Use of activated products as radiotracers for the development of environmental technologies

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Typical applications of radioactive substances in industry

SEALED SOURCES applications

- Gamma transmission techniques for measuring filling levels
- Radioisotope Gamma Scanning for columns and pipes
- Neutron backscattering for level and interface detection

UNSEALED SOURCES applications

- Measuring Residence Time Distribution (RTD) of material transport in continuously operating processes
- Measuring Mixing/homogenization time in vessels
- Leak detection in heat exchangers, e.g.
- Flow rate measurement of fluids (liquids or gasses) in pipelines
- …

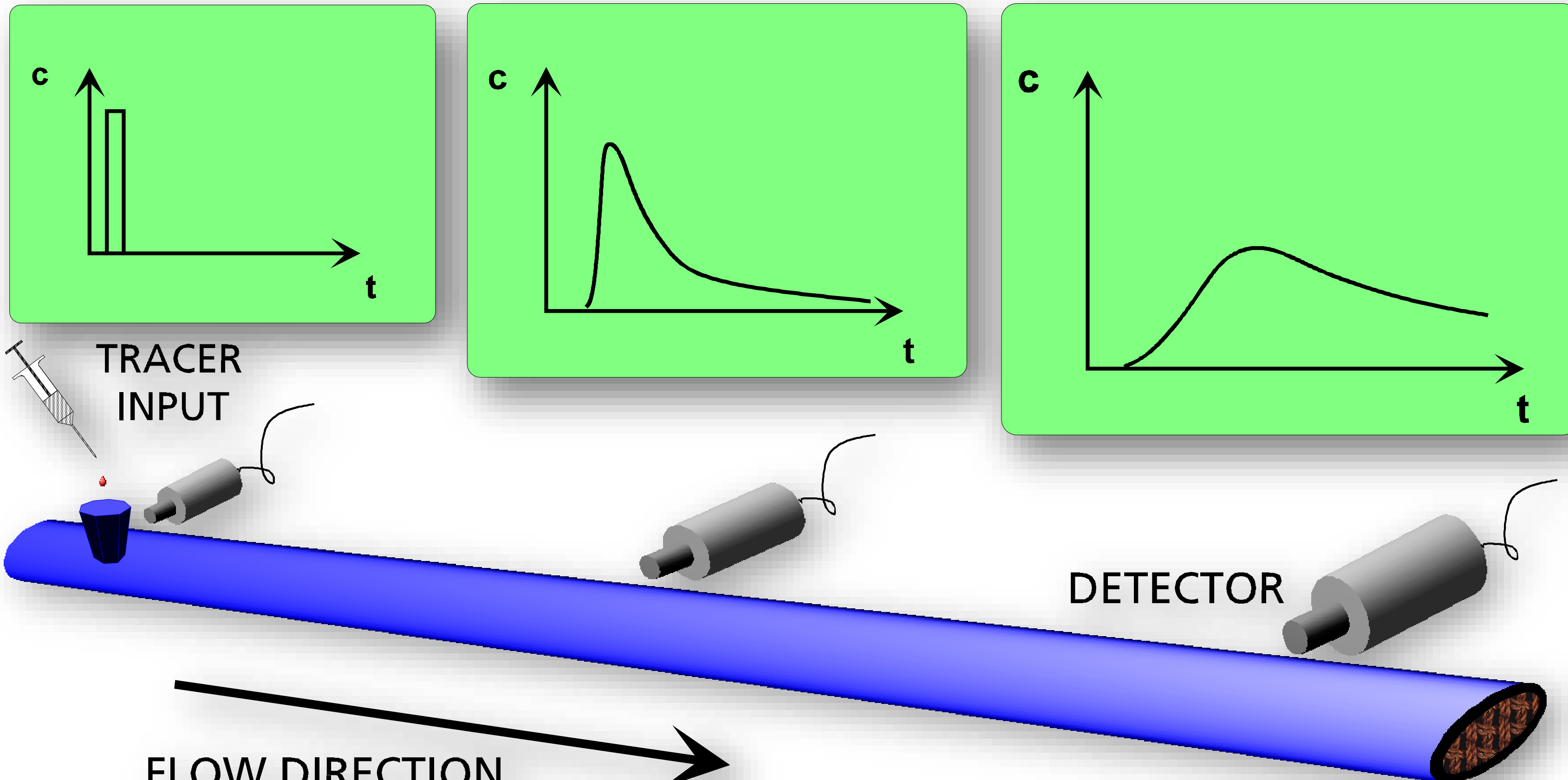
Typical unsealed radioactive isotopes used as radiotracers in industry (selection)

Isotope	Half-life	**Kind of Radiation:** Energy in MeV (intensity)
Sodium-24	***15 h***	***Gamma:*** *1.37 (100%); 2.75 (100%)*
Argon-41	***110 min***	***Gamma:*** *1.29 (99%)*
Scandium-46	***84 d***	***Gamma:*** *0.89 (100%); 1.12 (100%)*
Chromium-51	***28 d***	***Gamma:*** *0.320 (9.8%)*
Krypton-79	***35 h***	***Gamma:*** *0.26 (11%); 0.51 (15%); …*
Bromine-82	***36 h***	***Gamma:*** *0.55 (72%); … ; 1.32 (27%); 1.47 (17%)*
Technetium-99m	***6 h***	***Gamma:*** *0.14 (85%)*
Indium-113m	***1,7 h***	***Gamma:*** *0.392 (64%)*
Iodine-131	***8 d***	***Gamma:*** *...; 0.36 (82%); 0.64 (7%)*
Xenon-133	***5.3 d***	***Gamma:*** *0.03 (38%); 0.08 (37%)*
Lanthanum-140	***40 h***	***Gamma:*** *0.33 (21%); 0.49 (46%); 0.82 (22%); 1.60 (96%)*
Mercury-197	***2.7 d***	***Gamma:*** *0.07 (56%); 0.08 (35%)*
Gold-198	***2.7 d***	***Gamma:*** *0.41 (96%)*

