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Development of a method for plasma - induced combustion of intermediate to low-level radioactive waste

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List of Abbreviations

a	Atomic mass number
AC	Alternating Current
AGR	Advanced Gas Cooled Reactor
ALARA	As low as reasonably achievable
AtG	Atomgesetz
BfS	Bundesamt für Strahlenschutz
BWR	Boiling Water Reactor
DC	Direct Current
EBRD	European Bank for REconstruction and Development
eq.	equation
EW	Exempt Waste
fig.	figure
GCR	Gas Cooled Reactor
HDB	Hauptabteilung Dekontaminationsbetriebe
HEPA	High Efficiency Particulate Airfilter
HGW	Heat Generating Waste
HLLW	High level liquid waste
HLW	High level waste
i.e.	id est

IAEA	International Atomic Energy Agency
IATA	International Air Transport Association
ICAO	International Civil Aviation Organization
ICRP	International Commission on Radiological Protection
ILW	Intermediate level waste
IMO	International Maritime Organization
LILW	Low- and intermediate level waste
LILW-LL	Low- and intermediate level waste - long lived
LILW-SL	Low- and intermediate level waste - short lived
LLW	Low level waste
NHGW	Non-Heat Generating Waste
NPP	Nuclear Power Plant
NRC	Nuclear Regulatory Commission
PHWR	Pressurized Heavy Water Reactor
PWR	Pressurized Water Reactor
RF	Radiofrequency
SCR	Selective catalytic reduction
SNCR	Selective non-catalytic reduction
SRTF	Site Radwaste Treatment Facility
SRTF	Site radwaste treatment facility
StrlSchV	Strahlenschutzverordnung
tab.	table
TXRF	Total reflection X-Ray fluorescence analysis
UN	United Nations

VEK	Verglasungseinrichtung Karlsruhe	
VLLW	Very Low Level Waste	
VSLW	Very Short Lived Waste	
z	Proton number	
μ	attenuation coefficient	$\frac{cm^2}{g}$
ω_R	radiation quality factor	–
a	atomic mass number	–
D	dose	Gy
E	Energy	MeV
H	equivalent dose	Sv
I	Intensity	$a.u.$
m	mass	kg
R	Range	$\frac{g}{cm^2}$
t	thickness	$\frac{g}{cm^2}$
$t_{1/2}$	Half-life	s
z	atomic charge number	–
κ	electrical conductivity	–
λ	mean free path	–
σ	cross section	–
$\vec{E}$	electric field strength	–
$\vec{J}$	current density	–
b	mobility	–
g	statistical weight	–
n	density	$\frac{1}{m^3}$

Q	partition function	$-$
r	radius	m
T	Temperature	$K; °C$
u	drifting velocity	$-$
Z	impedance	$-$
A	activity	Bq
P	Power	W
t	time	s
T_{tot}	detraction efficiency	$-$
VRF	volume reduction factor	$-$
$\dot{m}$	throughput	$\frac{kg}{h}$
η	conversion factor	$-$
ρ	density	$\frac{g}{cm^3}$
C	total cost	$Euro$
c	specific cost	$-$
CU	capacity utilization degree	$-$

1 Introduction

The global demand for electricity is continuously increasing. Nuclear energy plays an important part in fulfilling this demand. Thirty-one countries are currently operating nuclear power plants (NPPs) and many more plan on building new plants in the next decades. This development is due to a variety of reasons, like for example a low resulting price for electricity, a small amount of needed fuel (Uranium) resulting in relatively low amounts of radioactive waste and essentially no emission of greenhouse gases, as well as a high stability of the electricity prices, and high safety for the public and reliability compared to other forms of energy production.

Radioactive waste is constantly generated throughout the operation as well as during the decommissioning of nuclear power plants. The generated waste is hazardous and therefore it has to be treated with care and responsibility. Each country by now has a strategy for the handling and storage of radioactive waste.

Radioactive waste is categorized by its specific activity and the handling and storage of it has to be undertaken according to the respective requirements of each category. Spent fuel and core components usually have a high activity and the corresponding waste is producing decay heat. The much larger amount is the non - heat producing waste which is usually categorized in intermediate level waste (e.g. spent ion exchange resins which are used for the filtering of radioactive streams) and low level waste. The highest amount of waste with the lowest radioactivity is the low level waste including all the mixed waste that is generated in operation and decommissioning of nuclear power plants, for example textile rags, plastic foil, overalls, gloves, etc.

Only little information on the detailed composition and content of low and intermediate level waste (LILW) produced in nuclear power plants is publicly available to date. In chapter 2 of this work an extensive survey on this topic is performed building a valuable database for LILW from NPP operations in different countries and types of power plants.

Chapter 3 first gives an overview of the existing regulations for radioactive waste for several countries as well as an overview of the current waste handling and treatment processes and technologies.
The waste from NPPs generally is processed with different methods depending on the type of the waste. The overall aim usually is to minimize the waste volume and to stabilize the waste, so that the final product is suitable for final disposal for a very long time without providing any hazard to the public. The reduction of the waste volume is necessary since the storage capacities of final repositories usually are limited and storage is expensive.
For the final storage of the waste the so-called deep geological repository is the favored option in Germany. In this method the processed and stabilized waste will be transferred to a former mine in geologically stable formations, so that no radioactivity will be released to the surface and to the ground water.
In most other countries the low and intermediate level waste will be stored in near surface repositories. However, storage space is also expensive for near surface solutions and a volume reduction of the material is still desired for this technology.

In chapter 4 the plasma induced combustion of the waste material is introduced as a method for volume reduction and stabilization. The idea behind employing plasma treatment methods for hazardous materials is to treat all material with very high temperatures. The combustible parts will incinerate and combust, and the non-combustibles will melt. The remnant of the treatment process is a mixture of molten slag and ashes, thus successfully achieving a volume reduction and a stabilization of the waste.
After covering the basic physical principles in plasma technology the currently available techniques for plasma treatment are discussed. Plasma treatment of hazardous material is still in a stage of development and prototyping, thus only a few fully functioning plants exist on an industrial scale. In this work the possibility of plasma treatment of low- and intermediate level waste material (LILW) will be analyzed with the aim of providing a comprehensive basis for decision making concerning a potential implementation of plasma treatment for LILW from NPPs.

Plasma treatment of waste can provide a significant advantage over conventional

treatment methods, since the prize per unit of packaged waste that has to be stored in final repositories is high. Moreover a high robustness of the material is desired with respect to a safe long term storage.
A design of a plasma oven for LILW is shown in chapter 5.2 which is based on the previously discussed waste compositions and plasma treatment technologies. To provide a proof of the principle characteristics of the plasma incineration method a small scale experimental device has been set-up for testing the principle design features and plasma specific properties related to radioactive waste treatment. The objective of the experiment is to verify the advantages of plasma technology on a small scale and, in addition, to identify possible problems and properties that can influence the design of a full scale facility. The analysis of the experiments performed with this laboratory plasma treatment device lead to an improved knowledge of the process for a full scale facility by upscaling the results.

Finally, a cost estimation is done and the result is presented in chapter 6 to present the cost effect of the volume reduction possibilities with respect to the final repository costs. Additionally, the boundaries for a an economical feasibility of a full scale plasma treatment plant are discussed.

This work demonstrates that the plasma - induced combustion of intermediate to low-level radioactive waste is a suitable method for volume reduction and stabilization. Weaknesses of existing facilities can be overcome with novel developments. Plasma treatment of LILW has a high economical advantage by volume reduction for storage in final repositories.

2 Radioactive Waste

2.1 General Considerations

Radioactive waste is any material containing significant amounts of radionuclides by activation or contamination from radioactive material and for which no further other use is foreseen. Radioactive waste is produced in different, non-nuclear fields, namely medicine, defense, industry and research.

The produced amounts, types and also physical forms and contained radionuclides of the waste vary strongly and depend on the specific nuclear application.
Radioactive waste, as defined in section 2.3, has to be stored and isolated from the environment in adequate repositories as long as the radioactivity is a threat to human life and health. Appropriate long - term storage (disposal) in repositories has to provide a high level of retention of the radioactive wastes, ideally without the need for further maintenance of the storage facility. To reach that goal, the waste has to be processed, conditioned packed in a proper way to allow for a secure handling of the containers. The volume of the radioactive wastes and therefore the volume of the disposal casks must be as low as possible due to limited space in the repositories. There must be no leaching of radioactive substances from the final packaged containers.
Various technologies for treating the waste are in use to achieve the necessary properties of the products to be handled and stored in a final repository that will be described for several countries in chapter 2.3.2. A detailed knowledge of the waste properties in terms of material composition is necessary to apply an appropriate treatment strategy.

However, unfortunately only few information on the composition and content of low and intermediate level waste (LILW) produced in nuclear power plants is publicly available to date. As part of this work an extensive investigation was conducted to collect the desired data from various sources. The results and the analysis of the database are discussed in this chapter.

2.2 Generation of Radioactive Waste

Waste including radioactive contaminants or activated material is generated in different situations that are not necessarily related to the nuclear fuel cycle. As an example, figure 2.1 depicts the distribution of waste generation by economic sector for the French radioactive waste inventory at the end of 2007 [1]. The distribution of radioacvtive waste production in countries that do not use nuclear material in defense is qualitatively comparable. Most of the waste is generated by the nuclear power industry, lesser quantities by research and by the non-nuclear power industry, as well as minor quantities by health applications. The distribution expected in Germany for the year 2040, which marks the end of nuclear power generation in Germany, contains 60 per cent of the total volume produced by the nuclear power industry [33]. Since the largest amount originates from nuclear power plants, the focus of the following chapters will be on this sector.

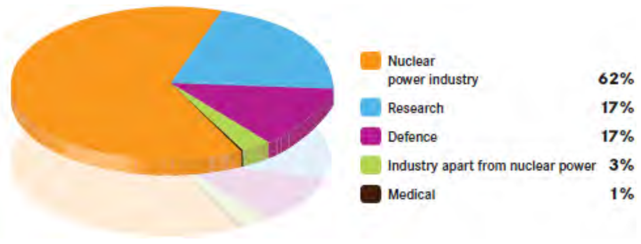

Figure 2.1: *Estimate of the volume distribution of radioactive waste existing in France at the end of 2007 by economic sector [1]*

2.2.1 Radioactive Waste Types

The radioactive waste can be measured and categorized by its activity content in terms of specific activity which is defined as activity per mass $[\frac{Bq}{kg}]$ or the activity per volume $[\frac{Bq}{m^3}]$. Of interest for this work are only the "low - and intermediate level wastes (LILW)", thus spent fuel and other heat generating wastes are excluded from this discussion. The LILW accounts for about 95% of the total radioactive waste volume, but only for 1% of the total stored activity content [34]. Typical LILW are contaminated waste material from works in radiation controlled areas. The exact type of the waste differs by respective application. In nuclear power plants all material that is used during operation, maintenance and decommissioning works, can get contaminated and therefore sorted out as waste. Typical waste from medical applications also includes organic material that was contaminated on purpose for cancer treatment. Radioactive waste arising in research can include a broad range of waste categories in terms of material and radioactivity content and is therefore not easy to classify.

Usually, the waste is categorized in terms of activity content to fulfill the regulatory requirements described in 2.3 and in addition, a categorization and sorting of the waste is made with respect to the applied treatment technology.

Gaseous radioactive material has to be collected by appropriate filters at the place of generation and is usually cached for some time. This has the effect that the radionuclides decay for a long enough time period for the activity content to be below the regulatory limit (decay storage). Gaseous radioactive waste forms only a small part of the total waste and has to be handled separately. Other main waste categories are liquid waste, wet solid waste and dry solid waste. These different categories are usually further sub-divided to fulfill the requirements of the foreseen treatment. The various treatment technologies currently in use are described in more detail in chapter 3.3.

2.2.2 Low - and Intermediate Level Waste in Nuclear Power Plant Operations

2.2.2.1 Origin of the Waste

Cooling circuits and condensate cleaning

The largest fraction of the radioactivity contained in LILW originates from the primary cooling circuit in nuclear power plants. Fission products and small fuel particles can be released through leakages of the nuclear fuel rods to the coolant. Solvent particles, chemical additives, corrosion products and the coolant can be activated by neutrons from the fission or from radioactive decay of the fission products [35]. Cleaning of the primary coolant is therefore necessary for radiological reasons.

The cleaning is usually done by passing the coolant through a combination of several mechanical and chemical filters. The mechanical filters are for the separation of larger particles to protect the chemical filters. The mechanical filter auxiliary materials such as cellulose, charcoal, silica gel, gravel or glass and metal fiber, can partly be cleaned by flushing with air and/or water. The resulting wastewater can have a significantly higher activity content than the average low level liquid waste from plant operations and is usually separately stored and treated due to concentration effects. The filters and the auxiliary materials themselves are to be seen as a separate waste stream and can bear intermediate level activities.

Chemical filters are used to clean the streams on a molecular level by chemical bonding [36], [37]. Ion exchange resins or inorganic zeolites are used for this. Ion exchange resins have to be handled separately because they usually have a higher specific activity than other wastes. They work by chemically exchanging soluble ions e.g. $H^+(aq)$, $OH^-(aq)$ from the resins with heavier radioactive ions (fission and corrosion products). Ion exchange resins exist either as powder or as small spheres (bead resins). In pressurized water reactors, the bead resins are generally being used, while in boiling water reactors the powder is preferred.

Wastewater from controlled areas

During operation of NPPs up to 50000 m^3 of low level radioactive wastewater is collected throughout the entire NPP lifetime [38]. Some parts originate from small leakages in the primary cooling circuit, auxiliary systems and the spent fuel pool. The wastewater can contain Boron and other chemical additives that can have an effect on the chosen waste treatment. Other sources for wastewater are the laundry, the showers and waters coming from the decontamination processes.

Due to the high volume of this type of waste, only the reduced volume after a first treatment step is mentioned in literature. This first step usually is the evaporation of the wastewater, as it shows best decontamination and highest volume reduction factors [39]. The majority of the contained radioactivity can be found in the so called evaporator concentrates, which can be intermediate level waste with typical specific activities of $10^9 - 10^{11}$ Bq/m^3 with densities of $1.14 - 1.21$ g/cm^3 [9],[27].

Maintenance work and revisions

The largest part of the produced waste in terms of volume is dry solid waste from maintenance work and revisions in the controlled area. During the revisions, where scheduled maintenance work takes place, a much larger waste volume is produced than during normal operation. The composition of this waste is essentially comparable with common industry waste. In some NPPs, there are efforts made to reduce the final waste volume. One approach is to substitute incombustible maintenance aiding material with combustibles, for example gloves or wrapping material.

The waste is usually collected loosely in waste bins and thus only has a typical density of $0.1 - 0.2\ g/cm^3$, but that can vary strongly. The waste can, with few exceptions, be categorized as LLW with specific activities in the region of $10^6 - 10^9\ Bq/m^3$ [9]. This waste is typically contaminated on the surface. Some special waste where a future use or recycling can be effective, e.g. electronics or cables, can be sorted out for further use when it is free of contamination. Special waste e.g. batteries, control fluids, lead, asbestos, etc. has to be sorted and treated separately.

For some waste decontamination is economically reasonable: Metal or concrete parts can be decontaminated. The cleaned material can be reused in a nuclear environment, or even as recycled material in other industries in the case of absence of radioactivity after the decontamination. The reuse in nuclear fields is more common in order to guarantee a complete radioactivity cycle. This does not include material that bears radioactivity from neutron activation, such as metal parts near the core, since these cannot be decontaminated.

Other sources

Further waste sources that have not been mentioned yet are gaseous wastes. These can occur during evaporation or other heat treatment and also be emitted in the reactor atmosphere. The gases are filtered and, for example with the use of retardation material like activated charcoal, kept until a significant decay of the activity. Due to the mainly short half-life of gaseous radionuclides the release to the environment can be kept to a minimum.

In heavy water reactors a reuse of the moderator material is intended since the heavy water is expensive to produce. Molecular sieves are being used for the cleaning of the coolant [40] which have to be treated separately when the end of lifetime is reached. Gas cooled reactors have an additional waste stream of chemical drying agents. Additionally, reactor graphite from these reactors and from graphite moderated reactors is usually a high activity bearing waste stream.

2.2.2.2 Waste Types

LILW can be categorized by different schemes. One suitable categorization is the sorting into three types: wet solid waste, dry solid waste and liquid waste. The advantage of this is that the waste can easily be separated since it is produced in different areas.
A "waste stream" is a grouped type of waste with a common origin or treatment that is measured in mass per time.

Wet solid waste

The "wet solid waste" stream consists of ion exchange resins, evaporator concentrates, sludges and sometimes filter concentrates. In general, the specific radioactivity is fairly high, because ion exchange resins and evaporator concentrates dominate this waste stream which both can be categorized as intermediate level waste. Sometimes these wastes are not mixed and ion exchange resins and evaporator concentrates are handled separately.
Wet solid waste is considered as the most difficult waste stream to treat due to its high specific activity. The amounts produced vary by reactor type, power of the reactor, country (and according waste handling policies) and time. Efforts are made to reduce the produced volume to a minimum. This trend can be seen when analyzing the produced amount of wet solid waste by time [27]. Averaged over a large number of sources and years, the produced volume of wet solid waste per nuclear power plant sorted by exemplary types and regions amounts to: 19.6 m^3/a (Western Europe PWR); 14 m^3/a (US PWR); 35.1 m^3/a (Western Europe BWR); 32.7 m^3/a (US BWR) [27].

Dry solid waste

Mixed dry solid waste is the largest volume fraction of LILW. The specific activity is low, though, and primarily results from surface contaminations. When analyzing the produced volume it has to be considered that the solid waste streams from different power plants can not be interpreted in the same way and have to be regarded with large uncertainties, due to different pre-treatment steps resulting in some volume reduction. This pre-treatment can be e.g. shreddering of waste or pre-densification by compaction. Another cause for divergence is a different classification of the waste in terms of waste type or packaging. For these reasons it became common to quantify the dry solid waste stream in mass per time and not in volume per time. In order to still be able to compare the amounts with other waste streams, the waste mass can be converted to volume by applying averaged densities.

Likewise, dry solid waste is already sorted in most nuclear power plants according to the intended treatment. In German plants the waste is distinguished into "combustible" and "incombustible".

For the mentioned exemplary NPP types and regions, the average produced waste amounts to [27]: $121m^3/a$ (Western Europe PWR); $355m^3/a$ (US PWR); $233m^3/a$ (Western Europe BWR); $426m^3/a$ (US BWR).

Liquid waste

Any liquid waste from NPP operations, for example from the laundry or from very small leakages in auxiliary systems, is usually evaporated and the residues are the concentrates that have to be handled as waste. These can also bear higher activities, depending on the evaporated stream. The volatile components Tritium and ^{14}C already vanish during the evaporation.

Liquid waste includes all the liquids that are not evaporated. In German power plants the liquid waste stream is limited to low level radioactive oils, which are incinerated. In other countries the non-evaporated waste waters may also be included in the liquid waste stream.

2.2.3 Waste Production Summary

All waste that is produced during the operation of nuclear power plants is usually declared as radioactive and has to be handled specifically. This includes mainly contaminated materials from the working environment, such as clothes, gloves, packing foils etc.. Another important part is waste from direct treatment of radioactive components, like spent ion exchange resins for radioactivity filtering and concentrates from evaporators which is "secondary waste". Figure 2.2 lists a common distribution of operational waste taken as a 10-year-average of a German PWR plant with 1300 MW$_{el}$ power output [2]. The produced amount of waste varies

Reactor type	∅ Net Power [MW_{el}]	∅ Waste [m^3/a]	∅ Waste [$\frac{m^3}{a \cdot MW_{el}}$]
PWR (Western Europe)	1103	143	0.13
PWR (WWER-1000)	963	246	0.26
PWR (USA)	1023	369	0.36
BWR (Western Europe)	1080	270	0.25
BWR (USA)	1110	459	0.41
GCR (AGR)	552	120	0.22
PHWR (CANDU)	634	143	0.23
Average		**250±127**	**0.265±0.092**

Table 2.1: *Production of low level waste in nuclear power plants. The values are averaged over a large number of sources [27].*

broadly and mainly depends on the factors type of reactor, nuclear waste policy of the country, waste policy of the plant operator and the plant outages strategy (revisions). Table 2.1 lists rough values for a first orientation sorted by the power plant types, which are averaged values from various sources [27]. The total average is 250 $\frac{m^3}{a}$ ± 127 $\frac{m^3}{a}$ or, if normalized on 1000 MW_{el} power output, 265 $\frac{m^3}{a \cdot GW_{el}}$ ± 92$\frac{m^3}{a \cdot GW_{el}}$ (see table 2.1). The produced waste amounts differ by region. In Western Europe, the lowest quantity of waste is produced, in the USA the quantity is almost double the value of the European waste produced by plants. Since the NPP's are the same type as in Europe (mostly PWR), an explanation for this difference is a stronger focus in Europe on waste minimization. The waste quantity per year from the Russian WWER-1000 is in between. Gas cooled reactors (GCR) and pressurized heavy water reactors (PHWR, CANDU) are special NPP types and therefore have different specific waste streams.

Wet solid waste mainly accounts for mainly ion exchange resins and evaporator concentrates. This waste stream is usually handled separately, because they are of intermediate level specific activity.

Liquid waste is generally evaporated and the resulting concentrates make up the wet solid waste. Other non-evaporated liquids, such as oils, are incinerated and of minor radiological concern due to a very low specific radioactivity.

Dry solid waste is the last and highest share of total waste. About 65% of all waste produced in NPPs is dry solid waste. To minimize waste production, as little as possible material is disposed of and, where possible, combustible material is used. The waste that is created during NPP operation can be divided into several subcategories, as indicated in figure 2.2.

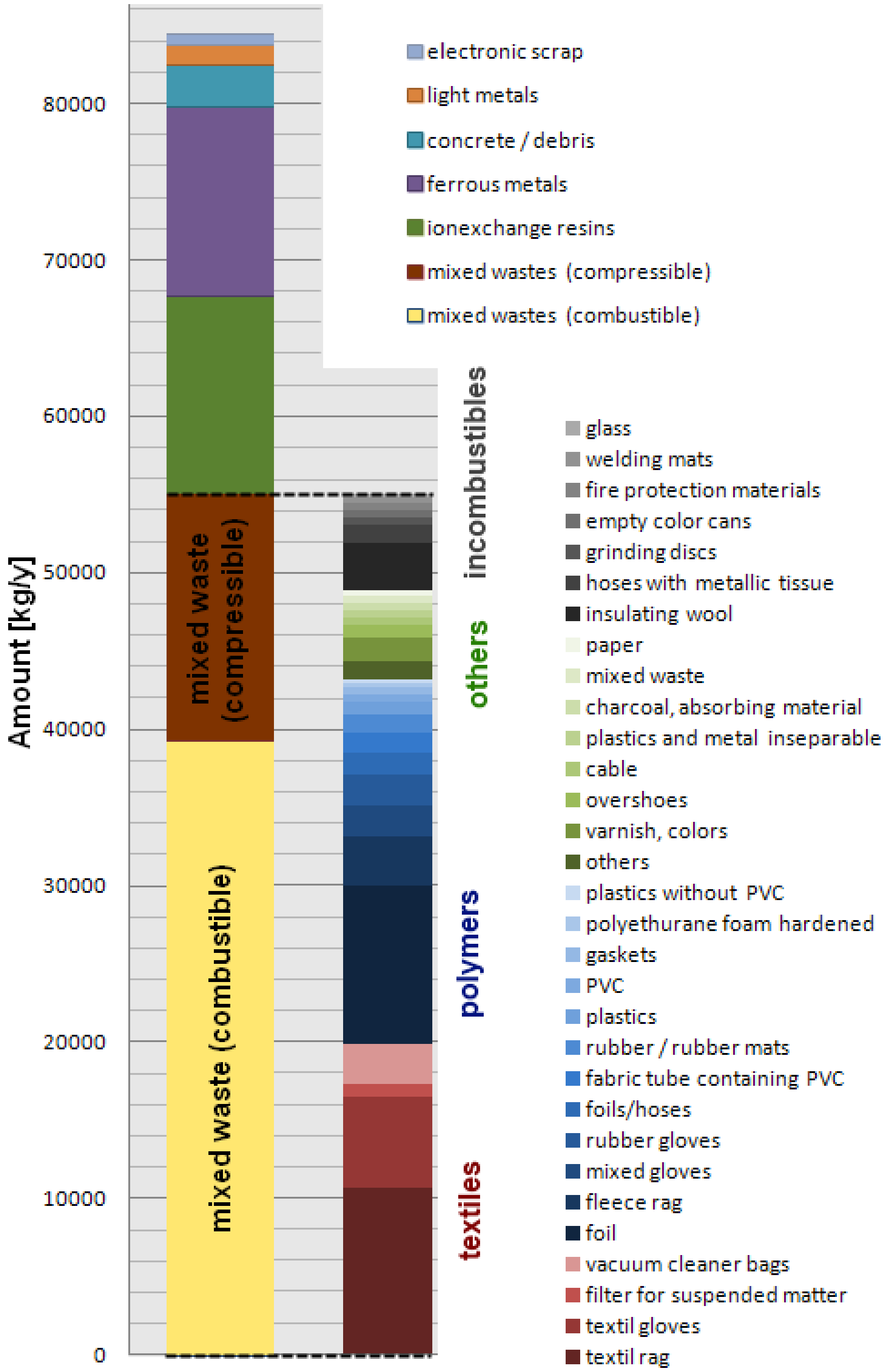

Figure 2.2: *Composition of waste from NPP operations, extended from [2]*

The bulk of 55000$\frac{kg}{a}$ is dry solid waste. In NPPs this bulk is typically divided in mixed waste - combustible and mixed waste - compressible. The bar in the right hand side of figure 2.2 shows the typical distribution of this mixed waste. As can be seen, the entirety of incombustible materials like glass, welding mats, insulation wool etc., constitutes for about 6000 kg/a of the produced waste. The remainders that are typically sorted out as incombustible are considered as more problematic waste when incinerated due to possible dangerous and hazardous off-gas production, which is categorized here as part of the polymers. PVC and material containing rubber is therefore usually treated together with the compressible waste. A lot of textiles can be found in the mixed waste stream that are easily combustible. The remainders are categorized as "others", which include special waste like varnish/colors, cable, charcoal and so on, which, when combusted, bear a special problem or enhancements. For example, charcoal is easily combustible, but when it is used for the filtering of radioactive substances it can have a high specific activity. Varnish/colors produce hazardous off-gas streams when combusted. Cable material (copper) can be recycled when it is not too contaminated.

As for wet solid waste, the ion exchange resins cosntitute about 15 % of the total produced waste in weight. With the typical density of 1.2 $\frac{g}{cm^3}$ (see table 2.2) this is a volume of 10.5 $\frac{m^3}{a}$.

Ferrous metals and light metals are recycled if possible and together they account for about 13400 $\frac{kg}{a}$ or roughly 3 $\frac{m^3}{a}$ (6 $\frac{g}{cm^3}$, 75% bulk density, see tab. 2.2). Around 1300 $\frac{kg}{a}$ debris waste is produced. Finally, there is the electronic scrap with a weight of 674 $\frac{kg}{a}$.

Table 2.2 lists the typical density and specific activity of the waste categories as averaged values [27]. In the category of mixed waste it is of course hard to define a typical density. However, the density of 0.2 $\frac{g}{cm^3}$ is averaged over a large number of sources. As a comparison a value of 0.33 $\frac{g}{cm^3}$ for combustible waste is recommended for rough calculations by the UK low level waste repository: "When converting from volume a typical density of 3m^3= 1te could be assumed for soft wastes" [41].

