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Note/Anmerkung

Essential parts of this diploma thesis were published by D. Boya, M. Nemetz, J.M. Welch, V. Rosecker, J.H. Sterba, D. Hainz, B. Feng, and G. Steinhauser in the “Journal of Radioanalytical and Nuclear Chemistry” under the title “Production of no-carrier-added aqueous $^{198}\text{Au}^{3+}$ ions in a Szilárd-Chalmers-like nuclear reaction” (DOI: <https://doi.org/10.1007/s10967-024-09705-1>). My own contributions included the construction, execution and analysis of the experiments mentioned, as well as the creation of all graphics and tables along with the writing of large parts of the publication. The paper is quoted under the relevant headings; except for chapter 4.4. and 5.4., which do not appear in the scientific paper. The experiments were carried out independently under the supervision of people who are experienced in radiation protection.

Wesentliche Teile dieser Diplomarbeit wurden von D. Boya, M. Nemetz, J.M. Welch, V. Rosecker, J.H. Sterba, D. Hainz, B. Feng, and G. Steinhauser im „Journal of Radioanalytical and Nuclear Chemistry“ unter dem Titel „Production of no-carrier-added aqueous $^{198}\text{Au}^{3+}$ ions in a Szilárd-Chalmers-like nuclear reaction“ (DOI: <https://doi.org/10.1007/s10967-024-09705-1>) publiziert. Die eigenen Beiträge waren der Aufbau, die Durchführung und die Auswertung der erwähnten Versuche, als auch die Erstellung sämtlicher Graphiken und Tabellen sowie das Verfassen großer Teile der Publikation. Diese wird im Rahmen der betreffenden Überschriften zitiert; bis auf die Kapitel 4.4. und 5.4., welche nicht in dem wissenschaftlichen abgehandelt Paper ab wurden. Die Experimente wurden unter Aufsicht strahlenschutzerprobter Personen eigenständig durchgeführt.

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Abstract

Preliminary experiments indicate the release of radiogold into an aqueous phase when gold foils that are submerged in water are irradiated with neutrons. Experiments verify this quantitatively, as the yield in ^{198}Au is ten-fold when gold foils are irradiated in H_2O compared to removal of said foils from the aqueous phase after mechanical shaking and prior to irradiation. Further experiments detail that there is no correlation between the activity of the gold foils with the amount of radiogold being released from the foils. However, the duration of the neutron bombardment is a key factor in the ejection of ^{198}Au into the aqueous phase, although the exact relationship between the duration of neutron irradiation and the yield of no-carrier-added ^{198}Au is not fully known. Changing the matrix in which the neutron irradiation takes place from an aqueous to a solid phase, leads to surprising, yet not fully conclusive results, where more radiogold is released into the solid phase, compared to the aqueous phase. Finally, the crystallization experiment conducted for speciation shows that the investigated nuclear reaction produces $^{198}\text{Au}^{3+}$.

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1. Introduction

Radioactivity has been a core topic of physics and chemistry since its discovery in 1896 by Antoine Henri Becquerel [1]. In the time since, many radionuclides and isotopes have been discovered via various methods and experiments while, often simultaneously, numerous nuclear reactions and effects were revealed to exist. After the discovery of the neutron in 1932 by Sir James Chadwick [2], it was the pioneering work of Enrico Fermi on the neutron capture in 1934 [3] that eventually led the Hungarian physicist Leó Szilárd and British scientist T.A. Chalmers to the observation of the so called Szilárd-Chalmers effect [4]. Chapter 2.3. will describe the experiment that lead both scientists to their discovery and the Szilárd-Chalmers effect itself. Even though the Szilárd-Chalmers effect has been used to create numerous radioisotopes by many scientists in the decades following its discovery [5], [6], [7], the exact details of how the change of certain parameters affects the outcome of this nuclear reaction are unclear.

There are numerous ways to produce anthropogenic radionuclides, however, some of these methods do not deliver the desired results. For example, processes that yield radioactive atoms surrounded by stable isotopes of the same element is not always the preferable outcome. The inactive material is called ‘carrier.’ The Szilárd-Chalmers effect offers a possibility to generate carrier-free radionuclides, which are highly valuable as precursor materials for radiopharmaceuticals. One example on how to generate a carrier-free radionuclide is the use of gold foils in conjuncture with the Szilárd-Chalmers effect to produce no-carrier-added radioactive ^{198}Au from stable ^{197}Au . This idea stems from the Applied Radiochemistry group at the TRIGA Center Atominstitut, where traces of radiogold were measured in the primary coolant of the TRIGA Mark-II research reactor during neutron flux density measurements using gold foils.

In this thesis, gold foils, which were placed in an aqueous phase, were used to investigate different aspects of the Szilárd-Chalmers effect in the production of no-carrier-added ^{198}Au . The activities from the gold foils released no-carrier-added radiogold of various experimental setups after irradiation with neutrons were analyzed and compared. First, it was necessary to confirm whether or not indeed neutron irradiation of gold foils in an aqueous medium leads to the release of significantly more gold into the liquid compared to shaking the gold foil in the aqueous phase prior to irradiation. Later, the roles of the activity of the gold foil in the experiments and the duration of the irradiation were examined in the Szilárd-Chalmers effect

regarding the release of radiogold. To investigate how the activity of the gold foil affects the yield of radiogold, gold foils were repeatedly irradiated multiple times. For the examination of how the duration of the neutron bombardment is connected to the amount of radiogold released, the same experimental setup was used, but with different irradiation durations. Afterwards, the results of the irradiations of gold foils in the aqueous phase were compared to irradiations of gold foils taking place in a frozen solid. Finally, a crystallization experiment based on [8] was conducted to understand which gold species are present in the aqueous phase after the irradiation process. In particular, the count rates of the characteristic gamma peaks of ^{198}Au at 411.8 keV were determined, analyzed and compared to each other.

2. Theory

2.1. Neutron induced radioactivity

While Marie and Pierre Curie were awarded the Nobel Prize in Physics in 1903 for their research in the field of radiation, it was their daughter Irène Joliot-Curie and her husband Frédéric Joliot-Curie who discovered induced radioactivity in 1934 [9]. Irradiating an aluminum foil with alpha particles lead to the creation of the radioisotope ^{30}P in an $^{27}\text{Al}(\alpha,n)^{30}\text{P}$ reaction, thereby showing the existence of artificial radioactivity [9].

Most commonly used nowadays is the activation of material via bombardment of neutrons. Since these particles are not electrically charged, they can be captured by the nucleus easily without Coulomb repulsion from electrons or protons [10], [11]. The target nucleus can absorb the neutrons during the bombardment in an effect called neutron capture and thereby increase its mass and create a new isotope, which in some cases is radioactive [10], [11], [12]. Initially, the new isotope is formed in an excited and unstable state (“compound nucleus”), from which it returns swiftly to its ground state, typically via emission of a gamma (γ) photon with a characteristic energy for each isotope [10], [11], [12]. This way, eventual alpha- or beta-decays are induced [10], [11].

The cross section σ of an isotope measures the probability of an interaction between the target nucleus and a neutron. Since this parameter is highly energy dependent, neutron energy is an essential factor in how likely a neutron is absorbed by a target nucleus [10], [11]. The lower the energy of the neutron, the higher the probability of the absorption of the neutron being absorbed by the nucleus becomes [10], [11]. Especially concerning reactor physics, there is a classification of neutrons based on their energy [10], [11]. If the kinetic energy of the free neutron is of the order of 0.025 eV, it is called a thermal neutron [10], [11]. From 0.025 eV to 0.4 eV neutrons are classified as epithermal [10], [11]. These can be grouped into the category of slow neutrons. Any neutrons with energies of the magnitude of MeV are called fast neutrons. In this thesis mainly slow neutrons will be looked at [10], [11].

2.2. Neutron Activation Analysis

Neutron Activation Analysis (NAA) was discovered and made famous by Nobel prize winner in chemistry George de Hevesy and Hilde Levi in 1935 [13] and 1936 [14], while previous work on this topic had already been done by Leó Szilárd and T. A. Chalmers in 1934 [4]. This analytical technique is based on the observation that the activity of a radionuclide that has been produced by bombardment with neutrons of the material that is proportional to the amount of target nuclei in the sample [13], [14], [15]. To this day, this technique is used, improved and innovated for the application in radiochemistry, archeology, forensics, biology, geology and many more disciplines involving the investigation of solid matter [15]. Isotopes produced upon neutron irradiation and their decays are subsequently analyzed [15]. The most important reactions mainly taking place in NAA are (n,γ) , (n,α) and $(n,2n)$ reactions [15]. Further important quantities involve the neutron cross section σ , which signifies the probability that a reaction takes place and the half-life of a radionuclide, which is the duration after which half of the starting material has decayed [15]. The activation of different nuclides, of which the target material is made of, leads to a multitude of different radionuclides, which naturally exhibit a variety of decay rates and radiations [15]. This can be used to determine the individual radioisotopes by measuring different decay intervals via high resolution detectors [15]. This procedure is called instrumental neutron activation analysis (INAA) when the sample is not damaged during his procedure [15]. If chemical methods are used to separate various radioisotopes of the irradiated material from each other, then the method is called radiochemical neutron activation analysis (RNAA) [15]. In general, any form of neutron activation analysis is used to calculate concentrations of unknown elements in the sample using a known standard sample where the concentration of the investigated radioisotopes is known [15]. In the end, this procedure delivers gamma-ray spectra of the measured radioisotopes and gives information about the concentrations and masses of the present nuclides in the examined material [15].

2.3. Szilárd-Chalmers effect

In 1934, Leó Szilárd and T. A. Chalmers, during their investigation of the so-called ‘Fermi effect’, later known as the neutron capture, irradiated liquid ethyl iodide with thermal neutrons. They observed the capture of the neutron by the iodine, yet did not expect what happened

afterwards [4]. Following the absorption of the neutron, the newly created ^{128}I was no longer found as a part of the organic compound, but extractable as an ion [4].

During the $^{127}\text{I}(n,\gamma)^{128}\text{I}$ reaction, the prompt γ photon emitted from the newly created radioisotope ^{128}I leaves behind a recoil [4]. Said recoil possesses an energy of approximately 100 eV, which exceeds the binding energy of the C-I bond of approximately 2.4 eV by far [4]. The difference by order of magnitudes between the recoil energy and the binding energy of the activated iodine atoms to the molecule leads to the release of the radioisotope ^{128}I from the ethyl iodide compound [4]. This process has been termed the ‘Szilárd-Chalmers effect’ and is nowadays used to create various no-carrier-added radioisotopes [16], [17]. The chemical changes after this nuclear reaction and how to fully take advantage of them is a topic of current research [18], [19].