Typical properties of radiotracers for application in industry

- Relatively **short half life** for rapid decay and rapid fall below the threshold values
- **Gamma emitters** because of large wall thicknesses and touchless data acquisition from outside without sampling
- **Activated products** usually produced in a nuclear research reactor by neutron capture

Typical application of radiotracers in industry: Measurement of Residence Time Distribution



Two fundamental types of (radio)tracers

Tracers

Extrinsic (or physical)

Intrinsic (or chemical)

Extrinsic or physical tracers

- are made up of atoms or molecules supposed to share the same physical (dynamic) characteristics and, in general, the same mass flow behavior as the investigated medium.

- For example, in case of water, $Na^{131}I$- and $^{99m}TcO_4$ are examples of extrinsic or physical tracers.

Extrinsic tracers are mostly used in industrial application

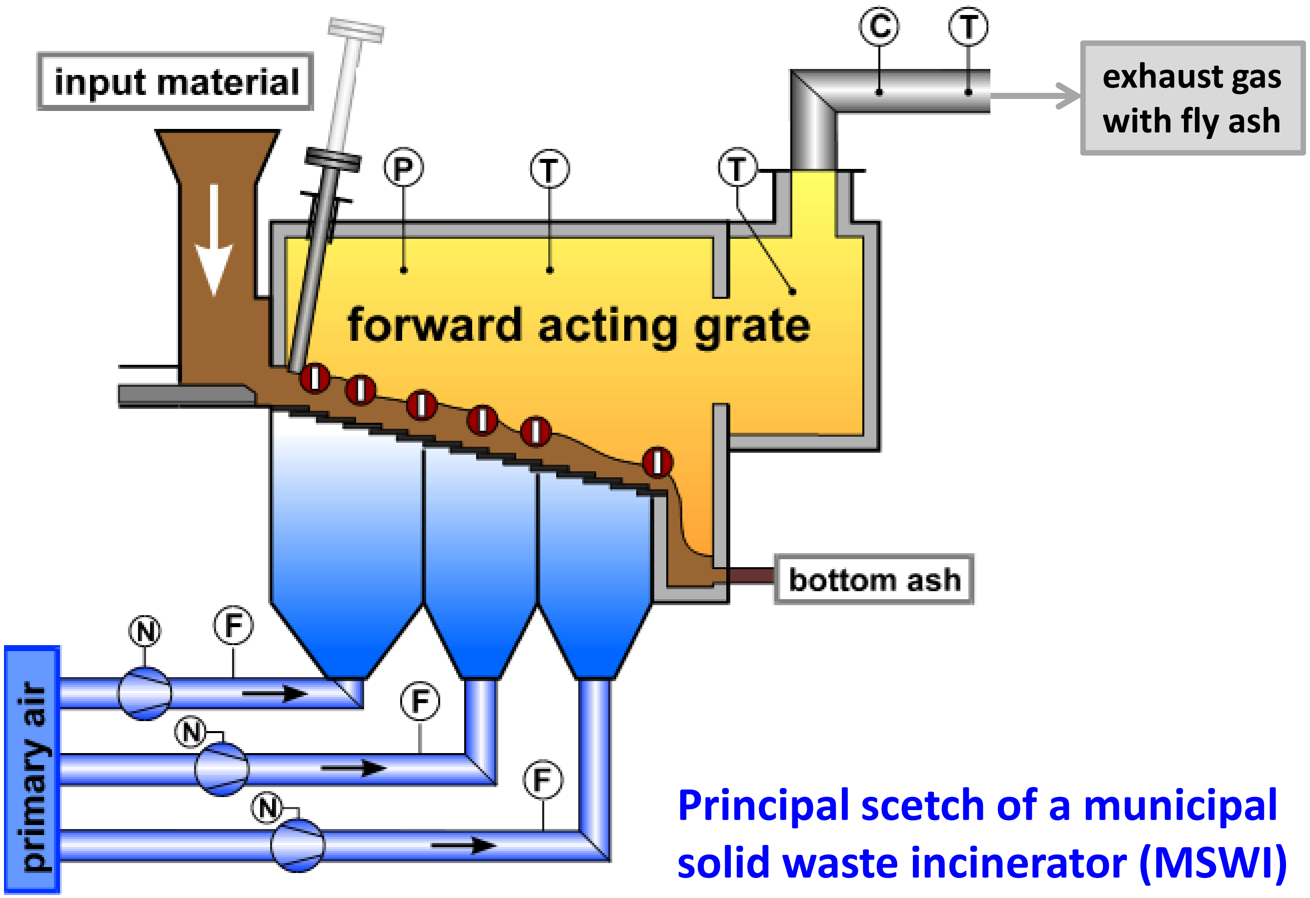
Intrinsic or chemical tracers

- are molecules containing an isotope of one of the molecule's natural elements.
- For example: water (H_2O): substitution of one hydrogen atom by Tritium (^3H) => $^1H^3H^{16}O$, (or of both => $^3H_2{}^{16}O$)
- Or in the case of nonradioactive labeling: Deuterium (^2H) => $^1H^2H^{16}O$ (or $^2H_2{}^{16}O$) (Deuterium = nonradioactive H-isotop)
- => Water molecule is traced "from the inside", in the intimacy of its nucleus.
- Consequently, the water tracer will practically follow all movements, phase changes and reactions of water itself.