The typical density of wet solid waste is known as 1.2 $\frac{g}{cm^3}$ for ion exchange resins and evaporator concentrates with 20% dry mass. As one can see, the typical specific radioactivity is high in comparison. As described in chapter 2.3 and especially in table 2.6, ion exchange resins usually do not account for low level waste in Italy and

	typ. density $[\frac{g}{cm^3}]$	spec. activity $[\frac{Bq}{m^3}]$	Comments
Wet solid waste			
Ion exchange resins	1.2 ± 0.1	$10^{10} - 10^{13}$	
Evaporator concentrates	1.2 ± 0.1	$10^{9} - 10^{11}$	with 20 % dry mass
Filter auxiliary material, slag	1.0 ± 0.2	$10^{6} - 10^{10}$	streams often intermixing
Dry solid waste			
Mixed waste	≈ 0.2	$10^{6} - 10^{9}$	density varying strongly
Filters	≈ 0.6	$10^{8} - 10^{12}$	density varying strongly
Debris	2.0 ± 0.5	$10^{6} - 10^{9}$	bulk density 50 - 100 %
Metal parts	6.0 ± 2.0	$10^{8} - 10^{12}$	bulk density 50 - 100 %
Liquid waste			
Oil, solvents	0.9	$< 10^{6}$	

Table 2.2: *Typical properties of different waste streams [27]*

the UK. In the US, it would be regarded as Class C low level waste and in Germany all LILW, also including spent ion exchange resins, are non-heat generating waste.

2.3 Radioactive Waste Regulations

Figure 2.3: *Waste management hierarchy [3]*

As a general guideline for the handling of radioactive waste a hierarchy of waste management has to be applied, (see figure 2.3) with the priority order of avoiding the waste, minimizing the amount, re-use, recycle and with disposal as the least favored option. In Europe this is established by law [3]. Radioactive waste, as defined by the International Atomic Energy Agency (IAEA) is "waste that contains or is contaminated with radionuclides at concentrations or activities greater than clearance levels as established by the regulatory body"[42]. This definition already includes a reference to national authorities and indicates that any legal regulations are defined by those. While this means that every country has their own regulations, the IAEA nevertheless issues guidelines on handling the radioactive waste for all member states.

2.3.1 Classification of Radioactive Waste

For the classification of radioactive waste the IAEA suggests a system consisting of six classes [4]:

- **Exempt waste (EW)**: Waste with negligible radioactivity that can be handled without additional care.
- **Very short lived waste (VSLW)**: Waste containing only short lived radionuclides, which can be stored until the activity contained falls below regulatory limits. This waste can then also be disposed without additional care.
- **Very low level waste (VLLW)**: Waste containing low amounts of radioactivity but also containing long lived radionuclides. This waste can be disposed in a near surface repository with limited control.
- **Low level waste (LLW)**: Waste containing low amounts of radioactivity with significant potential harm that have to be handled with care. The proposed disposal strategy is a near surface underground disposal.
- **Intermediate level waste (ILW)**: Waste containing significant amounts of radioactivity. Examples are spent ion exchange resins and concentrates from nuclear operations. This waste has to be handled with care and can be disposed in an underground disposal facility.
- **High level waste (HLW)**: Waste containing high amounts of radioactivity. HLW is heat generating waste. Examples can be spent fuel, waste from reprocessing and some waste from military operations. Disposal should be made with care in a deep underground disposal facility.

In this work the focus will be on LLW and ILW. The even lower waste classes EW, VSLW and VLLW can be handled or disposed without major safety and security provisions and therefore do not need any additional processing. The HLW however, needs special attention and special methods for processing, and is therefore also not considered in this work.

The IAEA definition gives a recommendation on how to classify radioactive waste,

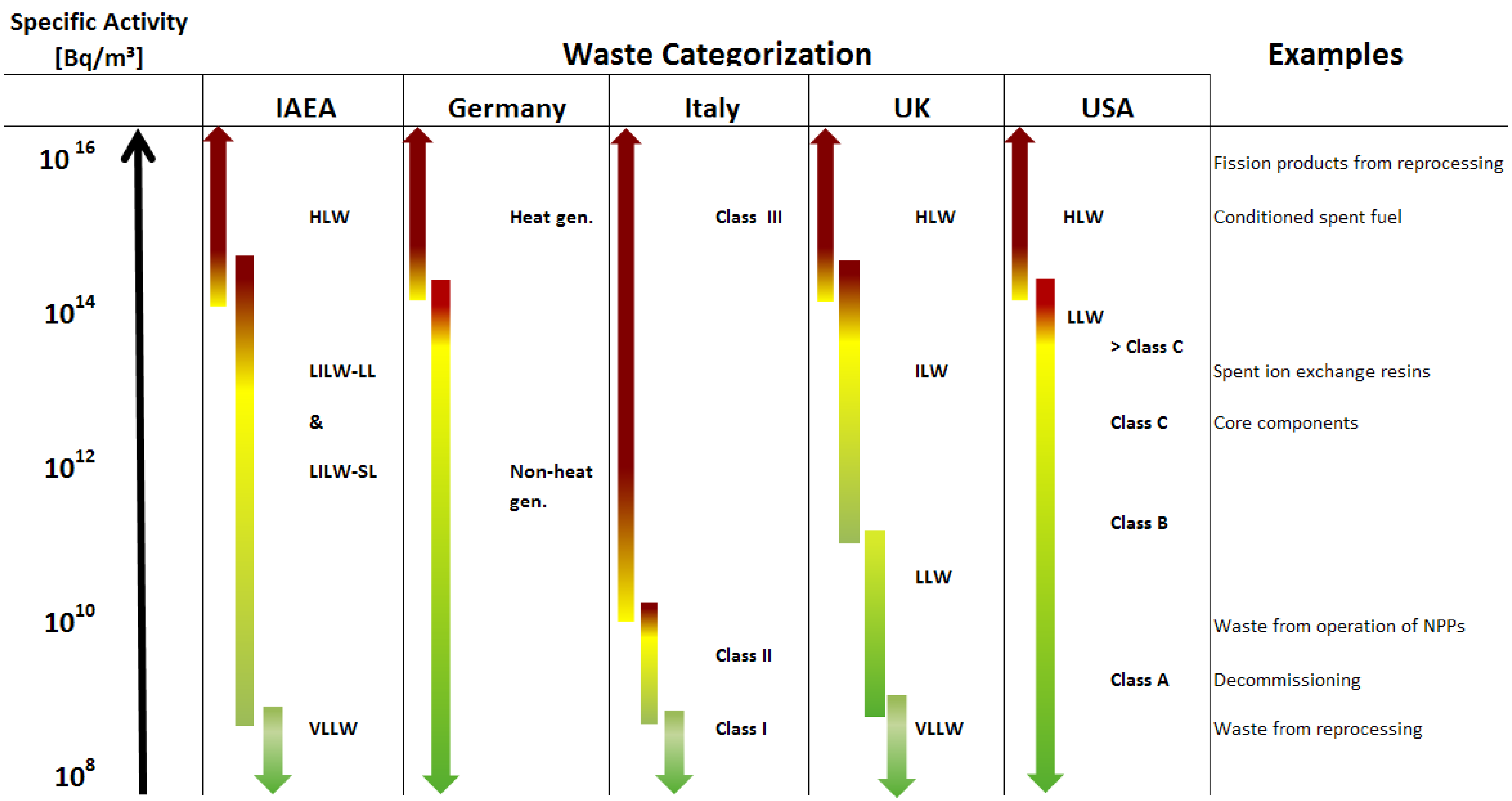

Figure 2.4: *Qualitative comparison of different waste categorization schemes of various countries and the IAEA. [4],[5],[6],[7],[8],[9]*

though the national regulations may differ. As an example the categorization and waste handling policies for Germany, Italy, UK, and USA are given and compared below and their final disposal strategies for this kind of waste are summarized. Figure 2.4 shows a qualitative comparison of the radioactive waste classification of these countries. Since there are not necessarily classification limits for the specific activity, the classification also depends on other parameters such as e.g. heat generating capacity or potential dose and therefore the graphic representation on the ordinate is not quantitative.

2.3.2 Waste Handling in Different Countries

2.3.2.1 Waste Handling Policies in Germany

In Germany a definition of radioactive waste is given in the "Atomgesetz" (Atomic Act, AtG [43]) and states that radioactive waste is radioactive material that has been produced or made radioactive by having being exposed to a process of the fabrication or the use of nuclear fuels associated radiation (Anlage 1, Absatz 4). The German legislation does not offer categories, but there exist special regulations for nuclear fuel and spent nuclear fuel. In practice, a categorization is made according to the processing and handling of the waste, so that the acceptance criteria for the final repository "Schacht KONRAD" [44] can be fulfilled. Guidelines for categorization are given by the Federal Agency for Radiation Protection (BfS, Bundesamt für Strahlenschutz) [8], with reference to the final repository acceptance requirements for KONRAD [44]. Final repositories in Germany for low and intermediate level waste will be deep geological repositories. The German final repository for storage of the currently produced waste will be "Schacht KONRAD", which will start operation around 2019 [45]. The waste will then be stored 800 m to 1300 m below ground in a former iron ore mine.

In general, the waste in Germany is categorized in negligible heat generating (NHGW) and heat-generating waste (HGW). The NHGW can then be further categorized in LILW-SL (90%) and LILW-LL (10%). All radioactive waste has to be documented and reported, namely in terms of volume or mass, activity, state of processing and type of waste amongst other criteria [46].

The quantified criteria for waste packages that can be delivered to KONRAD are given in the final repository acceptance requirements [28]. There are several requirements:

- *basic requirements on radioactive waste for disposal:* These mainly state that the delivered waste has to fulfill the statutory requirements of the AtG and the Ordinance on Radiation Protection (Strahlenschutzverordnung, StrlSchV [47]), as well as that the waste must not be mixed with other hazardous waste.
- *requirements on waste packages:* Waste packages have to fulfill basic requirements (for example on dose and contamination, inner pressure and physical state) as well as following requirements on the type of waste, the waste container, the radionuclide inventory and the masses of non-radioactive hazardous substances.
- *requirements on waste forms:* The packaged waste is sorted in different waste form groups according to its state of processing and content. There e 6 different groups with different quality aspects on pressure, thermal stability, mechanical stability and content. The different requirements in the waste groups are such, that a safe storage in defined final repository containers can be guaranteed. Basic requirements for all of the groups include that the forms have to be solid, must not foul or ferment, must not include liquids or gases and explosive substances, must not be able to reach criticality and solidified waste must fulfill criteria on the solidification (no possible chemical reactions, tightness).
- *requirements on waste containers:* Two classes of containers exist with different stability requirements.
- *activity limits:* Table 2.3 shows examples for limits of specific activities per waste cask for some radionuclides.
- *mass limits of non-radioactive hazardous substances:* There exist limits on non-radioactive substances, which are assigned in the annex and refer to other legal regulations.

Table 2.3 shows examples for limits of specific activities per waste cask for some radionuclides. The limits depend on the waste product type and chosen waste container class. The waste container class II fulfills more safety standards than class I and the waste product groups are chosen according to the state of conditioning and therefore the stability of the waste product.

Nuclide	Half life	Radiation type	Waste container class I	Waste container class II
^{3}H	12.3 y	β	$3.3 \cdot 10^{9} Bq$	$3.3 \cdot 10^{9} Bq$
^{60}Co	5.3 y	β	$5.0 \cdot 10^{9} Bq$	$1.2 \cdot 10^{14} Bq$
^{63}Ni	100.1 y	β	$7.0 \cdot 10^{11} Bq$	$1.7 \cdot 10^{16} Bq$
^{90}Sr	28.9 y	β	$8.6 \cdot 10^{8} Bq$	$2.1 \cdot 10^{13} Bq$
^{137}Cs	30.1 y	β	$5.1 \cdot 10^{9} Bq$	$1.3 \cdot 10^{14} Bq$
^{241}Pu	14.3 y	α, β	$1.7 \cdot 10^{10} Bq$	$4.3 \cdot 10^{14} Bq$

Table 2.3: *Chosen radionuclide specific activity limits per waste package for the KONRAD repository [28]. The values for waste form group 01 in waste containers class I are shown, as well as the values for waste form group 01-06 for class II containers.*

2.3.2.2 Waste Handling Policies in Italy

In Italy three classes of radioactive waste are defined according to the radioisotope characteristics and concentrations [7].

Class I waste, the lowest category, includes radioactive material with a short half-life, so it decays in a few months to a radioactivity level below safety concerns [48].

Class II waste includes radioactivity of higher concentration. "Second category wastes are in particular characterized by a radioactivity concentration that, following possible treatment and conditioning processes, shall not exceed at disposal the values listed in tab. I." [29], with the referenced tab. I below as table 2.4:

RADIONUCLIDES	CONCENTRATION
α emitters $T_{1/2} > 5$ years	^{a} 370 Bq/g (10 nCi/g)
β / γ emitters $T_{1/2} > 100$ years	^{a} 370 Bq/g (10 nCi/g)
β / γ emitters $T_{1/2} > 100$ years in activated metals	3.7 KBq/g (100 nCi/g)
β / γ emitters $5 < T_{1/2} \leq 100$ years	37 KBq/g (1 μCi/g)
^{137}Cs e ^{90}Sr	3.7 MBq/g (100 μCi/g)
^{60}Co	37 MBq/g (1 μCi/g)
^{3}H	1.85 MBq/g (50 μCi/g)
^{241}Pu	13 KBq/g (350 nCi/g)
^{242}Cm	74 KBq/g (2 μCi/g)
Radionuclides $T_{1/2} \leq 5$ years	37 MBq/g (1 mCi/g)

Table 2.4: *Concentration limits for second category conditioned wastes*
^{a}*values must be intended as average values referred to the whole of the wastes contained in the disposal repository, taking into account that the limit value for each package cannot exceed 3.7 KBq/g (100 nCi/g) [29]*

Categorised as Class III waste is all radioactive waste that exceeds the given limits. This typically includes high level waste such as spent fuel or waste from reprocessing.

Class I and II waste are considered to be suitable for near-surface disposal, whereas class III waste is to be stored in deep geological disposal [48].

2.3.2.3 Waste Handling Policies in the United Kingdom

In the UK radioactive waste is categorized according to its specific activity and heat production. The categories are [49]:

- VLLW: 0.1 m^{3} of waste containing less than 400 kilobecquerels (kBq) of total activity or single items containing less than 40 kBq of total activity. This is a subcategory of LLW.

- LLW: "radioactive waste having a radioactive content not exceeding four gigabecquerels per tonne (GBq/te) of alpha or 12 GBq/te of beta/gamma activity." [6]
- ILW: radioactive waste exceeding the limits for LLW, but with negligible heat generation.
- HLW: heat generating radioactive waste.

Final disposal repositories for LLW and ILW already exist as near surface disposal facilities (LLWR Cumbria (open), Dounreay (closed)).

2.3.2.4 Waste Handling Policies in the United States of America

In the USA waste is divided in HLW and LLW and the additional classes transuranic waste (TRU) and uranium tailings. The categorization of the waste is chosen according to its subsequent treatment strategy. The LLW is distributed in 3 classes, where Class A waste is the least radioactive waste and can be disposed in a near surface repository, whereas Class B and C waste have to be disposed progressively deeper. The required depth for Class C waste disposals is only 5 m below the surface, and the disposal facility has to be constructed "with intruder barriers that are designed to protect against an inadvertent intrusion for a least 500 years" [50]. Table 2.5 shows the limits for each LLW class given by the Nuclear Regulatory Commission (NRC) [50]. Several disposal facilities exist and refer in their waste acceptance criteria mainly to the NRC regulation.

2.3.2.5 Comparison of the Requirements for the Different Countries

Table 2.6 shows a comparison of the limits for total specific activity of the discussed countries.

To be able to better compare the LLW radioactivity limits of the countries all values are converted into Bq/m^3. The limits are given for waste that is considered to be allowed in final disposal facilities for non high level waste.

Some limit values are given per weight unit. These are converted using an average density of 500 kg/m^3, which for example is in between the suggested densities for rough calculations in [41] of combustible waste ($\frac{1}{3}t/m^3$) and metallic waste ($1t/m^3$).

Radionuclide	Class A (Ci/m^3)	Class B (Ci/m^3)	Class C (Ci/m^3)
Total of all nuclides with less than 5 year half-life	700	No limit	No limit
H-3 (Tritium)	40	No limit	No limit
Co-60	700	No limit	No limit
Ni-63	3.5	70	700
Ni-63 in activated metal	35	700	7000
Sr-90	0.04	150	7000
Cs-137	1	44	4600
C-14	0.8		8
C-14 in activated metal	8		80
Ni-59 in activated metal	22		220
Nb-94 in activated metal	0.02		0.2
Tc-99	0.3		3
I-129	0.008		0.08
Alpha emitting transuranic nuclides with half-life greater than 5 years	10 nCi/g		100 nCi/g
Pu-241	350 nCi/g		3500 nCi/g
Cm-242	2000 nCi/g		20000 nCi/g

Table 2.5: *US NRC limits for the specific radioactivity of LLW*

There is no total specific activity limit for LLW in the USA. Practical limitations are given by waste that exceeds the criteria for class C waste in terms of specific activity of chosen radionuclides, and also for heat-generating waste.

The limits for Germany are to be seen as average values, according to the final disposal requirements, which means that on average the disposed waste must not exceed these values. Single packages may exceed these values but are bound to the specific activity limits of the radionuclides contained.

The limits for the USA and Germany seem to be in the same range, due to the fact that all low - and intermediate level waste (LILW) can generally be disposed

Country	β/γ limit	α limit	β limit , converted to $[Bq/m^3]$
Germany	$1.6 \cdot 10^{13} Bq/m^3$	$4.9 \cdot 10^{11} Bq/m^3$	$1.6 \cdot 10^{13}$
Italy	$3.7 \cdot 10^{9} Bq/t$	$3.7 \cdot 10^{8} Bq/t$	$7.4 \cdot 10^{9}$
UK	$1.2 \cdot 10^{10} Bq/t$	$4 \cdot 10^{9} Bq/t$	$2.4 \cdot 10^{10}$
USA	$2.59 \cdot 10^{14} Bq/m^3$		$2.59 \cdot 10^{14}$

Table 2.6: *Comparison of limits of specific radiation for disposal of low level waste*

in the same disposal facilities in these countries. These are basically all non-heat-generating wastes. In the UK the ILW are handled separately and in Italy the waste categorization scheme differs largely, as seen qualitatively in figure 2.4.

2.4 Relevant Radionuclides in Waste

2.4.1 Leading Nuclides

The occurring radionuclides in waste treatment are limited to a set of nuclides with a half-life of over one year, so that the nuclides are present in waste that is stored for a few years. The radioactivity is mainly caused by contamination and by neutron activation. Therefore activation products, fission products and transuranic elements may be present.

For each group by definition there is a "'Leading Nuclide"'. The selection of these leading nuclides is guided by easy measurements. Based on reference measurements with defined fractions of the activity of each nuclide to the relevant leading nuclide at a given time, the total activity of each occurring radionuclide can be estimated. This spectrum of radionuclides is called the nuclide vector, which is characteristic for a radioactive compound and its specific history. The leading nuclides are ^{60}Co for activation products, ^{137}Cs for fission products and ^{241}Am for transuranium elements. Most of the other occurring nuclides are already mentioned in section 2.3, since some countries have regulations focusing on leading nuclides.

Some of these nuclides need to be addressed in more detail.

Tritium (^{3}H) is produced by activation in the reactor water. Since it is usually chemically bonded in the form of water, it cannot be held back from being emitted.

Carbon-14 (^{14}C) is also an activation product and binds chemically to gaseous organic substances or CO_2, which makes it as hard to control as ^{3}H.

Cobalt-60 (^{60}Co) is produced in the reactor by activation of Cobalt-59 in some steel compounds. Due to its half-life of 5.3 y it is considered as one of the more important nuclides to control.

Nickel-63 (^{63}Ni) has a half-life of 100.1 years and is also produced by neutron activation of steel.

Strontium-90 (^{90}Sr) is a fission product with a half-life of 28.78 years. Strontium behaves chemical similar as Calcium and can therefore be biologically absorbed in bones. Therefore it is considered as a nuclide with a high radiotoxicity. During the nuclear disaster of Chernobyl in 1986 ^{90}Sr was set free during the graphite burning of the core and was among the most important isotopes regarding threats for the health.

Iodine-131 (^{131}I) has a high radiotoxicity and is gaseous or attached to aerosols, thus it can be inhaled. Its half-life is short though($T_{1/2}$ = 8 days), and therefore it does not have to be addressed in radioactive waste treatment because it completely decays during storage.

Caesium-137 (^{137}Cs) is another fission product with a high radiotoxicity. Caesium is chemically important, since its boiling point is 671°C and therefore it has to be considered as an aerosol and has to be filtered in the off-gas stream of any thermal treatment in which a high temperature can occur. Caesium is an alkali metal and therefore has a high solubility in water.

Americium-241 (^{241}Am) is a leading nuclide for other transuranium elements because it can be easily measured. The amount of the other transuranic elements is in defined proportions to the amount of ^{241}Am. These nuclides are generally α - emitters, consequently they are easily shielded but are dangerous when ingested.

2.4.2 Airborne Radionuclides

A special threat exists by radionuclides that are in gaseous or particulate form. This includes aerosols containing Tritium and Carbon-14, other activated gaseous elements like Nitrogen and especially gaseous fission products like halogens and noble gases. Table 2.7 lists these Isotopes with their main properties. Listed are only the relevant nuclides that are not decayed after a storage of several months [51].

These radionuclides have to be filtered and controlled because of their potentially harmful effects by submersion or ingestion. The human skin protects the body from external radioactivity, but internal radioactivity ingested by breathing or intake is unshielded.

2.4.3 Application Examples

Radioactive waste from medical applications and from research other than nuclear energy can generally be specified based on the contained radionuclides. The reason is that, other than in nuclear energy applications, the medical procedures are mainly

Radionuclide	Half-Life	Comment
^{3}H	12.3 y	Chemically binds in Water
^{14}C	5700 y	Chemically binds as organic material
^{85}Kr	10.7 y	Noble gas. Hard to filter due to higher Xenon concentration
^{106}Ru	371 d	Naturally not gaseous, but binds chemically as RuO_4
^{129}I	$15.7 \cdot 10^6$ y	Fission product, Halogen, long half-life
^{134}Cs	2.1 y	Fission product
^{137}Cs	30.1 y	Fission product, long time contamination from nuclear disasters
^{222}Rn	91.8 h	Noble gas, naturally occurring material from the ^{238}U decay chain

Table 2.7: *Airborne radionuclides*

based on one specific radionuclide, so all possible contaminations can be classified accordingly. Often the half-lives of the applied radionuclides are short enough to allow for a "decay storage". A typical medical radioisotope applied to patients in the field of radiopharmacology would be Tc-99m, which is used for tumor diagnosis, with a half-life of 6 hours. Tc-99m can be gained from a Mo-99 source ($T_{1/2}$ = 66 hours) which was created by fission. The occurring waste products then decay over a controllable period of time of a few weeks.

In nuclear energy research a multitude of radionuclides is generated by nuclear fission and any of those products or a mixture of them can occur in contaminations. However, most of the contamination are corrosion products from activated materials such as Co-60. The fission products typically have a broad range of half-lives, which excludes the application of "decay storage". One of the leading nuclides is the fission product Cs-137 with a half-life of 30.17 years. A decay storage typically requires up to about 10 half-lives of the contained nuclide. A decay storage of Cs-137 containing material would therefore take 300 years, which is not practicable.

3 Low - and Intermediate Level Waste Treatment

3.1 Purpose of Radioactive Waste Treatment

The collected radioactive waste is treated and processed with the aim to reduce its volume and to achieve a final product which can be stored and disposed without the risk of releasing radionuclides to the environment.
The objective of volume reduction is to minimize the needed costs for transport and storage.

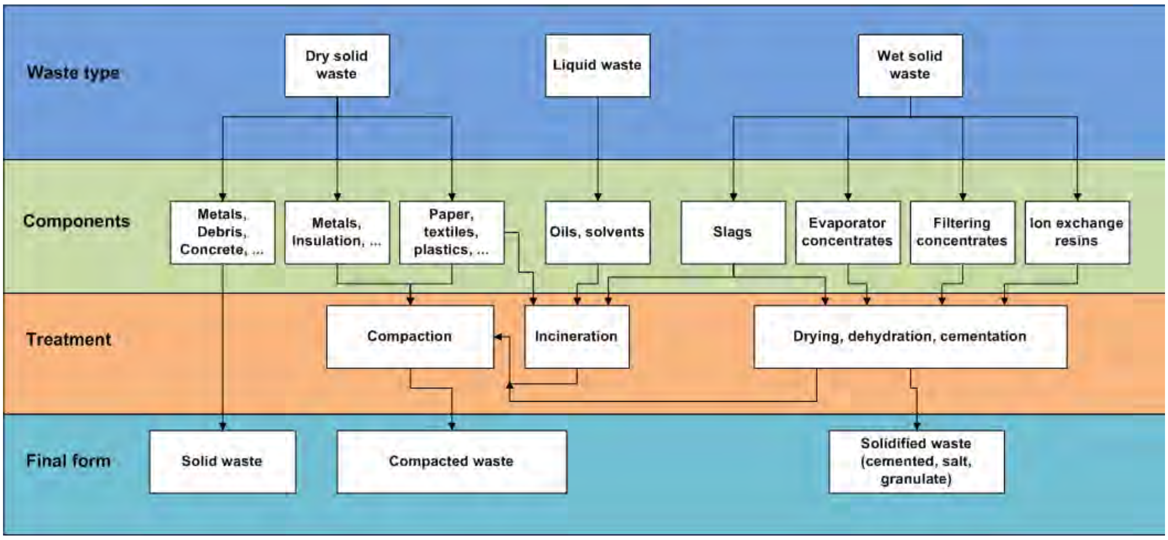

Figure 3.1: *Example of optimized waste processing paths [9]*

Table 3.1 lists a comparison of existing treatment methods with the general properties and application. Except for the "Plasma" treatment method, all mentioned technologies are routinely performed for waste treatment. For an overall waste treatment, a combination of several methods is performed. A typical flow diagram of the waste and the treatment products is shown in figure 3.1 [9]. The depicted

			Applicability						
Treatment method	Main Purpose	Volume reduction	Combustible	Incombustible	Liquids	Metals	Resins	Concentrates	Concrete/Debris
Incineration	Vol.red., Drying	100	X						
Compaction	Vol.red.	2-6	X	X					X
Supercompaction	Vol.red.	4-10	X	X		X			X
Vitrification	Solid. of. liquids	20-50			X				
Metal melting	Vol.red. of metals	20				X			
Pyrolysis	Wet ILW treatment	3					X	X	
Grouting/Cementation	Solid.	0.5	X	X	X		X	X	X
Drying/Evaporation	Wet treatment	2-100			X		X	X	
Decontamination	Recycling	7	X	X		X			X
Plasma	Vol.Red., Solid.	5-50	X	X	X	X	X	X	X

Table 3.1: *Methods for treating radioactive waste and applicability on different waste streams [27], [24].*

technologies and paths are the usually applied radioactive waste treatment methods in Germany for the three mentioned main LILW types. The main treatment is a combination of incineration and (super-)compaction, where applicable. The final form must fulfill all requirements for final disposal and is therefore solidified and packaged.
Not shown in the process chart are the waste handling steps, namely transport, decay storage, interim storage and final storage.

