of tellurium oxide, which is less volatile by itself. Therefore, the slight increase (from the humid conditions) of tellurium fraction reaching the filter in humid conditions when CsI was added could be explained by the formation of a more volatile cesium-tellurium species. However, this behavior is not clearly indicated based on the available data.

Overall, the transported fraction of tellurium was lower when analyzed with INAA, which detects the actual elemental content instead of the bare total accumulated mass, which was obtained by weighing with a scale (Table 3). Based on the INAA results, the tellurium transport ranged from 13% to 16% of the released tellurium mass in inert conditions. The tellurium transport was higher in oxidizing conditions, ranging from 50% to 63%. In reality, the transport could be lower, since the metallic tellurium precursor was oxidized in the crucible during the experiment. The oxidation increases the mass of the remaining precursor and thus decreases the obtained results for the tellurium release (analyzed by weighing).

4.1.3. Mass transport

In all experiments, the geometric mean diameter of the number size distribution of formed particles was ca. 100 nm and this decreased during the experiments (measured with the combination of DMA and CPC). The mass concentration of the transported aerosol particles was online analyzed using TEOM. The obtained data was converted into the form of cumulative mass fraction and is shown in Figure 4. The curves were overlapping in general. There was a clear difference in the shape of the curves between the oxidizing and inert atmospheres. In the experiments T1 to T3 in an air atmosphere, the transport of particles was high at the beginning of experiments, with subsequent decrease in the aerosol transport. This was concluded to originate from the oxidation of metallic precursors in the crucible over the course of the experiments. Visual observation of the powdery precursor verified the change in color from black to grey, indicating oxidation of the precursor. After the oxidation, the higher temperature would have been needed to release the oxidized tellurium (likely in the form of TeO₂) in the crucible. In the experiments T4 to T6 in a nitrogen atmosphere, the transport of tellurium aerosol was quite stable (the slopes of curves were closer to linear). From ca. 20 minutes onward, the transport of particles was higher than in the experiments in the air atmosphere. As a general observation, the aerosol transport decreased over time in all experiments, thus supporting the above-mentioned decrease in the particle mean size.



Figure 4. Cumulative mass fraction of the transported aerosol particles in experiments T1 to T6, presented as a function of time.

4.2. Tellurium spray experiments

Filter, sodium hydroxide liquid traps, and online data were acquired from the spray experiment part of the TETRA project. In this report, an overview of the most important results is presented. For detailed analyses of all the results, more information can be attained in the different articles from the TETRA project that present the spray experiments (to be submitted in June 2020, planned journal: Annals of Nuclear Energy) and the INAA analysis (submitted 2020 to Microchemical Journal).

4.2.1. Mass difference

Observations about the release behavior of different tellurium precursors can also be made from the mass loss during the experiments (Table 4). The crucibles and the masses of the precursors were weighed before and after each experiment and the mass difference was calculated. The overall mass loss in experiments 1, 2, and 3 is relatively consistent. The mass difference during the experiments varied between 0.2 and 0.3 g of the initial 1.26 g - although the duration was slightly different between the experiments. The release increased according to the mass loss from dry to humid air to CsI additive, which coincides with the removal efficiencies obtained from INAA filter analyses (See Section 5.2.2.).

The mass difference in experiments 4, 5, and 6 with metallic Te in air had a very low loss, or even a slight increase, compared to the initial mass of 1 g. In dry air the mass difference was 0.05 g, which is 5% of the initial mass of the precursor. In humid air and with CsI additive, the mass of the precursor increased 0.009 g and 0.07 g, respectively. These results conflict with the filter results, which suggests that in humid air the release is significantly lower than in the presence of CsI. The observed low mass difference could again be due to the oxidation of metallic tellurium precursor (likely into tellurium dioxide) in the crucible during the experiments. The oxidation increases the mass of the remaining precursor.

The largest mass difference was in experiments with Te in nitrogen atmosphere (Exp. 7, 8, and 9). In Exp. 7 the mass loss was 0.8 g of the initial 1.0 g of the precursor. Experiments 8 and 9 were performed back to back, thus increasing the duration of the experiment, which makes it difficult to differentiate the loss in the individual parts. However, the overall mass loss was 0.9 g, which is 90% of the initial amount of the precursor.

Experiment#	Initial Mass, [g]	Mass Release, [g]
1	1.26	0.199
2	1.26	0.255
3	1.26	0.295
4	1.02	0.051
5	1.00	-0.009
6	1.01	-0.069
7	1.00	0.789
8-9	1.00	0.883

Table 4. Precursor mass and release in the experiments (TeO₂ in Experiments 1 to 3; Te in Experiments 4 to 9).

4.2.2. Spray Removal Efficiency

The removal efficiencies of the sprays were determined from the INAA filter analyses. The efficiencies were calculated from the reference filter sample, taken from the steady flow without any sprays and from the filters trapping the aerosols coming through the system collected during each spray. The calculated efficiencies are presented in Figure 5, 6, and 7 for Experiments 1-3, 4-6, and 7-9, respectively. The presented values are not corrected for the actual spray coverage of the spray chamber cross-section; full coverage is assumed. However, the trends in removal efficiencies between the various experiments can be compared. For the TeO₂ precursor, the removal efficiency was very high in all three conditions and with each spray. Water spray removal efficiency varied between 83% and 91% whereas with the chemical sprays the efficiency was even higher. Both chemical sprays removed 96-97% of the tellurium species in both dry (Experiment 1) and humid conditions (Experiments 2 and 3).