Applications of radiotracers for the development of environmental technologies

- Measurement of heavy metal release at a municipal solid waste incineration process in pilot plant scale
- Measurement of the residence time of the gaseous phase in a High Pressure Partial Oxidation (HP-POX) reactor

Measurement of heavy metal release at municipal solid waste incineration



Principal scetch of a municipal solid waste incinerator (MSWI)

Measurement of heavy metal release at municipal solid waste incineration

Because of the **HEAVY METAL CONTENT**

(copper, zinc, lead, …)

in the municipal solid waste incineration (MSWI) residues

(**bottom ash,** fly ash)

<u>simple</u> reuse of mineral residues (ashes) is a

PROBLEM

(that means without any further processing)

Measurement of heavy metal release at municipal solid waste incineration

Proposal for solution:

Separation of heavy metals already during the incineration process!!!

Reduction of the heavy metal content **in the bottom ash** by volatilization of them by primary measures

➔ **Enrichment** of heavy metals **in the fly ash**

Advantages:

- **simple reuse of bottom ash** (without additional processing)
- **retrieval of heavy metals** from the fly ash

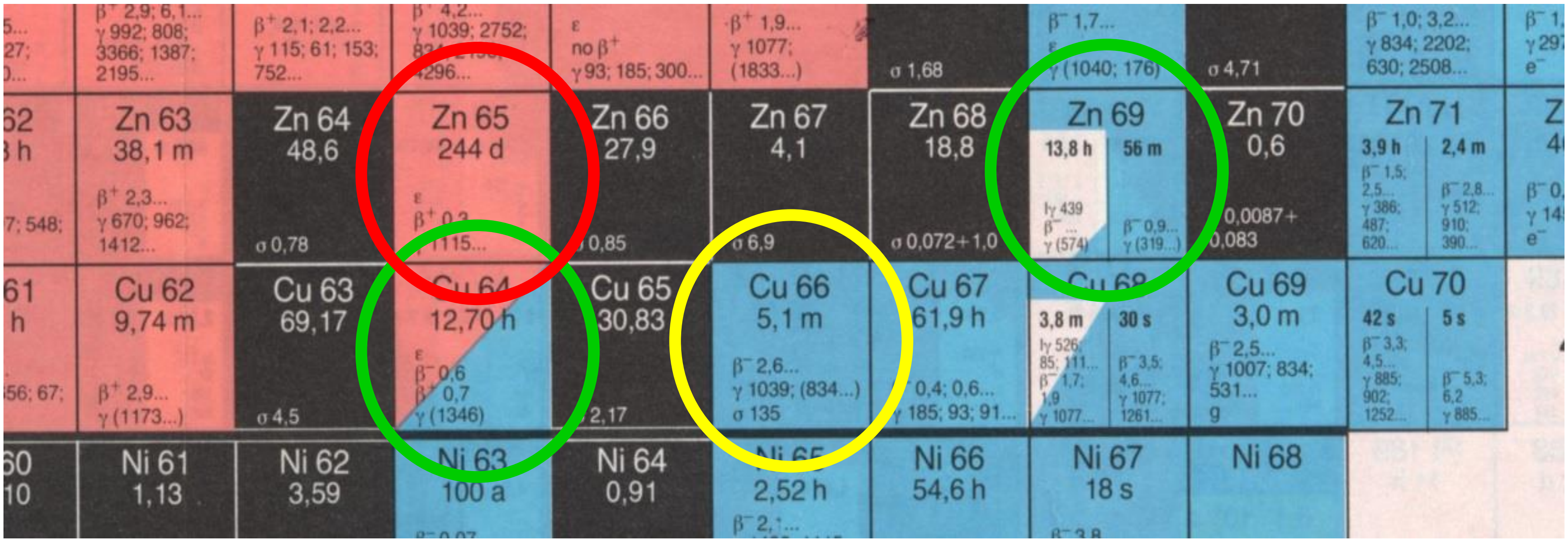
Measurement of heavy metal release at municipal solid waste incineration

Objectives of radiotracer measurements:

- **Firstly**, investigation of the behavior of the **metallic forms**
- Localize the place in the incinerator **where** the evaporation occurs
- **Verification of hypotheses** for the volatility of Zn & Cu: (from theory and lab-scale experiments)
  - A good zinc evaporation occurs at:
    - High temperatures &
    - Reducing conditions
  - Copper is negligibly evaporated

Measurement of heavy metal release at municipal solid waste incineration

[Tracer selection:]



Detail of a nuclid chart:

Black boxes: stable isotopes contained in the natural isotope mixture

Red boxes: positron (and gamma radiation) emitters

Blue boxes: electron (and gamma radiation) emitters

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Measurement of heavy metal release at municipal solid waste incineration