3.2 Waste Handling

Waste handling includes the transport and storage of radioactive waste and waste treatment products. Defined requirements are given for each step.

3.2.1 Decay Storage

After the waste is generated, it is collected in bags or filled in drums or containers and stored this way. Storing the waste for some months ensures that the short-lived radionuclides have decayed. This way, a significant part of the radioactivity content of the waste is avoided in the treatment chain. Typically, the waste is classified afterwards into the activity categories described in section 2.3.

3.2.2 Transport

Mode of transportation	International/ regional organization	Name of regulation/ agreement code
All	IAEA	Regulations for the Safe Transport of Radioactive Material, TS-R-1
All	UN	Recommendations on the Transport of Dangerous Goods
	UNECE	European Agreement concerning the International Carriage of Dangerous Goods by Road (ADR)
Sea	IMO	International Maritime Dangerous Goods Code (IMDG Code) International Code for the Safe Carriage of Packaged Irradiated Nuclear Fuel, Plutonium and High-Level Radioactive Wastes on Board Ships (INF Code)
Air	ICAO	Technical Instructions for the Safe Transport of Dangerous Goods by Air (TI)
	IATA	Dangerous Goods Regulations (DGR)

Table 3.2: *Safety Regulations for the Transport of Radioactive Material [30]. The organization abbreviations are: International Atomic Energy Agency (IAEA), United Nations (UN), United Nations Economic Commission for Europe (UNECE), International Maritime Organization (IMO), International Civil Aviation Organization (ICAO), International Air Transport Association (IATA).*

Radioactive wastes have to be transported to another site for reasons of treatment outsourcing or disposal. Depending on the mode of transportation, internationally applied regulations are given by the responsible institution. Table 3.2 lists these guidelines [30].
The regulations apply to the activity content, the nuclide content and the labeling to the packages used, which have to be qualified in a specific way.

3.2.3 Intermediate Storage

As a rule, the waste has to be stored until a final disposal is available. For this the waste is stored either on-site or at a central storage facility in barrels or containers, already in the final form required for disposal.

3.2.4 Final Repository

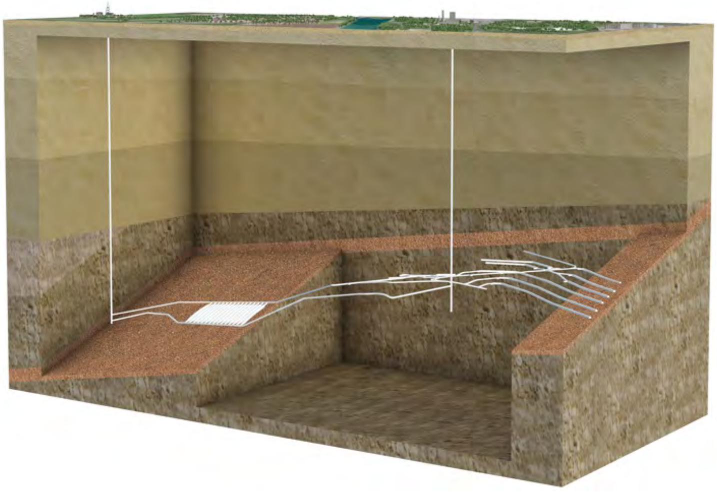

Figure 3.2: *Schematic view of the planned final repository for LILW "Schacht KONRAD". The repository is 800-1100m below surface in a former ore mine. [10]*

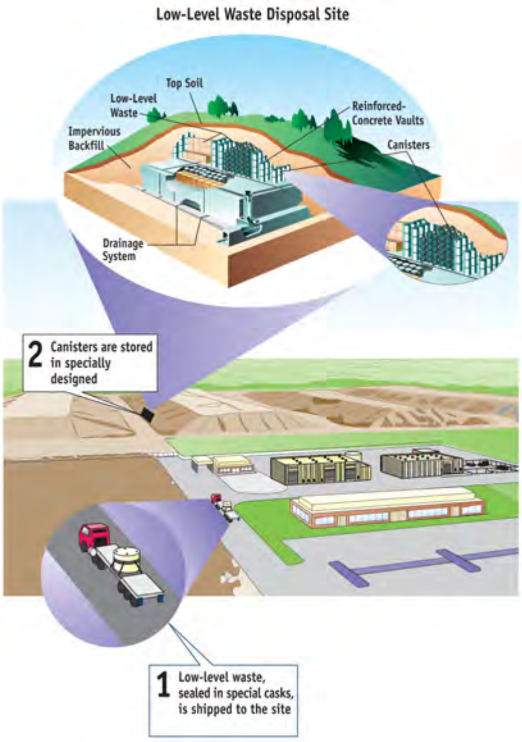

Figure 3.3: *NRC Graphic of a Low - Level Waste Facility [11].*

LILW is recommended to be stored as final disposal in near surface repositories (see chapter 2.3). The different countries may deviate from this recommendation and follow their own strategy for radioactive waste disposal. In Germany for example the LILW will be stored in a deep geological repository, "Schacht KONRAD". The criteria the waste has to fulfill usually are defined by the final repository, e.g. the German acceptance requirements for "Schacht KONRAD" [28].

Picture 3.2 shows a sketch of the German final repository for LILW [10]. "Schacht KONRAD" is designed to hold 303000 m^3 packaged waste, which is expected to be Germany's total production of LILW until 2080 [33]. The packaged waste will be stored $800\ m - 1100\ m$ below the surface in a former ore mine. As a comparison, picture 3.3 shows a sketch of a near surface low level waste repository by the NRC [11] as planned and in operation in the United States and similarly in other countries. This approach is in agreement with the IAEA guidelines on final disposal of LILW.

One approach for a complete waste treatment can be the direct final storage of the waste as produced. The waste is then stored untreated in appropriate containers. This approach of course has the disadvantages of a high final waste volume. It is applied in countries where final repositories with large space are available and the treatment infrastructure is not developed to treat the waste effectively.

In recent times, the retrieval of the stored waste from the final repository is being discussed, especially in Germany [52]. The reasons for this are that there could be the possibility to recycle the waste if needed and treat or recondition it with better methods in a new way. In the final repository "Asse II" in Germany, a closed research facility for the final disposal of LILW, the waste is stored in 700-1100 m depth in a former salt mine. Ground water intrudes into presumably 2 storage chambers. Some barrels rusted and leaked and some activity content was washed out and can be collected inside the mine. Currently, the plan for handling this situation is to retrieve the complete inventory of the "Asse II", as well as the contaminated salt, and provide some reconditioning [52].

The possibility of returning the waste from final repositories may have significant advantages for high level waste. If the HLW is stored unconditioned, without further reprocessing treatment, the waste still bears enough fissile material when stored to be economically recyclable. However, LILW is usually stored already treated and conditioned in a solidified way. It contains few materials that are in general recyclable. The effort of retrieving this waste for recycling is very high. Therefore the later retrievability of the LILW should not be considered during treatment.

3.3 Standard Methods for Radioactive Waste Treatment

Table 3.1 (see page 27) lists the commonly applied treatment methods for LILW. One applicable and often used path is shown in figure 3.1, with deviations possible depending on the installed and outsourced treatment methods. These technologies are the currently used technologies. With a suitable combination of these it is possible to treat the whole spectrum of low - and intermediate level waste.

3.3.1 Decontamination

Decontamination of the waste material is a favored method for material that can be recycled or is only weakly contaminated. Different methods for decontamination exist, depending on the material. Chemical decontamination and mechanical decontamination are well known and broadly applied techniques. Since both produce significant amounts of secondary waste, new technologies have been developed e.g. by using high-power LASERs [53]. Hereby the surface of the contaminated material is cleaned spot wise with high intensity lasers.
For larger parts considered as waste decontamination of the surfaces is usually the preferred processing technique in order to be able to recycle the material, as according to the general waste management guideline (see fig. 2.3).
In a German facility at the Karlsruhe Institute of Technology (KIT) called "Central Decontamination Facility" (Hauptabteilung Dekontaminationsbetriebe, HDB) all mentioned methods except the laser decontamination are applied, especially for radioactive waste parts from decommissioning of nuclear facilities. Applicable methods at the HDB are [54]:

- "physical procedures such as steam or sand blasting,
- mechanical surface removal such as grinding, planning, milling and turning,
- chemical procedures such as chemical stripping or leaching,
- physical-chemical procedures such as dry ice blasting"

These points summarise the current level of advancements in decontamination methods.

3.3.2 (Super-)Compaction

Solid waste can be treated or pre-treated by (Super-)Compaction to reduce the volume. The waste is packed into compactable drums and pressed with a force of up to 2000 Mg [9]. The result is a compacted pellet that is stacked and stored in larger drums. Alternatives are bailing pressing and in-drum compaction. Compaction is a generally applied method for volume reduction of combustible waste as a pre-treatment, for incombustible waste, and also combusted waste.

3.3.3 Vitrification

Vitrification describes a treatment method of liquid waste in a way that the waste is solidified within a glass-like compound. This is achieved by melting glass particles together with additives and the liquid waste. An amorphous, solid ingot is created that is suitable for final disposal. Vitrification is usually not applied to all liquid waste, since the technical effort is very high and low level radioactive liquids are more easily treatable by evaporation and by drying the concentrates. For high level radioactive liquid waste though, vitrification is the most advanced and currently employed treatment method. One example is the vitrification plant in Karlsruhe, pictured in 3.4 ("Verglasungseinrichtung Karlsruhe", VEK). This plant was constructed to treat all German high level liquid waste (HLLW) that was generated in research. High level liquid waste occurs in reprocessing of nuclear fuel. Commercial reprocessing activities for German fuel was performed in France, where the generated HLLW was sent back to Germany as vitrified products.

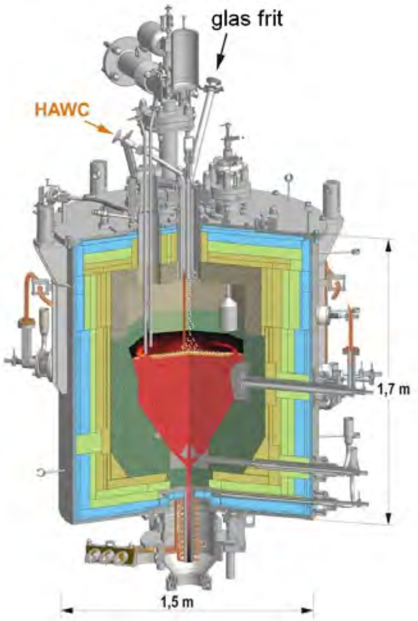

Figure 3.4: *Design of the VEK melting furnace, [12]*

3.3.4 Metal Melting

All not too highly contaminated metallic wastes from NPP operations and decommissioning can be collected for recycling by metal melting. The waste is reduced in volume and the radioactivity content is divided in the melting process: the input is separated into molten metals, slag and dust where the radioactivity is collected in the slag and dust and securely encased in the metal melt [55]. The metal product can then be recycled in the nuclear industry for storage containers, shielding or other metallic constituents.

3.3.5 Pyrolysis

Pyrolysis is a flameless thermal process which is performed to treat organic material. Pyrolysis is generally applied together with steam reforming or incineration, since the products are charcoal and combustible gases [24]. Pyrolysis plants are capable of processing ILW with doses of up to 1 $\frac{Sv}{h}$ and can particularly be used for treatment of ion exchange resins. These are typically contaminated with inorganic radionuclides, remaining in the solid residue after the pyrolysis process, and thus being separated from the volatile organic constituents. The result is a granular, mainly inorganic material. A volume reduction of approximately 70% can be achieved, though a possible limit on specific activity for the outcome has to be considered [56].

3.3.6 Grouting/Cementation

To solidify the pre-treated waste, particularly ash from incineration and wet waste processing, cementation is performed to achieve a solid, storable final product. The input is mixed in a proper fraction with cement. The result is non leaching and stable and can therefore easily be handled and used for final storage.

3.3.7 Drying/Evaporation

There are several methods for drying and evaporating wet and liquid waste. A frequently used method is the in-barrel-drying. The stored liquid waste is transferred to a barrel where it is heated continuously. The accrued steam is drawn off, condensed and collected.

For the larger liquid streams, evaporators are used, resulting in an evaporator concentrate with a solid fraction of 15% to 30% [9].

3.3.8 Incineration

Incineration of low level radioactive waste is a well proven technology and in use in many states for treatment of combustible LILW [24]. Basically, the organic material is oxidized and transformed into dry ash residues. For additional stabilization of the remaining ashes it can be solidified, for example by grouting/cementation, or added to and mixed with other types of waste for more efficient packing.

Depending on the chemical and radiological composition of the waste, some special treatment or processing may be necessary.

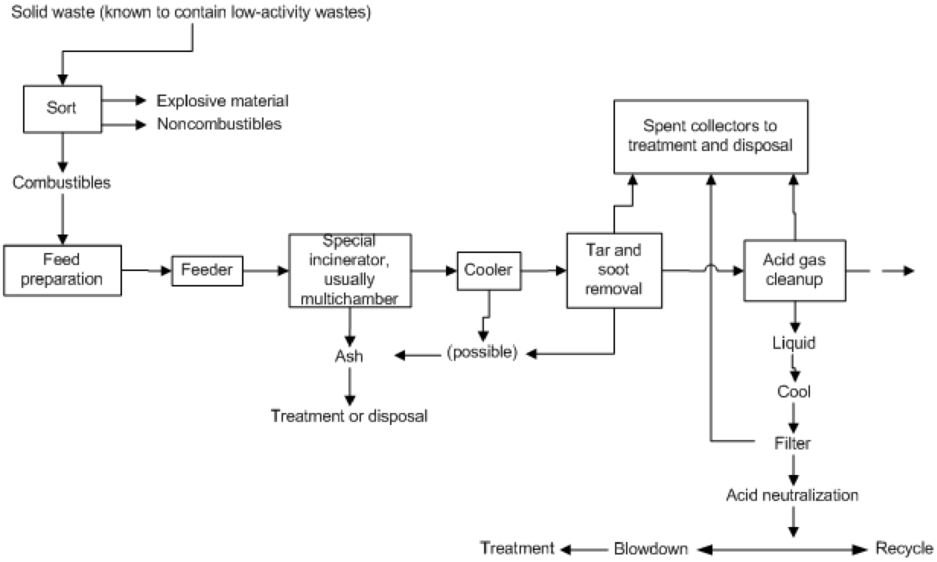

Figure 3.5: *Generic flow schematic for an incinerator for "Low - Level Wastes Composed of Cellulosics, Animal Remains, Plastics (including PVC), Rubber Gloves, and Tygon Tubing (Materials that Produce Acid Gases upon Incineration)" [13]*

The low level waste from NPP operations can contain for example significant amounts of PVC. During the incineration of this waste acid gases or other hazardous gases are produced in the off-gas stream and have to be treated and filtered accordingly. In figure 3.5, taken from [13] an exemplary schematic flow chart for an incinerator of low level wastes containing special byproducts from NPP operation and research is given. The most important steps are the sorting out of untreatable material and the application of a thorough off-gas treatment, which generally includes a secondary incineration chamber for the full oxidization of the off-gas.

Incineration of LILW is a common treatment for all combustible waste produced in NPP operations since the volume reduction factor is the highest (see table 3.1) and combustible solid waste includes most of the generated waste (see figure 2.2).

Plasma treatment is a method comparable with incineration but with a higher treatment temperature. The effect is that in addition to the burning of the combustible material, the incombustible material can melt. Plasma treatment will be investigated further in this work.

3.3.9 Summary of Treatment Technologies

Relevant for LILW are the mentioned decontamination, incineration, (Super-)Compaction, metal melting, pyrolysis, grounting/cementation and drying/evaporation. Vitrification is only performed with HLW. Current radioactive waste treatment plans foresee a treatment method for each waste according to its radiological, chemical and mechanical type. Solid materials that can be reused are decontaminated. Typically combustible materials are treated with incineration and incombustible materials are treated with compaction. Spent resins, which have a comparatively high specific radioactivity, are treated with drying or pyrolysis. A comprehensive plan that includes all types of waste has to be performed when treating the waste.

3.4 Waste Treatment Facilities

Usually, the entire waste produced is sorted and treated in a facility at the site of the plant or in a central facility for several power plants. New nuclear power plants are generally planned together with combined waste treatment facilities.

As an example, the suggested treatment facilities for the Areva "EPR" in the UK includes 3 different main waste treatment paths. A distinction is made between the

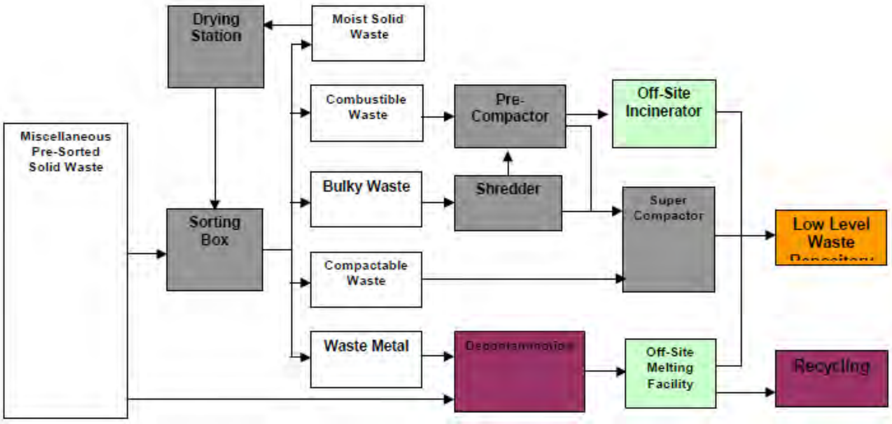

Figure 3.6: *Suggested LLW treatment path for the Areva UK EPR [14]*

"liquid effluent treatment system", the "storage and treatment of solid LLW" and "storage and treatment of solid ILW streams" [14]. The liquid waste treatment steps are mainly the evaporation and dewatering. The solid waste treatment steps are the drying, shredding, pre-compaction, super-compaction and decontamination. Incineration of combustible solids and melting of LLW metals is planned in a facility off-site. Figure 3.6 pictures the solid LLW flow chart.

In comparison the comprehensive waste treatment facility "site radwaste treatment facility" (SRTF) is planned as a treatment facility for 6 Westinghouse AP1000 units in Sanmen, China. The LILW of the 6 units is collected during the operation and treated in a combined central plant. "The final product of the SRTF-treatment is a 200-L-drum with cemented waste or waste packages for storage in a local storage" [15]. The entire treatment will be executed on-site. There is no intention for incineration and melting, only supercompaction of the dry solid waste. In addition, stations for sorting, drying, evaporation of the liquids, grouting and cementation are implemented.

Figure 3.7 pictures the process diagram for the SRTF facility in Sanmen (China). The different collected waste streams are appropriately merged and treated according to their main properties.

Compressible dry radioactive waste, non-compressible dry radioactive waste and mixed dry radioactive waste are all collected in the plants in "yellow bags", which are opened by a shredder. The shredded material is manually sorted in a sorting box into 160-L drums. The sorting criteria are e.g. metallic waste, wood, plastic, glass and textile [15]. After drying in a 12-Drum-Dryer the drums are supercompacted. The design of the supercompactor is pictured in figure 3.7. The collected ventilating and air conditioning filters are also supercompacted after precompaction.

In figure 3.7 at the top left a sketch of a trailer with a shielding cask is shown, mounted on a movable platform belonging to the filter cartridge processing system. The trailer transfers the spent filter cartridges to the SRTF. The 200-L drum is directly transferred to the grouting station without further treatment.

Liquid radioactive waste mainly consists of samples from the chemical analysis of the reactor coolant system, decontamination water and wash water [15]. The liquids are evaporated to increase the solid concentration and treated in an In-Drum-Dryer. In a last step the residual is grouted in a 200-L-drum.

Ion exchange resins are transferred to a conical dryer, in which drying and heating of the resins is achieved. The hot resins are packed in 160-L drums and compacted in the supercompactor.

The final result in the SRTF process is a storable grouted 200-L-drum which is stored until release for final disposal from the treatment building in the drum storage system.

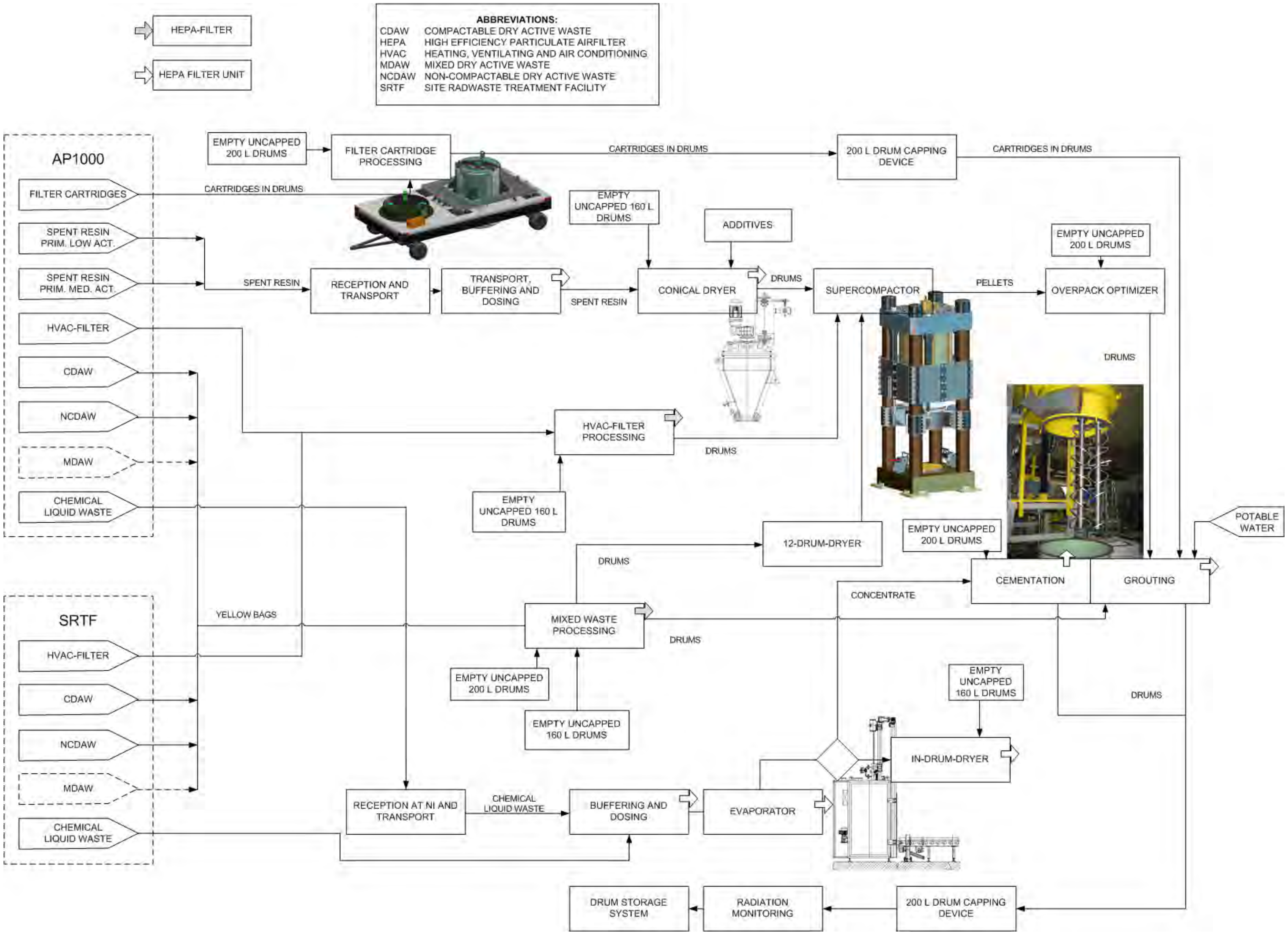

Figure 3.7: *Process diagram for the planned site radwaste treatment facility for the AP 1000 in Sanmen, China. Pictured in detail are the trailer for the filter cartridge processing system, the conical dryer, the supercompactor, the in-drum-dryer and the cementation device [15], [16]. Not included in the chart are the evaporator concentrates.*

4 Aspects of Plasma Treatment

4.1 Plasma Technology

A plasma is a partly ionized and therefore electrically conducting gas with a high temperature. Any gas can become a plasma by heating above the ionization temperatures of the atoms. If the plasma is directly put into contact with to a target material, thermal energy is transferred to the target which is high enough to destroy the molecular bonds of the target on a microscopic scale and melt or incinerate the target on a macroscopic scale. A plasma for thermal treatment of materials can be created by an electric discharge. The ions and electrons in the plasma are in thermal equilibrium in the order of 10000 K up to 20000 K under atmospheric density.

4.1.1 Plasma Physics

Since plasmas occur in very different phenomena and applications, the field of plasma physics is very extensive. In this work only a basic introduction to the field which is of use for this specific application is discussed.

A common physical definition of a plasma is: "A plasma is a quasineutral gas of charged and neutral particles which exhibits collective behaviour." [57]

Quasineutrality basically means that the bulk of the gas is free of large electric potentials or fields - the number density of electrons is about the same as the number density of ions. There still exist electromagnetic phenomena due to microscopic charge inequalities and the plasma is not completely neutral, either.

Collective behavior of the plasma is achieved by its charged particles. The resulting Coulomb force is long ranged and therefore plasma particles affect each other in motion. The motion is not only dominated by local effects like simple collisions, the state of the plasma in remote regions is important as well.

A classification is made between "thermal" and "non-thermal" plasmas. In thermal plasmas the electrons and heavy particles have the same temperature - they are in thermal equilibrium.

4.1.1.1 Saha - equation

The equation which describes the quotient of the ionized particles to neutral particles is called the "Saha - equation" and can be derived from thermodynamics [17]. The Saha equation (eq. for the first ionization in local thermal equilibrium conditions (the temperature of ions and electrons in a local environment is the same) looks like the following.

$$\frac{n_e n_i}{n} = \frac{2Q_i}{Q_0}(\frac{2\pi m_e kT}{h^2})^{\frac{3}{2}} exp(-E_i/kT) \tag{4.1}$$

Here n_e, n_i and n are the densities of the electrons, ions and neutral particles, respectively; Q_i and Q_0 are the partition functions of the ions and neutral particles; m_e is the electron mass, h is Planck's constant, k is Boltzmann's constant, T is the temperature and E_i is the specific ionization energy for the first ionization of the atoms.

The partition functions Q_i and Q_0 are the sum over all states in the denoted configuration:

$$Q_i = \sum_s g_{i,s} exp(-E_{i,s}/kT) \tag{4.2}$$

where $g_{i,s}$ is the statistical weight of the respective particle species, i.e. the corresponding degeneracies of the energy levels $E_{i,s}$ of the excited states.

The factor 2 in equation 4.1 is the value for the electron partition function which is 2 due to two possible electron spin orientations.

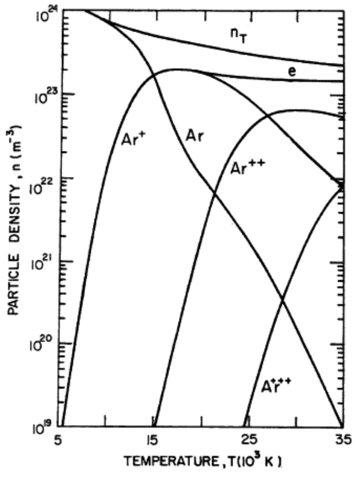

Figure 4.1: *Composition of an argon plasma at 100 kPa [17]*

The Saha - equation can be considered a "law of mass action" for the ionization process [17]. For a given pressure and gas species constants the plasma composition can be calculated as a function of the temperature. Figure 4.1 pictures the composition of an argon plasma at a pressure of 100 kPa. The plasma behavior of Argon is of special interest in this work, since later experiments are performed with Argon gas.