One such possibility to use the Szilárd-Chalmers effect would be to create no-carrier-added radiogold, which later may be used as a radiopharmaceutical [20]. The idea consists of irradiating a gold foil in aqueous phase with neutrons, creating radiogold in the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction. During irradiation, thermal neutrons would be captured by the stable gold atoms in the foil, predominantly from the first few atomic layers, leading to the release of radiogold into the aqueous phase, following the recoil of the emitted γ photon. After extracting the now activated gold foil, the liquid should contain ^{198}Au atoms, which can be detected using a gamma detector.

However, certain aspects of this reaction remain unclear. For one, it is unknown if the activity of the gold foil in the experiment has an actual effect on the amount of radiogold released in the Szilárd-Chalmers effect. If this was the case, other effects, such as the beta-decay, would play a role in the release of radiogold. Additionally, the duration of irradiation and the amount of radiogold released in this reaction are linked, however, the exact connection is not known. The theory of the Szilárd-Chalmers effect and further linked parameters and their effect on the outcome is therefore not fully understood.

3. Materials and methods

3.1. Materials used in experiments

Gold consists of only one stable isotope (^{197}Au), which has a cross section for thermal neutron capture of 98.65 b. To produce ^{198}Au ($T_{1/2} = 2.694$ d), disc-shaped gold foils of the purest commercially available grade of 99.9 % with a radius of 3 mm and a thickness of 0.02 mm made by Goodfellow GmbH, Germany, were used. The aqueous phase, in which the gold foils were submerged during the experiments, consisted of in-house produced triple distilled water to keep the matrix as simple as possible. The liquid is needed to capture the activation products which are created during the course of the experiments. Two different types of screw top polyethylene vials, as shown in Figure 1, served as containers for the liquid and gold foils for the irradiation and measurement.

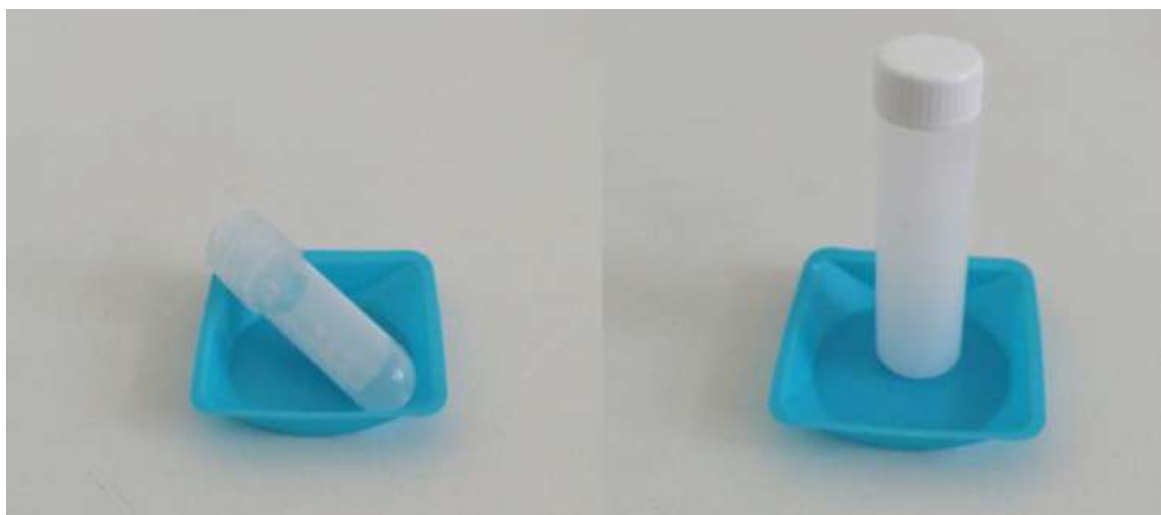


Figure 1 Vial used for irradiation holding liquid and gold foil (left) and vial used for measurement holding liquid after irradiation (right).

The vial, used for holding the gold foil during the irradiation, displayed in Figure 1, can fit up to 2 mL of liquid inside of it, has a diameter of 1.3 cm and a length of 4.8 cm. For the experiments, the vials containing the gold foils and liquid were placed in irradiation capsules, also called rabbits. These capsules, shown in Figure 2, are 12 cm long and have a diameter of 2.1 cm. Before closing these containers, a tampon was placed on top of the vial and moistened with triple distilled water, to fixate the position of the vial inside the irradiation capsule and soak up possible aqueous radiation products in case of leakage. This way, the consistent placement of the vial containing the gold foil and liquid are ensured during the irradiation process for every replicate of each experiment. Due to a vertical neutron flux gradient at the

irradiation location, the issue of consistent positioning of the vial inside the irradiation capsule was of utmost importance.



Figure 2 Irradiation capsule used to hold vial with gold foil in liquid during irradiation.

Each experiment starts by thoroughly washing and drying the gold foils and vials with triple distilled water. Afterwards, the vial used for irradiation is filled with 1.1 mL of triple distilled water, with the gold foil being immersed in the liquid. Before placing the sample inside the rabbit and sealing it with a damped tampon, the vial with the gold foil has to be cleaned again from the outside to wash away potential ubiquitous contamination of the surface by unwanted isotopes, which during irradiation, produce radioisotopes that inhibit the counting during the measurement. The same procedure has to be done with the irradiation container holding the vial with the gold foil immersed in the aqueous phase after being screwed shut. After washing, each vial and capsule was only being moved or touched using tongs and tweezers, to prevent said ubiquitous contamination.

For the first experiment, a table shaker, as seen in Figure 3, was used to shake the samples. Compared to other ways of shaking the gold foil immersed in the liquid, such as an ultrasonic bath, the shaking table was not able to destroy the surface of the gold foil or the gold foil itself.



Figure 3 Shaking table used to shake gold foil in liquid via rotation of vial.

To freeze the triple distilled water containing the gold foil during the experiment to compare the radiogold release in aqueous and solid phases to each other, in-house produced liquid nitrogen was used.

For the crystallization experiment gold(III) chloride trihydrate ($\text{H}[\text{AuCl}_4] \cdot 3 \text{H}_2\text{O}$, Sigma Aldrich 99+ % metal basis), ammonium nitrate (NH_4NO_3 , Sigma Aldrich p.a.) and ammonia vapor (NH_3) were used to verify the chemical nature of the via irradiation released radiogold.

3.2. Gamma-ray detector

After irradiation and preparation of the measurement vials, a low-level high-purity germanium (HPGe) detector, consisting of a 226 cm^3 germanium crystal with a 2.0 keV resolution at the 1332 keV ^{60}Co peak; 52.8 % relative efficiency, connected to a PC-based multi-channel analyzer with preloaded filter was used to analyze the spectra of the activation product [21]. The crystal has a diameter of 65 mm and a length of 68 mm and is shielded by layers of lead, zinc, cadmium and copper, which are necessary due to various absorptions of ionizing particles and emissions of x-rays [22]. The procedure of measuring the spectrum of a radioactive substance via its emission of gamma rays is called gamma spectroscopy [23], [24]. This method displays the radioisotopes present in the measured substance, since each gamma ray emitted from the radioactive sample has a characteristic, discrete energy which, after detection, can be assigned to the corresponding isotope [23], [24]. The count rates in all experiments were

measured and determined using the characteristic gamma peak of ^{198}Au at 412 keV [25]. To minimize errors due to slight fluctuations of the samples, the distance between the sample and the detector was set to 4 cm.

For visualization of the measured data by the gamma detector, the spectroscopy software Genie2000 Gamma Acquisition and Analysis by Canberra Industries was used. The preliminary count rates were determined via this software and afterwards corrected using the equations

$$cts(t) = cts_0 \cdot e^{-\lambda t}$$

and

$$\frac{cts}{s} = \frac{cts_0 \cdot \lambda}{1 - e^{-\lambda t_m}}$$

The first equation uses the net peak area given by the software (cts_0), the time between the end of the irradiation and the start of the measurement (t) and the decay rate constant (λ ; for gold: $\lambda = 2.977 \cdot 10^{-6} \text{ s}^{-1}$) to produce the corrected net peak area. The second equation gives the corrected count rate using the time of measurement (t_m). An example of a spectrum visualized by the spectroscopy software is shown in Figure 4.

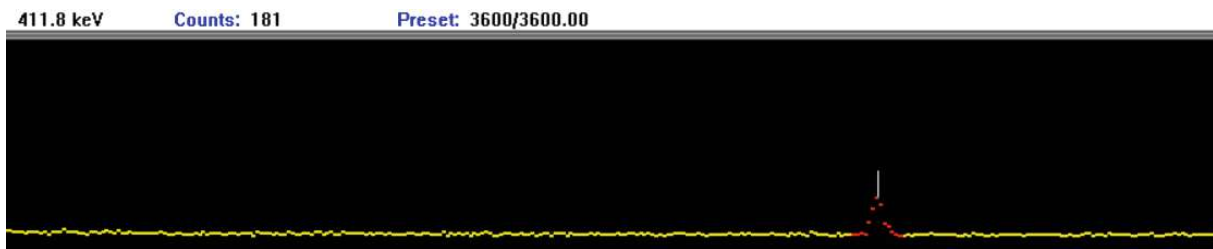


Figure 4 Typical spectrum with resulting ^{198}Au peak for the experiments.

3.3. TRIGA Mark-II research reactor

Built and installed over the course of three years starting from 1959 by General Atomics, USA, the TRIGA (Training, Research, Isotope, General Atomics) Mark-II research reactor in Vienna is designed as a research reactor for the purposes of isotope production and training [26], [27], [28], [29], [30]. The maximum power output is 250 kW_{th} [27], [28], [30]. The temperature of the fuel elements during the operation of the reactor is about 200 °C [27], [29], [30]. To control the neutron flux, three manually and automatically adjustable control rods, made of boron carbide, act as neutron absorbers [27], [29], [30]. At the central irradiation tube, the neutron flux density is $1 \times 10^{13} \text{ cm}^{-2}\text{s}^{-1}$ [27], [30], [31]. Heat production of the 250 kW_{th} reactor is very

moderate. Using a channel of the Danube Canal, the produced heat during the operation of the reactor is released into the river via a primary and secondary coolant circuit [27], [30], [31]. A homogeneous mixture of 91 wt% zirconium, 8 wt% uranium and 1 wt% hydrogen makes up the fuel [26], [27], [30]. The zirconium hydride in the fuel itself is used as the moderator, due to its negative temperature coefficient of reactivity, which results in the reactor having a self-moderating fuel [26], [27], [30]. Due to this property of the moderator, rising fuel temperature would result in a major decrease of reactivity [26], [27], [30].

The core of the reactor comprises of 90 core elements around the central irradiation tube [26], [27], [30], [31]. Five rings (B, C, D, E, F) of these elements surround the central irradiation tube at the position A1, as seen in Figure 5 [26], [27], [30], [31], [32], [33].