Tracer production by neutron capture in a nuclear research reactor

$^{68}Zn(n,\gamma)\ ^{69m}Zn$

$T_{1/2} = 13.8$ h

$E_{\gamma} = 439$ keV

metal sheets

(1 - 2 mm)

$^{63}Cu(n,\gamma)\ ^{64}Cu$

$T_{1/2} = 12.7$ h

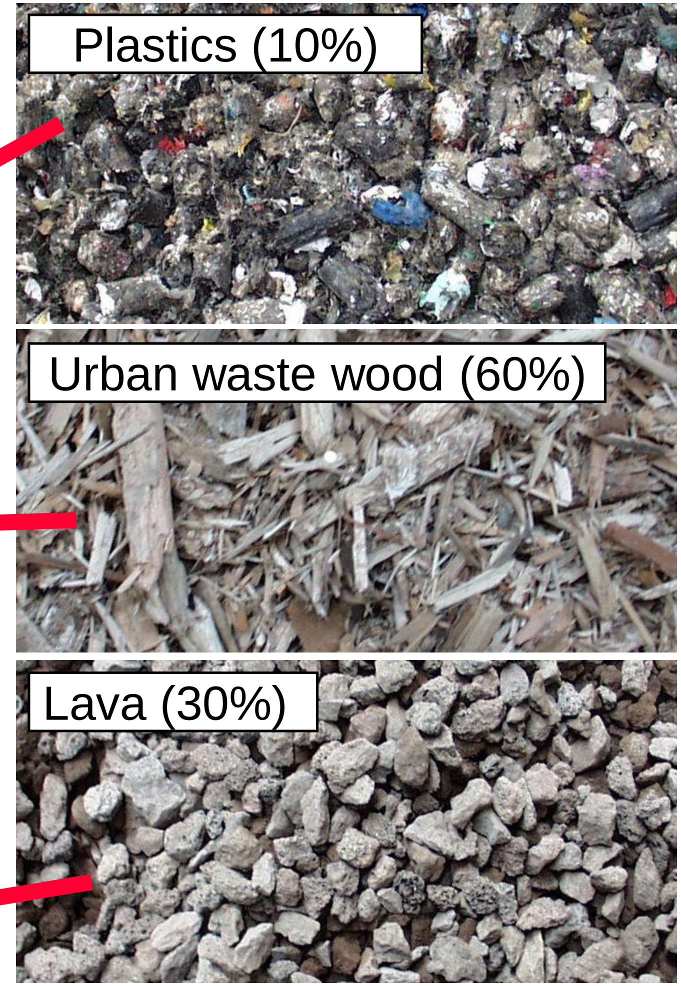
$E_{\gamma} = 511$ keV

metal spheres

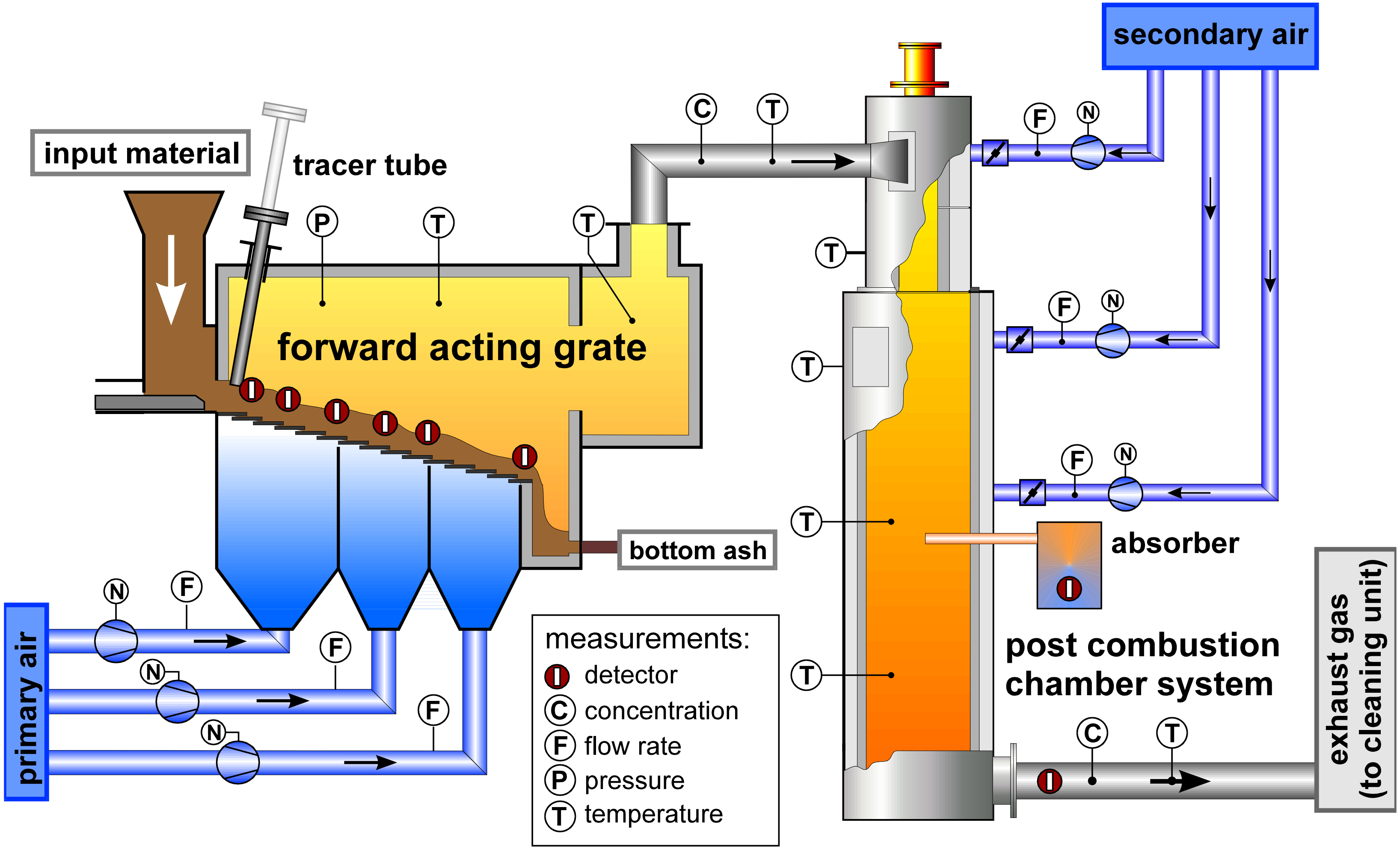
(0.2 - 0.6 mm)