Between the temperature range of 5000 K and 15000 K the density of the single ionized Argon increases up the point of an equilibrium in densities at about $2 \cdot 10^{23} m^{-3}$. The total density $n_T = n_i + n_e + n$ decreases since the pressure is kept constant. At 15000 K the temperature and thus the energy is sufficiently high to achieve a second ionization of the Argon. At the point where the density of neutral Argon is the same as the density of Ar^{++} around 20000 K, the Ar density is already negligible compared to the amount of ions and the plasma can be regarded as fully ionized.

4.1.1.2 Electrical Conductivity

The value of the electrical conductivity of a plasma can be important for modeling and creating a plasma due to it being present as a proportionality factor in Ohm's law. It is an essential property of any plasma and is created by the drifting movement of the charged particles. The electrical conductivity κ is calculated as in equation 4.3:

$$\kappa = e \cdot n_e \cdot (b_e + b_i) \tag{4.3}$$

with the electron charge e, electron density n_e and mobility of the electrons and ions b_e and b_i:

$$b_j = \frac{e\lambda_j}{m_j \vec{u}_j} \tag{4.4}$$

where λ_j is the mean free path of a particle, m_j the mass and $\vec{u}_j$ the drifting velocity of the particle [58]. The mobility of electrons is much larger than the mobility of ions due to the difference in mass. Therefore the term b_i in 4.3 can be neglected. The mean free path of the electrons is calculated with:

$$\lambda_e = \frac{1}{n_i \sigma_{scatter}}. \tag{4.5}$$

The scattering cross section $\sigma_{scatter}$ is weakly temperature dependent but can be approximated with $\sigma_{scatter} = \pi \cdot r_i^2$, where r_i is the ion radius (for example Argon: $r_i = 158\ pm$). n_i is the particle density of ions in the plasma. The scattering on neutral particles can be neglected due to the absence of Coulomb forces and the electron - electron scattering is neglected due to the cross section being very small compared to electron-ion scattering.

Consequently, the current density is with Ohm's law $\vec{J} = \kappa \vec{E}$ with the electric field strength $\vec{E}$:

$$\vec{J} = e^2 n_e \frac{1}{m_e n_i \pi r_i^2} \frac{1}{\vec{u}_e} \vec{E} \tag{4.6}$$

The electric conductivity is mainly dependent on the particle density and the electron drift velocity. In the case of single-ionization plasmas where the electron and ion density is the same, the electric conductivity is even simpler when the densities in 4.6 cancel each other out.

Since the process dominating the mean free path is the ion-electron scattering i.e. the recombination, a plasma can only keep its state under continuous new ionization by a steady energy input.

4.1.2 Creation of Thermal Plasmas

To create a plasma a steady energy input is necessary to ionize a gas. The easiest way to generate a plasma on a technical level is by creating an electric discharge. The electric arc forms a plasma. A sufficient number of charge carriers must be generated in the gas to achieve electrical conduction. Flashes during thunderstorms are an example for the electrical breakdown pheonmenon.

In the simplest way, a plasma is created by the electric arc between two electrodes through a carrier gas. Two main options exist for the application as a thermal treatment device. When one of the electrodes is the treated material, thus the electric arc is directly targeting the material, the method is called "transferred". Alternatively the carrier gas between two electrodes can be used for treatment of the material by directing it through the electric arc spot onto the material. This method is called "non-transferred".

In RF Plasmas the plasma is created by inductively coupled RF coils. This method is independent of electrodes. The discharge is supported by a time-varying magnetic field.

A newer sub kind of non-transferred plasma creation is the microwave-induced plasma. In this kind, the plasma is created by applying a strong electrical field to a limited area, which is strong enough to ionize significant numbers of atoms to form a plasma. The electrical field is produced by a microwave signal. The plasma is maintained by operation at the resonance frequency of the conductors. It has to be taken into account that by ignition of a plasma the electrical properties of the setup are changed: The plasma provides additional charge carriers which form an electrical body which is the plasma column. Therefore, different resonance frequencies at ignition and operation of the torch exist.

In a practical example the ignition is achieved by a three staged impedance transformation, called Γ-transformer [59]. The microwave signal with 2.45 GHz enters the transformation network, which is implemented as the conductors in the plasma torch used. An impedance transformation from $Z_0 = 50\Omega$ to approximately $0.5 \cdot 10^6\Omega$ is achieved with this Γ-transformer, which allows a very high power efficiency of the torch [60].

4.2 Advantages and Limitations of Plasma Treatment

The general principle of plasma treatment of hazardous materials is the heating of all waste to very high temperatures. The combustible parts will incinerate and combust, and the non-combustibles will melt. The residual is a mixture of molten slag and ash. On a microscopic scale the plasma treatment provides sufficient energy to break the molecule bonds in the non-combustible parts of the treated material. After the direct treatment in the plasma focus, the atoms can then fuse, i.e. they form a new amorphous structure. This structure would be glass-like and of high stability and leaching resistance.

However, the need for electricity as the power source is also a big disadvantage, since it is an expensive form of energy and the economic feasibility of any plasma treatment installation has to be in terms of operation costs [61]. Another disadvantage is the small distribution and therefore little experience with plasma treatment plants on an industrial scale.

4.3 Plasma Torch Technology

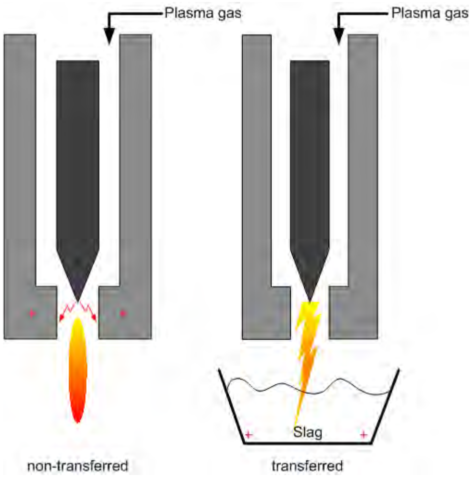

Figure 4.2: *Basic principle of non-transferred and transferred plasma torches*

As shown above, plasmas can be created either by an electric arc or a radio frequency discharge. The plasma creation by electric arcs dominates in the waste treatment sector due to a better insensitivity to changes in plasma conditions [61]. The major difference in possible application design, when the plasma is created by an electric arc, is the type of the torch, which can be "transferred" or "non-transferred". In the transferred design, the plasma arc is seated directly between an anode and the target, which has the role of the cathode. In this configuration the complete energy of the arc is transferred to the target material. Peak temperatures of the plasma arc are typically 12000 K to 20000 K [61]. In the non-transferred design, an electric arc is built up inside a chamber into which a gas is injected. The gas is heated up and ionized by the arc and then applied to the target as a plasma jet. The resulting treatment temperature is lower than in the transferred design with peak temperatures of 10000 K - 14000 K [61], but still high enough to produce the intended effects on mixed waste material. In contrast to the transferred design, the treated material does not have to be electrically conducting itself. The lifetime of the electrodes is typically higher for the non-transferred design and can reach several thousands of hours [62]. In figure 4.2 the basic functionality of the types is sketched.

4.3.1 Transferred Torches

Transferred arc torches can have multiple rod electrodes and can operate in DC, AC or three-phase mode. However, newer installations are preferably equipped with a DC torch [17]. The reason for this is that in the high power regions AC torches are more difficult to operate and the current development aims for a high torch power. Transferred torches are a standard device in metallurgy, where the treated material is metallic and conducting. The plasma arc from the torch directly transfers the

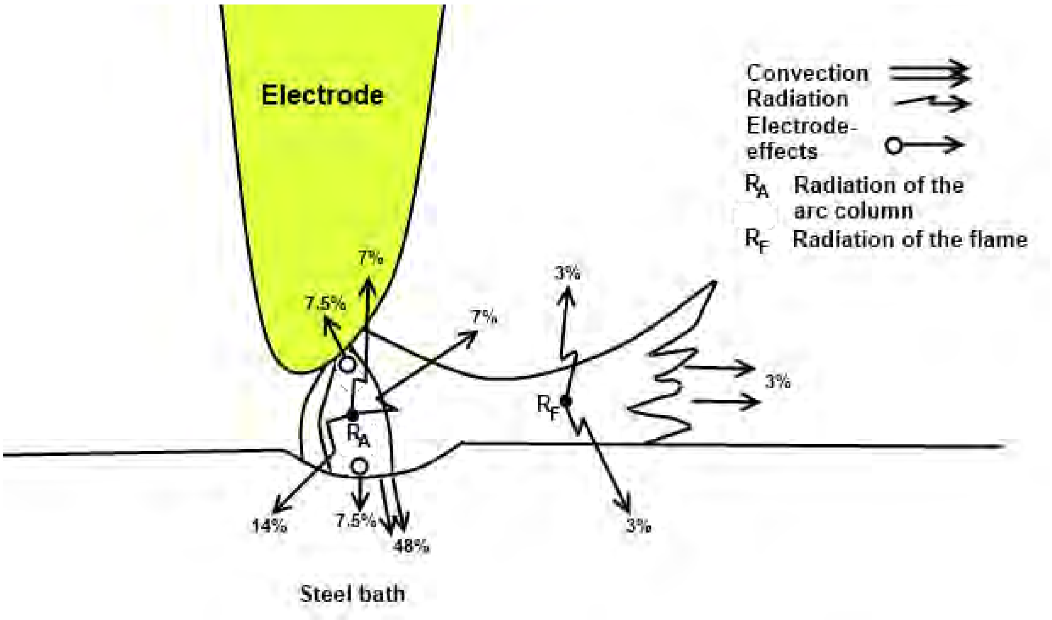

Figure 4.3: *Experimental power balance of an electric arc with* 7 *kA and* 143 *V [18]*

plasma energy to the material, melting it. In case of a non conducting material, as in the case of typical waste from nuclear power plant operations, an additional way to achieve a conducting surface and therefore the ignition of the plasma arc has to be constructed.

Transferred torches exist in a wide range of power levels of up to several megawatts power. The losses are low if the used material is an electrode.

Picture 4.3 pictures the measured power balance for an electric arc on molten steel with 7 *kA* current and 143 *V* Voltage and therefore 1 *MW* power taken from [18]. In this example 72.5 % of the total power transferred to the cast. The loss effects are: convection heat transfer back to the electrode and to the oven wall, radiative heat transfer to the adjacent surfaces and electrode effects on the electrode and the steel bath.

4.3.2 Non-transferred Torches

Unlike transferred arcs, where the arc length is of minor importance to the temperature at the impact point, the temperature in a non-transferred plasma decreases with increasing distance from the plasma creation spot.

4.3.3 RF - Torches

In a Radio Frequency (RF) induction plasma reactor the plasma is induced by magnetic coils surrounding the reactor chamber. The plasma has no contact with the tip of an electrode thus there is no electrode erosion in this setup. However, an RF-plasma is comparatively sensitive to disturbances in the electric power supply. Therefore only clearly defined components can be introduced in a RF-chamber. RF-plasma torches have been used for treatment of hazardous liquids that are injected into the RF-torch [61].

4.3.4 Microwave Torches

So far, microwave torches are employed in plasma technology only for specific low power purposes, since dimensions of the needed energy supply are very large and usually the same disadvantages as in the case of RF-plasmas apply.
Recently, a potential-free microwave plasma torch has been developed at atmospheric pressures with abilities comparable to arc torches [63]. Here the electrode is built as a small hollow cannula and the plasma gas flows directly through the electrode. At the tip the gas is ignited and forms the plasma. The dimensions of the electrode are such that the electric field created at the electrode tip is maximal, which is sufficient to ionize the plasma gas atoms. The plasma is created outside of the electrode, which allows for the use of electrode materials with lower melting points than tungsten such as for example copper. The cannula is sufficiently cooled by the plasma gas stream.
As described in chapter 4.1.2, the electric field properties change after the plasma ignites at the tip of the cannula. Therefore the resonant operating frequency deviates from the ignition frequency. One limitation of present microwave plasma applications is that either the frequency has to be adjustable or one fixed frequency is taken with significant losses due to no optimization. Figure 4.4 pictures the general setup of such a microwave plasma torch. λ is the wavelength of the microwave signal.

During this work, an enhancement of a microwave plasma torch has been developed, which allows an operation at a single fixed frequency and with this an increase of the power supply in the order of several kW by the use of magnetrons as a microwave generator. This was made possible by a change in the inner topol-

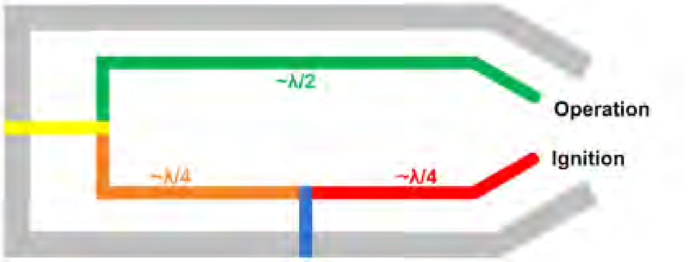

Figure 4.4: *Inner topology of the plasma torch with ignition and operation path [19]*

ogy of the plasma torch. In figure 4.4 this is pictured as the green electrode being the operating electrode and the red electrode as the ignition electrode. Basically, the lengths of the electrode had to be fitted in a way that during ignition the resonance frequency is met with the ignition electrode and during operation the resonance frequency is met with the operation electrode. The electrical conductivity of the plasma column had to be calculated as illustrated in a simplified description in chapter 4.1.1.2. This development is explained in more detail in [19].

4.4 Industrial Applications of Plasmas

4.4.1 Metallurgy

The application of thermal plasma, or more exact the use of electric arcs for metallurgical purposes has been performed for over 100 years. Typically, conventional arc furnaces are equipped with consumable electrodes, i.e. electrodes made of e.g. graphite that erode during operation. Newer developments make use of non consumable electrodes with various advantages: in addition to the elimination of graphite electrode wear, the so called arc-plasmas or plasma furnaces show reduced noise levels and higher yields [17].

Plasma metallurgy is applied for melting and remelting applications as well as extractive metallurgy. Remelting is performed for melting of scrap for recycling purposes. Other melting/remelting applications are alloying, iron melting in cupolas and tundish[1] heating. In general, DC transferred arc plasmas are used with power levels up to 10 MW. The conducting material serves as an electrode and the major

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[1] Tundish: Container in metal melting used to feed molten metal into an ingot. The temperature of the tundish is controlled to achieve a high quality cast.

energy input is directly at the impact point of the electric arc.
Extractive metallurgy is the extraction of metals from their respective ores. Though because of the high energy demand of this applications, few industrial scale applications exist. For the desired throughput above 3 $\frac{ton}{h}$ a furnace capacity of 100 MW is desired, which is beyond present technology [64]. However, due to the very high peak energy of plasmas, a number of potential extractive applications are possible , including the smelting of virgin ores, preheating of gases as part of a complete process and specialized systems for recovery of metals from waste assemblies [64].

4.4.2 Treatment of Hazardous Substances

The decomposition of materials by exposure to a plasma is a valid application in the field of hazardous waste treatment: A chemical destruction of the hazardous waste is desired to achieve a stabilized product and the respective materials are possible to treat with plasma. Compared to other thermal treatment technologies it has various advantages:

- Very high treatment temperatures above the melting point of any material undergoing treatment. It has the effect that the material is decomposed into its constituents and can melt after treatment into a new, amorphous mass. The product is reduced in volume compared to the original material, since the combustible parts were burned and the melting deleted any cavities and unused space within the material.

- Moreover, the result of the treatment can be vitrified by adding appropriate additives to the material feed and the hazardous material can be encapsulated in a non-leaching stable material.

- Plasma treatment reaction chambers (Plasma reactors) have a high energy density. In comparison with other thermal treatment methods, a small footprint of the installation with a comparable throughput is obtainable. They can even be used as mobile installations. The high energy density and the small size also allows for a rapid start-up and shutdown of the system.

- No combustible gas is required due to the use of electric arcs, either as the direct energy carrier in transferred operation, or for the heating of the non-combustible process gas in non-transferred operation. Therefore the destruction of expensive gas is avoided. The process gas can be chosen from a wide range of gases, e.g. Nitrogen, Air or Argon, which results in a better control of the occurring process chemistry.

4.4.2.1 Medical Waste Treatment

As an example, thermal plasma treatment of hazardous wastes is applicable for medical waste. Medical wastes can be infectious and thus have special requirements for collection and disposal. The amount of medical waste that is produced in a hospital is typically usually around $200\frac{t}{a}$ to $500\frac{t}{a}$ [65]. The amount of radioactive waste produced in a nuclear power plant is around $90\frac{t}{a}$ (see chapter 2.2.3), thus these problem statements are comparable.

Few plasma plants exist for the treatment of medical waste from hospital operations. The devices developed at the Russian academy of science [66] and at the Jeonju University Incheon Korea [67] are two examples which have a different approach: The Russian plant is working with a 3-Phase AC - plasma torch with 500 kW power and the oven is designed as a rotary kiln. A throughput of $200 - 300\frac{kg}{h}$ with a specific energy requirement of $0.8 - 1\frac{kWh}{kg}$ is achieved. The Korean plant is a shaft furnace equipped with a DC - torch than can be operated in transferred- and non-transferred mode with a plasma power of 40-85 kW. The throughput is in the order of $50 - 100\frac{kg}{h}$.

However, plasma torch treatment is not a standard application in medical waste treatment either, therefore no large scale facilities exist yet. It seems economically suitable for an annual waste amount of $100t$ and above as found out by Fiedler [65].

4.4.2.2 Syngas

In economic considerations for plasma treatment plants, the possibility of producing syngas as a valuable by-product is often taken into consideration. Syngas is typically rich on combustible hydrocarbons. The reuse of this syngas, which is produced with the off-gas, eliminates the typical drawback of a high energy consumption. Some processes already include a mechanism to generate the needed electricity for the plasma torch out of syngas combustion.

However, in the case of radioactive waste treatment, this option is not desired. The produced syngas could contain the radionuclides Tritium and Carbon-14, thus a separate combustion or collection would need additional effort for radiation protection and a limitation on the release to the environment.

5 Improvement of the Plasma Method for the Treatment of Radioactive Waste

5.1 Existing Facilities

The treatment of radioactive waste with plasma torches seems economically suitable, since the costs for disposal are very high even when compared with other hazardous wastes. Up to now, there exist two prototype facilities which were designed for the treatment of radioactive waste, and some full-size facilities are presently under construction.

The plasma melting furnace at the ZWILAG in Würenlingen, Switzerland [21] has been in operation the longest. A small scale pilot plant is in operation at the research center RADON in Russia [31]. Under construction are a plasma plant at the nuclear power plant in Belene which is based on the RADON design, and in Kozloduy a plant is being constructed which is loosely connected to the ZWILAG design. Below, these (known) plants are being shortly introduced and their specific design choices are being pointed out.

5.1.1 The ZWILAG Plasma Treatment Plant

The plasma plant at the ZWILAG has been in operation since 2004. Construction, which started in 1997, was afflicted with major problems on the side of the construction company, which at the end went bankrupt in the wake of the complications. The ZWILAG finalized the plant themselves.

Figure 5.1 pictures the sketch taken from the patent file on the ZWILAG plasma melting furnace [20], modified with the descriptions. The design principle is based on a rotary kiln in vertical operation. The melting furnace is equipped with a

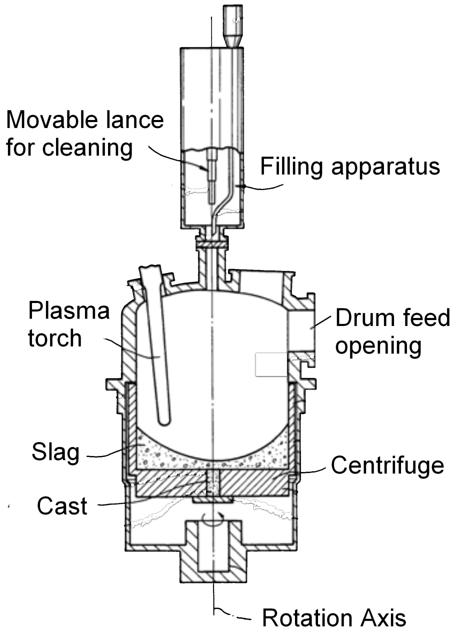

Figure 5.1: *Patent sketch for the ZWILAG plasma melting furnace, modified with description [20].*

1200 *W* DC transferred torch. Due to this setup and that the waste material is not conducting, additional engineering was needed to enable the ignition of the plasma. This was realized by adapting the oven ground plate with graphite pins that build up the first electric arc discharges. The treatment process is as follows:

- A waste drum is fed into the oven. During feeding the drum is continuously cut in slices with a propane burner positioned at the oven inlet. The feed is designed to process whole waste drums automatically, as pictured in figure 5.2.

- During the feeding the oven is heated with the plasma torch. The gas that is used for ignition is Helium, while in operation nitrogen can be used as the plasma gas [21].

- Up to five barrels are fed and molten. The barrels are automatically taken from a storage system in a manner that optimizes the slag properties with respect to the barrel contents. The slag is heated for several hours. The needed treatment time is gained from experience. On average one 200-L-barrel is treated per hour [65].

- When the slag is homogeneous and all solids are molten, the slag is cast. This is done by adjusting the rotation speed of the centrifuge so that the slag can drop out in a controlled way in the middle of the centrifuge through an outlet, which is filled during the operation stage with a special sealing system employing sand as a closure [21]. This way a possible cladding of the outlet channel by molten material is prevented. In case of an occurring cladding, a movable lance for cleaning the outlet mechanically can be used which is installed at the top of the oven (see fig. 5.1).

- The treated material is cast as an ingot, which is stored for up to 24 hours in a storage system for cooling down. Afterwards a new batch can be treated.

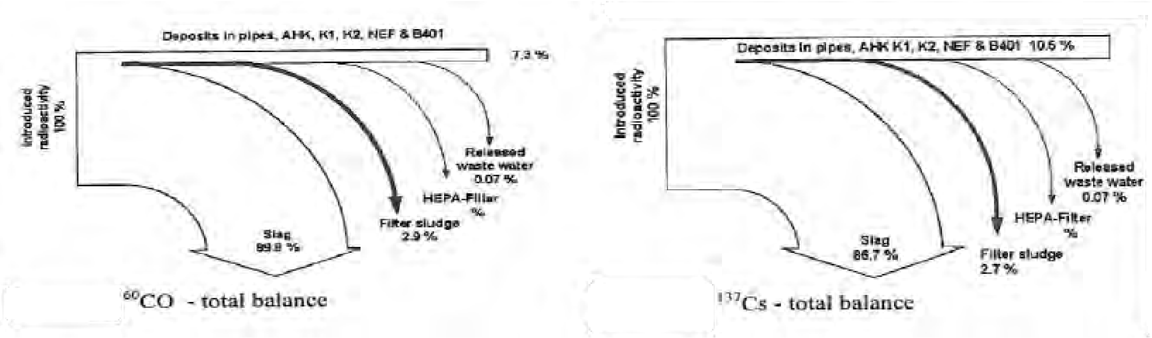

Figure 5.3: *^{137}Cs and ^{60}Co total balance at the ZWILAG plasma melting furnace [21]*

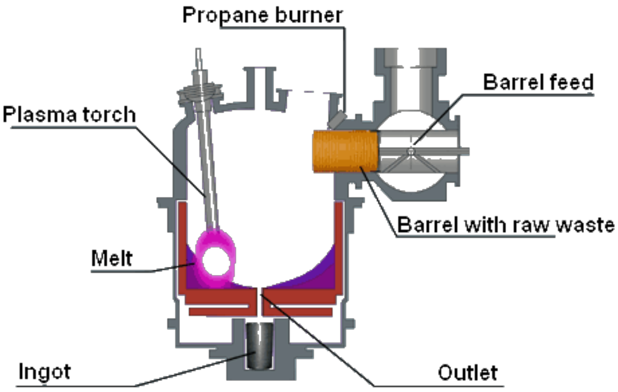

Figure 5.2: *Schematic of the ZWILAG melting furnace [22]*

Typically two treatment campaigns of several weeks are performed each year. After the startup in 2004 the amount of treated barrels and cast ingots per campaign increased strongly. The number of treated barrels in total up to 2011 was around 6000 and in the year 2011 1075 barrels were treated [68]. A mean volume reduction factor of 4 is achieved. It is important to note, that the waste drums that are delivered to the ZWILAG already contain compressed material, thus the volume reduction factor from raw waste material would be significantly higher: The typical volume reduction in compression is factor 3 to 6 which would mean an average volume reduction in the plasma plant from the raw material of 18.

One necessary requirement as named by the ZWILAG is the need of a specifically trained operating crew with technical capabilities. The plasma plant can not be regarded as a maintenance-free and easy to use device.

In the course of the licensing of the facility the quantitative flow of Cobalt-60 and Caesium-137 was measured [21]. The results show a full retention of these nuclides (see fig. 5.3), including a retention of volatiles in the off-gas system. The gaseous emissions from the off-gas treatment of the plant were measured to be free from Cs and Co. However, 7.3% of the introduced Cobalt and 10.5% of the Caesium could

Figure 5.4: *Maintenance work at the plasma torch at the GEKA Munster*

not be retrieved and are assumed to deposit in the pipes of the off-gas and post-treatment systems. The bulk of 89.8% of the Co and 86.7% of the Cs is to be found in the slag, as intended.

There are two plants with a technically identical basis to the ZWILAG plasma plant: One facility in Tsuruga, Japan for the treatment of the waste from the Tsuruga nuclear power plant operations and one facility at GEKA Munster, Germany for the treatment of chemically hazardous soil. The Tsuruga plant is still under development and still not functioning as intended. The plant in Munster was adapted to the standard design with a higher power plasma torch of 2 MW, which helped in igniting the plasma and achieving a constant treatment of the waste. Figure 5.4 pictures a photograph of maintenance work of the plasma torch. In difference to the plants working with radioactive material, the maintenance work at the GEKA Munster facility is possible with fewer security arrangements than in facilities operating with radioactive materials.

5.1.2 Other Commercial Radioactive Waste Plasma Treatment Devices

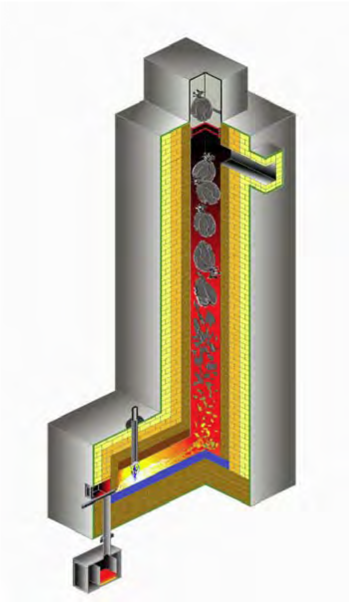

Figure 5.5: *Sketch of the PLUTON plasma shaft furnace at SIA RADON [23]*

One new commercial construction is a combined waste treatment plant at the site of the nuclear power plant in Belene, Bulgaria. The plant design is based on a shaft furnace which was designed and operated as a prototype at SIA RADON, Moscow, Russia. Table 5.1 shows the basic properties of the research device called PLUTON, taken from [31]. Picture 5.5 pictures a sketch of the plant. The waste bags fall through the shaft into a bed of molten material, which is created by the two plasma burners. The slag is collected at the bottom. The created off-gas passes through the shaft and exits at the top to the off-gas treatment chain.
Worth for mentioning is the significant "^{137}Cs loss" of 7-9% (tab.5.1).