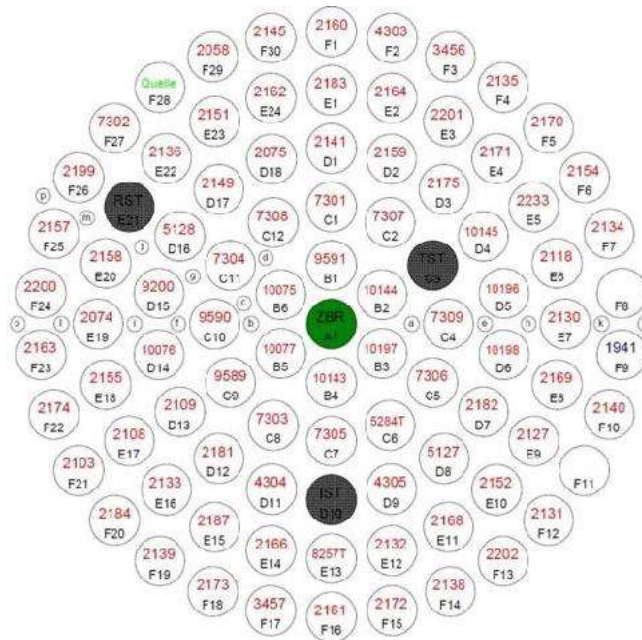


Figure 5 Schematic arrangement of core elements of the TRIGA Mark-II research reactor in Vienna [33].

A Sb-Be photo neutron source, emitting 6×10^6 neutrons per second, is used for the start of the nuclear reaction [26], [27], [30], [31]. Samples can be irradiated using the central irradiation tube, one of the five reflector irradiation tubes, one of the four neutron beam holes, the thermal column, the neutron radiography facility or the pneumatic transfer system [26], [30], of which the latter was used mostly for the experiments conducted that are described in this thesis.

The pneumatic tube system, also called rabbit system, has a transfer and is directly connected to the radiochemical laboratory, which allows working with the irradiated sample right after irradiation, without leaving said laboratory. It is installed in the F-ring positions F08 and F11 [33], [34], [35], seen in Figure 5. Temperature of the coolant in this position is around 60 °C.

Due to the short irradiation duration of mostly 120 seconds in the experiments, the high specific heat capacities of the water inside the vial and the surrounding air and finally the thermal shielding from the rabbit, effects that temperature may have had during the experiments are deemed negligible. A sample is delivered to the irradiation location in three seconds, which is the same time it takes for the container to return to the laboratory after irradiation [30]. At the irradiation location, the sample gets irradiated at a neutron flux density of $3.8 \cdot 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$, with a thermal-to-fast neutron flux density ratio of 2:1 in this position. The pneumatic transfer system and its connecting tubes to the radiochemical laboratory can be seen in Figure 6.



Figure 6 Entrance and starting point of the pneumatic transfer system.

The dry irradiation tube on the other hand, which was used particularly for the crystallization experiment, is located on the edge of the reactor core, in the graphite reflector [35]. Here, the sample needs to be guided manually to the irradiation location with the irradiation capsule containing the sample being lowered from the entry point to final location via a string, which is attached to said capsule. The irradiation takes place with a neutron flux density of $2 \cdot 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ [35].

4. Experiments

4.1. Comparison of radiogold production in aqueous phase prior and after neutron irradiation [36]

To investigate the degree of the extent of radiogold releases from the gold foil in the course of neutron irradiation, the first of the experiments was designed to compare the amount of liberated gold from the gold foil via irradiation to mechanical shaking in liquid. For this, two samples were prepared with a gold foil each placed inside a vial containing 1.1 mL of triple distilled water. The first vial was irradiated immediately for 120 seconds using the pneumatic transfer system and placed on the table shaker, seen in Figure 3, for exactly 60 minutes. The sample was shaken at a frequency of 200 rounds per minute (3.33 Hz). After the shaking process, 1 mL of the liquid was taken for gamma-ray measurement on the gamma detector. The second vial on the other hand was placed on the table shaker for 60 minutes first. Afterwards, the gold foil was taken out of the vial before the vial was ultimately irradiated for 120 seconds. Finally, 1 mL of the irradiated liquid was extracted for gamma ray measurement. Each extraction of liquid was conducted via a pipette. For this, after every transfer of liquid, the pipette tip was changed to negate cross-contamination of the samples.

Shaking the gold foil in the aqueous phase would result in gold atoms being detached mostly from the surface of the gold foil. While the first sample had already been irradiated with neutrons, this would mean that the gold atoms liberated from the surface of the foil via mechanical shaking would mostly be ^{198}Au . The second sample however, which was placed on the shaking table first, would only have released stable ^{197}Au into the aqueous phase. Since the gold foil was taken out of the second vial prior to irradiation, this would mean that these stable gold atoms will have been mostly activated due to the bombardment of neutrons and converted to ^{198}Au . The difference in activated gold atoms in the aqueous phase of both samples determined using gamma-ray measurement indicates the amount of the activation product being released into the liquid via irradiation with neutrons. If the count rate of ^{198}Au is substantially bigger in the case of the gold foil being irradiated with neutrons in the liquid prior to shaking compared to shaking the sample first and removing the gold foil from the vial before neutron irradiation, this would suggest a neutron induced process, such as the Szilárd-Chalmers Effect, is the cause for the liberation of radiogold.

The reason for using the shaking table with a low frequency instead of different high frequency sources, e.g. an ultrasonic bath, for mechanical movement of the gold foil immersed in the aqueous phase relates to the fragility of the gold foil. As a consequence of its dimensions, the gold foil is more likely to be ruptured or destroyed when using higher frequencies. In a preliminary experiment, the same experiment was conducted using an ultrasonic bath, which in the end lead to the destruction of the gold foil. Therefore, the gentle shaking environment created by the shaking table was the most viable option to move the gold foil immersed in liquid continuously and evenly for a longer period of time, without damaging the gold foil.

4.2. Investigation of radiogold release via cyclic irradiation processes [36]

A possible parameter determining the amount of released radiogold is the activity of the gold foil itself. Examination of the influence that the buildup of activity of the gold foil may have on the release of radiogold took place using cyclic irradiation of the same gold foil in fresh triple distilled water. For this, four vials with each 1.1 mL of triple distilled water were prepared with one gold foil was placed in one of them. The vial with the gold foil was irradiated inside the irradiation container for two minutes using the pneumatic transfer system. Following each irradiation cycle, the gold foil was taken out of the vial and placed in the next one containing 1.1 mL of fresh triple distilled water, which was subsequently placed in a new rabbit for another two-minute irradiation. This process was repeated until the same gold foil was ultimately irradiated four times, each for two minutes. After each irradiation, 1 mL of the vial containing the gold foil was pipetted from the vial into a separate measurement vial and taken for gamma measurement. Pipette tips were changed after each extraction of liquid to negate cross-contamination of the samples.

With each neutron irradiation, the activity of the gold foil increases. This means that the decay rate is becomes bigger, leading to more decays in the gold foil. If the measured count rate of ^{198}Au in the irradiated liquid becomes bigger with each subsequent irradiation of the gold foil, this would indicate, that due to the resulting higher activity of the gold foil, beta and gamma radiation of the gold foil would play a part in the detachment of radiogold from the gold foil. If however, the yield for no-carrier added ^{198}Au stays largely unaffected by the higher activity of the gold foil, the observed phenomenon is mostly taking place due to the neutrons themselves.

4.3. Examination of linear connection between radiogold release and irradiation duration [36]

One could raise the activity of the gold foil not only via cyclic irradiation but simply also by increasing the duration of neutron irradiation. However, this would lead to a longer neutron bombardment of the sample, which is why it is not a viable option to investigate the influence of the activity on the detachment of ^{198}Au from the gold foil. By increasing the duration of the sample sitting at the irradiation location by manifold, the count rate of ^{198}Au during measurement should, in theory, be a multiple of the original yield of ^{198}Au . This linear relationship between the release of radiogold from the gold foil and the irradiation duration was scrutinized by irradiation of two samples for different amount of time periods.

Two vials with a gold foil inside of them, each filled with 1.1 mL of triple distilled water were prepared. Both were placed sequentially in a rabbit and irradiated via the pneumatic transfer system. The first was bombarded with neutrons for two minutes. The other one was irradiated with neutrons for ten minutes. In both cases, 1 mL of the liquid, in which the gold foils were irradiated in, was extracted and taken for gamma-ray measurement. Every extraction of irradiated liquid happened via pipetting and using a new pipette tip for each sample. If a linear connection of the release of ^{198}Au and the duration of irradiation with neutrons exists, then the yield of ^{198}Au should be five times bigger in the case of the sample being irradiated for ten minutes compared to the sample being irradiated for two minutes, since the amount of time of the second sample at the irradiation location increased fivefold. Non-linearity would indicate different neutron-based processes taking place, such as sputtering due to fast neutrons hitting the surface of the gold foil. In case the former experiment regarding the activity of the gold foil shows significant influence of the activity on the release of radiogold, beta and gamma radiation could also contribute to the detachment of ^{198}Au from the gold foil, if no linear connection can be found between the irradiation duration and the release of ^{198}Au into the aqueous phase.

The decision to increase the irradiation duration to ten minutes was made based on the fact that a fivefold increase in yield of radiogold would be big enough to be contributed to the possible linear connection between irradiation duration and release of ^{198}Au . A longer period of time at the irradiation location would lead to a possible saturation of the activation of the gold foil, which would distort the results.

4.4. Comparison of radiogold release via irradiation in aqueous and solid phase

Having investigated the behavior of gold foils being irradiated with neutrons in an aqueous phase for the release of radiogold, the question of how this phenomenon occurs when the gold foil is fixated in a solid matrix. Since all previous experiments were conducted using triple distilled water, the simplest and most comparable matrix the gold foil can be placed in is ice, made from frozen triple distilled water.

For this experiment, two vials were prepared, each filled with 1.1 mL of triple distilled water and a gold foil placed inside of them. One of them was irradiated inside the rabbit using the pneumatic transfer system for two minutes. Afterwards, 1 mL was taken from the vial for gamma counting. The second vial was frozen first using liquid nitrogen in a container. The vial was held vertically for one minute inside the canister containing the freezing agent via tongs, as to freeze the gold foil at the bottom of the vial. Following the freezing procedure, the vial was placed inside the irradiation container and irradiated with neutrons using the pneumatic transfer system for two minutes. Finally, after letting the ice inside the vial melt after irradiation, which took a few minutes, 1 mL of the irradiated liquid was taken for gamma-ray measurement.