Measurement of heavy metal release at municipal solid waste incineration

Synthetic model waste

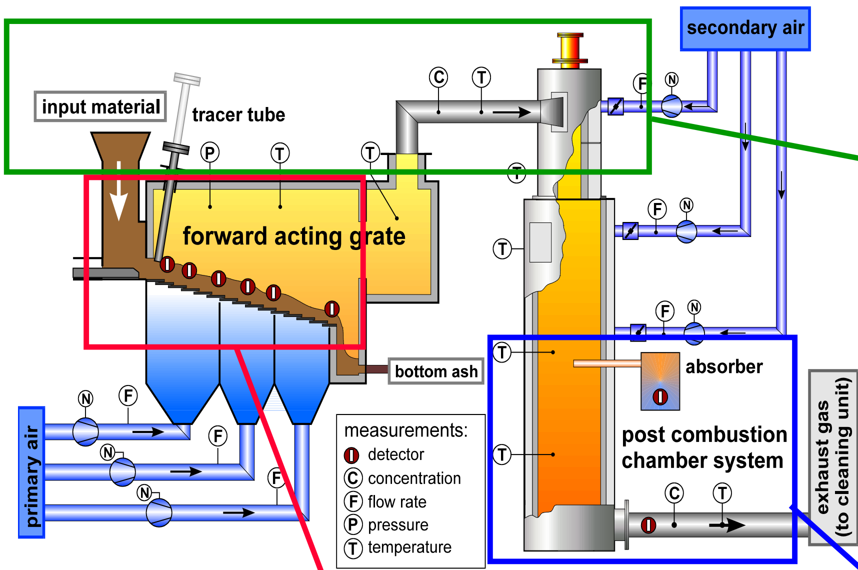


Measurement of heavy metal release at municipal solid waste incineration

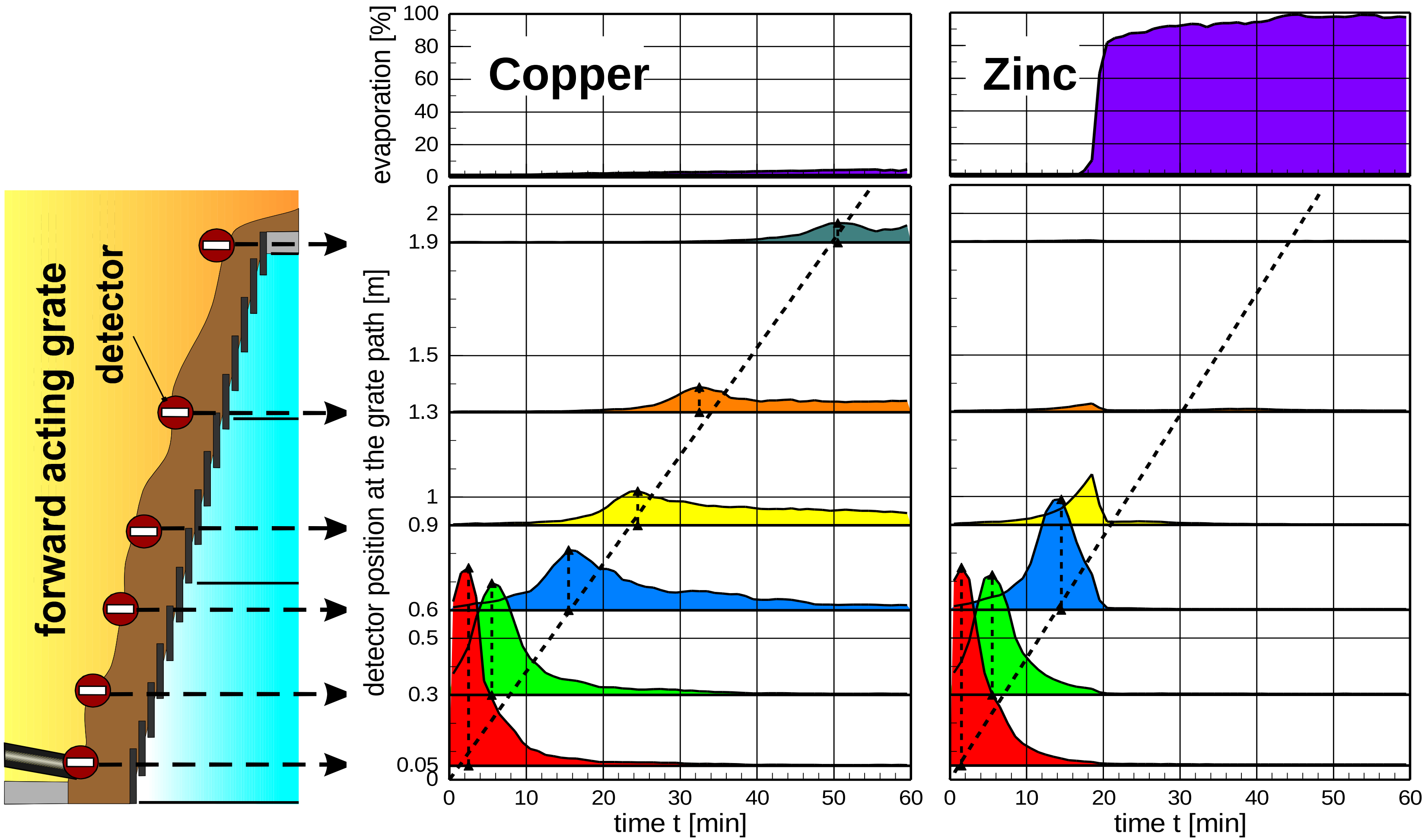


Scetch of pilot plant scale incinerator with detector positions

Installation for the radio tracer measurements



Measurement of heavy metal release at municipal solid waste incineration



Typical results of the radiotracer experiments

Applications of radiotracers for the development of environmental technologies

2nd example:

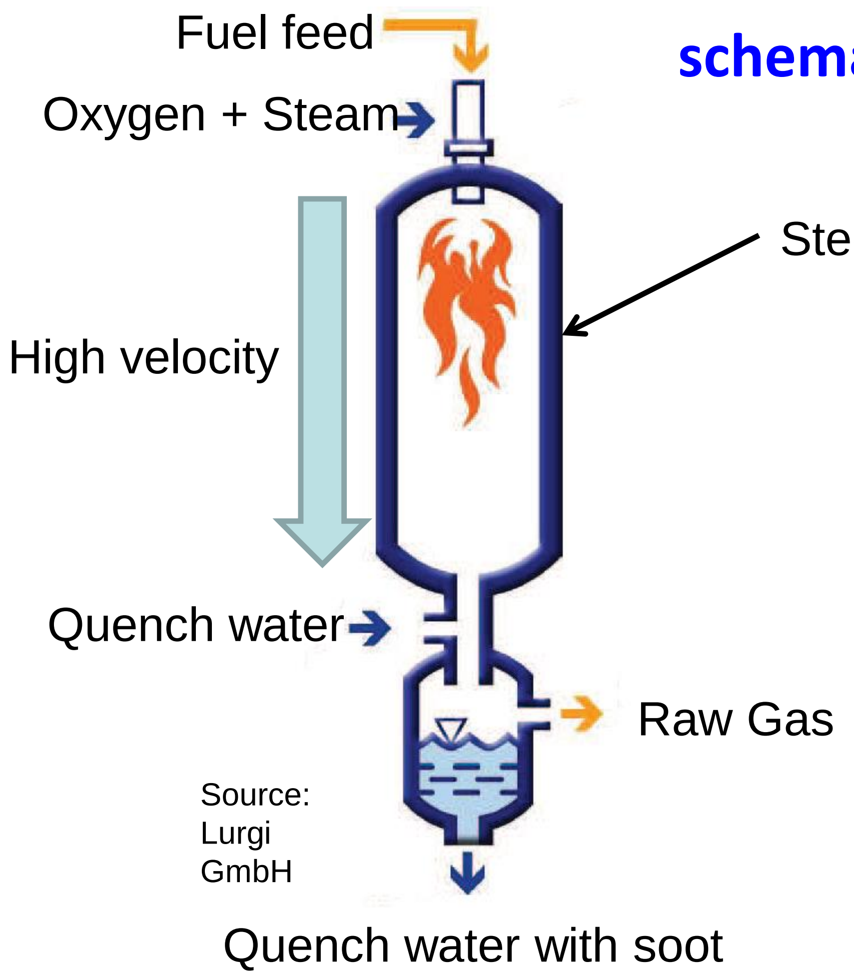
Measurement of the residence time of the gaseous phase in a High Pressure Partial Oxidation (HP-POX) reactor

Measurement of the residence time distribution of the gaseous phase in a HP-POX reactor

Purpose of a HP-POX reactor:

- Conversion of hydrocarbonic residuals (gases, liquids, slurries) from chemical processes to synthesis gas ($CO + H_2$) by autothermal reforming (ATR) and partial oxidation (POX) of gases or by multi purpose gasification (MPG) of liquids and slurries.
- HP-POX reactor in pilot plant scale:
  - Investigation and understanding of the reactor behaviour
  - Acquisition of data for process modelling
  - Measurement of the residence time distribution of the gaseous phase for better process understanding

Measurement of the residence time distribution of the gaseous phase in a HP-POX reactor



schematic sketch of a HP-POX-reactor

Extreme Conditions:

- high pressures up to 100 bar
- high temperatures up to 1,300 °C
- high-velocity gas stream
- massive, thick shell (some cm of steel)