The plant at Belene is under construction and expected to be completed in 2014 [69]. A throughput of 6.5 tons per week or 250 tons/year is aimed for [62]. The purpose of the plant is the treatment of operational and historic radioactive waste from the plant's site. It has an estimated average volume reduction factor of 40 [69].

Another planned plasma plant is under construction at the site of the Kozloduy NPP, Bulgaria. The constructors are the company Belgoprocess, who operate the

Solid waste capacity, kg/h	200-250
Overall dimensions, m	$12 \times 18 \times 12$
Number of plasmatrones	2
Electric power of plasmatrone, kW	100-150
Specific power expenses, kW·h/kg	0.5-1
^{137}Cs loss*	7-9 %

Table 5.1: *Plasma plant parameters of the PLUTON plant at SIA RADON [31]. *:depends on waste composition*

Belgian waste treatment and storage facility in Mol, and Iberdrola Ingeniería y Construcción in a joint venture. The plant is funded by the European Bank for Reconstruction and Development (EBRD) with 70 % and the Bulgarian state with 30% [70]. The total costs to build the turnkey plant are 30 Million Euro [71].
The plant is equipped with a 500 kW non-transferred plasma torch. The throughput to be achieved is 6.5 tons per week / 250 tons per year. The off-gas treatment is planned similarly to the off-gas chain at the Belgoprocess incinerator site in Mol, Belgium. The facility is scheduled to start up in 2014.
The plant is loosely based on the ZWILAG design, in the sense that the waste is treated batch wise in an oven with one central plasma torch. The experiences made at the ZWILAG plant lead to this different basic design.
The purpose of the plant is the treatment of operational and historic radioactive waste from the plant's site. The volume reduction factor for a mixture of organics and inorganics is estimated to be 6, whereas the volume reduction factor for primarily organic waste should be 100.

5.2 Lessons Learned for Radioactive Waste Plasma Treatment

The only existing facility for plasma treatment of low level radioactive waste on an industrial scale is the ZWILAG in Switzerland. Various difficulties occurred during construction and operation of the facility (see chapter 5.1.1). Most of the problems are due to the plant being the first of its kind. The design was completely based on previous plasma ovens for the treatment of completely different materials. However, the plant is now in full operation with increasing success [68] due to various back fitted adaptions of the requirements. Some basic design decisions cannot be taken back though, thus some basic work on plasma treatment of radioactive waste material can be useful for a review of the technology before designing a new plant. An optimized plant would have several requirements resulting from the review in chapter 4.
Any operating industrial scale plasma treatment facilities aim for a very high throughput of above 250 kg/h. With 4400 operating hours per year (50%) this could include the waste produced by 16 nuclear power plants per year with the typical production

of 68500 $\frac{kg}{a}$. The goal of this work is to show that a small-scale plasma treatment facility for the treatment of the LILW from the operations of one NPP is technically and economically feasible. In difference to existing plasma waste treatment facilities the small-scale facility has the aim of treating the waste originating from NPP operations as produced on the site of the NPP without any additional pre-treatment and sorting. To that end a design study was undertaken, accounting for all relevant subsystems.

Some functionalities of plasma treatment are possible to observe in small scale experiments. An experimental setup was built at the FH Aachen / Jülich for testing of important design characteristics and plasma specific properties related to radioactive waste treatment. The main aim is to verify the known advantages of plasma technology on a small scale, as well as to identify problems and properties that can influence the design of a full scale device.

A description of the required components and properties of a plasma treatment oven can be made. In the chapters 2.1, 3 and 4 the motivation for using plasma technology for thermal treatment of radioactive wastes was described: LILW from operations of nuclear power plants are hazardous and have to be treated before final storage. Plasma treatment technology has some advantages over conventional combustion of wastes. Basically, all material can be treated, which avoids the need for an extensive sorting, the time and costs for transport are reduced and the possibility of replacing a waste treatment with several different treatment installations is given.

5.3 New Concept for a Plasma Oven

An oven designed after the experience gained from existing facilities as presented in chapter 4.4 is described in the following. The key properties and components are identified with a reason given for the chosen technology.

5.3.1 Basic Requirements

Some initial assumptions have to be made which are based on the experiences collected so far.

5.3.1.1 Throughput

The plasma oven would be designed in a way that the waste from NPP operations from one nuclear power plant can be treated. As pointed out in chapter 2.2.3, the average volume of produced waste is $250 \pm 127 \frac{m^3}{a}$. With the typical density of $0.2 \ \frac{g}{cm^3}$ for dry solid waste and $1.2 \ \frac{g}{cm^3}$ for wet solid waste (see table 2.2) the respective content amounts to $68500 \ \frac{kg}{a}$.

To fully treat the ongoing waste production, the oven must therefore meet the throughput of $68500 \ \frac{kg}{a}$. Taking into account a 50% degree of capacity utilization ($4400 \ \frac{h}{a}$) the continuous oven throughput is $15.7 \ \frac{kg}{h}$. This is notably below the throughput of existing facilities which is above $250 \ \frac{kg}{h}$. This reduced capacity results in a much more compact design.

5.3.1.2 Composition of the Treated Material

The composition of the respective material is of major importance. 20% of the weight and 4% of the total volume would be wet solid waste and 80% of the weight and 96% of the expected volume is dry solid waste (see chapter 2.1). This can be categorized according to chemical properties which are relevant for the combustion of the material, when regarding the typical properties of the waste as indicated in figure 2.2 as shown in my data collection from NPPs etc. The dry solid waste consists of textiles, polymers, incombustibles and others. The combustibles incinerate in the oven and provide additional heat energy according to their caloric value.

Table 5.2 lists all dry solid waste material with their weight fraction and caloric value which are taken from [32].

By weighting the caloric value of each material with its weight fraction and summing up, the average caloric value can be estimated. In total, an additional energy of $5.58 \ \frac{kWh}{kg}$ is added to the process by combusting the waste material.

Material	weight %	type	Caloric value [$\frac{kWh}{kg}$]	relative caloric value [$\frac{kWh}{kg}$]
textile rag	19.29	T	4.5	0.87
textile gloves	10.72	T	4.5	0.48
filter for suspended matter	1.43	T	4.5	0.06
vacuum cleaner bags	4.57	T	4.5	0.21
foil	18.57	P	8.5	1.58
fleece rag	5.72	P	8	0.46
mixed gloves	3.57	P	5.5	0.20
rubber gloves	3.57	P	6.4	0.23
foils/hoses	2.57	P	9.3	0.24
fabric tube containing PVC	2.28	P	9.3	0.21
rubber / rubber mats	2.14	P	10.5	0.23
plastics	1.43	P	7.8	0.11
PVC	0.86	P	6.2	0.05
gaskets	0.86	P	5.8	0.05
polyethurane foam hardened	0.57	P	7.6	0.04
plastics without PVC	0.29	P	7.8	0.02
others	2.14	O	0	0
varnish, colors	2.86	O	8.1	0.23
overshoes	1.43	O	4.9	0.07
cable	0.86	O	5	0.04
plastics and metal inseparable	0.86	O	5.2	0.04
charcoal, absorbing material	0.86	O	9.3	0.08
mixed waste	0.71	O	6.2	0.04
paper	0.71	O	4.5	0.03
insulating wool	5.43	N	0	0
hoses with metallic tissue	2.28	N	0	0
grinding discs	0.86	N	0	0
empty color cans	0.86	N	0	0
fire protection materials	0.86	N	0	0
welding mats	0.57	N	0	0
glass	0.29	N	0	0
total				**5.58**

Table 5.2: *Caloric values of the waste material taken from [32]. The "type" letters indicate if the material is categorized as a textile (T), polymers (P), non-combustibles (N) or others (O). The "relative caloric value" is the weighted caloric value multiplied by the share of the material. The total is the sum of the relative caloric values.*

5.3.1.3 Radioactivity Inventory

The activity of the material is in the range of $10^9 - 10^{13}\frac{Bq}{m^3}$ in the case of wet solid wastes and in the range of $10^6 - 10^9\frac{Bq}{m^3}$ in the case of the mixed dry solid waste (see table 2.2). Expected leading radionuclides are those listed in chapter 2.4, except for ^{131}I, which alreadydecays before the treatment. The amount of each radionuclide varies strongly, mostly depending on the origin of the waste.

5.3.2 Components

A complete plasma treatment facility consists of several components: The oven itself is the reactor where the material is treated. The oven is equipped with a plasma torch. The design of the material feed to the oven and the outlet of the oven have to be discussed. Finally the off-gas treatment is described which is a vital part for the complete process.

5.3.2.1 Plasma Torch and Burner

The choice of the type of the plasma used falls on a non-transferred plasma torch due to several reasons.

The standard torch technique is in the analogy to metallurgy the transferred torch. Transferred torches are proven and tested in various fields where the main advantage of a very high peak temperature is desired (see also chapter 4.3). However, there are two major disadvantages to non-transferred torches that are relevant to this application: the treated material to needs to be electrically conducting, thus a solution is needed to start up the transferred torch, and the primary electrode wears off during operation and is therefore maintenance-intensive.

Non-transferred torches are the preferred choice for this application. In addition to the mentioned advantages it is possible to apply a process gas of choice, for example air, via the non-transferred plasma torches. When using a microwave torch powered by a magnetron (see section 4.3.4), the torch itself requires no maintenance and still fulfills the requirement of a high treatment temperature above $5000°C$, which is high enough to melt and treat all difficult material.In addition to the plasma torch the majority of energy is transferred to the oven by conventional heating (e.g. gas burners or inductive heating) and the torch does not have to serve as the main firing device. The conventional heating region in the oven can be easily controlled in terms of temperature. It is situated above the plasma torch, so that the heating via gas burners or induction in the plasma region is of minor importance.

5.3.2.2 Oven

The oven is constructed of standard firebricks. A steel hull is covered on the inside with refractory brick. The shape of the oven is more important and a result of considering the other aspects and components. At the bottom of the oven the plasma torch is placed with the intention that the treated material has to pass through the plasma focus before it can reach the outlet. Therefore the oven has a conical shape at the bottom. An addition would be the option to allow a second gas stream to enter from the bottom for stirring and for better control of the gas flow.

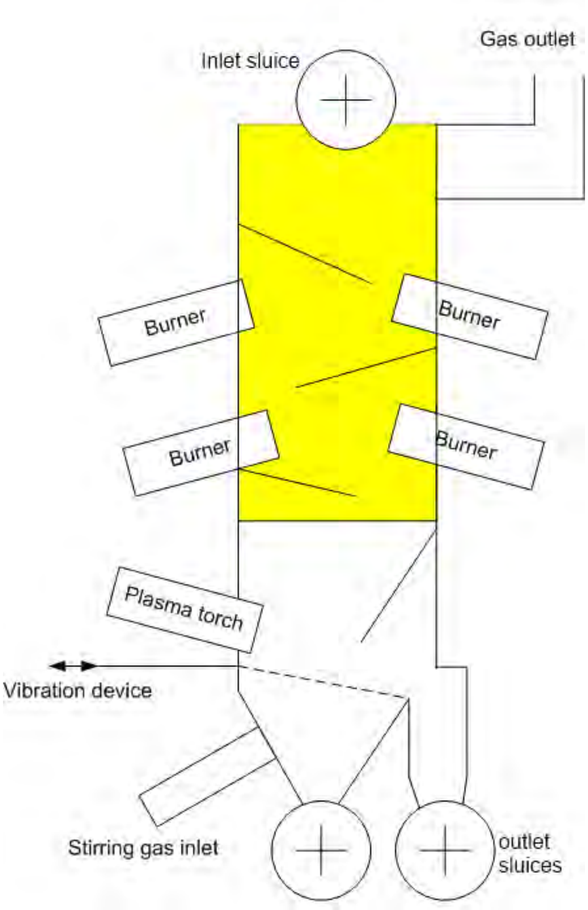

Figure 5.6: *Basic design concept of a plasma treatment device with two combustion chambers.*

Figure 5.6 shows a principal sketch of the components: The oven chamber is divided into two parts where the upper (yellow) part is conventionally heated and the bottom part is equipped with a plasma torch. The exposure time of the waste to the heat is increased with built-in components shaped in a way to decelerate the material and allow for a longer treatment time. The alternative to providing the conventional heating via several gas burners is an inductive heating of the upper oven part (yellow).

5.3.2.3 Feed

The material feed to the oven is split in two streams that are intermixed at the inlet. The wet solid wastes and the dry solid wastes are fed separately as the two streams. The activity inventory of each stream is continuously measured. The feed is controlled so that it is possible to mix the two streams. The goal is to optimize specific activity for the complete batch below the regulatory limits. The feed is continuous and the ratio of the streams is adjusted according to the collected experience on the volume reduction factor:

$$x[\frac{Bq}{m^3}] > (a \cdot A_{DSW}[\frac{Bq}{m^3}] + b \cdot A_{WSW}[\frac{Bq}{m^3}])/VRF \tag{5.1}$$

Here x is the regulatory limit on low level waste in $\frac{Bq}{m^3}$, a and b are the respective feed shares for dry solid waste and wet solid waste with $a + b = 1$, A_{DSW} and A_{WSW} are the measured specific activity contents of the two feed streams and VRF is the expected total volume reduction factor for the given material feed composition.
For cases where x is a constant in units of $\frac{Bq}{kg}$, this can be performed in the lines of this way with $VRF = 1$ in formula 5.1.

The feed is working with a continuous sluice, which is best realized by using a sealed rotary feeder, a common and well established feeding technology. By adjusting the rotation speed, the needed amount for the feed can be controlled.

5.3.2.4 Outlet

The most important improvement is that the outlet should allow for continuous treatment. The advantages of this requirement are that no material is treated for longer than necessary and that there is no need for a waiting time for starting up and cooling down, which significantly increases the efficiency of the process.
The material is collected in the oven on a temperature resistant rocking motion sieve. By the vibration, the material is transported either through the sieve or to the rim of the sieve and is then collected independently. The material that passes the sieve is defined as the fully treated waste and the material that falls over the rim is not yet treated completely and is refed into the oven. The treated material is transported out of the oven through a sluice.

5.3.3 Off-gas Treatment

Though the off-gas treatment process is state of the art and known from conventional incineration processes of LILW (see chapter 3.3.8), the design of the off-gas process is essential for a plasma treatment plant. During the plasma treatment some volatile radionuclides become gaseous and it is possible that there is a release to the environment without off-gas treatment. It has to be assured, that the resulting release is below any regulatory limits.

A sufficient design basis is the off-gas treatment of existing plasma treatment plants and incineration plants for radioactive waste treatment. Figure 5.7 pictures the off-gas treatment process of the ZWILAG plasma plant and figure 5.8 pictures the off-gas treatment of an incineration plant for combustible LLW in Belgium [24]. The key components are in both cases a secondary burner, a quencher, an acidic scrubber a HEPA filter and an additional filter (fabric filter / electrostatic filter). The ZWILAG process has an additional DE-NOX installation, due to Nitrogen being the process gas in the plasma oven. These key components are described in more detail below.

An example for an optimized off-gas treatment process for a plasma plant is given below.

5.3.3.1 Preliminary Assumptions and Parameters

In addition to the general assumptions for the oven design in 5.3.1 some more information is needed for the dimensioning of the off-gas system.

It is assumed that the off-gas consists of fly ash with a mass bulk flow below 5 $\frac{kg}{h}$ with the intended throughput of 15.7 $\frac{kg}{h}$. The fly ash is estimated to have no untypical parameters in terms of average diameter and density. The initial off-gas temperature is expected to be below 1000 $^{\circ}C$, based on referenced waste incineration facilities and plasma facilities that allow a conventional off-gas treatment (see above).

5.3.3.2 Components

Secondary burner

A secondary burner is commonly used and necessary to fully oxidize the newly formed volatile chemicals. In the chosen optimized design, the secondary burning

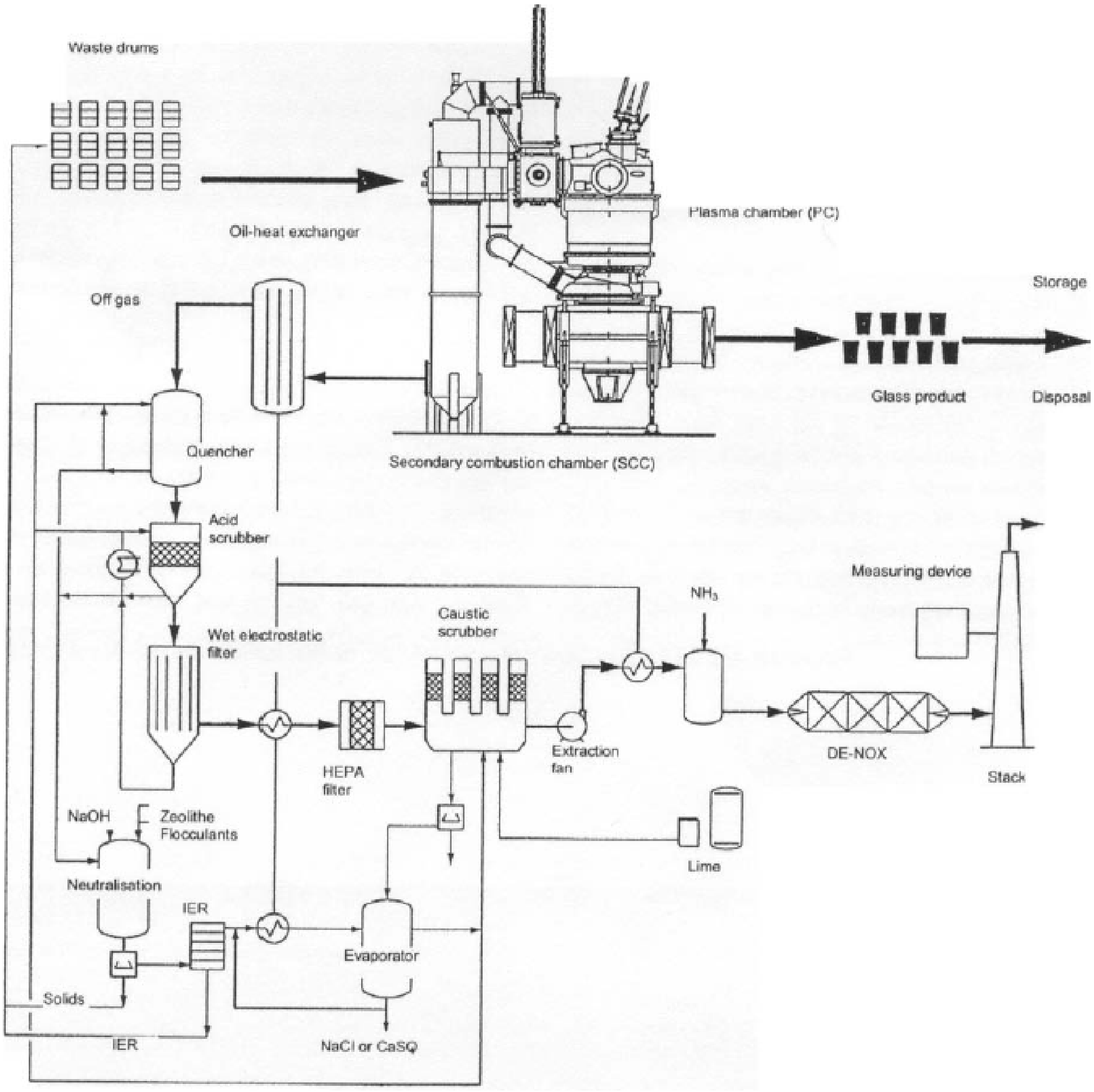

Figure 5.7: *The ZWILAG process, including in detail the off-gas treatment [24].*

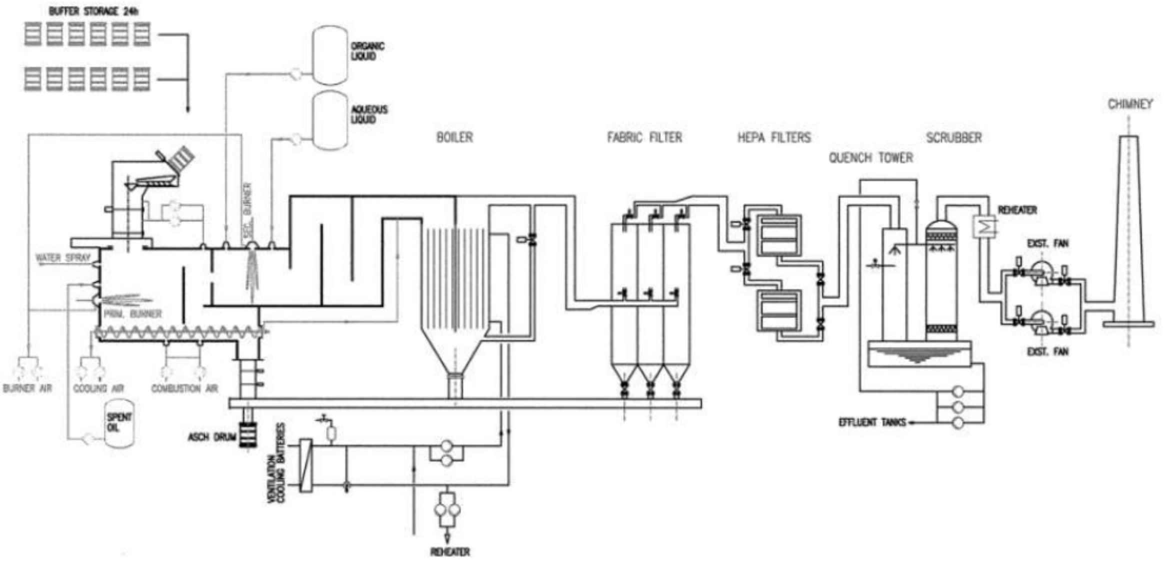

Figure 5.8: *CILVA low level waste incinerator facility, Belgoprocess, Belgium [24].*

is already performed in the plasma oven.

Cyclone

Though not a component of the referred off-gas processes shown in figures 5.7 and 5.8, a hot gas cyclone is a beneficial part of a newly designed off-gas process. Its advantages are a broad operating range regarding the temperature of up to 1000 $^\circ C$, it is a robust system and the dust particles are separated as an easily extractable stream [72].

Figure 5.9 pictures a schematic diagram of a cyclone. The incoming gas flows in tangentially, so that it is forced on a circular path. Due to the difference in density in the dust particles and the (clean) gas, the dust particles are transported downwards by gravity and are then extracted. The almost dust-free gas can exit through the dip pipe.
For the dimensioning of a cyclone according to the preliminary assumptions, it is necessary to know the off-gas volume and mass stream. The resulting dimensions for a off-gas volume of 550 m^3/h and a mass below 5 kg/h are already given in the description in 5.9. With the additional assumption of a pressure loss in the dip pipe of 2500 Pa the total extraction efficiency can be calculated as $T_{tot} = 0.969$ [25].

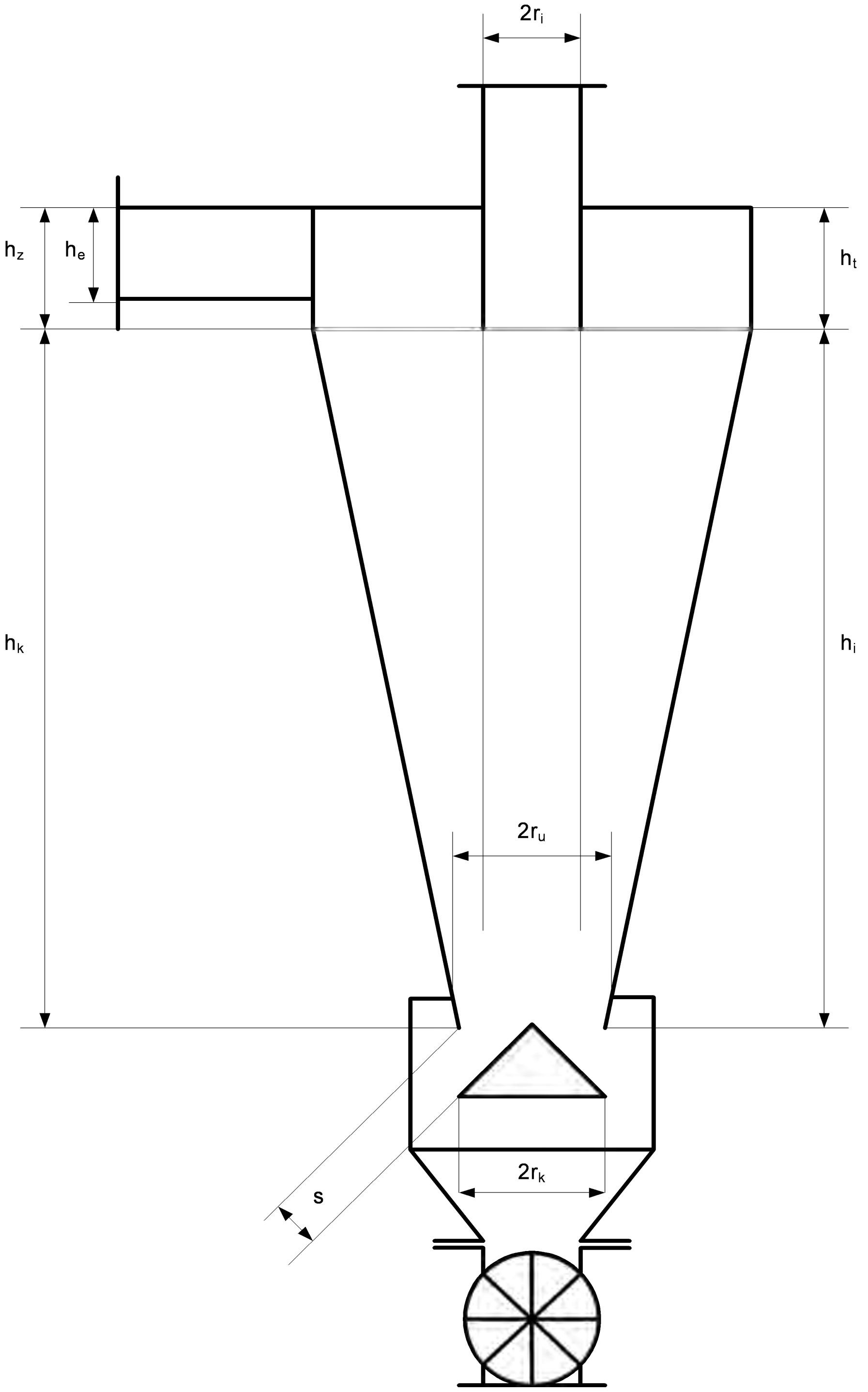

Figure 5.9: *Sketch of a cyclone with the calculated dimensions:* $r_i = 0.053m$, $r_a = 3r_i = 0.0159m$, $r_e = 2.5r_i = 0.132m$, $r_u = r_i = 0.053m$, $r_k = 1.5r_i = 0.08m$, $h_i = 16r_i = 0.849m$, $h_z = 4r_i = 0.212m$. $h_t = 3.5r_i = 0.185m$, $h_e = 3r_i = 0.159m$, $S = r_i = 0.053m$, $h = h_i + h_t = 1.036m$ *[25].*

HEPA Filter

High Efficiency Particulate Airfilters (HEPA) are fiber filters used for retention of fine dust, particulate matter and aerosols. They are in use where a very high filter efficiency is needed, for example in medical applications, as clean room filters and in nuclear technology applications.