Further irradiation using frozen triple distilled water took place, to understand the influence the crystallization process of the triple distilled water has on the detachment of gold atoms from the gold foil and especially its surface. For this control experiment the gold foil was placed inside the irradiation vial inside 1.1 mL of triple distilled water and frozen using liquid nitrogen, as described in the paragraph above. However, this time prior to irradiation the sample was left at room temperature in the laboratory for the ice to melt. After liquifying the ice made from triple distilled water, the gold foil was taken out of the vial. Finally, to imitate the initial experiment regarding frozen solutions, the vial placed inside the container with liquid nitrogen to freeze the liquid again and irradiated using the pneumatic transfer system for two minutes. In the end, 1 mL of the irradiated liquid was taken for gamma-ray measurement. This way, possible surface damage sustained by the gold foil leading to the detachment of gold atoms from the foil due to the crystallization of the surrounding aqueous phase can be accounted for and be analyzed.

4.5. Crystallization experiment for speciation of produced ^{198}Au [36]

Identifying the species of radiogold created by the irradiation of gold foils immersed in triple distilled water with neutrons was a further and final step in exploring the details and properties of the results of the conducted experiments. Gamma-ray measurements showed that indeed neutron bombardment of the gold foil produced ^{198}Au , however, it is not obvious if this radiogold is ionic or not. This is an important characteristic that has to be investigated if the via neutron irradiation produced radiogold is used further for different purposes other than research, e.g. as a radiopharmaceutical. For this, a crystallization experiment using the ^{198}Au species created during neutron irradiation while placed in triple distilled water, was carried out. It was hypothesized that the released radiogold in this process was aqueous $^{198}\text{Au}^{3+}$. To verify this assumption, the crystallization of $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$ was carried out using a well-established and highly reproducible synthesis of a gold(III) compound [8], [37].

The process of the synthesis in this experiment is potentially dangerous, if not conducted exactly according to protocol. Therefore, protective measures are recommended [38]. For this synthesis, $\text{H}[\text{AuCl}_4] \cdot 3 \text{H}_2\text{O}$ (254.16 mg, 0.645 mmol; Sigma Aldrich, 99.9 + % metal basis) were dissolved in 6.354 mL triple distilled water, of which 1.092 mL contained ^{198}Au atoms due to this amount of liquid being used in a prior irradiation process involving a gold foil being irradiated immersed in 1.1 mL triple distilled water for 30 minutes using the dry irradiation tube. The solution had a gold-concentration of 20 mg/mL and was later saturated with NH_4NO_3 (Sigma Aldrich, p.a.) at room temperature. This process is needed to avoid the onset of explosive fulminating gold upon contact with NH_3 with Au^{3+} [8], [39]. Via filtration (washing with fresh saturated NH_4NO_3 solution) the residual NH_4NO_3 was removed from the solution. Afterwards, NH_3 , which was taken from a bottle of 25 % solution vapor was added to the solution via a syringe until the yellow color disappeared. This process takes a while; however, the disappearance of the color is sudden. Just shortly after this procedure, crystallization of white needle-like shapes of $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$ commenced. After 60 minutes, when the crystallization was allowed to develop, the white product $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$ was filtrated and washed with cold (0 °C) H_2O and cold (4 °C) methanol (Sigma Aldrich, p.a.). Following this, the product was dried at 50 °C to ultimately weigh it. In the end, the yield of $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$ was 1650.09 mg, which equates to 57%. From $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$, 88.39 mg were mixed and dissolved with 5 mL of H_2O at room temperature for low-level gamma-ray measurement. For

the comparison at the end of the experiment, 5 mL of the mother liquor, which after crystallization had a volume of 12.8 mL, were measured for via neutron irradiation of a gold foil produced $^{198}\text{Au}_{\text{aq}}$. Finally, INAA was used to determine the stable gold content in both the final solution of $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$ and the mother liquor.

5. Results

5.1. Yield of ^{198}Au through neutron irradiation [36]

The experiment described in section 4.1. was conducted in four replicates. Table 1 displays the measured masses of the gold foils used for the replicates. By knowing the weights of the gold foils, further mathematical calculations are possible regarding the release of gold atoms via irradiation with neutrons, such as out of which atom layer of the gold foil said detached atoms stem from. Table 2 and Table 3 show the measured masses of the irradiation and measurement vials used in the experiment for each replicate with and without liquid. Weighing each vial with and without the liquid ensures the consistency and accuracy of pipetting the right amount of triple distilled water.

Table 1 Masses of gold foils used in the experiment concerning the shaking table. Gold foil No. 1 was placed in the vial, which was irradiated first and then shaken. Gold foil No. 2 was placed in the vial for shaking first and taken out afterwards.

	Replicate No. 1	Replicate No. 2	Replicate No. 3	Replicate No. 4
Gold foil No. 1	9.52±0.01 mg	8.60 mg±0.01	9.36±0.01 mg	9.14±0.01 mg
Gold foil No. 2	9.26±0.01 mg	9.02±0.01 mg	9.37±0.01 mg	8.84±0.01 mg

Table 2 Masses of vials with and without 1.1 mL of triple distilled water used for the experiment including the shaking table.

	Irradiation vial No. 1 (empty)	Irradiation vial No. 1 (with 1.1 mL triple distilled water)	Irradiation vial No. 2 (empty)	Irradiation vial No. 2 (with 1.1 mL triple distilled water)
Replicate No. 1	2.12920±0.00001 g	3.23785±0.00001 g	2.14006±0.00001 g	3.24546±0.00001 g
Replicate No. 2	2.14787±0.00001 g	3.24790±0.00001 g	2.17216±0.00001 g	3.27407±0.00001 g
Replicate No. 3	2.14484±0.00001 g	3.26950±0.00001 g	2.14155±0.00001 g	3.29788±0.00001 g
Replicate No. 4	2.12956±0.00001 g	3.27059±0.00001 g	2.14643±0.00001 g	3.26943±0.00001 g

Table 3 Masses of vials with and without 1 mL of triple distilled water used for the experiment including the shaking table.

	Measurement vial No. 1 (empty)	Measurement vial No. 1 (with 1 mL triple distilled water)	Measurement vial No. 2 (empty)	Measurement vial No. 2 (with 1 mL triple distilled water)
Replicate No. 1	3.60827±0.00001 g	4.60859±0.00001 g	3.64745±0.00001 g	4.64490±0.00001 g
Replicate No. 2	3.65400±0.00001 g	4.65217±0.00001 g	3.66031±0.00001 g	4.65729±0.00001 g
Replicate No. 3	3.57701±0.00001 g	4.52883±0.00001 g	3.64918±0.00001 g	4.60911±0.00001 g
Replicate No. 4	3.63947±0.00001 g	4.52552±0.00001 g	3.66124±0.00001 g	4.61755±0.00001 g

As Table 4 shows, production of ^{198}Au via neutron irradiation of a gold foil immersed in triple distilled water is larger by more than one order of magnitude compared to mechanical shaking of a gold foil in the same aqueous phase and activation of only the liquid. This significant result is also displayed in Figure 7, where the individual replicates of this experiment and their respective count rates of ^{198}Au are portrayed. The count rates have been corrected using the equations (1) and (2) mentioned in Section 3.2. Due to this result being consistent in the order of magnitude considering the difference in the count rate of ^{198}Au , it is safe to assume that the irradiation of the gold foil with neutrons is the reason for the liberation of activated gold atoms. However, subsequent effects, such as the beta-decay, cannot be excluded from having an impact on the release of ^{198}Au yet. This will be topic of the following experiment.

Table 4 Comparison of results of the experiment regarding the shaking table. Corrected count rates of ^{198}Au in the aqueous phase of each replicate.

	Replicate No. 1	Replicate No. 2	Replicate No. 3	Replicate No. 4
Irradiation taking place prior to shaking the sample	$(3.205 \pm 0.351) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(2.320 \pm 0.208) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(1.931 \pm 0.303) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(3.403 \pm 0.342) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$
Shaking of the sample with removal of gold foil prior to irradiation	$(6.194 \pm 0.543) \cdot 10^{-3} \frac{\text{cts}}{\text{s}}$	$(1.582 \pm 0.380) \cdot 10^{-3} \frac{\text{cts}}{\text{s}}$	$(3.206 \pm 0.374) \cdot 10^{-3} \frac{\text{cts}}{\text{s}}$	$(4.153 \pm 0.396) \cdot 10^{-3} \frac{\text{cts}}{\text{s}}$

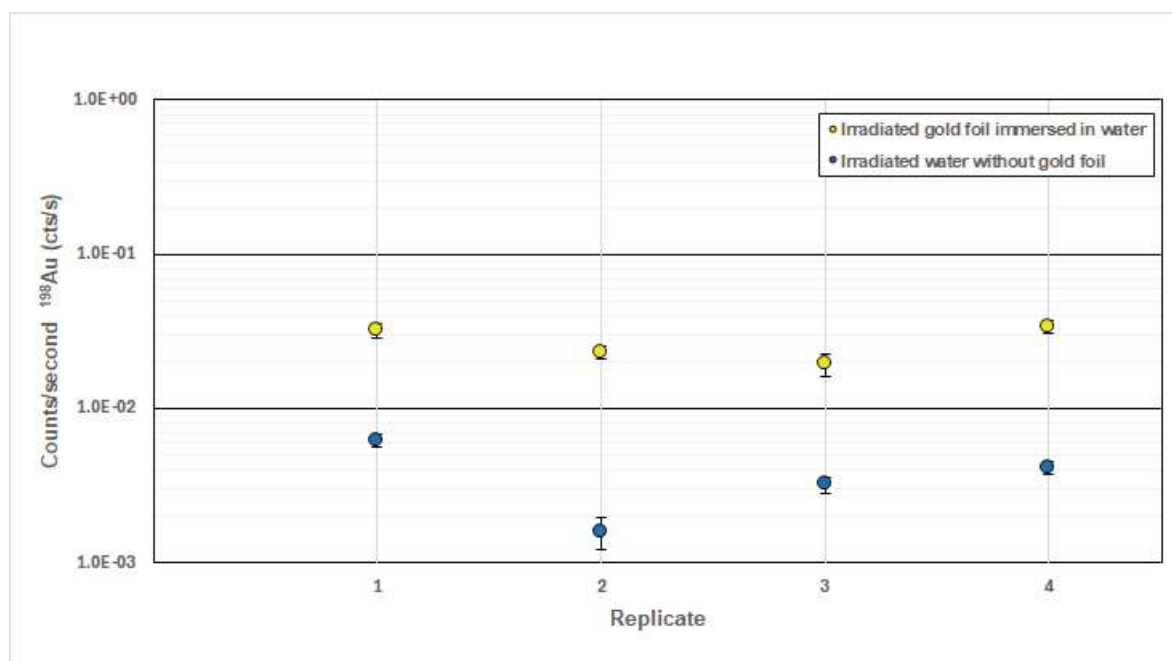


Figure 7 Visualization of the results regarding the experiment conducted using the shaking table. The blue dots display the corrected count rate of the samples, which were irradiated without the gold foil inside the vial, while the yellow dots represent the count rate of the irradiation of the sample containing a gold foil inside the vial.