Measurement of the residence time distribution of the gaseous phase in a HP-POX reactor

Photographs of the pilot plant scale HP-POX reactor

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Measurement of the residence time distribution of the gaseous phase in a HP-POX reactor

Choice of the radiotracer:

^{41}Ar

- relatively short halflife — $T_{1/2} = 110$ min
- relatively high gamma energy — $E_{\gamma} = 1,293$ keV
- relatively easy to produce in a nuclear research reactor by neutron capture — $^{40}Ar(n,\gamma)\ ^{41}Ar$

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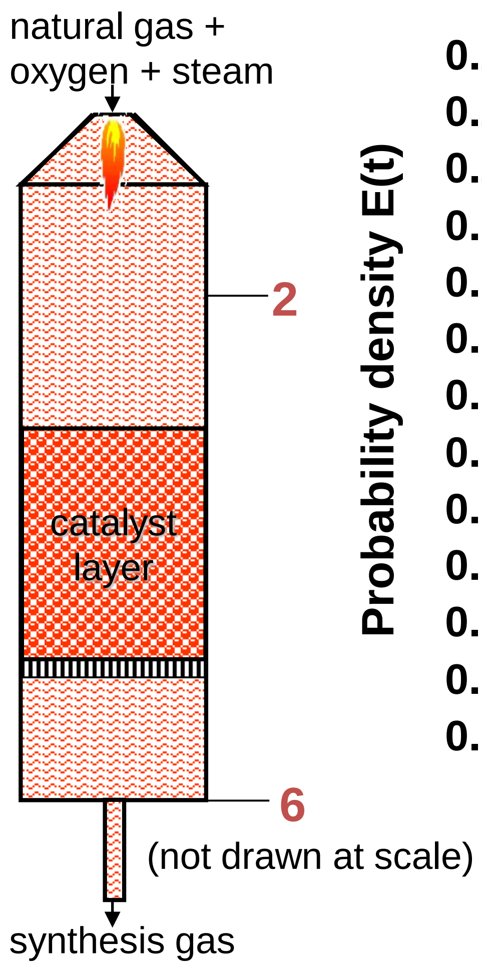
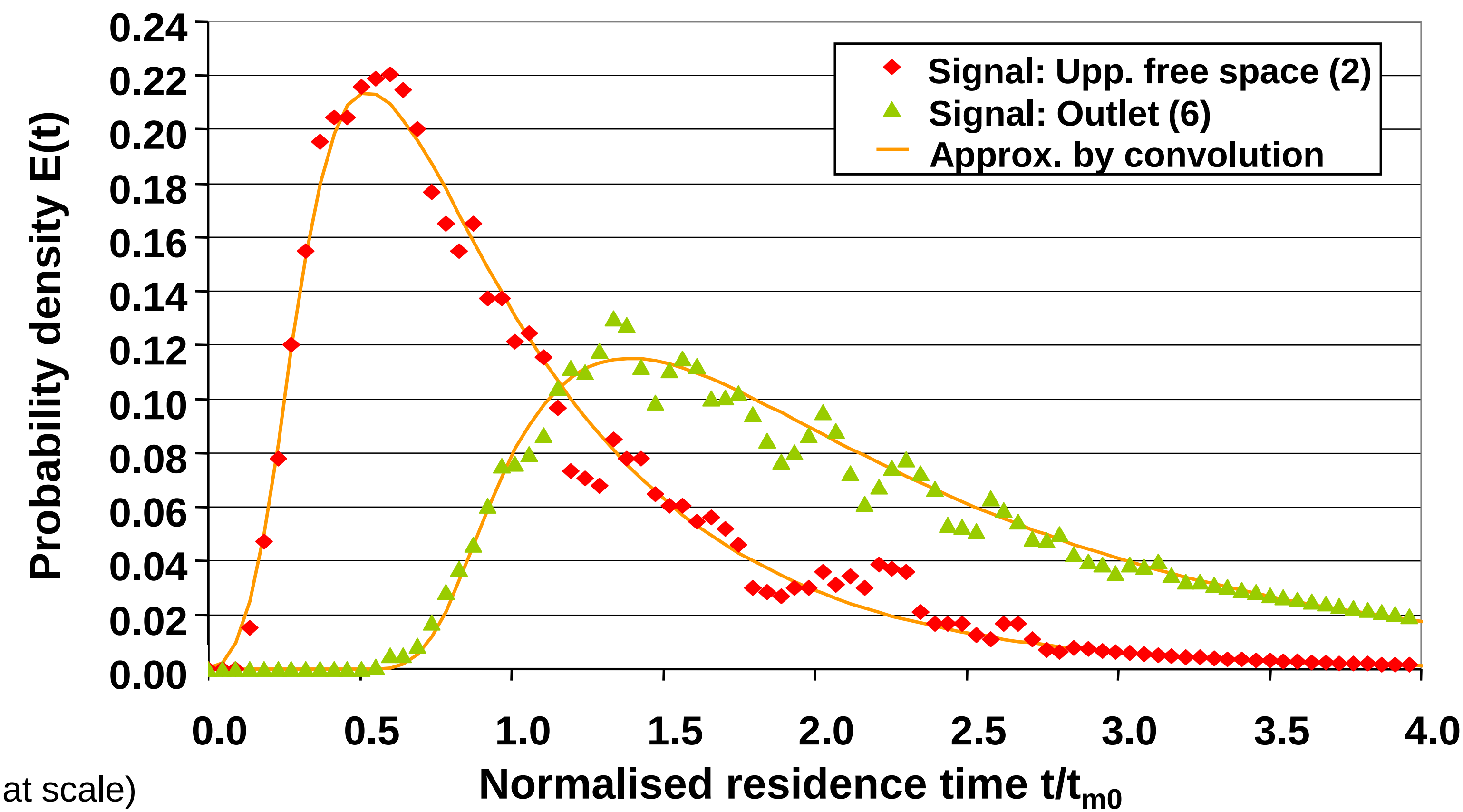
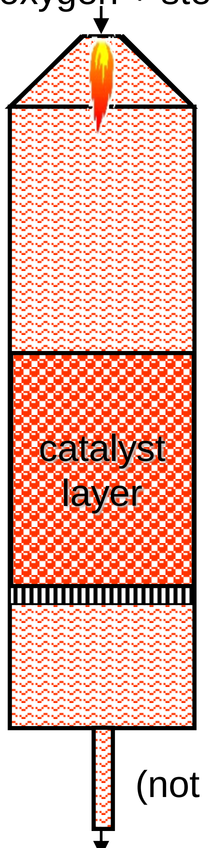
Measurement of the residence time distribution of the gaseous phase in a HP-POX reactor