A HEPA filter is an array of fiber layers. Figure 5.10 pictures a sketch of a HEPA Filter. The filtration is achieved by the following three mechanisms:

- Interception: the particles adhere to the fibers while in range,
- Impaction: larger particles collide with the fibers,
- Diffusion: the small particles diffuse along the air flow and intercept or impact with the fibers.

Due to not only the mechanical sieving being of importance, but also the particle adherence according to van der Waals force, smallest particles are separated as well.

An appropriate HEPA Filter for the intended application would be a HEPA H14 Filter (filter efficiency $T_{totHEPA} = 0.99995$), with a maximum load of 1200 g after which is has to be exchanged. The load is controlled with a Δp - measurement. Important for the choice are the particle size, the desired separation efficiency and the type of the dust to be separated.

One requirement of the filter is a low temperature of the medium to filter. Therefore a heat exchange is necessary in front of the HEPA filter.

Scrubber

The off-gas can contain some hazardous substances as HCl, HF and SO_x. These are absorbed in a two-step scrubber.

In the first step the off-gas is cooled down in the so-called Quench step. Mercury and halogen compounds are absorbed by contact with water droplets in the scrubber region after the quench. The water droplets are introduced as a spray. The off-gas flows through the spraying region and the water with the absorbed hazardous substances is collected [73].

The detailed reactions in the scrubber for HCl and HF are:

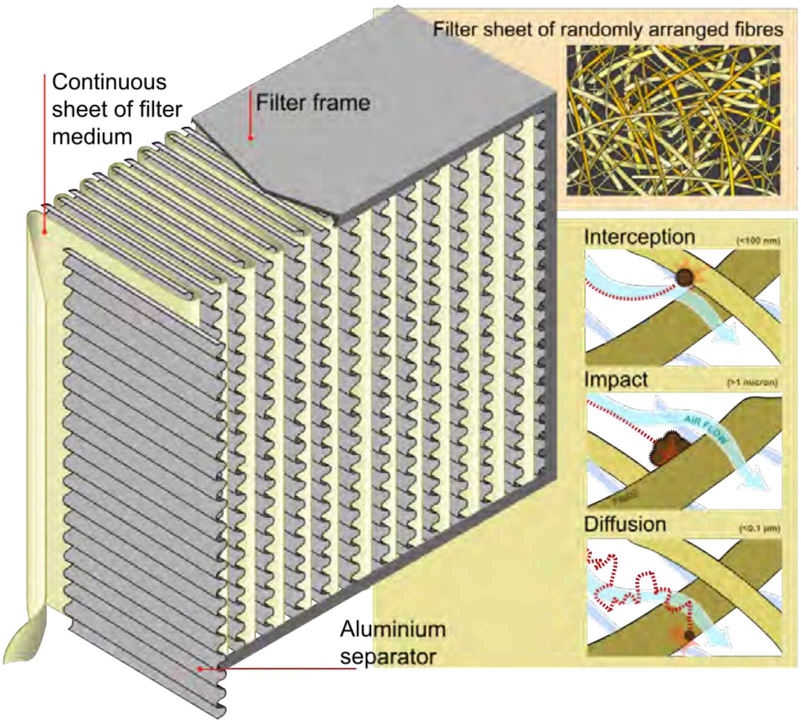

Figure 5.10: *Diagram showing the main parts of a HEPA filter [26]*

$$HF(g) + H_2O(l) \rightarrow HF(aq) + H_2O(l) \tag{5.2}$$

$$HCl(g) + H_2O(l) \rightarrow HCl(aq) + H_2O \tag{5.3}$$

An option is to add lime water to produce salts as the output:

$$2HF(aq) + Ca(OH)_2(aq) \rightarrow CaF_2(s) + 2H_2O \tag{5.4}$$

$$HCl(aq) + Ca(OH)_2(aq) \rightarrow CaCl_2(aq) + 2H_2O \tag{5.5}$$

Mercury is found in municipal waste incineration facilities at 850°C predominantly (95%) as $HgCl_2$ [73] which is easily absorbed by water:

$$HgCl_2(g) + H_2O \rightarrow HgCl_2(aq) + H_2O \tag{5.6}$$

In the second scrubber step the SO_x are absorbed. Here caustic soda solution is introduced dissolved in water as the reactant. The reactions in the second scrubber are shown for SO_2 as example:

$$SO_2(g) + H_2O \rightarrow H^+ + HSO_3^- \tag{5.7}$$

$$H^+ + HSO_3^- + \frac{1}{2}O_2 \rightarrow SO_4^- + 2H^+ \tag{5.8}$$

$$HSO_3^- + Na^+ \rightarrow NaHSO_3(aq) \tag{5.9}$$

$$SO_4^- + 2Na^+ \rightarrow Na_2SO_4(aq) \tag{5.10}$$

However it is possible, that the off-gas from the plasma oven does not contain the aforementioned hazardous substances due to a possible high-temperature destruction of hazardous substances and reforming as syngas. Usually this effect is considered in the production of syngas (see chapter 4.4.2.2). There is no information available about this issue on radioactive waste treatment though, other than that in the ZWILAG treatment plant a scrubber is being used as well. Therefore this issue has to be reviewed for an industrial scale device.

DeNOx

A device to decrease nitrogen oxides (NOx) in the off-gas, called DeNOx, can be necessary due to the high burning temperature in the oven and a possible addition of nitrogen or air as process gas. However, as described for the scrubber, it is possible that the off-gas does not contain NO_x in significant amounts, which would make an additional DeNOx device unnecessary.

The DeNOx device can be designed comparatively simple in the case of a high off-gas temperature. In the case of temperatures above 760 °C the selective non-catalytic reduction (SNCR) process can be used. This basically works by adding ammonia or urea to the off-gas, where it reacts with the NO_x to nitrogen, carbon dioxide and water.If the gas temperatures are not high enough, the selective catalytic reduction (SCR) has to be used, where an additional catalyst is needed to

enhance the reaction efficiency.
In the case of urea, the chemical reaction with NO is:

$$NH_2CONH_2 + 2NO + \frac{1}{2}O_2 \rightarrow 2N_2 + CO_2 + 2H_2O \tag{5.11}$$

In the case of ammonia it is:

$$4NH_3 + 4NO + O_2 \rightarrow 4N_2 + 6H_2O \tag{5.12}$$

Heat exchanger

Located in the off-gas chain at the beginning, a heat exchanger can be useful to both cool the off-gas stream down as well as to reuse the off-gas heat. One has to be careful though, if the heat exchanger is located before the first off-gas treatment step (the cyclone in this case), dust particles and radioactive particles can deposit in the heat exchanger and the radioactivity could accumulate in the heat exchanger. Therefore, depending on the particle load in the off-gas, a heat exchanger should be located after the cyclone. The off-gas would still be hot enough there to reuse a significant amount of heat.
The transferred heat could be used directly in the off-gas system for preheating before a DeNOx.

5.3.3.3 Process

In figure 5.11 the process for the complete off-gas treatment is shown. The mechanical components, the cyclone and the HEPA filter, are required, while the chemical components, the scrubber and the DeNOx, may be added optionally depending on the composition of the produced off-gas.

The first treatment of the off-gas from the oven and the afterburner is executed in the cyclone, where dust particles are separated with an efficiency of 96.9 % [25]. A heat exchanger is placed directly after the cyclone to avoid any contamination inside the heat exchanger. The chemical treatment in the scrubber needs already cooled off-gas. Additional cooling is performed in the quench step. The final bar-

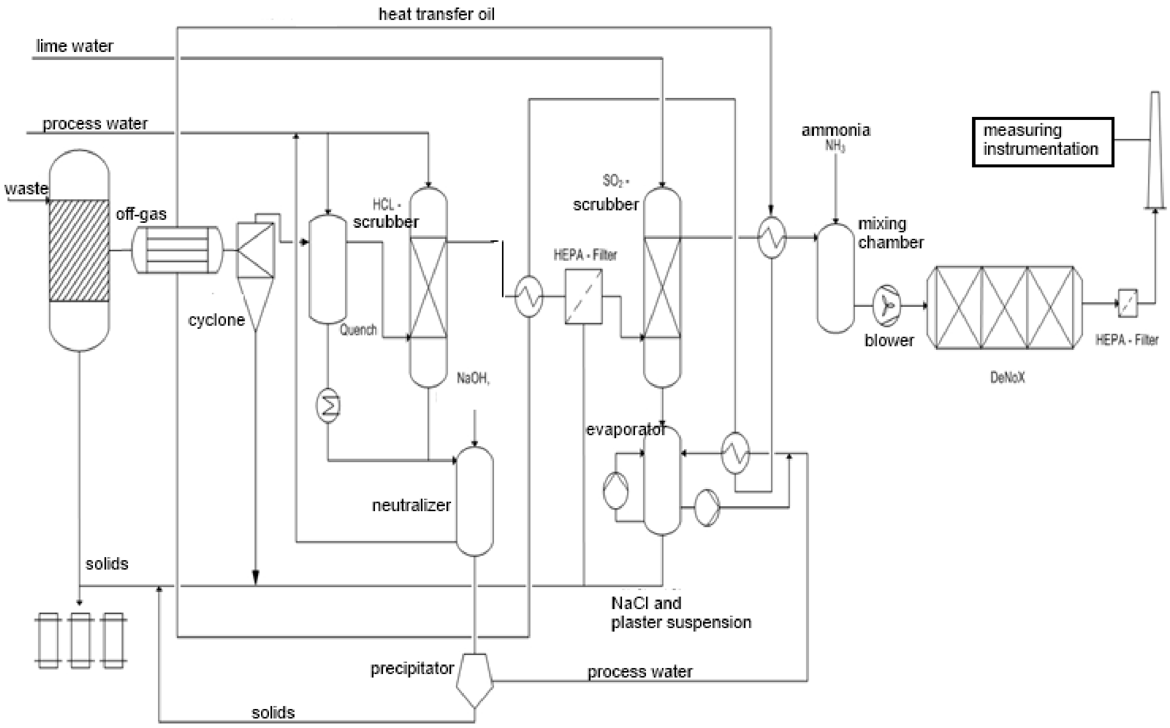

Figure 5.11: *Optimized off-gas treatment process for a plasma treatment plant with* 15.7 *kg/h throughput.*

rier after the scrubber is the HEPA filter with avery high efficiency, so that any resulting activity emissions are below regulatory limits. After the HEPA filter the gas stream is heated up again and the nitrogen oxides are removed in the DeNOx. All these components are standard technology and can be adjusted according to the requirements of the combustion process.

The oven for plasma incineration with a MW-powered plasma torch needs to be investigated in detail.

5.4 Laboratory Setup

Some functionalities of plasma treatment and the discussed components are possible to observe in laboratory scale experiments. An experimental setup was built at the FH Aachen / Jülich for testing of important design characteristics and plasma specific properties related to radioactive waste treatment.

The opportunity was taken to make use of a newly designed microwave plasma torch developed at the FH Aachen by HHFT [63] (see also 4.3.4). Major advantages over common plasma torches apply for this specific application: the torch provides a high thermal efficiency and needs no further maintenance. In addition a process gas of choice can be applied to allow e.g. combustion in air.

The laboratory setup of a plasma oven for experimental treatment of various materials was planned with the need for a simplistic and versatile model. The aim was for the setup to be easily amendable to adapt to any new experimental results. Picture 5.12 shows the general idea for a modular oven with one or more burning chambers in an assembly of fire bricks. The main modifications are an inlet for the plasma torch, one optional inlet for a bunsen burner, an off-gas tube and an inlet for the specimen.

The setup of the experiment was varied in the beginning to find a suitable setup with a minimum complexity and to allow for a representative and reproducible environment for the experiments.

For the oven part fire bricks were used, into which holes were drilled which form the combustion chamber. The burning chamber is therefore surrounded by fireproof material. Preliminary tests revealed, that the thermal power output of the plasma torch is low enough to allow metal parts in the oven which do not melt. As an

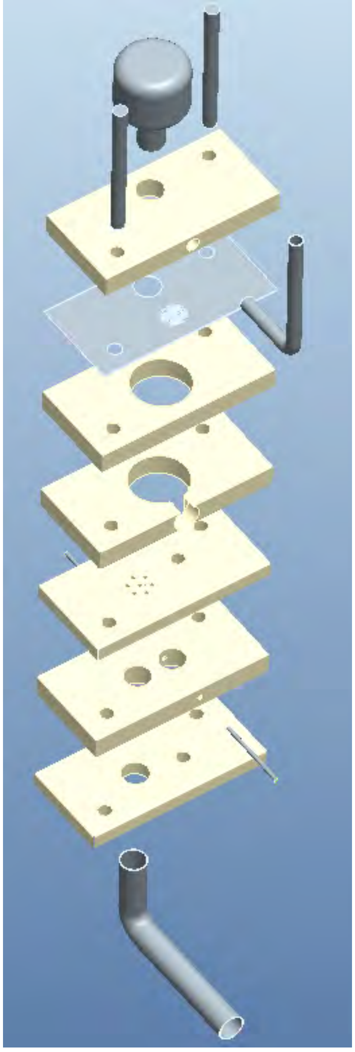

Figure 5.12: *First design of a modular laboratory device with several combustion chambers. The fire bricks can be assembled in various ways for different experiments.*

oven part a cylindrical piece of a steel tube was used and two bore holes were added. Figure 5.13 shows a photograph of the oven piece. Shaped aluminum foil could be used as a bed to hold the specimen.

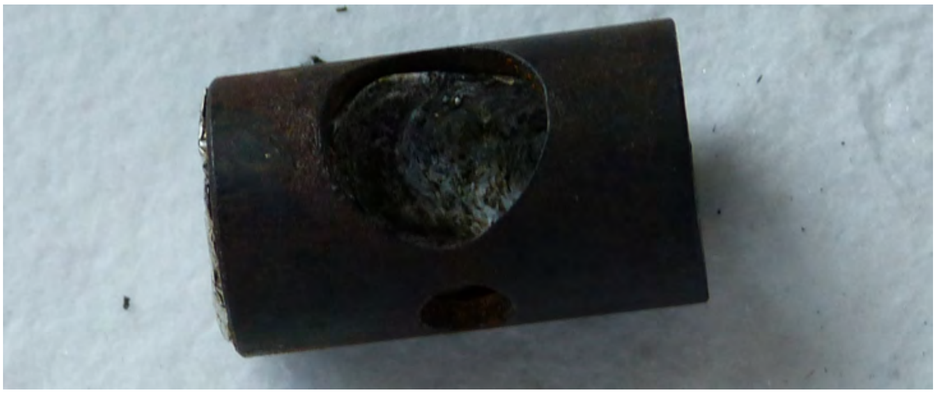

Figure 5.13: *The used steel oven piece with an aluminum bed for the specimen*

5.4.1 Plasma Torch

A newly developed microwave plasma torch and high frequency generator by HHFT was used. The torch is driven by a 2.45 GHz microwave signal. The input power can be regulated up to 200 W. The torch is built up with a hollow copper cannula as the electrode. The length of the copper electrode is $\lambda/2$, with $\lambda = 12.23\ cm$ being the wavelength of the signal. The plasma gas flows through the cannula and is ignited at the tip where it gets ionized by the high electric field power. For the torch used the ignition frequency is 2.40 GHz and the operating frequency is 2.44 GHz as found by calibration. Typically a ratio between the input and output energy of 97.5% or $-6dB$ is reached, which can be observed on a display. The plasma gas flow has to exceed 0.2 l/min. Welding protective gas can be used, which is Argon or an Argon 98% / Hydrogen 2% mixture. In these experiments the mixture called "Hydrostar" by the supplier Praxair was used. A second gas is used for cooling the cannula and focusing the plasma flame. For this gas pressurized air was used with a high flow above 2 l/min.

Figure 5.14 shows a photograph of the torch in operation. The Hydrostar supply flows through the front hose and the air through the back hose.

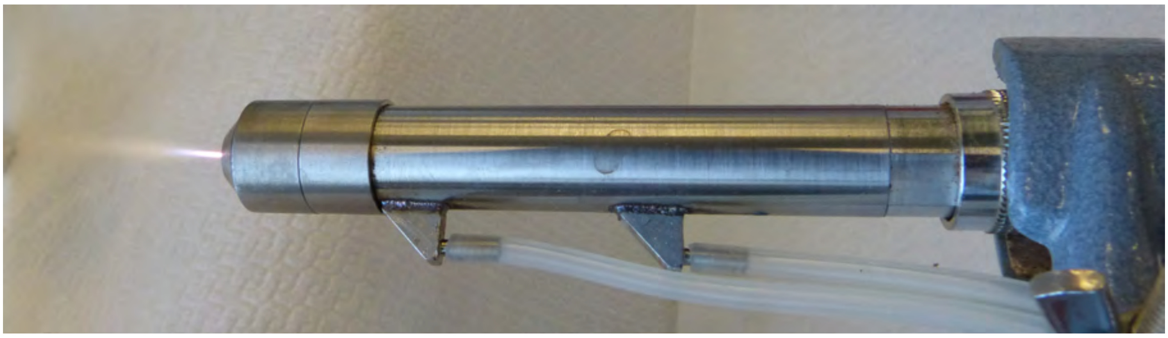

Figure 5.14: *Photograph of the used 200 W plasma torch in operation*

5.4.2 Experiments

Several experiments were performed with the laboratory device. Figure 5.15 pictures the oven as used in all experiments, except for the qualitative comparison of gas incineration and plasma incineration, for which an assembly of two burning chambers for the plasma and the gas burner was used. On the upper oven outlet an off-gas quartz tube was installed and tightened with aluminum foil. An exhaust suction was placed directly above the quartz tube. For a tight fitting of the pieces, a layer of high-temperature insulation wool was used in between the fire bricks. In the diagonal corners of the fire bricks, holes were drilled and a threaded rod was put inside to allow for tightening with screw nuts.

5.4.2.1 Short Time Plasma Treatment of Various Materials

Motivation

In this experiment various materials with a broad range of properties were treated in the plasma. Of interest was the effect of the plasma on the material. The aim was to examine the correctness of the claim that generally all material is treatable and observable, especially on this low scale. In addition, it should be investigated what effect the size of the specimen has on the treatment time and result. The assumption, that the required treatment time and energy decreases with higher fragmentation of the specimen should be confirmed. In additional experiments, glass particles were employed with the aim to find out if any vitrification effects occur.

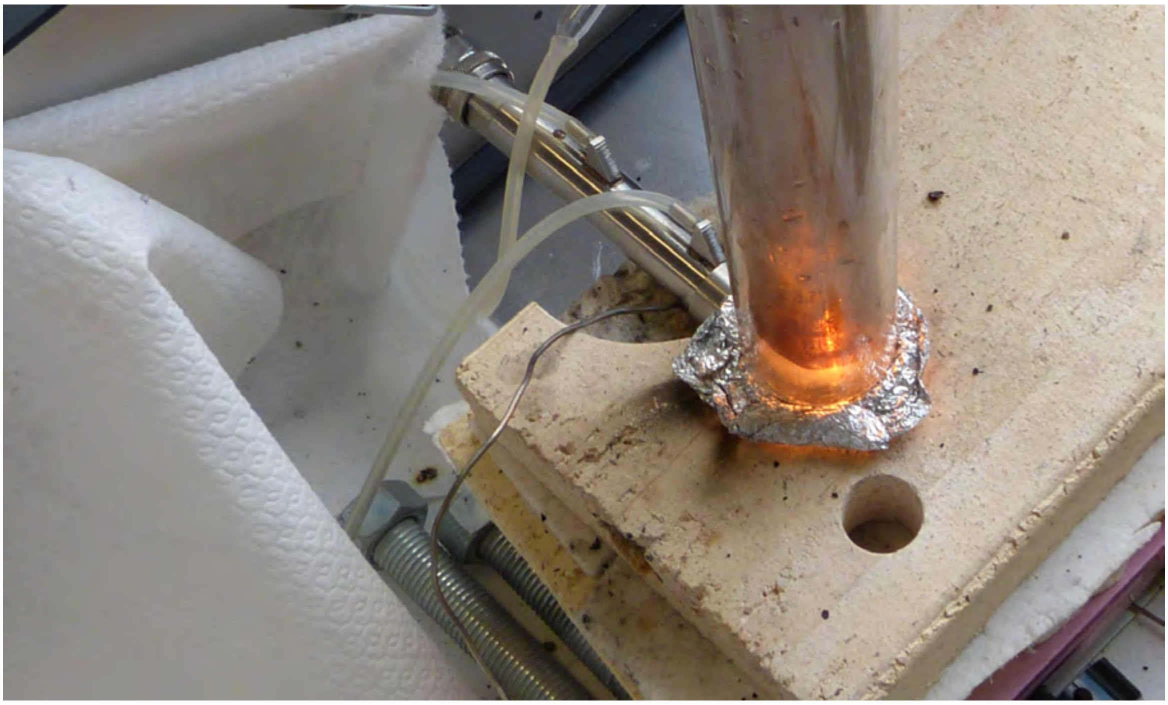

Figure 5.15: *The assembled oven in operation*

Experimental setup

The plasma oven and torch are set up as described above. The specimen are placed on a filter paper by Macherey Nagel - "Schwarzband", which guarantees for ash-free combustion. The filter paper is then folded to hold the specimen and then introduced in the oven. The time for full treatment is measured manually with a stop watch. The temperature at the tip of the plasma torch is measured with a temperature probe which is fixated in a position near the tip to prevent overheating. The "Hydrostar" - gas flow is set to $0.2 l/min$. The sample can be observed during treatment directly through an observation side hole in the burning chamber, which can be plugged for better thermal isolation.

Execution of the test and analysis

Table 5.3 lists all tested materials together with their experimentation parameters. In general it can be noted that combustible material combusts quickly with small to no residue. Non-combustibles all show an effect though the input power is too low to completely melt down any larger specimen, especially metal pieces. In principle all materials show an effect when treated with plasma.

Material	Mass [g]	Time [s]	Observations and remarks
Activated charcoal	0.2	120	Completely vanishes, Melting was observable, see fig. 5.16
Paper	0.2	60	No residue
Quartz wool	0.15	60	Volume reduction
Tin plates	0.4	300	No effect
Latex gloves	0.15	60	Glued to oven walls, a lot of smoke production, no residue
Glass particles	0.7	30	Melts together to a single droplet
Brazen chipping	0.3	300	Melts together
PE	0.2	90	Ash, smoke
Textiles	0.2	60	High flame, no residue
Cable	0.3	60	Isolation burned, metal melt where flame touched
Wood	0.3	120	Burns down, orange flame
Mixture	0.5	300	Burns down
Mixture + Glass	0.7	600	Combustibles partially melt together in glass droplet
Brazen chipping + Glass	0.75	460	Melt together to brown, grainy particle
Aluminum chipping	0.33	20	partially molten
Iron chipping	0.25	50	Sparks, partially molten
Stainless steel chipping	0.67	80	Sparks, partially molten
PVC	0.15	120	Lots of dark smoke, residue on walls
Acrylic glass	0.16	10	Melts away
Rock salt	0.2	30	Breaking of structure, melting
Tungsten wire	-	40	Exp. in air, flame on a spot, wire melts down on that spot
Ceramic tile	-	30	Exp. in air, where the flame touches the ceramic melts

Table 5.3: *List of experiments with short time plasma treatment*

Activated charcoal was expected to burn away. In addition some bubble forming on the surface of the charcoal could be observed (5.16). Besides of evaporation of volatile species formed in the production of charcoal, the peak temperature in the plasma may have been so high, that not only (chemical) burning but physical melting and gasification occurred (melting point: 3550 °C, gasification point: 4027 °C [74]).

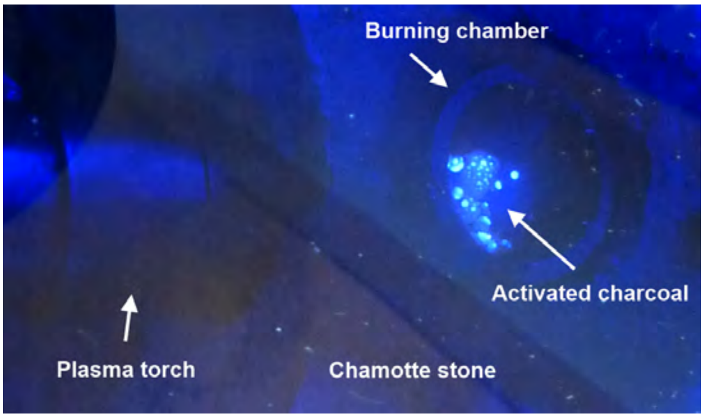

Figure 5.16: *Photograph during burning of charcoal as viewed through a colored cobalt glass.*

Quartz wool showed a significant volume reduction. This is being further examined in the following experiment.

Some materials that are incombustible, but are sorted to the non-combustible waste in 2.2 like PE, PVC and acrylic glass have been treated with plasma and burn away. Under unfavorable conditions these materials may form toxic compounds in the flue gas. Hence, detailed investigations and analysis of the flue gas should be performed to establish conditions avoiding their formation.
The various metal chippings showed an effect where the plasma flame touched the material: a spherical melting point was visible at these spots. Some particles were also sintered.

Figure 5.17: *Treated mixture of glass particles and combustible waste. On the top of the glass droplet some dirt spots can be seen.*

The so called "Mixture" in table 5.3 is a typical scaled down composition of combustible material from nuclear power plant operations as described in chapter 2.2.2. In detail the mixture used in this experiment consisted of 0.1g textiles, 0.17g PVC and 0.08g latex glove (polymers), 0.12g charcoal (other). In a subsequent experiment 0.23g glass (incombustibles) was added. The mixture was easily treatable and in the experiment with glass some dirt spots could be found on the molten glass droplet, which indicate a partial vitrification of the combustibles (see fig. 5.17).

When treating a mixture (50:50) of brass grains and glass particles, it could be observed that a large grainy, brown droplet had formed (fig. 5.18) in addition to some residual brass grains.
Tungsten wire was examined to see if the effective temperature is high enough to melt even high temperature resistant material (melting point: 3422 °C [75]). When treating a wire fixated in front of the plasma torch, it broke after about 40 *s* of treatment. A very bright glare was seen through a cobalt glass to protect the eyes. During the treatment of ceramic tiles, only a local effect around the plasma focus was observed: the surface melts with an appearance similar to molten glass.
Some materials were chosen for further experimental series.

The experiments listed in table 5.4 were chosen because of a potential interesting behavior when treated with plasma. It could be seen that the brazen chipping melts

Material	Mass before[g]	Mass after[g]	Time [s]	Remarks
Brazen chipping	0.2	0.2	20	No visible changes
Brazen chipping	0.2	0.16	30	Green flame, loosely molten
Brazen chipping	0.2	0.18	40	1 larger molten piece
Brazen chipping	0.2	0.18	50	1 larger molten piece
Glass particles	0.2	0.11	10	Molten where touched
Glass particles	0.2	0.15	20	Molten in several beads
Glass particles	0.2	0.15	40	Several pieces, one large piece
Glass particles	0.2	0.19	50	One large piece
Glass particles[1]	0.2	0.15	40	Melt together to one piece
Glass particles[1]	0.2	0.14	40	One larger piece, few small pieces
Glass particles[1]	0.2	0.15	40	Two larger molten pieces, rest as input
Glass particles[1]	0.2	0.17	40	Only partially melt where flame touched
Rock salt	0.2	0.2	10	Nothing to see
Rock salt	0.2	0.17	20	Molten where touched by flame
Rock salt	0.2	0.16	30	1 piece, grainy
Rock salt	0.2	0.15	50	Broken in many pieces

Table 5.4: *Treatment series of some chosen materials. [1]: increasing grain size in this series from many tiny parts to few larger fragments.*

together with increasing time. Some sparks were visible in the experiments, which probably account for the missing mass.