With metallic tellurium precursor, the removal efficiency varied between the different experimental conditions. In dry air atmosphere, water spray removed 82% of tellurium species. However, when the humidity of the air was increased the removal percentage with water decreased to 6.4%. The reason for this decrease in removal efficiency is unknown and the possibility of an experimental error is considered. With CsI added to the humid air atmosphere, the removal of tellurium species increased again to approximately 92%. The trend continued with both chemical sprays, where in dry and humid CsI atmospheres the removal efficiency was 99%, whereas in humid air the removal was around 73% for both chemical sprays.

In inert atmosphere (Experiment 7-9), the removal of tellurium species was generally lower compared to the experiments performed in air atmosphere. Tellurium released in dry nitrogen atmosphere was removed with 63%, 72%, and 75% efficiency with water spray, chemical spray without and chemical spray with sodium thiosulphate, respectively. In contrast, the removal efficiency decreased with increasing chemical composition of the spray solution for tellurium in humid N₂. The percentages were 70%, 64%, and 60% for water, borate buffer without, and borate buffer with sodium thiosulphate. Finally, the efficiency increased again for Experiment 9 where metallic tellurium was released in humid nitrogen with CsI addition. The removal efficiencies were 89-94% with all three spray solutions.



Figure 5. Removal efficiencies for TeO₂ in air.



Figure 6. Removal efficiencies for Te in air.



Figure 7. Removal efficiencies for Te in nitrogen.

4.2.3. Liquid samples

INAA and ICP-MS were used to analyze tellurium concentration in the sodium hydroxide traps to determine whether any gaseous species containing tellurium had passed through the filter. However, both of the chosen methods were unable to determine the concentration of tellurium in the liquid traps, due to the concentration being too low for detection.

The sump samples collected from the bottom of the spray chamber after each spray were analyzed by ICP-MS for tellurium and cesium concentration (Experiment 3, 6, and 9). Overall the concentration of tellurium in the sump was low compared to the mass difference of the precursor. When using metallic tellurium as a precursor in nitrogen atmosphere, the sump had silvery black precipitation (Figure 8). This indicates that tellurium species, which were removed from the containment atmosphere, were possibly in the form of elemental tellurium. Before analysis, the precipitation was left to oxidize in air and dissolve. The concentrations of cesium stayed consistently low in all three experiments (3, 6, and 9).



Figure 8. The beaker where the sump from experiment 7 was collected before flushing.

4.2.4. Aerosol properties

The average mass size distributions of the fed aerosol inside the spray chamber under dry conditions (before the initiation of first spray solution injection) in Experiments 1 to 9 are presented in Figures 9 to 11. The data is based on the ELPI measurements (number data converted to mass data). In all experiments the mode of the aerosol mass size distribution was below 1 μ m (aerodynamic diameter), although the existence of a second aerosol mode of coarse particles was indicated in some experiments. In the case of the TeO₂ precursor in an oxidizing atmosphere (Experiments 1 to 3), the mass size distribution was unimodal in Experiments 1 and 2. The second aerosol mode appeared in Experiment 3. The second mode corresponded to the fraction of coarse particles (>1 μ m) and this was notably higher for experiments 4 to 9

performed with the metallic Te precursor. Based on the SEM analysis (see below), agglomerated particles were observed. The formation of large agglomerate particles possibly corresponds to the second aerosol mode. The agglomerate formation seemed to be pronounced in oxidizing conditions (Experiments 4 to 6). The mass concentration was decreased in the case of inert conditions (Experiments 7 to 9). The mass concentration in Experiment 5 was very low.



Figure 9. Average mass size distributions of the fed aerosol inside the spray chamber under dry conditions in Experiments 1 to 3 (TeO₂ precursor in an oxidizing atmosphere).



Figure 10. Average mass size distributions of the fed aerosol inside the spray chamber under dry conditions in Experiments 4 to 6 (metallic Te precursor in an oxidizing atmosphere).



Figure 11. Average mass size distributions of the fed aerosol inside the spray chamber under dry conditions in Experiments 7 to 9 (metallic Te precursor in an inert atmosphere).

4.3. SEM imaging of tellurium aerosol

Samples of the aerosol particles formed from the metallic and oxide forms of tellurium precursors (Te and TeO₂) in dry air and nitrogen atmospheres were collected during the transport and spray experiments. Further SEM analysis of the collected particles verified the morphology of the particles formed from the powdery precursors heated in a furnace under simple oxidizing and reducing conditions; tellurium dioxide in air, tellurium in air, and tellurium in nitrogen, see Figures 12, 13 and 14.