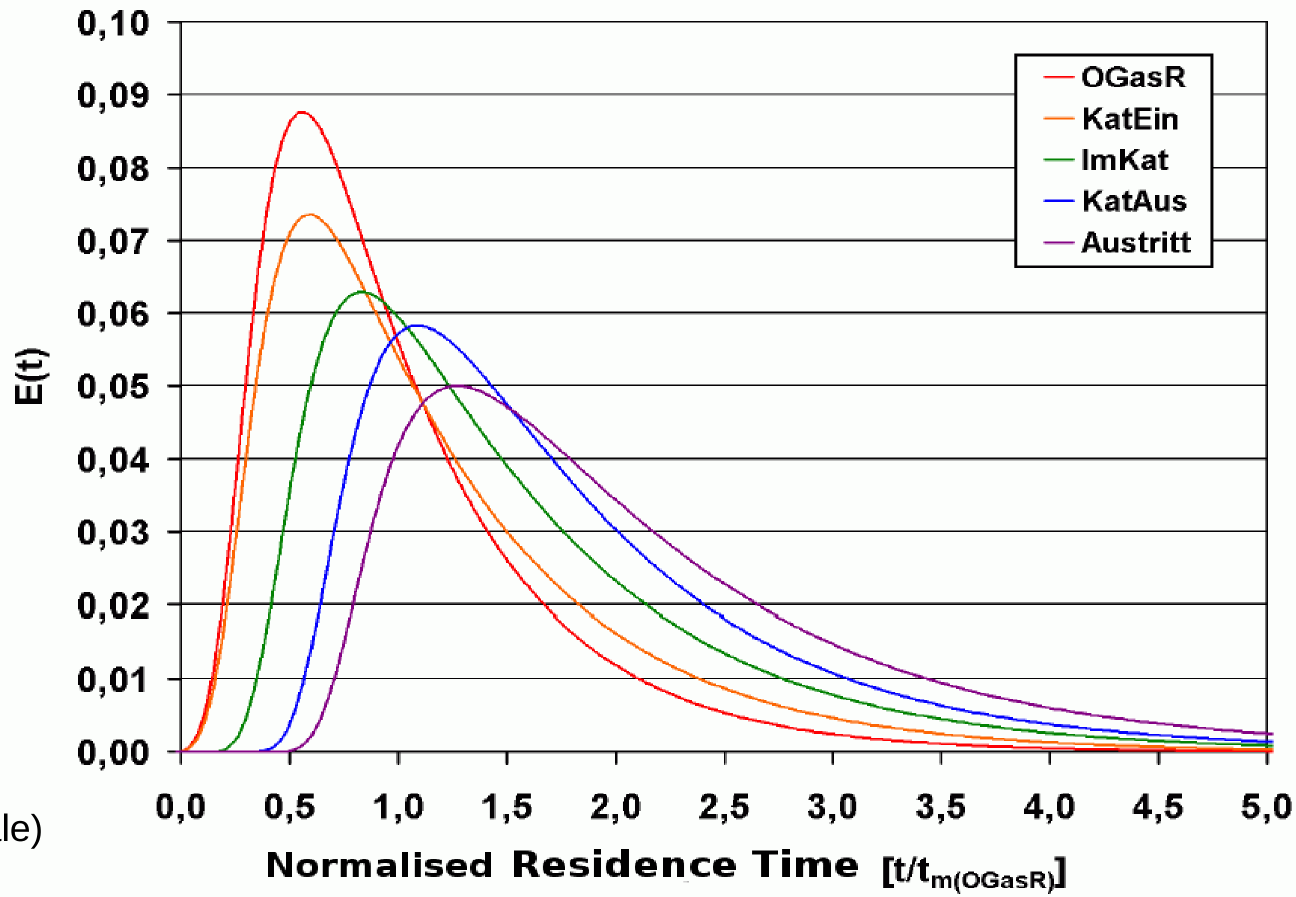
Measurement of the residence time distribution of the gaseous phase in a HP-POX reactor

natural gas + oxygen + steam



(not drawn at scale)

synthesis gas



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