Figure 5.18: *Treated mixture of glass particles and brazen chipping.*

Glass particles were the most interesting material, therefore two series of experiments were performed with it. In the first series, again the effect of the treatment time was examined. The longer the glass is in the plasma, the larger the output particles grow. In figure 5.19 the input and output of this series can be seen. In the second glass series, the effect of the grain size of the glass particles was examined. For this some glass was crushed and the residuals were sorted by size, which were then taken as specimen. The last experiment in this series was only one larger glass piece of 0.2 g. It could be observed that the melting proceeds faster when the particle size is smaller.

Rock salt is no typical waste material. The intention was to investigate the plasma effect on the salt, related to possible actual final repository problems, where ra-

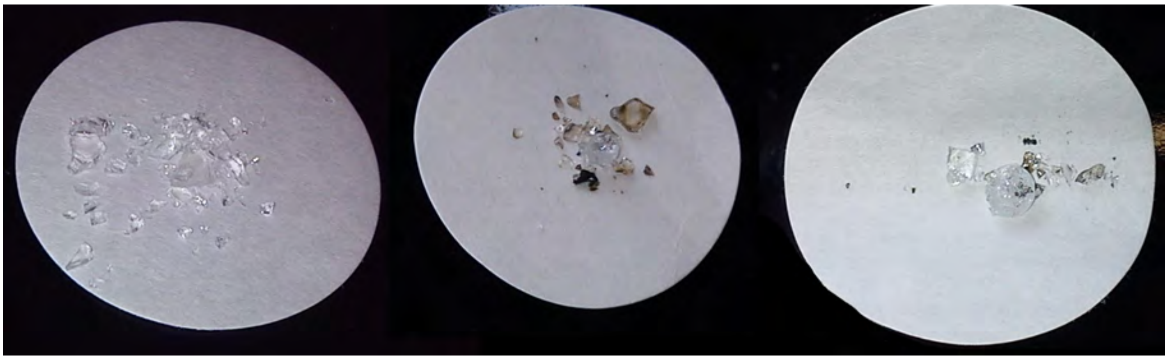

Figure 5.19: *The sample on the left pictures the untreated sample. The second is the glass treated for 20 seconds and the third picture is the glass treated for 50 seconds. For comparison, the diameter of the filter paper is 30 mm.*

dioactive waste is stored in rock salt formations. It became clear that in principle this material is treatable, though with different effects depending on the particle size and treatment time. Some parts broke under the influence of the plasma and some parts melted. No clear trend was observable.

Results

All materials that are contained in typical waste streams can be treated by plasma with visible effects. Especially all materials constituting waste from nuclear power plant operations are treatable. In general the combustible materials burn and the non-combustible materials melt. A partial vitrification was observed when mixing material with glass particles. An effect is visible after a few seconds of treatment and the longer the treatment, the more distinct is the effect. Metals are also treatable, though in the experiments the effect was not significant due to the low scale power of the plasma torch. It was observed that melting is easier to achieve with small particle sizes in the samples. The most intense effect or the starting point of the combustion was always in the focus of the plasma. Therefore to achieve a complete treatment, the plasma flame has to touch all surfaces for a short time. This, too, is more easily achieved with small grains as the used material to increase the surface.

When compared with the typical waste distribution from nuclear power plant operations as laid out in section 2.2.3, all occurring material can be treated. An effect of volume reduction by burning or melting is observable with most combustible and non-combustible mixed waste. Metals are treatable too, though no volume reduction could be observed. The treatable material excluding the metals and concrete/debris accounts for 96% of the total produced volume and 80% of the total mass.

5.4.2.2 Qualitative Comparison of Gas Incineration and Plasma Incineration

Motivation

When applying plasma treatment on LILW, it must be considered that the performance should be better as compared with conventional combustion. A treatment effect should be visible on non-combustible material. Combustible material should incinerate and combust quickly. In addition to testing various materials in experiment 1, here a comparison between the effect of the plasma torch and the conventional burning is made for selected materials. Finally an experiment is run with a combination of both. The aim is to find out if the combination of the treatment method leads to an enhanced burning, namely if the bulk of the energy can be introduced via conventional burning while the peak temperature is created with the plasma for difficult material.

Experimental setup

Firstly, the oven is assembled with the plasma burning chamber and the off-gas tube on top. The burning chamber is replaced with another one which has an inlet for a bunsen burner for the conventional combustion experiments.

For the combined experiment, the oven is assembled with two burning chambers: one is the standard plasma burning chamber and on top of that the conventional burning chamber is placed. The off-gas tube is placed on top of the outlet of the conventional burning chamber with an additional fire brick in between.

Execution of the test

Various waste materials have been examined. Remarkable effects could be observed with the following materials: Glass, quartz wool, brazen chipping and rock salt.

In general it was observed when treating combustible material that there is no difference in outcome between plasma incineration and gas induced combustion, though the time needed until incineration and burning down was much lower in plasma incineration. Combustibles caught fire in an instant when touched by the plasma flame, whereas this material caught fire after some seconds in gas induced combustion when in touch with the flame. Also the time needed for a complete burning of the material was similarly enhanced in plasma treatment. However, only qualitative observations could be made with no clear time measurements, since the burning chamber for the gas burner does not allow for a visual observation of the experiments and therefore no clear treatment time could be measured. It could be seen when treating material in air without an oven, that any effect, e.g. melting of glass or combustion, occurred in plasma treatment directly when the material was in the plasma focus whereas a significant time in the range of 20 seconds to a few minutes passed until the effects could be observed with a gas burner. The combination of a plasma and a gas burner worked. It was observable, that the off-gas - tube was less smoke filled in experiments where smoke producing material like PVC were inserted. The "afterburner" already combusted some of those particulate residuals. In addition, most thermal effects (except melting of metals) were achieved faster.

Analysis

With regard to the motivation, the most remarkable effect could be seen when treating quartz wool. Whereas with conventional burning no visible effect could be observed, the plasma flame burned the wool down. Where the plasma flame touched the material it shrank down and its volume was highly reduced.
Glass melts down in the plasma stream in a few seconds. An orange flame was visible. Due to the plasma being very sharp, holes were burned in glass tiles. In comparison it took the conventional gas burner a minute to partially melt a glass tile and until the flame was colored orange.
Brazen chipping, when placed in the plasma focus, melts together. No effect at all was visible when placed in a gas flame since the typical temperature in gas flames is below 800 °C.
Rock salt melts locally on the spot of interaction with the plasma and forms droplets. Again, no effect could be seen when treated with the gas burner.

Results

Compared to conventional gas burners, plasma burners have the advantage of a very high peak temperature. This is the main cause for a number of additional thermal effects, such as melting of heat resistant material and material with high melting points. The combined treating via plasma and conventional burning is possible with the aim of reducing the treatment time. This application has the positive side-effect of partial off-gas cleaning.

5.4.2.3 Caesium Distribution in Plasma Oven

Motivation

Caesium-137 is a radionuclide with a half-life of 30.1 years volatilizing at 671°C (see section 2.4.2). ^{137}Cs is a leading nuclide for all radioactive waste material. Due to its low gasification point, it is a special threat in heat treatment. The aim of this experiment is to find out the effect of the plasma on Caesium and analyze the distribution of the Caesium in the oven after treatment. By analyzing the amount left in the experimental device and comparison with the input, the amount that gets lost in the off-gas system can be estimated. In addition the amount after treatment in the burning chamber and in the off-gas tube is compared to see what part is deposited locally and what part travels away from the burning chamber.

Experimental setup

The plasma device is set up as usual, with the burning chamber inside a fire brick, the off-gas tube on top and the observation hole plugged. The focus of the plasma flame is in the center of the burning chamber, where a preferential holding spot for the specimen was created in the aluminum foil bed. The treatment time and the temperature at the tip of the plasma burner were controlled to prevent any damage to the equipment. The quantitative analysis is undertaken with a total reflection X-ray fluorescence analysis (TXRF) Bruker Picofox S2.

Execution of the test

In every experiment, the plasma burner was set-up with an input power of 180 W at an operating frequency of 2.44 GHz. The air flow was set to a high level (maximum throughput) and the "Hydrostar" - flow was set to 0.2 l/min. For the specimen, 10

μl of a solution of dissolved CsCl were taken and put on a filter paper.
The specimen was put in the plasma and treated for about a minute. Afterwards, the off-gas tube was removed and the burning chamber was taken out and put in a beaker with 20-50 ml distilled water, to dissolve the remaining Caesium. The off-gas tube was cleaned with distilled water collected in a beaker. Both solutions were then analyzed with the TXRF. For the quantitative analysis a Bismuth solution and a Gallium solution were available as the internal standard. The concentration in the original solution was 1.266 $g/(100\ ml)$ CsCl to achieve a Caesium concentration in the solution of 10 g/l Cs.

Analysis

The results from the experiments are listed in table 5.5.
In the first experiment, 38.9 μg Caesium were on the specimen and 36.9 μg + 2.5 μg = 39.4 μg were found in the solutions afterwards. The amount which was found in the analysis is therefore higher than the amount that was originally put in. Qualitatively, 6% of the Cs were found in the Off-Gas tube and 94% in the oven piece. In this experiment the Caesium did not travel far. Possibly the oven temperature was low enough to allow for a deposition of Caesium on the walls away from the plasma focus. The increased amount compared to the input can be a result of remnants of the preliminary tests which could be stuck in the oven piece. Additionally, a high number of other elements could be identified in the analysis spectrum which can also adulterate the quantification. The other elements were expected to be in the sample, because in this first experiment the distilled water could not be used, but instead the remnants in the oven piece were solved in tap water.
In the second experiment, 75.4 μg Caesium were on the specimen and 50.0 μg + 23.1 μg = 73.1 μg were found in the solutions afterwards. 97% of the put in sample were found again in the solutions. This time, 32 % were found in the Off-gas tube and 68 % in the oven piece. Almost all Caesium was found again with a realistic distribution in the same order of magnitude in off-gas and oven. Only a small fraction was not recovered, which possibly became volatile and was lost through the exhaust.
Figure 5.20 pictures the TXRF spectrum taken from the original source in this experiment with a Bismuth - standard. As one can see, both the Bi and Cs peaks are dominant peaks and the fitted line for the internal analysis is in accordance with the measurements, which is a sign for a good quantification.

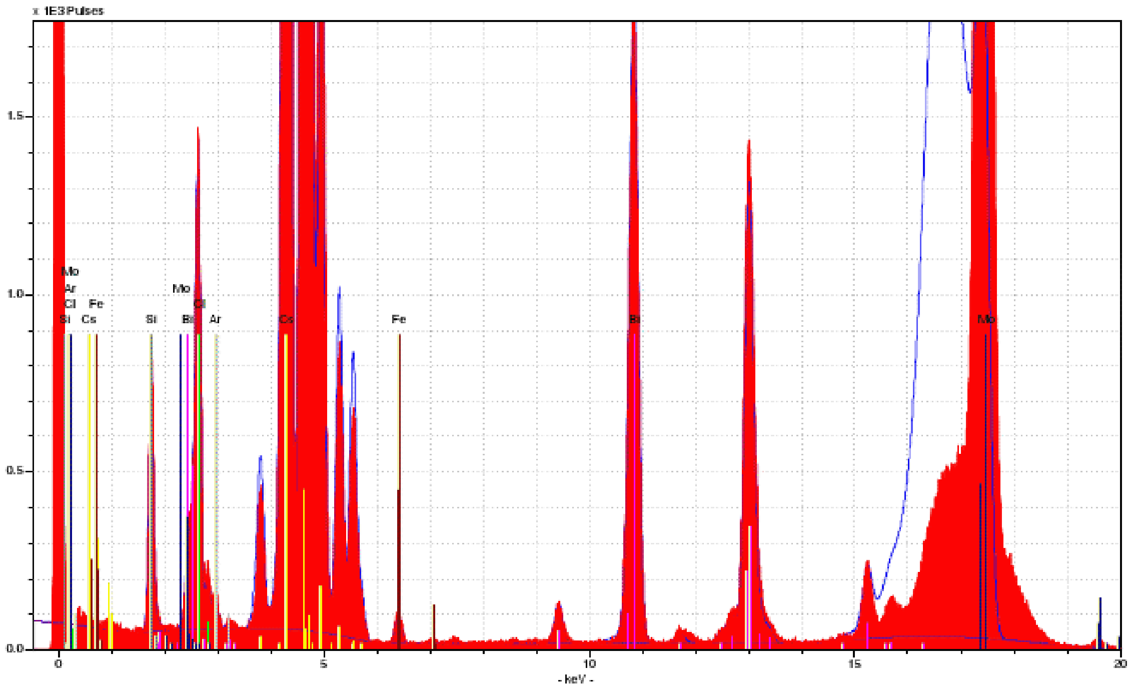

Figure 5.20: *TXRF spectrum of the original source in experiment 2:* 1.266g *CsCl in* 100ml *water +* 10μl *Bi (*10g/l*)*

In the third experiment, 80.5 μg Caesium were on the specimen and 47.4 μg + 31.3 μg = 78.7 μg were found in the solutions afterwards. This can be easily compared to the second experiment, where the same internal standard and the same sample solution were used. The measured distribution is comparable (60% / 40% oven / off-gas compared with 68% / 32%) and the quantitative results are in the same region. 98 % of the Caesium were recovered. The missing amount can be due to leakages or gone in the exhaust.

In the fourth experiment, 43.9 μg Caesium were on the specimen and 74.1 μg + 48.3 μg = 122.4 μg were found in the solutions afterwards. These measurements are highly off and therefore the experiment is regarded as not successful. The qualitative distribution is again (as in 2 and 3) 61% to 39% oven to off-gas.

In the fifth experiment, 46.0 μg Caesium were on the specimen and 18.0 μg + 35.2 μg = 53.2 μg were found in the solutions afterwards. Qualitatively the distribution is swapped (34% oven / 66% off-gas). This alone is not a sign for a failed experiment, since it is probable that the distribution depends on the local temperature in the oven piece and the plasma effect on the sample, which is not necessarily constant due to different positioning of the sample. The collected Caesium is again more than the input, probably due to the same reasons as in experiment 1.

Number	Solution	Standard	Cs-concentration	Cs-amount
1	Origin	20 μl Ga ($1g/l$)	$3.89 \pm 0.09 g/l$	38.8 μg
1	Oven	20 μl Ga ($10mg/l$)	$0.74 \pm 0.09 mg/l$	36.9 μg
1	Off-gas	20 μl Ga ($10mg/l$)	$0.06 \pm 0.02 mg/l$	2.5 μg
2	Origin	10 μl Bi ($10g/l$)	$7.54 \pm 0.2 g/l$	75.4 μg
2	Oven	20 μl Bi ($10mg/l$)	$2.45 \pm 0.05 mg/l$	50.0 μg
2	Off-gas	20 μl Bi ($10mg/l$)	$0.44 \pm 0.02 mg/l$	23.1 μg
3	Origin	10 μl Bi ($10g/l$)	$8.05 \pm 0.02 g/l$	80.5 μg
3	Oven	20 μl Bi ($10mg/l$)	$0.95 \pm 0.03 mg/l$	47.4 μg
3	Off-gas	20 μl Bi ($10mg/l$)	$0.78 \pm 0.02 mg/l$	31.3 μg
4	Origin	20 μl Ga ($1g/l$)	$4.39 \pm 0.10 g/l$	43.9 μg
4	Oven	20 μl Ga ($10mg/l$)	$1.48 \pm 0.04 mg/l$	74.1 μg
4	Off-gas	20 μl Ga ($10mg/l$)	$0.97 \pm 0.03 mg/l$	48.3 μg
5	Origin	20 μl Ga ($1g/l$)	$4.60 \pm 0.13 g/l$	46.0 μg
5	Oven	20 μl Ga ($10mg/l$)	$0.90 \pm 0.07 mg/l$	18.0 μg
5	Off-gas	20 μl Ga ($10mg/l$)	$1.60 \pm 0.05 mg/l$	35.2 μg

Table 5.5: *Analysis results*

All experiments taken together it is remarkable that, with the exception of experiment 4, all showed a Caesium recovery of about 100 %. The two experiments measured with the Bismuth standard showed similar results, whereas the experiments measured with the Gallium standard all showed a Caesium recovery of above 100 %. This indicates a systematic error in the measurements with the Gallium standard.

Results

The purpose of this experiment was to find out the part of Caesium that becomes volatile and does not deposit locally in plasma incineration. It was found that at least 97% of the Caesium was found again and therefore only 3% or less are lost in the off-gas or through leakages. The most important reason for that is, that Caesium is known for depositing on a relatively cold surface. This behavior is therefore confirmed and reproduced for high temperature plasma combustion. In a setup with a high temperature in a larger volume, such as an industrial oven, one has to account for a foreseen spot to collect the volatile Caesium, as the first cold surface in the off-gas stream.

About two thirds of the total Caesium is deposited locally in the burning chamber and one third travels with the off-gas stream in the region of the off-gas tube. These numbers are not very reliable though, since only experiments 2 and 3 showed these results and experiment 4, which verifies these, was in quantification far off of the expectations and has to be regarded with care. This distribution however might be more a characteristic of this specific plasma setup and not of plasma incineration in general.

5.4.2.4 Time Dependence of Melting a Metal Wire

Motivation

This experiment is performed to roughly characterize the specific amount of energy needed to treat a sample until it is completely molten. It is known, that a large part of the applied energy is not used for heat treatment of the material, but also heating up the local environment such as the oven material. in addition, also the target material loses heat by heat transfer and heat radiation. In this experiment it is intended to find out what amount of applied energy is sufficient for a full plasma treatment of the sample material. It should be measured if there is any volume or weight reduction after the treatment.

Preliminary tests

This experiment is a series of treatment of one specific material, to achieve the necessary statistical certainty on the measured parameters. First, several materials e.g. glass, wire and rock salt were tried and it was visually observed which would work as a reproducible testing material. The choice was a piece of metal, due to it being incombustible and the final treatment point being clearly observable, when all material is molten. To have an easy and reproducible testing scenario, iron paper clips were chosen as the specimen.

Additionally, it was necessary to find out a way of treating the paper clip in the plasma so that it would melt down without much loss of energy. It was decided on manually shoving clip through the plasma.

Experimental setup

Other than in the other experiments, no oven was used here. The plasma torch was burning in free air. The sample was held with tongs and placed directly in the plasma focus. The whole experiment was recorded with a camera, to analyze the needed amount of time with the recorded movies. The plasma parameters were as before 180 W input power and 2.44 GHz operating frequency. The weight and the volume of the paper clips was taken. The molten droplets from treatment were collected with a ceramic cup.

Execution of the test

Figure 5.21 pictures a capture of the running experiment. The paper clips melt down to several droplets which were collected in the ceramic cup. Many sparks emerged during treatment. As one can see, the paper clip is held directly in the plasma focus and continuously pushed forward when it is visually observed that the treatment point is already molten. A treatment series of four paper clips was performed.

As can be seen in figure 5.22 a problem occurred in experiment three: One droplet was hot enough to break the ceramic cup due to an extreme heat difference. All droplets could be recovered though, so there is no effect on the outcome of the experiments.

Analysis

Four experiments were performed. The measured parameters are listed in table 5.6. The volume could be measured for all 4 clips together as 0.4 ml. After the experiments the total volume of all droplets also amounts to 0.4 ml.

The specific energy is calculated as seen in eq. 5.13:

$$\frac{E}{m}[\frac{J}{g}] = P_{plasma}[W] \cdot t[s]/m[g] \tag{5.13}$$

Where P_{plasma} is the plasma energy (180 W), t is the treatment time and m the mass of the paper clip. The result can be compared to the theoretical minimal energy

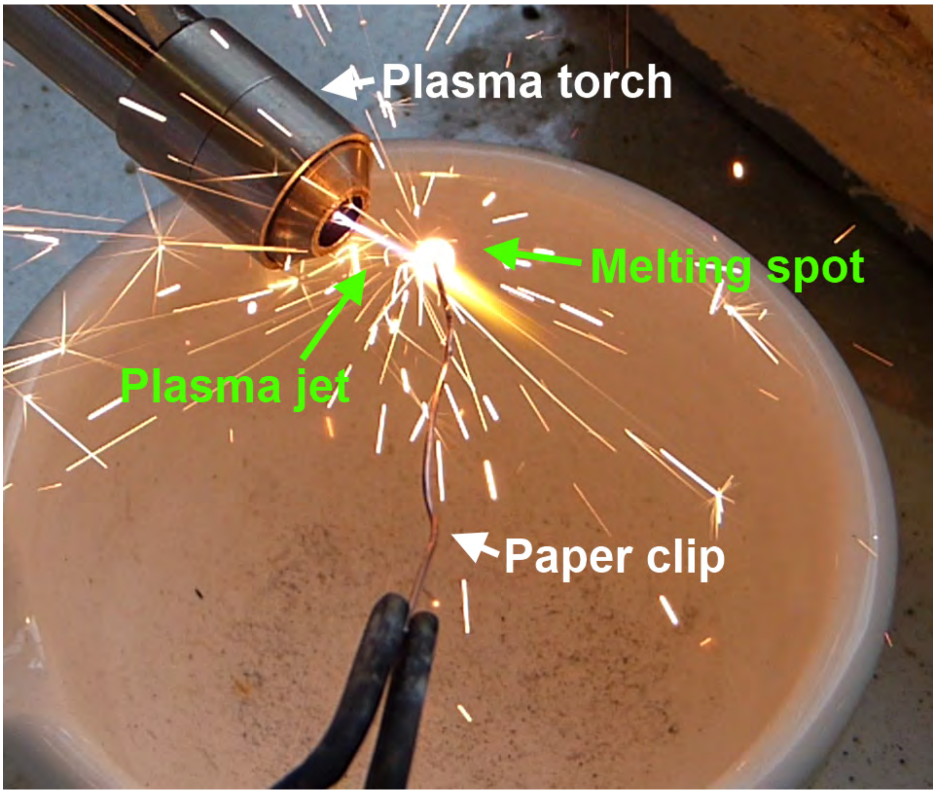

Figure 5.21: *Photograph of the running experiment of melting a paper clip*

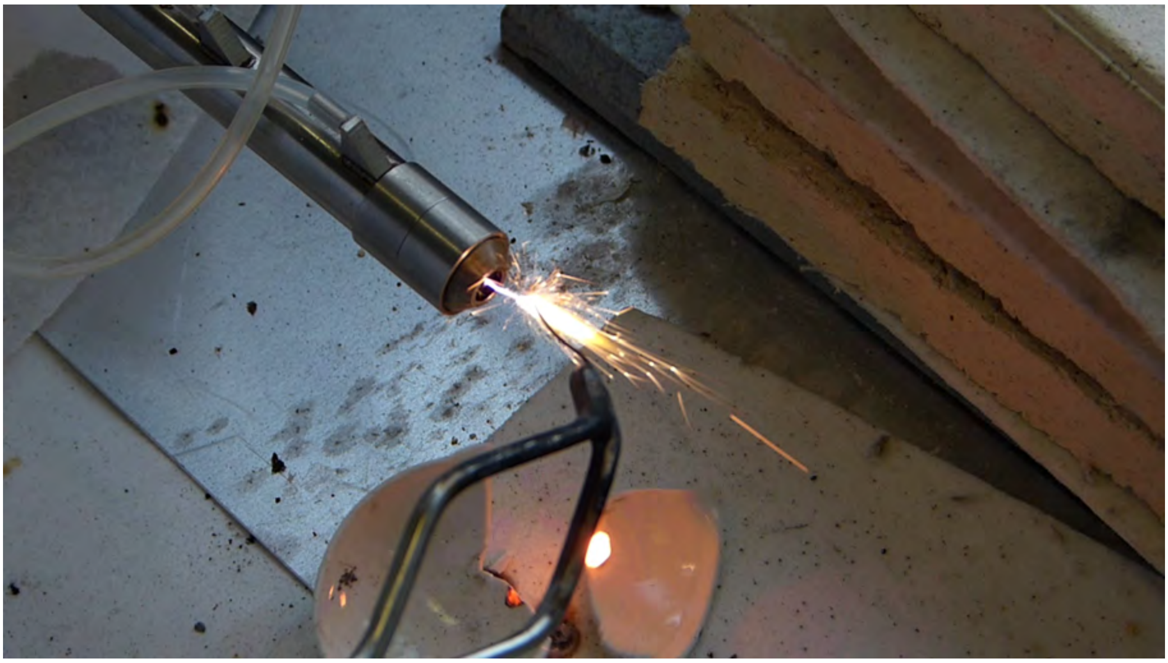

Figure 5.22: *Capture of a metal droplet breaking the ceramic cup*

needed to melt iron. The melting point of iron is at 1811 K, its effective heat capacity is 0.449 $\frac{J}{gK}$ and the melting heat is 13.8 $\frac{kJ}{mol} = 268 \frac{kJ}{kg}$ [76]. With these values one can calculate the specific energy needed to heat a piece of iron from room temperature (20 °C) to its melting point and melt it:

$$0.449\frac{J}{gK} \cdot (1811K - 293.15K) + 268\frac{J}{g} = 949.5\frac{J}{g} \tag{5.14}$$

The energy needed in the experiment is therefore about a factor of 10-14 higher than the minimum needed energy, or about 90% of the energy is not used to heat and melt the paper clip.

The weight of the droplets is a bit less than the original weight of the paper clip. This is probably due to some fraction carried away as Fe_2O_3 in the sparks during the experiment: this can account for such a low loss of 0.2-0.3 g.

Number	Weight before [g]	Weight after[g]	Treatment time [s]	Energy [kJ/g]
1	0.37	0.36	23	11.2
2	0.39	0.36	30	13.8
3	0.4	0.38	20	9.0
4	0.36	0.33	26	13.4

Table 5.6: *Experimental results of the paper clip plasma burning*

Results

The volume of the metal had not changed after treatment, but the shape. The treated material is no longer bulky, but molten in a drop. This can be of advantage for the treatment of wastes. The mass is slightly reduced in the treatment, but the missing part is probably deposited elsewhere as sparking of Fe_2O_3 since mass should be conserved.

The needed energy for the complete melting of an iron piece is about 10 times higher than the theoretical minimum needed energy. This result is in accordance with the expectations, since a high amount of heat is needed to keep the temperature in the metal, losses occur by heat radiation and not all the heating energy goes into the specimen but into the environment. Considering these effects, the energy that was applied is still quite low and the plasma torch seems to work efficiently. This can be compared with higher power applications, like for example a 1.2 MW torch for treatment of radioactive wastes (see chapter 5.1.1), where the throughput is one 200 l barrel per hour. Assuming a material density in the barrel of 2 $\frac{g}{cm^3}$ the specific energy would be 10.8 $\frac{kJ}{g}$. This is in the same range as the energy needed in the laboratory experiment. Since in the experiment only iron is treated which is a material consuming heat, the specific energy result demonstrates a good efficiency of the plasma setup.

5.4.3 Conclusions

From the experiments, several conclusions can be drawn for the upscaling of the effects and development of a full scale plasma treatment device for radioactive wastes.