Interestingly, even though the duration of the samples being irradiated for were identical and the neutron flux densities were the same (considering small fluctuations), the resulting corrected count rates seen in Table 4 and Figure 7 are not completely reproducible. The irradiations took place on different days, which would slightly vary the neutron flux density at the terminal irradiation location, however these fluctuations are minor and unlikely to impact the measurements in such a drastic way. One possibility is that during the transfer of the sample to the irradiation location, the gold foil emerges from the aqueous phase and, for example, sticks

to the top of the lid due to the speed at which the sample is being transferred with. Therefore, the positioning of the gold foil would not be uniform for each replicate, leading to different amounts of neutrons hitting the gold foil due to the vertical gradient regarding the neutron flux at the end of the rabbit system. To gauge this problem, a bigger amount of replicates would be needed to understand if the issue can be solved statistically.

5.2. Influence of activity of sample on radiogold release [36]

The experiment concerning the cyclic irradiation of the same gold foil in fresh triple distilled water, which was described in Section 4.2. was conducted in two replicates. Each replicate consisted of four samples, which means the same gold foil was irradiated four times. Each irradiation took place straight after the prior one, as to minimize the involvement of possible fluctuations of the neutron flux density. Tables 5 and 6 show the measured masses of the vials used for irradiation for each replicate in an empty and a with 1.1 mL of triple distilled water filled state. Tables 7 and 8 show the measured masses of the vials used for gamma-ray measurement for each replicate with and without 1 mL of the irradiated liquid. Finally, Table 9 shows the weighed masses of the gold foils used for each replicate.

Table 5 Weighed masses of empty irradiation vials used for the experiment concerning cyclic irradiation of the same gold foil.

	Irradiation vial No. 1 (empty)	Irradiation vial No. 2 (empty)	Irradiation vial No. 3 (empty)	Irradiation vial No. 4 (empty)
Replicate No. 1	2.14080±0.00001 g	2.13751±0.00001 g	2.17432±0.00001 g	2.17705±0.00001 g
Replicate No. 2	2.13140±0.00001 g	2.15864±0.00001 g	2.17434±0.00001 g	2.17325±0.00001 g

Table 6 Weighed masses of irradiation vials containing 1.1 mL of triple distilled water used for the experiment concerning cyclic irradiation of the same gold foil.

	Irradiation vial No. 1 (with 1.1 mL triple distilled water)	Irradiation vial No. 2 (with 1.1 mL triple distilled water)	Irradiation vial No. 3 (with 1.1 mL triple distilled water)	Irradiation vial No. 4 (with 1.1 mL triple distilled water)
Replicate No. 1	3.25767±0.00001 g	3.27629±0.00001 g	3.24736±0.00001 g	3.29801±0.00001 g
Replicate No. 2	3.24988±0.00001 g	3.27362±0.00001 g	3.28734±0.00001 g	3.28698±0.00001 g

Table 7 Weighed masses of empty measurement vials used for the experiment concerning cyclic irradiation of the same gold foil.

	Measurement vial No. 1 (empty)	Measurement vial No. 2 (empty)	Measurement vial No. 3 (empty)	Measurement vial No. 4 (empty)
Replicate No. 1	3.65139±0.00001 g	3.61955±0.00001 g	3.67730±0.00001 g	3.56361±0.00001 g
Replicate No. 2	3.57946±0.00001 g	3.61768±0.00001 g	3.64016±0.00001 g	3.60816±0.00001 g

Table 8 Weighed masses of measurement vials filled with 1 mL of neutron-irradiated triple distilled water used for the experiment concerning cyclic irradiation of the same gold foil.

	Measurement vial No. 1 (with 1 mL triple distilled water)	Measurement vial No. 2 (with 1 mL triple distilled water)	Measurement vial No. 3 (with 1 mL triple distilled water)	Measurement vial No. 4 (with 1 mL triple distilled water)
Replicate No. 1	4.57295±0.00001 g	4.60307±0.00001 g	4.54644±0.00001 g	4.64408±0.00001 g
Replicate No. 2	4.57280±0.00001 g	4.57768±0.00001 g	4.58138±0.00001 g	4.50750±0.00001 g

Table 9 Masses of the gold foils used for each replicate of the experiment concerning cyclic irradiation of the same gold foil.

	Replicate No. 1	Replicate No. 2
Gold foil	9.27±0.01 mg	9.00±0.01 mg

Due to the cyclic irradiation of the same gold foil, activity of the sample increases to a degree, where high precision and caution is needed while working with long-distance tools, such as pincers. The resulting corrected count rates of the released ^{198}Au are shown in Table 10 and displayed in Figure 8. Due to a technical error, the count rate for ^{198}Au in the aqueous phase after the second irradiation cycle was not measurable. This, however, did not impede the following irradiations. Both Table 10 and Figure 8 show fluctuations regarding the count rate with each irradiation cycle throughout the series, which are within the error margin of the measurement.

Table 10 Corrected count rates of the respective irradiation cycle of both replicates. Please note that due to a technical difficulty, the count rate of the second irradiation cycle of the first replicate was not measurable.

	Count rate after 1 st irradiation cycle	Count rate after 2 nd irradiation cycle	Count rate after 3 rd irradiation cycle	Count rate after 4 th irradiation cycle
Replicate No. 1	$(4.925 \pm 0.400) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	-	$(4.634 \pm 0.369) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(4.611 \pm 0.377) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$
Replicate No. 2	$(3.132 \pm 0.337) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(2.988 \pm 0.317) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(3.064 \pm 0.319) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(2.615 \pm 0.301) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$

The x-axis in Figure 8 represents the number of irradiations, while the y-axis gives the count rate of ^{198}Au measured in the aqueous phase. The development of the count rate is given by the dotted connection of the measured points. While the first replicate shows a steady and slow decline with each irradiation, the second replicate shows an unstable behavior. Here, the count rate of ^{198}Au in the aqueous phase decreases first when the gold foil is being irradiated for a second time and increases when being irradiated for a third time. The final irradiation cycle led to the measurement of a way smaller count rate of ^{198}Au in the aqueous phase, compared to the

first one measured in this replicate. For some reason, the first irradiation cycle of both replicates yields the highest count rate of ^{198}Au in the aqueous phase of the respective replicate.

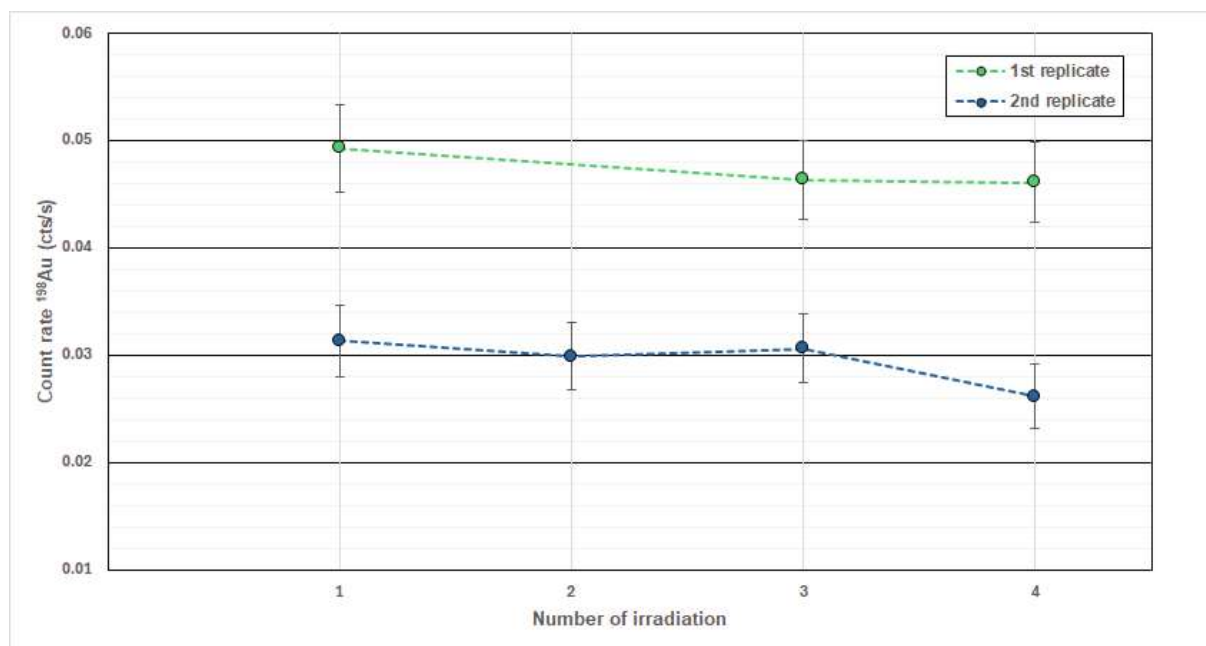


Figure 8 Evolution of the corrected count rate of the liquid with gold foil inside after each irradiation in fresh triple distilled water. Please note that due to technical difficulties the result for the second irradiation cycle of the first replicate was not measurable.

The diagram shows the very clear and big difference between the count rates of the replicates. This means that even though both replicates of the experiment were conducted with almost identical gold foils in the same manor, the different amounts of radiogold are released from each gold foil. The yield of carrier-free ^{198}Au in the aqueous phase using the gold foil in the first replicate is consistently higher than the yield of the other gold foil. The exact reason for this behavior is unclear, however the slight fluctuation of masses of the used gold foils and differences in texture of the surfaces may lead to these observations.

The experiment shows that with increasing activity of the gold foils due to quick successive irradiation cycles the count rate does not significantly and not consistently increase, which means that the yield in no-carrier-added ^{198}Au in the aqueous phase is not affected by the increase in activity. Since not significantly more radiogold atoms are detached from the gold foils due to subsequent irradiations and no consistency can be found regarding this fact, it is safe to assume that the neutron irradiation itself provides the observed results. Beta- and gamma-radiation on the other hand seem not to have a measurable or significant impact on the release of radiogold from the gold foils.

The first two experiments indicate that the observed phenomenon concerning the release of radiogold from the gold foil stems mainly from the neutron irradiation. However, in Section 2.1. the difference between thermal and fast neutrons is mentioned. Therefore, it is still unclear which class of neutron is mainly responsible for the observed effect.