In all conditions, a significant fraction of the analyzed particles were long agglomerate chains composed of primary particles with a diameter of less than 100 nm. In the case of tellurium in air, the particles seemed to be somewhat fused together. In general, the formed aerosols were polydisperse in nature. Further analyses of the particle properties obtained in this study are summarized in the different articles from the TETRA project presenting the transport experiments and the spray experiments (to be submitted in June 2020).



Figure 12. Micrograph of tellurium containing aerosol - precursor TeO2 vaporized in dry air atmosphere.



Figure 13. Micrograph of tellurium containing aerosol - precursor Te vaporized in dry air atmosphere.



Figure 14. Micrograph of tellurium containing aerosol - precursor Te vaporized in dry nitrogen atmosphere.

5. Conclusions

The objective of the TETRA project was to study the tellurium chemistry in a severe nuclear power plant accident. The experiments were focused on the tellurium transport in a primary circuit and on the tellurium retention by the containment spray system. Based on the data from the transport experiments with metallic tellurium precursor, it can be concluded that the released tellurium will have a higher transport through the RCS under oxidizing conditions compared to inert conditions. However, the release of tellurium was significantly higher in inert conditions. The tellurium release increasingly slowed down over the course of the experiment due to the oxidation of tellurium precursor in oxidizing conditions. Moreover, the addition of humidity decreased the transported amount of tellurium slightly under inert conditions and increased it under oxidizing conditions. In the remaining oxidizing condition, the addition of CsI resulted in a significant drop in the released amount of the precursor, but the fraction of the released tellurium reaching the filter decreased only moderately. However, the CsI additive may have been reacting with the precursor and deposited on the filter, thus increasing the mass of both (hence, affecting the tellurium fraction). Under inert conditions, the addition of CsI increased the tellurium fraction on the filter (compared to the humid inert conditions). Indicating that the CsI had an effect on the transported tellurium amount.

In terms of spray efficiency, it was found that the spray system is effective in removing tellurium-containing aerosols from the containment. The efficiency was rather high in all conditions used. However, the removal efficiency was lower when using metallic tellurium as the precursor. Moreover, the removal was more effective for metallic tellurium when CsI was added to the flow. This might indicate a change in the form or speciation of tellurium in the Te-Cs-I system.

Chemicals used in the spray improved the removal of tellurium-bearing aerosol. This was especially apparent in the removal efficiency of TeO_2 when it was used as a precursor, as the chemical sprays removed up to 97% of the tellurium aerosols. The effect of chemicals was less noticeable when using metallic tellurium as the precursor.

The TETRA experiments provide information about the transport of tellurium in the RCS, as well as the effectiveness of the spray system under different conditions. In addition, the effect of chemicals in the sprays was investigated. The results obtained from this study provide new information on the behavior of tellurium in a severe nuclear power plant accident. The atmosphere conditions seemed to have an effect on tellurium transport. The applicability of the containment spray system for tellurium removal was verified. The use of chemicals to further enhance tellurium removal was indicated. These new observations should be noted when the tellurium source term is defined in the safety analysis.

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7. Disclaimer

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8. References

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Title	Main Outcomes of the TETRA Project on Tellurium Chemistry in Severe Accident				
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in the NKS-R TETRA activity (2019) was carried out to investigate the tellurium transport in the primary circuit system and the effect of the different spray solutions used in the containment spray system for the tellurium removal efficiency. Tellurium is considered to be one of the volatile fission products released in a severe nuclear power plant accident. The experimental work was carried using two different experimental setups at VTT. One setup utilized two furnaces; one used to volatilize (540 °C) the precursor (Te-metallic) and another one used as the reaction zone (1227 °C). This setup was used to observe the transport behavior of tellurium under the different investigated atmospheres (oxidizing and inert), both dry and humid. For the emergency spray solution investigation (water or two different alkaline borate solution), another setup with one furnace and spray chamber was used. The furnace was used to volatilize (840 °C and 540 °C) the two different precursors used (TeO₂ and Te-metallic) and the volatilized species were transported to the spray chamber by the carrier gas establishing the experimental conditions (oxidizing and inert), both dry and humid. Both the transport experiments and the spray experiments also investigated the effect of CsI on Te behavior under humid conditions (oxidizing and inert).

The transport experiments indicated the highest transport of the released Te through the experimental setup under oxidizing conditions compared to inert conditions. However, the highest release fraction of Te occurred under inert conditions. Increasing the humidity content of the atmosphere resulted in an increase in the fraction of tellurium transported under oxidizing and a decrease under inert conditions. The addition of CsI reduced the amount of tellurium transported to the filter and also the released amount of tellurium under oxidizing conditions. Moreover, CsI increased the tellurium fraction on the filter in humid inert conditions in comparison to the humid conditions without CsI.

The containment spray system experiments showed that the tellurium bearing aerosols were generally efficiently removed under both oxidizing and inert conditions. Although, a decrease in the removal was observed when metallic Te precursor was used instead of TeO₂. The airborne CsI increased the removal efficiency of tellurium. Furthermore, the addition of chemicals to the spray solution did have a noticeable effect when TeO₂ was used as a precursor and less apparent when metallic Te was used.

Key words Tellurium, Cesium iodide, Containment spray system, Aerosol, Primary circuit, Severe Accident, Source Term