With the use of plasma it is possible to treat all kinds of material. The temperature is in principle high enough to burn and melt material with a high melting point and heat capacity, such as iron, tungsten and ceramics. The thermal power output of the torch is an important factor to consider. In the experiments it was not possible to melt down larger particles of metal due to the rather low power of the plasma torch (200 W). In addition to increasing the power, it is helpful to also decrease the size of the respective material by shreddering or other cutting technologies. In conclusion, for 80% of the total mass and 96% of the total waste volume produced during nuclear power plant operations a volume reduction can be achieved.

Another advantage of plasma treatment compared to conventional combustion is a faster processing. The combination of a plasma torch and a conventional burner can improve the burning process even more and, depending on the layout of the oven, the conventional burner can be used as an afterburner for the off-gas stream.

Caesium gets volatile under the influence of the plasma. However, only a small fraction of the total Caesium gets lost in the exhaust system. It can be found locally around the treatment spot and deposits on a cold surface. Therefore it is recommended for the design of a plasma plant for radioactive waste to plan for a collection point for the Caesium in the off-gas stream. The results are in agreement with findings at the prototype plants where about 90% end up in the solid (see chapter 5.1.1).

The specifically used microwave induced plasma proved to be useful for these experiments. Estimating, the total input energy for melting a metal wire resulted in a thermal efficiency of 9% compared to ideal melting. This is especially remarkable, since the experiment took place in free air without any oven or other thermal insulation.

5.5 Upscaling from Research to an Industrial Scale Facility

The results of the experiments would alter the optimization of the plasma oven described in chapter 5.3.

It is of major importance that all waste to be treated passes the plasma region and stays for a certain time in the plasma. A possible change in the design for adjusting the residence time would be the installation of several plasma torches which together form a layer of a plasma flame, which is then situated directly above the holding sieve.
The waste material can be treated better when the specific grain size of the feed is small. Therefore a shredder has to be included as a pretreatment of the material. The grain size should be as small as possible, so the shredder has to be of high performance.
Regarding the Caesium experiment, the results show that a Caesium collecting device is needed in the off-gas stream. Cs settles at the first cold surface and would contaminate the tubes in the off-gas process. It is also possible that a large portion of the Caesium will stick to the oven walls. To avoid accumulation in the oven, the design has to be adjusted in a way that this effect becomes less probable. This can be done by broadening the oven so that the actual treatment point is a certain distance away from the oven walls. A gas flow streaming along the oven walls adds to the Caesium retention.

The power of the plasma torches should be high enough to melt solid material. For a "worst - case scenario" if all the material treated for an hour (15.7 kg) would be iron, the energy required for a complete melt is 14.8 MJ (see equation 5.14). The total energy of all plasma torches should therefore be 165 MJ, if the conversion factor is 9% of the input. For a installed plasma torch this would mean a plasma torch power of 45.6 kW is required:

$$P_{Plasma} = \cdot \frac{165MJ}{1h} = 45.6kW \tag{5.15}$$

Detailed analysis of flue gases for toxic compounds, aerosols and particulate matter are still required.

6 Estimate of Economic Feasibility

The costs for installation and operation of a plasma treatment facility for radioactive waste depend on various factors. Due to any new installment still being in a prototype stage, a high risk and therefore a broad possible variation of the costs must be taken into account.

For economic considerations it is important to note, that a plasma plant would make costly sorting infrastructure and other waste treatment methods redundant. In scenarios where the whole waste stream is produced or stored at the plasma plant site, transportation costs are omitted as well.

The main economic advantage of plasma treatment of LILW is the efficient volume reduction. Disposal costs in final repositories are typically given in $\frac{e}{m^3}$ and vary by disposal country and site.

Table 6.1 lists the cost for disposal of mixed solid low level waste in final repositories in the countries that were already discussed in chapter 2.3.

To allow a comparison of the specific disposal costs as in table 6.1, some requirements and explanations have to be given on the displayed countries. The estimated costs per m^3 for the German final repository KONRAD differ from the official 2.2 billion € / 303000 m^3 = 7260 €/m^3 [80]. In comparison to the other countries, where in difference to Germany no costly deep geological repository is planned, the costs are half of the amount. An internal study at Westinghouse [77] showed that more realistic cost estimations also include e.g. the costs for the special containers plus an increase due to an additional limitation in the packaging degree by a desired

Country	Site	Disposal costs [€/m^3]
Germany	KONRAD	17500 [77]
Italy	Protex Italia Spa	15000 - 20000 [78]
United Kingdom	LLW Repository Ltd	2007 [41]
United States of America	Barnwell, SC	16000 - 25000 [79]

Table 6.1: *Disposal costs in different final repositories for LILW.*

shielding of the containers. The 17500€ in table 6.1 reflect these considerations.
Italy does not have nuclear power plants in operation. The radioactive waste is usually collected and disposed of in standardised 60-L containers. The costs are as high as in Germany.
In the United Kingdom at the LLW Repository Ltd close to the West Cumbrian coastline in the North West of England the low level waste is stored in a near surface repository. The costs to dispose of LLW are about a factor 10 below the costs of the compared countries.
In the US at the Barnwell site the costs differ dependent on the package density whith which the waste is delivered [79]. In addition the costs rise when disposing of waste with a higher activity content.

Vital for the design of a plasma plant is an extensive knowledge of the waste feed to treat. This includes the amount and also the composition as laid out in chapter 2.1 for the operational waste from nuclear power plants.
Plasma treatment is in principle applicable to all kinds of intermediate to low level radioactive waste. From the discussion in chapter 5 is a plant with a small plasma torch and therefore a low possible throughput as compared to existing plants. Plasma torches are mainly engineered to meet high throughputs. Typically plasma torches for thermal treatment start at power levels of about 150 kW. It is necessary for a proper waste treatment to achieve the high peak temperatures, which is not a given for lower plasma torch powers.
However, in chapter 5.5 it was already argued that for a low scale facility for the treatment of the operational waste produced in one nuclear power plant, a throughput of $15.7 kg/h$ and a total plasma power of at least $45.6\ kW$ must be achieved. The scenario for the cost estimation therefore is the treatment of operational waste from one NPP.

An average volume reduction factor of 20 is assumed, which is reasonable according to thealready existing plasma treatment plants and the discussion in chapter 5. With these data an estimation of the cost advantage dependent on the volume reduction and the disposal costs can be performed. The results are listed in table 6.2.

Name	Variable	Unit	Value
Plant throughput	$\dot{m}$	kg/h	16
Average density of input	ρ_{ave}	kg/m^3	300
Volume reduction factor	VRF	–	20
Degree of Capacity Utilization	CU	%	50
Annual cost advantage by vol. red.			
Germany		M€/a	7.3
Italy		M€/a	7.3
United Kingdom		M€/a	0.8
United States of America		M€/a	8.4

Table 6.2: *Parameters for the cost evaluation with estimated values*

The annual cost advantage by volume reduction was calculated as in eq. 6.1:

$$costs \ = \ average\ disposal\ costs \cdot \frac{\dot{m}}{\rho_{ave}} \cdot CU \cdot hours/year \cdot \frac{VRF-1}{VRF} \qquad (6.1)$$

The average disposal costs mutliplied with the annual outcome volume and the volume reduction compared to no treatment equals the cost advantage.
The result is that a very high cost advantage is given by the high volume reduction, especially in countries with high costs for disposing of LLW. As a comparison, the order to build a large scale plasma treatment facility in Kozloduy was awarded with 40 M€ [71]. Of course this is only an indication that plasma treatment of LILW is economically advantageous to conventional treatment of waste. However, it seems obvious that a large part of the costs in the waste treatment chain are the disposal costs which can be significantly reduced by decreasing the volume of the waste. Plasma treatment of LILW can be a economic advantage due to a high volume reduction factor which can be achieved.

7 Summary and Conclusions

The aim of this work was the development of a method for a small-scale plasma-induced combustion of low- and intermediate level waste (LILW) to provide a technological basis for a potential implementation of plasma treatment of LILW from nuclear power plant operations.

Firstly, generation of radioactive waste was analyzed and the waste streams produced in nuclear power plant operations studied in detail. This research produced a valuable database on the generation of LILW that did not exist before with data taken from different countries and types of NPPs. It was found out that the bulk of the volume is combustible or compressible low level waste. The waste components include combustible material such as textiles, synthetics such as foils and plastics, incombustibles such as insulation material and few other material. The typical amounts were summarized. Based on these data, a simulated non-radioactive waste material could be designed with the typical characteristics of the generated waste for further practical investigations.
In the next step, the state of the necessity for treatment of low- and intermediate level waste was described. The state of the art in LILW treatment was summarized to allow a comparison and to have an outline of the methods typically undertaken and the technical requirements of handling and treatment.

Plasma treatment of hazardous material is a technology that is not yet applied on a broad scale though some prototype facilities exist that are operating successfully. The plasma torch technology is technically mature and various applicable technological solutions for plasma torch treatment exist.
The plasma treatment of radioactive waste is considered an advanced technique and up to now only one industrial scale prototype facility exists. Based on an analysis of its strengths and weaknesses additional consideration on the design of a small scale plasma treatment of radioactive waste material was performed..

An experimental device based on microwave plasma induction was set-up at the FH Aachen / Jülich to test different design features. Specific properties of the plasma torch related to radioactive waste treatment were investigated. The main aim was to verify the known advantages of plasma technology on a laboratory scale and, in addition, to identify problems and properties that can influence the design of a full scale device.

Experiments were performed with different purposes: The treatment of typical material occurring in nuclear power plant operations resulted in a verification of the fact that in principle 80% of the total LILW mass and 96% of the total LILW volume undergo the desired changes, e.g. combustion or melting. Typical times to achieve volume reduction for a variety of materials were determined.

Some materials could be mixed with glass particles as additives which allowed for a vitrification and therefore a solidification of the material, though the volume was significantly increased compared to the raw waste treatment without the additives.

Nonradioactive Caesium was introduced in the treatment device in one experiment and the distribution of the Caesium particles in the oven and the off-gas stream was analyzed. Caesium retention in the off-gas stream is an issue that has to be addressed when constructing a LILW plasma treatment device.

When exposing metal in air it was found that an efficiency of the plasma torch of 9% as compared to ideal melting could be achieved. This conversion factor could be used for an estimation of the needed plasma power for a given throughput.

The experimental results allow for an upscaling to larger scale devices regarding the respective phenomena. A fundamental set-up of a plasma treatment facility was undertaken to identify the important components of such a plant.

Finally, an estimation of the cost advantage of plasma treatment compared to direct storage was performed, basing on the disposal costs in various countries and the volume reduction by plasma treatment. It was found that a plasma treatment device has a high economic advantage in a scenario where the costs for waste disposal are comparatively high. However, a risk in investment has to be taken into account since any new plasma treatment plant would be an exclusive design without any comparable reference devices at this point.

Statement of Authorship

Except where reference is made in the text of this dissertation, this dissertation contains no material published elsewhere or extracted in whole or in part from other works.
No other person's work has been used without due acknowledgement in the main text of the dissertation.
This dissertation has not been submitted for the award of any other degree or diploma in any other tertiary institution.

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Frederik Nachtrodt

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References

[1] FRENCH NATIONAL AGENCY FOR RADIOACTIVE WASTE MANAGEMENT ANDRA: *National Inventory of Radioactive Materials and Waste 2009: In summary.* Website/Print, 2009

[2] DIEKMANN, P: *Measurement of Tritium and Carbon-14 in radioactive waste*, FH Aachen, Diplomarbeit, 2009

[3] EUROPEAN COMMISSION: *2008/98/EC (Waste Framework Directive).* `http://ec.europa.eu/environment/waste/framework/index.htm`

[4] IAEA: *Classification of radioactive waste.* 2009

[5] IAEA: *NEWMDB Country Waste Profile Report for United States of America, Reporting year: 2008*

[6] IAEA: *NEWMDB Country Waste Profile Report for UNITED KINGDOM, Reporting year: 2007*

[7] IAEA: *NEWMDB Country Waste Profile Report for Italy, Reporting year: 2008*

[8] BUNDESAMT FÜR STRAHLENSCHUTZ: *Richtlinie zur Kontrolle radioaktiver Abfälle mit vernachlässigbarer Wärmeentwicklung, die nicht an eine Landessammelstelle abgeliefert werden.* 1989

[9] VGB: *Arbeitskreis Abfallmanagement des VGB PowerTech e.V.: Entsorgung von Kernkraftwerken - Eine technisch gelöste Aufgabe.* Information booklet, 2004

[10] BUNDESAMT FÜR STRAHLENSCHUTZ (BFS): *Endlager Konrad. Antworten auf die meistgestellten Fragen.* `http://www.endlager-konrad.de/cln_351/nn_1073436/SharedDocs/Downloads/DE/konrad-faq,templateId=raw,property=publicationFile.pdf/konrad-faq.pdf`, mar 2011

[11] NRC: *NRC Graphic of Low-Level Waste Facility*. `http://www.nrc.gov/images/reading-rm/photo-gallery/20071115-007.jpg`, 2010. – [Online; accessed 20-nov-2012]

[12] KARLSRUHE INSTITUTE OF TECHNOLOGY: *Verglasungseinrichtung Karlsruhe*. `http://www.ine.kit.edu/540.php`. Version: 2012. – [Online; accessed 20-nov-2012]

[13] PERKINS, B.L.: *Incineration facilities for treatment of radioactive wastes: a review*. `http://www.osti.gov/energycitations/product.biblio.jsp?osti_id=7181109`. Version: 1976

[14] GREENWOOD, Sarah ; ROSENBERGER, Stefan ; BONNET, Claude ; THOUVENOT, Rémi ; BRODT, Alfred: *SOLID RADIOACTIVE WASTE STRATEGY REPORT (SRWSR)*. `http://www.epr-reactor.co.uk/ssmod/liblocal/docs/SupportingDocuments/SolidRadioactiveWasteStrategyReport.pdf`. Version: 2008

[15] LOHMANN, Peter ; (WESTINGHOUSE ELECTRIC GERMANY GMBH): Implementation of Treatment Systems for Low and Intermediate Radioactive Waste at Site Radwaste Treatment Facility (SRTF), PR China. In: *KONTEC*, 2011

[16] WESTINGHOUSE ELECTRIC GERMANY GMBH: *GBRH007457*. 2012. – internal document

[17] BOULOS, M.I. ; FAUCHAIS, P ; PFENDER, E: *Thermal plasmas: fundamentals and applications*. Bd. 1. Plenum Press, 1994

[18] ECSC CONVENTION NO. 6210.93/8/801: *Basic properties of high-intensity electric arcs used in steelmaking*. 1973

[19] SCHOPP, C. ; NACHTRODT, F. et. a.: A novel 2.45 GHz/200 W Microwave Plasma Jet for High temperature Applications above 3600 K. In: *Proceedings of HTPP12*, 2012

[20] INEICHEN, Robert: *Hochtemperatur-Ofen*. patent application, may 2005. – EP 1 533 568 A1

[21] HEEP, W. ; INEICHEN, R.: The ZWILAG Incinerator and Melting Furnace. In: *Proceedings of GLOBAL*, 2005, S. 9–13

[22] ZWILAG ZWISCHENLAGER WÜRENLINGEN AG: *Plasma-Anlage*. `http://www.zwilag.ch/upload/cms/user/plasma-anlage2.jpg`, November 2012. – [Online; accessed 20-nov-2012]

[23] GORBUNOV, V.A. et. a.: PLASMA TECHNOLOGIES: THE ENHANCEMENT OF THE JOINT TREATMENT OF COMBUSTIBLE AND NONCOMBUSTIBLE SOLID RADIOACTIVE WASTES. In: *Proceedings of KONTEC 2011*, S. 593–596

[24] IAEA-TECDOC-1527: *Application of Thermal Technologies for Processing of Radioactive Waste*. 2006

[25] FASTNER, André: *Verfahrenstechnische Auslegung der mechanischen Komponenten einer Abgasbehandlungsstrecke zur Plasmaverbrennung von radioaktiven Abfällen*, Hochschule Mannheim, Bachelors thesis, 2011

[26] PUBLIC DOMAIN, WIKIPEDIA: *HEPA filter diagram*. `http://en.wikipedia.org/wiki/File:HEPA_Filter_diagram_en.svg`, 2008. – [Online; accessed 20-nov-2012]

[27] BLAESIUS, Christoph: *Zusammensetzung schwach- und mittelradioaktiver Betriebsabfälle aus Kernkraftwerken*, RWTH Aachen, Bachelors thesis, 2011

[28] BRENNECKE, P: *Requirements on Radioactive Waste for Disposal (Waste Acceptance Requirements as of October 2010) - Konrad repository*

[29] POI ENEA-DISP ED INFINE ANPA POI DIVENTATA APAT CNEN-DISP: *La Guida Tecnica n. 26 - La gestione dei rifiuti radioattivi*. 1987

[30] WORLD NUCLEAR TRANSPORT INSTITUTE: *Fact sheet 1 - Safety Regulations Governing Radioactive Materials Transport*. `http://www.wnti.co.uk/media/3742/1.pdf`, aug 2010

[31] SOBOLEV, IA et. a.: *High Temperature Treatment of Intermediate-Level Radioactive Wastes-Sia Radon Experience*. United States. Department of Energy, Waste Management Symposia 2003, 2003

[32] BAUFORUM STAHL: *Brandbelastungen und Heizwerte - Heizwerte in alphabetischer Reihenfolge*. `http://www.bauforumstahl.de/upload/documents/brandschutz/kennwerte/Heizwertalpha.pdf`, 2012. – [Online; accessed 20-nov-2012]

[33] BUNDESAMT FÜR STRAHLENSCHUTZ (BFS): *Abfallprognosen*. `http://www.bfs.de/de/endlager/abfaelle/prognose.html`. Version: November 2012

[34] INTERNATIONAL ATOMIC ENERGY AGENCY: *Net-Enabled Waste Management Database (NEWMDB)*. `http://newmdb.iaea.org/`. Version: Februar 2002

[35] NIEDER, R.: *Chemie und Wasseraufbereitung im Kernkraftwerk* (Lehrhefte für die Ausbildung zum Kraftwerker)

[36] BORLEIN, Markus: *Kerntechnik* -. 2. überarbeitete Auflage. Würzburg : Vogel Business Media, 2011. – ISBN 978–3–834–33253–0

[37] EFREMENKOV, V.M.: Radioactive waste management at nuclear power plants. In: *IAEA BULLETIN, 4/* (1989)

[38] *Program on Technology Innovation: Technical Support for GE Economic Simplified Boiling Water Reactor (ESBWR) - Radwaste System Design*. 2006

[39] USNRC TECHNICAL TRAINING CENTER: *Reactor Concepts Manual - Boiling Water Reactor Systems*. `http://www.nrc.gov/reading-rm/basic-ref/teachers/03.pdf`. Version: nov 2012

[40] OEHME, C.: Ionenaustauscher und Ionenaustauschverfahren bei der Abwasserbehandlung. In: *Pure Appl. Chem.* (1973)

[41] LLW REPOSITORY LTD: *Service Pricing - Overview*. `http://www.llwrsite.com/UserFiles/File/Customers/Version2-0/WSC-SPR-OVR-Version2-0-Jan2011.pdf`. Version: jan 2011

[42] IAEA: *Radioactive Waste Management Glossary*. 2003

[43] *"Atomgesetz in der Fassung der Bekanntmachung vom 15. Juli 1985 (BGBl. I S. 1565), das zuletzt durch Artikel 5 Absatz 6 des Gesetzes vom 24. Februar 2012 (BGBl. I S. 212) geändert worden ist"*. `http://www.gesetze-im-internet.de/atg/BJNR008140959.html`

[44] BRENNECKE, P: *Anforderungen an endzulagernde radioaktive Abfälle (Endlagerungsbedingungen, Stand: Dezember 1995) - Schachtanlage Konrad -*

[45] BUNDESAMT FÜR STRAHLENSCHUTZ (BFS): *Radioaktive Abfälle beim Rückbau von Kernkraftwerken*. `http://www.bfs.de/de/endlager/publika/rueckbau.html`, November 2012

[46] BUNDESAMT FÜR STRAHLENSCHUTZ: *Richtlinie zur Kontrolle radioaktiver Reststoffe und radioaktiver Abfälle*. 2008

[47] *"Strahlenschutzverordnung vom 20. Juli 2001 (BGBl. I S. 1714; 2002 I S. 1459), die zuletzt durch Artikel 5 Absatz 7 des Gesetzes vom 24. Februar 2012 (BGBl. I S. 212) geändert worden ist"*. `http://www.gesetze-im-internet.de/strlschv_2001/BJNR171410001.html`

[48] OECD/NEA: *Radioactive Waste Management Programmes in OECD/NEA Member Countries*. `http://www.oecd-nea.org/rwm/profiles/`. Version: 2013

[49] DEFRA, DTI ; DEVOLVED ADMINISTRATIONS the: *Policy for the Long Term Management of Solid Low Level Radioactive Waste in the United Kingdom*. March 2007

[50] NUCLEAR REGULATORY COMMISSION, U S.: *NRC Regulations § 61.55 Waste classification*. `http://www.nrc.gov/reading-rm/doc-collections/cfr/part061/part061-0055.html`,-

[51] HAEFNER, D ; LAW, J: System Design Description and Requirements for Modeling the Off-Gas Systems for Fuel Recycling Facilities. In: *INL-2010, Idaho National Laboratory* (2010). `http://www.inl.gov/technicalpublications/Documents/4655297.pdf`

[52] BUNDESAMT FÜR STRAHLENSCHUTZ (BFS): *Asse II - Maßnahmen und Perspektiven*. `http://www.endlager-asse.de/SharedDocs/Publikationen/DE/2_broschueren/asse_massnahmen_perspektiven.pdf?__blob=publicationFile`, feb 2012

[53] MIRZAKHANIAN, A.: *Surface Decontamination using a high performance Laser*, FH Aachen, Diplomarbeit, 2011

[54] WIEDERAUFARBEITUNGSANLAGE KARLSRUHE RÜCKBAU- UND ENTSORGUNGS-GMBH: *Decontamination*. `http://www.archive-de.com/page/29641/2012-05-30/http://www.ewn-gmbh.de/ewngruppe/`

`wak/waste-disposal/technical-facilities/decontamination.html?L=1`, aug 2010

[55] QUADE, U ; KLUTH, T: Recycling metallischer Reststoffe: 20 Jahre Betrieb der Schmelzanlage CARLA durch Siempelkamp Nukleartechnik GmbH. In: *Atw. Internationale Zeitschrift für Kernenergie* 54 (2009), Nr. 10

[56] NUKEM TECHNOLOGIES GMBH: *Pyrolysis of Radioactive Organic Waste*. 2007. – Information Booklet

[57] CHEN, Francis F.: *Introduction to plasma physics and controlled fusion*. Plenum Press, New York, 1984

[58] GOLDMAN, K.: Electric arcs in argon. In: *Physica* 29 (1963), Nr. 9, S. 1024–1040

[59] HEUERMANN, Holger: *Hochfrequenztechnik*. 2009. Aufl.2. Wiesbaden : Vieweg, 2005. – ISBN 978–3–8348–0769–4

[60] HEUERMANN, H.: *Technisch erzeugte Plasmen*. `http://www.hhft.de/files/service/Mikroplasma.pdf`, 2012

[61] HEBERLEIN, J ; MURPHY, A B.: Thermal plasma waste treatment. In: *Journal of Physics D: Applied Physics* 41 (2008), S. 53001

[62] DECKERS, J.: Treatment of Low-Level Radioactive Waste by Plasma: A Proven Technology? In: *Proceedings of the 13th International Conference on Environmental Remediation and Radioactive Waste Management*, 2010

[63] HOLTRUP, S. ; HEUERMANN, H.: Fundamentals and ignition of a microplasma at 2.45 GHz. In: *Microwave Conference, 2009. EuMC 2009. European* IEEE, 2009, S. 1607–1609

[64] COMMITTEE ON PLASMA PROCESSING OF MATERIALS AND NATIONAL MATERIALS ADVISORY BOARD (U.S.): *Plasma processing of materials*. National Academy Press, 1985 `http://books.google.de/books?id=AzkrAAAAYAAJ`

[65] FIEDLER, J.: *Grundlagen für die Entwicklung eines Verfahrens zu plasmachemischen Vernichtung humanmedizinischer Abfallstoffe*, Martin-Luther-Universität Halle-Wittenberg, Diss., 2007

[66] RUTBERG, P.G.: Plasma pyrolysis of toxic waste. In: *Plasma physics and controlled fusion* 45 (2003), Nr. 6, S. 957

[67] PARK, H.S. ; LEE, B.J. ; KIM, S.J.: Medical waste treatment using plasma. In: *J. Ind. Eng. Chem* 11 (2005), Nr. 3, S. 353–360

[68] ZWILAG ZWISCHENLAGER WÜRENLINGEN AG: *Geschäftsbericht 2011*. `http://www.zwilag.ch/upload/cms/user/geschaeftsbericht_2011.pdf`, 2012

[69] BÜTTNER, K.: Neue Lösungen für Abfallbehandlungszentren bei Neubauten von Kernkraftwerken russischen Typs. In: *Atw. Internationale Zeitschrift für Kernenergie* (2010), Mai

[70] DECKERS, J.: *Plasma treatment of radioactive waste*. `http://www.bgns.bg/web/pub/bgns/22_paper.pdf`, September 2010

[71] IBERDROLA ENGINEERING AND CONSTRUCTION: *BULGARIA: Kozloduy Waste Management*. `http://www.iberdrolaingenieria.com/ibding/proyectos.do?op=det&id=31&despliega=3`, November 2012. – [Online; accessed 20-nov-2012]

[72] HEMMING, Werner ; WAGNER, Walter: *Verfahrenstechnik* -. 10. korrigierte Auflage. Würzburg : Vogel Verlag Und Druck, 2008. – ISBN 978–3–834–33113–7

[73] BUNDESMINISTERIUM FÜR LAND- UND FORSTWIRTSCHAFT, UMWELT UND WASSERWIRTSCHAFT: *Stand der Technik bei Abfallverbrennungsanlagen*. `http://www.umweltbundesamt.at/fileadmin/site/umweltthemen/industrie/pdfs/endversion_deutsch.pdf`, 2002

[74] WOLFRAM | ALPHA: *Wolfram Alpha LLC*. `http://www.wolframalpha.com/input/?i=activated+charcoal`, 2012. – [Online; accessed 20-nov-2012]

[75] WOLFRAM | ALPHA: *Wolfram Alpha LLC*. `http://www.wolframalpha.com/input/?i=tungsten`, 2012. – [Online; accessed 20-nov-2012]

[76] WOLFRAM | ALPHA: *Wolfram Alpha LLC*. `http://www.wolframalpha.com/input/?i=iron`, 2012. – [Online; accessed 20-nov-2012]

[77] D. BLASIUS, WESTINGHOUSE ELECTRIC GERMANY GMBH: *Endlagerkosten KONRAD*. 2012. – internal document

[78] PROTEX ITALIA SPA: *private communication*. `http://www.protexgroup.com/gruppo.htm`, 2013

[79] SOUTH CAROLINA ENERGY OFFICE: *Uniform Schedule of Maximum Disposal Rates for Atlantic Compact Regional Waste*. `http://www.energy.sc.gov/publications/MURS2013.pdf`. Version: jul 2012

[80] ENDLAGER KONRAD: *Kosten und Kostenverteilung des Endlagerprojekts Schacht Konrad*. `http://www.endlager-konrad.de/cln_351/nn_1072870/DE/Themen/Kosten/__node.html?__nnn=true`, aug 2012