5.3. Connection between release of radiogold and duration of irradiation [36]

Following the previous experiment, described in section 4.2. and 5.2., examining the yield of ^{198}Au with the increase of activity of the gold foil and coming to the conclusion, that only the neutron irradiation is responsible for the release of radiogold from the gold foil, it is now possible to investigate the effects that longer periods of neutron irradiation of the sample have on the release of radiogold. The third experiment, which was described in Section 4.3., was conducted to investigate if a linear connection exists between the irradiation duration of the sample and the yield of no-carrier-added ^{198}Au in the aqueous phase. For this, the experiment was carried out three times. Every replicate was conducted on the same day and each irradiation of one replicate was carried out subsequently, as to minimize the involvement of possible fluctuations of the neutron flux density. For each replicate, there were two gold foils, each immersed in triple distilled water. One of these samples was irradiated for two minutes, the other for ten minutes. The masses of the vials used for irradiation and gamma-ray measurement with and without liquid inside of them, are given in Table 11 and Table 12, while the masses of the gold foils used in this experiment are given in Table 13.

Table 11 Masses of the irradiation vials in an empty state and filled with 1.1 mL of triple distilled water. Irradiation vial No. 1 describes the container, which was irradiated for two minutes. Irradiation vial No. 2 describes the container, which was irradiated for ten minutes.

	Irradiation vial No. 1 (empty)	Irradiation vial No. 1 (with 1.1 mL triple distilled water)	Irradiation vial No. 2 (empty)	Irradiation vial No. 2 (with 1.1 mL triple distilled water)
Replicate No. 1	2.13751±0.00001 g	3.25490±0.00001 g	2.13628±0.00001 g	3.25321±0.00001 g
Replicate No. 2	2.14040±0.00001 g	3.25365±0.00001 g	2.13393±0.00001 g	3.24762±0.00001 g
Replicate No. 3	2.15756±0.00001 g	3.26712±0.00001 g	2.13667±0.00001 g	3.24465±0.00001 g

Table 12 Masses of the measurement vials in an empty state and filled with 1 mL of neutron-irradiated triple distilled water. Measurement vial No. 1 is the vial containing the liquid, which was irradiated for two minutes. Measurement vial No. 2 is the vial containing the liquid, which was irradiated for ten minutes.

	Measurement vial No. 1 (empty)	Measurement vial No. 1 (with 1 mL triple distilled water)	Measurement vial No. 2 (empty)	Measurement vial No. 2 (with 1 mL triple distilled water)
Replicate No. 1	3.62860±0.00001 g	4.60814±0.00001 g	3.60250±0.00001 g	4.59341±0.00001 g
Replicate No. 2	3.61711±0.00001 g	4.61698±0.00001 g	3.64469±0.00001 g	4.643051±0.00001 g
Replicate No. 3	3.57181±0.00001 g	4.57110±0.00001 g	3.58516±0.00001 g	4.58371±0.00001 g

Table 13 Masses of the gold foils used in the experiment regarding different irradiation durations. Gold foil No. 1 describes the sample, which was irradiated for two minutes. Gold foil No. 2 describes the sample, which was irradiated for ten minutes.

	Replicate No. 1	Replicate No. 2	Replicate No. 3
Gold foil No. 1	9.40±0.01 mg	9.12±0.01 mg	9.40±0.01 mg
Gold foil No. 2	9.17±0.01 mg	9.54±0.01 mg	9.38±0.01 mg

Hypothetically, if there was a linear connection between the period of time which the gold foil was irradiated with neutron while placed in triple distilled water and the yield of ^{198}Au , then the count rate of ^{198}Au from measured the irradiated liquid must increase proportionally to the increase in irradiation duration. In this case, where the irradiation duration of one gold foil was two minutes, while the other one was irradiated for ten minutes, the resulting difference in count rates should roughly be a factor of five, since the second gold foil was irradiated five times as long as the first one. The results of the three replicates carried out in the course of this experiment are given in Table 14 and visualized in Figure 9. The count rates have been corrected using the equations (1) and (2) mentioned in Section 3.2. Both show mainly unreproducible, mixed behavior.

Table 14 Corrected count rates of respective replicates regarding irradiation duration of two minutes and ten minutes.

	Replicate No. 1	Replicate No. 2	Replicate No. 3
Two-minute irradiation	$(3.772 \pm 0.368) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(4.545 \pm 0.395) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(3.423 \pm 0.352) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$
Ten-minute irradiation	$(7.801 \pm 0.553) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(11.140 \pm 0.620) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(16.707 \pm 0.076) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$

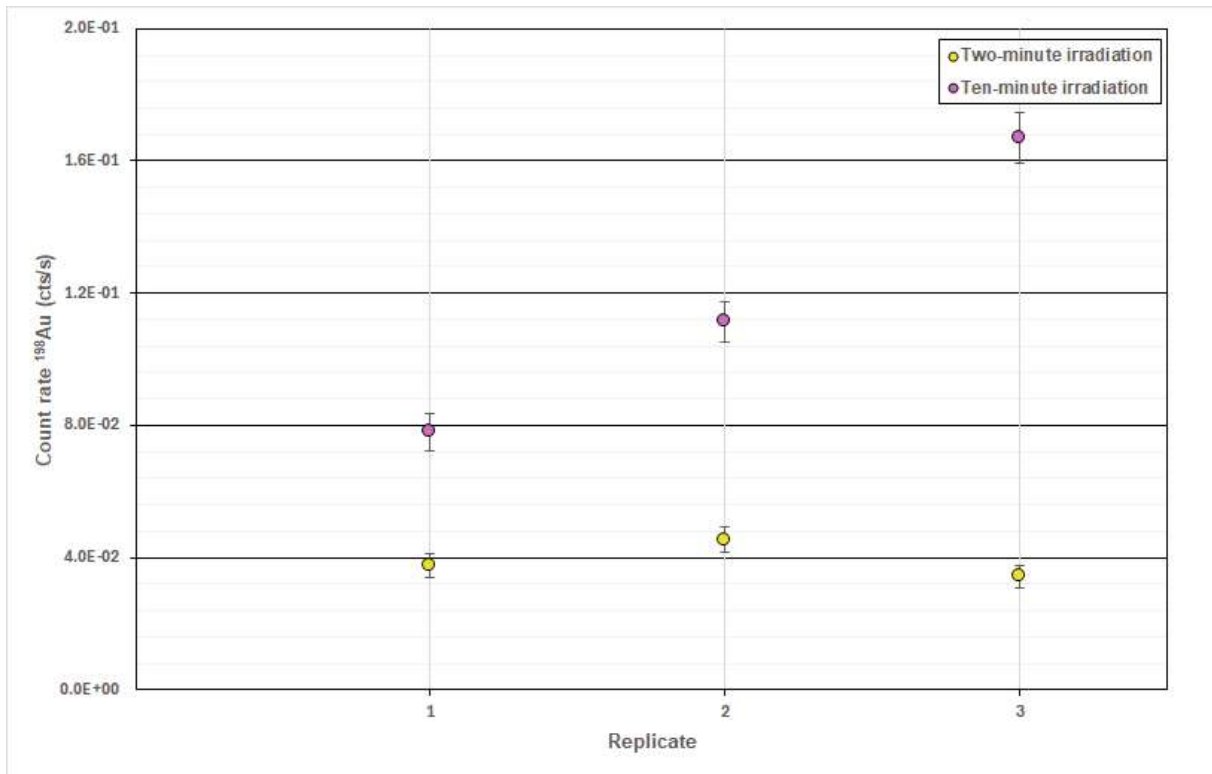


Figure 9 Comparison of the corrected count rates of the released ^{198}Au into the aqueous phase from a gold foil irradiated for two minutes (yellow) and ten minutes (pink).

While the first two replicates show no factor of five between their two-minute irradiation count rates and their respective ten-minute irradiation count rates, the count rate for ^{198}Au of the ten-minute irradiation of the third replicate roughly corresponds to being five times as big as its two-minute counterpart, as Table 14 shows.

These mixed results display that a linear connection between the release of radiogold from the gold foil via neutron irradiation and the duration of the irradiation is possible, as can be seen from the results of the third replicate. However, these results are not reproducible, as observed with the other two replicates. Therefore, slight uncontrollable circumstances during the irradiation process, such as the gold foil not being fully immersed in the liquid due to the transport to the irradiation location via the pneumatic transfer system and possible interference of changing neutron flux densities during a longer irradiation especially concerning the impact of thermal and fast neutrons on the sample lead to this non-linearity and non-reproducibility.

5.4. Yield of ^{198}Au in solid phase compared to aqueous phase

Up to this point, every replicate of each experiment was conducted using liquid matrix in which the gold foil was immersed in. This choice made measuring the activity of the aqueous phase after neutron irradiation simple, due to the transfer of the liquid from the irradiation vial to the measurement vial being done via pipetting. Now that it is known that radiogold is released from a gold foil into an aqueous phase when being irradiated with neutrons, the question arises what would happen if irradiation of the gold foil takes place in a solid phase. In this case, frozen triple distilled water was used.

The weights of the measurement and irradiation vials used in this experiment are given in Table 15 and Table 16. The weights of the gold foils used in this experiment are shown in Table 17. The masses of the irradiation and measurement vial and the gold foil used for the control experiment are displayed in Table 18.

Table 15 Masses of the irradiation vials concerning the solid phase experiment. Irradiation vial No.1 corresponds to the sample with liquid used as the matrix. Irradiation vial No.2 corresponds to the sample with ice used as the matrix.

	Irradiation vial No. 1 (empty)	Irradiation vial No. 1 (with 1.1 mL triple distilled water)	Irradiation vial No. 2 (empty)	Irradiation vial No. 2 (with 1.1 mL frozen triple distilled water)
Replicate No. 1	2.13848±0.00001 g	3.24226±0.00001 g	2.14545±0.00001 g	3.25392±0.00001 g
Replicate No. 2	2.16887±0.00001 g	3.27448±0.00001 g	2.14426±0.00001 g	3.24979±0.00001 g

Table 16 Masses of the measurement vials concerning the solid phase experiment. Measurement vial No.1 corresponds to the sample with liquid used as the matrix. Measurement vial No.2 corresponds to the sample with ice used as the matrix.

	Measurement vial No. 1 (empty)	Measurement vial No. 1 (with 1 mL triple distilled water)	Measurement vial No. 2 (empty)	Measurement vial No. 2 (with 1 mL triple distilled water)
Replicate No. 1	3.61953±0.00001 g	4.60722±0.00001 g	3.58512±0.00001 g	4.569892±0.00001 g
Replicate No. 2	3.68426±0.00001 g	4.65014±0.00001 g	3.63924±0.00001 g	4.62903±0.00001 g

Table 17 Masses of the gold foils used in the solid phase experiment. Gold foil No. 1 refers to the sample using liquid as the matrix. Gold foil No. 2 refers to the sample using ice as the matrix.

	Replicate No. 1	Replicate No. 2
Gold foil No. 1	9.36±0.01 mg	9.41±0.01 mg
Gold foil No. 2	9.44±0.01 mg	9.16±0.01 mg

Table 18 Masses of the irradiation and measurement vials and gold foils used in the control experiment for the solid phase experiment.

	Irradiation vial (empty)	Irradiation vial (with 1.1 mL triple distilled water)	Measurement vial (empty)	Measurement vial (with 1 mL of triple distilled water)	Gold foil
Replicate No. 1	2.14557±0.00001 g	3.23581±0.00001 g	3.59583±0.00001 g	4.56338±0.00001 g	9.38±0.01 mg
Replicate No. 2	2.14901±0.00001 g	3.23004±0.00001 g	3.55720±0.00001 g	4.52928±0.00001 g	9.39±0.01 mg

Due to the melting process of the ice starting straight after the sample was frozen via liquid nitrogen, the preparation of the irradiation capsule had to happen very fast. The time between the end of the freezing process of the sample and start of the irradiation differed for the two replicates for about six seconds. This amount of time is small compared to the duration it takes for the ice to melt. Therefore, the mentioned difference in time which occurred during the preparation of the samples between the two replicates is negligible and has little to no effect on the results of the experiments. The corrected count rates of the released ^{198}Au from the gold foils are listed in Table 19 and displayed in Figure 10.

Table 19 Corrected count rates for ^{198}Au measured from the liquid being used as the matrix for the gold foil and ice being used as the matrix for the gold foil.

	Irradiation using ice as matrix	Irradiation using water as matrix
Replicate No. 1	$(21.154 \pm 0.796) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(3.109 \pm 0.033) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$
Replicate No. 2	$(8.708 \pm 0.507) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$	$(2.724 \pm 0.030) \cdot 10^{-2} \frac{\text{cts}}{\text{s}}$

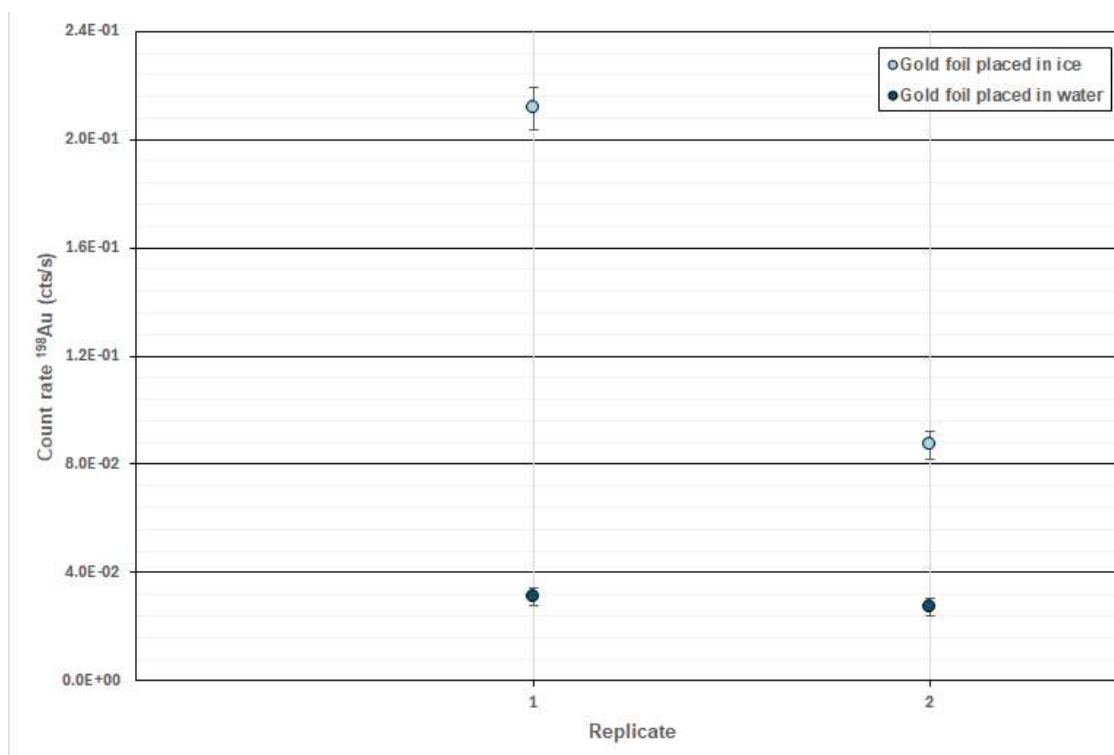


Figure 10 Corrected count rates of the samples using ice as a matrix for the irradiation (light blue) and using water as a matrix for the irradiation (dark blue).

As Figure 10 shows, the corrected count rates of the gold foil being immersed in liquid for both replicates are similar to each other and to the results of the former experiments, where the irradiation of the same preparation took place. The irradiation of the sample where the gold foil was placed in ice however shows drastically and non-reproducible results, which are not similar to each other. In fact, the ratio of the corrected count rates seen in Table 19 for each replicate are vastly different. The first replicate shows a corrected count rate for the solid phase which is bigger compared to the corrected count rate for the liquid phase by a factor of roughly 6.8, while the factor between the corrected count rates of the second replicate is 3.1. In both cases, the

yield in ^{198}Au released by the gold foil via neutron irradiation is higher when the sample is frozen.

Although the exact reason(s) for this observed phenomenon is unclear, the idea of the crystallization of water when changing phase to a solid and thereby damaging the surface of the gold foil can be eliminated. The presumption is based on the idea that during the freezing process, the ice crystals formed “dig” into the gold foil, damaging the surface of the gold foil and thereby releasing a greater amount of gold atoms into the matrix. These liberated gold atoms then get subsequently activated during neutron irradiation, causing the count rate of ^{198}Au to the frozen sample to increase by a large amount. However, the control experiment, where the sample was frozen, left to thaw and finally the gold foil was extracted from the vial prior to irradiation showed that almost no gold atoms were activated. Since no ^{198}Au was found in the solution during the gamma-ray measurements of both replicates, this means that the crystallization process during freezing of the sample does not release gold atom from the foil into the matrix.

One aspect that could influence the higher yield of radiogold is the thermalization of fast and thermal neutrons due to the ice. The neutrons would slow down and lose energy while passing through the frozen solution, leading to the fast neutrons becoming thermal neutrons and thermal neutrons becoming even slower and having a higher probability of being captured by atoms of the gold foil. However, this effect is likely to be small, since it would take a longer distance for the neutrons to pass through to be significantly slowed down. Another factor leading to a higher activity of the solution when the sample is irradiated in a frozen state could be the higher strain and tension of the gold foil is suffering from when being frozen. This would make the surface of the gold foil more brittle and more susceptible to fast neutrons knocking gold atoms out of the first few atomic layers via sputtering. Finally, since the gold foil is fixated in ice inside the vial for the whole duration of the irradiation without it being able to move unlike when placed in liquid, activated atoms at the surface of the foil cannot be transferred to the walls of the vial due to the foil moving freely inside the vial during the transportation of the sample from the irradiation location to the entrance of the rabbit system. This would mean that there are liberated gold atoms from the gold foil sticking to the walls of the vial when the gold foil is irradiated while immersed in water, which are not factored in the gamma-ray measurement of the samples, since the liquid is usually transferred from the irradiation vial to the measurement vial after irradiation. However, that would not be the case, if the gold foil is placed in ice, since the gold foil would never touch the walls of the vial during irradiation, leading to a higher concentration

of released radiogold inside the matrix. The atoms themselves would not be able to cross the solid phase to get to the walls due to the properties of a solid, while in a liquid, this process would be much easier because of the movement of its particles.

5.5. Speciation of resulting gold atoms following neutron irradiation [36]

The mass of the gold foil used for irradiation, of which the released ^{198}Au was used for the co-crystallization with the gold(III) complex $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$ is given alongside the masses of the used irradiation vial in an empty state and filled with 1.1 mL triple distilled water in Table 20.

Table 20 Masses of gold foil and irradiation vial (with and without liquid) for the speciation experiment.

Gold foil	Irradiation vial (empty)	Irradiation vial (with 1.1 mL of triple distilled water)
9.29±0.01 mg	2.14662±0.00001 g	3.27421±0.00001 g

In order to gauge the distribution of ^{198}Au released from the irradiation of the gold foil in liquid between the product and the mother liquor, the ratio between ^{198}Au and stable gold in both was determined. Supposing the radiogold released into the aqueous phase during the irradiation process of the gold foil with neutrons is $^{198}\text{Au}^{3+}_{\text{aq}}$, the activated ^{198}Au atoms received from the gold foil would distribute evenly between the product and the mother liquor. If, however, other species of gold are involved, such as Au^+ or colloidal gold, differing ratios of stable to ^{198}Au are expected when comparing the product to the mother liquor. In the latter case, the mother liquor would show a higher activity compared to the product, since different gold species than $^{198}\text{Au}^{3+}$ would be excluded from the lattices during the crystallization process of $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$. This way, the released ^{198}Au from the irradiation of the gold foil in the aqueous phase is used as a tracer to determine its own species.

For this process 10 μL of each product and mother liquor was transferred to an irradiation vial and diluted with 100 μL of triple distilled water. Via INAA, using an in-house created material for reference, which was made of a diluted $\text{H}[\text{AuCl}_4]$ solution, the analysis for stable gold

commenced for both vials. It is important to mention the fact that the amount of ^{198}Au created from the irradiation of the gold foil in the aqueous phase preexisting inside the two vials prior to the neutron irradiation is negligible compared to the much higher activity caused by the samples being bombarded with neutron for 30 seconds inside the dry irradiation tube.

Using the data from the crystallization and the low level measurements, the ratio of $^{198}\text{Au}/^{197}\text{Au}$ in was $(1.82 \pm 0.07) \cdot 10^{-4} \text{ cts s}^{-1} \text{ mg}^{-1}$ in $[\text{Au}(\text{NH}_3)_4](\text{NO}_3)_3$, while the mother liquor showed a ratio of stable gold to ^{198}Au of $(1.47 \pm 0.15) \cdot 10^{-4} \text{ cts s}^{-1} \text{ mg}^{-1}$. These results are similar enough to be deemed as the same. Therefore, it is safe to assume that the radiogold produced via neutron irradiation in aqueous phase consists entirely of $^{198}\text{Au}^{3+}_{\text{aq}}$.

6. Conclusion

Szilárd-Chalmers-like reactions can be observed when gold foils are irradiated with neutrons inside a vial containing an aqueous phase. Compared to gold foils only being immersed and shaken inside the aqueous phase and irradiation taking place after the removal of said gold foil, the radiogold release upon neutron irradiation, while the gold foil is still placed inside the liquid, is one order higher in magnitude.

The activity of the gold foil itself does not contribute to a higher release of ^{198}Au into the aqueous phase, as the experiment with cyclic irradiation of the samples shows. Therefore, beta- and gamma radiation from the sample itself are unrelated to the amount of ^{198}Au released in the observed phenomenon.

Different irradiation durations, however, lead to varying, non-linear behavior between the time the sample spends at the irradiation location and the amount of ejected ^{198}Au from the gold foil into the aqueous phase. The ten-minute irradiations conducted for this experiment show that the yield of ^{198}Au in the aqueous phase is not always five times bigger compared to the two-minute irradiation. These ambiguous, unreproducible results are a product of certain circumstances. For one, the gold foil can move freely in the vial during the transfer of the sample to the irradiation location, at the end of which it may be placed at a different position inside the vial at the terminal location for irradiation. Therefore, the gold foil is being bombarded with a different numbers of neutrons every time, its location changes inside the vial. Additionally, fast neutrons also present during the irradiation may lead to non-linear sputtering effects, which can dominate when the duration of the neutron bombardment of the sample increases. This can produce random amounts of released, activated gold atoms.

Comparing neutron irradiation of gold foils in liquid to neutron irradiation taking place inside a solid phase leads to further unreproducible results. The activity inside the solid phase is much higher compared to the liquid, however, it is unclear how exactly the connection between the phase of the matrix and the yield of radiogold works. What is apparent at the present stage is that the crystallization of the liquid around the gold foil is not responsible for the higher activity found in the ice. It is, nevertheless, possible that the strain of the frozen gold foil in ice increases the release of gold atoms via sputtering. The previously used argument concerning the movability of the gold foil inside the vial could also play a role in these results, since in the case of a solid state, the gold foil cannot move freely during the transfer of the sample to the

irradiation location and return. This would be possible when liquid is being used as a matrix, leading to radiogold being transferred to the inside walls of the irradiation vial and not being measured.

Speciation of the released radiogold during the neutron irradiation of the gold foil immersed in an aqueous phase via a co-crystallization proved quantitatively that the liberated gold atoms during the Szilárd-Chalmers-like reaction are $^{198}\text{Au}^{3+}_{\text{aq}}$ ions.

The topic surrounding the observed Szilárd-Chalmers-like reaction is far from sufficiently researched. Uncertainties such as the exact effect of fast neutrons on the release of radiogold into the aqueous phase, impurities in the gold foil, parameters concerning the neutron and gamma dose rates, the formation of colloidal gold at the walls of the vial, the effect of hydrogen peroxide that may be created during the irradiation on the gold foil and many more have to be investigated to gain a complete understanding of the observed phenomenon.

7. References

- [1] H. Becquerel, “Sur les radiations émises par phosphorescence,” *Comptes rendus de l’Academie des Sciences, Paris*, vol. 122, pp. 420–421, 1896.
- [2] J. Chadwick, “Possible Existence of a Neutron,” *Nature*, vol. 129, no. 3252, p. 312, 1932, doi: 10.1038/129312a0.
- [3] E. Fermi, E. Amaldi, O. D’Agostino, F. Rasetti, and E. Segrè, “Artificial radioactivity produced by neutron bombardment,” *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character*, vol. 146, no. 857, pp. 483–500, 1934.
- [4] L. Szilard and T. A. Chalmers, “Chemical separation of the radioactive element from its bombarded isotope in the Fermi effect,” *Nature*, vol. 134, no. 3386, p. 462, 1934.
- [5] G. A. Brinkman, “Chemical Effects of Nuclear Transformations,” in *Advances in Inorganic Chemistry*, vol. 28, H. J. Emeleus and A. G. Sharpe, Eds., Academic Press, Cambridge, 1984, pp. 101–133. doi: [https://doi.org/10.1016/S0898-8838\(08\)60206-8](https://doi.org/10.1016/S0898-8838(08)60206-8).
- [6] G. Harbottle and N. Sutin, “The Szilard–Chalmers Reaction in Solids. Part I. Chemical Effects of Nuclear Recoil,” *J Phys Chem*, vol. 62, no. 11, pp. 1344–1351, Nov. 1958, doi: 10.1021/j150569a002.
- [7] H. MÜLLER, “Hot Atom Chemistry in Inorganic Solids — The Present Status and Future Aspects,” vol. 28, no. 4, pp. 233–240, 1981, doi: 10.1524/ract.1981.28.4.233.
- [8] G. Steinhauser, J. Evers, S. Jakob, T. M. Klapötke, and G. Oehlinger, “A review on fulminating gold (Knallgold),” *Gold Bull*, vol. 41, pp. 305–317, 2008.
- [9] F. JOLIOT and I. CURIE, “Artificial Production of a New Kind of Radio-Element,” *Nature*, vol. 133, no. 3354, pp. 201–202, 1934, doi: 10.1038/133201a0.
- [10] H. Abele, “The neutron. Its properties and basic interactions,” *Prog Part Nucl Phys*, vol. 60, no. 1, pp. 1–81, 2008.
- [11] J. Byrne, *Neutrons, nuclei and matter: an exploration of the physics of slow neutrons*. North Chelmsford: Courier Corporation, North Chelmsford, 2013.
- [12] G. A. Bartholomew, “Neutron capture gamma rays,” *Annu Rev Nucl Sci*, vol. 11, no. 1, pp. 259–302, 1961.

- [13] G. Hevesy and H. Levi, "Artificial radioactivity of dysprosium and other rare earth elements," *Nature*, vol. 136, no. 3429, p. 103, 1935.
- [14] G. Hevesy and H. Levi, "Action of slow neutrons on rare earth elements," *Nature*, vol. 137, no. 3457, p. 185, 1936.
- [15] R. R. Greenberg, P. Bode, and E. A. D. N. Fernandes, "Neutron activation analysis: A primary method of measurement," *Spectrochim Acta Part B At Spectrosc*, vol. 66, no. 3–4, pp. 193–241, 2011.
- [16] G. Harbottle and A. G. Maddock, "Chemical effects of nuclear transformations in inorganic systems," (*No Title*), 1979.
- [17] L. J. Arsenault, A. J. Blotcky, M. L. Firouzbakht, and E. P. Rack, "Preparation of radiopharmaceuticals by Szilard-Chalmers labelling and radioprotection in an ice lattice," *Journal of Radioanalytical Chemistry*, vol. 57, pp. 543–551, 1980.
- [18] V. Pichler, J. M. Welch, and J. H. Sterba, "Radiochemical effects of thermal neutron capture in Cr (tmhd) 3: method development," *J Radioanal Nucl Chem*, vol. 331, no. 12, pp. 5067–5079, 2022.
- [19] N. M. Quach *et al.*, "Extraction of 99Mo hot atoms made by a neutron capture method from α -MoO₃ to water," *J Radioanal Nucl Chem*, vol. 332, no. 10, pp. 4057–4064, 2023.
- [20] R. Kannan *et al.*, "Functionalized radioactive gold nanoparticles in tumor therapy," *Wiley Interdiscip Rev Nanomed Nanobiotechnol*, vol. 4, no. 1, pp. 42–51, 2012.
- [21] G. Steinhauser and A. Musilek, "Do pyrotechnics contain radium?," *Environmental Research Letters*, vol. 4, no. 3, p. 034006, 2009.
- [22] C. Raith Christina, "Neutron activation analysis measurement in different energy ranges at the TRIGA Mark II reactor for determination of the neutron spectrum," 2015. doi: 10.34726/hss.2015.22222.
- [23] G. Gilmore, *Practical gamma-ray spectroscopy*. John Wiley & Sons, Hoboken, 2008.
- [24] G. F. Knoll, *Radiation detection and measurement*. John Wiley & Sons, Hoboken, 2010.
- [25] G. Steinhauser, S. Merz, F. Stadlbauer, P. Kregsamer, C. Strelj, and M. Villa, "Performance and comparison of gold-based neutron flux monitors," *Gold Bull*, vol. 45, pp. 17–22, 2012.
- [26] G. Atomic, "TRIGA Mark II Reactor General Specifications and Description," *General Atomic Company, USA*, 1964.

- [27] E.-M. Furtmüller, “Study and analysis of reactor core configurations of TRIGA Mark II with SERPENT calculation code,” Wien, 2019. doi: 10.34726/hss.2019.51060.
- [28] M. Villa *et al.*, “The new I&C system of the TRIGA Mark II reactor Vienna,” in *RRFM European Research Reactor Conference 2016*, European Nuclear Society, 2016, pp. 496–503.
- [29] H. Böck and M. Villa, “The TRIGA Reactor Vienna within the World of TRIGA Research Reactors,” in *Österreich in einer nuklearen Welt*, Bundesministerium für Landesverteidigung und Sport, 2016, pp. 349–357.
- [30] H. Boeck and M. Villa, “TRIGA reactor characteristics,” Vienna University of Technology, 2007.
- [31] H. Böck, M. Villa, M. Bichler, M. Tschurlovits, and G. Röhr, “Sicherheitsbericht für das Atominstitut der Österreichischen Universitäten,” 2006.
- [32] R. Khan, S. Karimzadeh, H. Boeck, and M. Villa, “Neutronics analysis of the current core of the TRIGA Mark II reactor Vienna,” 2010.
- [33] R. Khan, “Neutronics analysis of the TRIGA Mark II reactor core and its experimental facilities,” 2010.
- [34] S. Merz, M. Djuricic, M. Villa, H. Böck, and G. Steinhauser, “Neutron flux measurements at the TRIGA reactor in Vienna for the prediction of the activation of the biological shield,” *Applied Radiation and Isotopes*, vol. 69, no. 11, pp. 1621–1624, 2011, doi: <https://doi.org/10.1016/j.apradiso.2011.05.019>.
- [35] V. Rosecker, “Chemical and physical impact on the Szilárd-Chalmers-effect: using the example of molybdenum,” Wien, 2017. doi: 10.34726/hss.2017.27560.
- [36] D. Boya *et al.*, “Production of no-carrier-added aqueous $^{198}\text{Au}^{3+}$ ions in a Szilárd-Chalmers-like nuclear reaction,” *J Radioanal Nucl Chem*, pp. 1–6, 2024.
- [37] E. Weitz, “Über die Stickstoffverbindungen des Goldes,” *Justus Liebigs Ann Chem*, vol. 410, no. 1-3, pp. 117–222, 1915.
- [38] T. M. Klapötke, B. Krumm, F. X. Steemann, and G. Steinhauser, “Hands on explosives: safety testing of protective measures,” *Saf Sci*, vol. 48, no. 1, pp. 28–34, 2010.
- [39] E. Weitz, “Über die Stickstoffverbindungen des Goldes,” *Justus Liebigs Ann Chem*, vol. 410, no. 1-3, pp. 117–222, 1915.

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