

## Quantitative tar pollutant production and water solubility prognoses in the frame of Underground Coal Gasification (UCG)

Quantitative Modellprognosen zur Teer- Schadstoff Produktion und Wasserlöslichkeit im Rahmen der Untertagevergasung (UTV) von Kohle

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### Kurzfassung

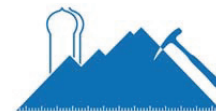
Unter Berücksichtigung der globalen Verteilung fossiler Energiereserven birgt die Untertagevergasung von Kohle (UTV) ein langfristiges Potenzial zur Sicherung des weltweiten Energiebedarfs durch wirtschaftliche Nutzung anderweitig nicht abbauwürdiger Kohlevorkommen. Im Hinblick auf die langfristige Wirtschaftlichkeit und Umweltverträglichkeit stellt die Minimierung Teer-assoziierter organischer Schadstoffe (BTEX, PAK, Heterozyklen) im Gashaushalt einen entscheidenden Faktor dar, wie verschiedene Grundwasserschadensfälle durch Gasverluste im Rückblick vergangener Feldstudien belegen. Die Evaluierung einer potenziellen Einflussnahme auf diesen unerwünschten Effekt bedarf zunächst der Kenntnis quantitativer Auswirkungen einzelner Prozessparameter (u.a. Vergasungsmittel, Betriebstemperatur) auf die Entstehung verschiedener Teerfraktionen im Prozessgas. Vor diesem Hintergrund erfolgte im Rahmen des Forschungsprojektes CO2SINUS die Entwicklung eines thermodynamischen Modells zur Quantifizierung der Teer-Schadstoff Produktion in UTV-Prozessgasen. In einem aufbauenden Modellansatz wurden exemplarisch potenzielle Teer-Schadstoff Wasserlöslichkeiten für zukünftige UTV-Tiefenlagen evaluiert.

**Schlüsselworte:** UTV, Teer, organische Schadstoffe, BTEX, PAK, Heterozyklen, Gashaushalt, CO2SINUS, Teer-Schadstoff Produktion, Teer-Schadstoff Wasserlöslichkeit, Thermodynamisches Modell

### Abstract

In view of the global fossil fuel reserves Underground Coal Gasification (UCG) bears the potential to guarantee long-term energy supply for the future markets by conversion of otherwise non-mineable coal seams. Besides the production of medium to high calorific syngas for various industrial applications (e.g. electricity generation in the frame of the Integrated Gasification Combined Cycle - IGCC Concept; or Coal-To-Liquid- CTL procedures like Fischer-Tropsch synthesis), UCG provides an option for effective greenhouse gas reduction through CO<sub>2</sub> storage in converted coal seams. With regard to long-term economic and environmental sustainability however minimization of tar related organic pollutants in the process gases is a key factor, as past projects were often confronted with organic groundwater pollution issues of BTEX, PAHs and heterocyclic compounds due to condensation of tar-loaded gas losses. With focus on potential tar reducing strategies the present CO2SINUS study thus investigated quantitative single process parameter effects (amongst others gasification agent flux & composition, pyrolysis temperature) on total tar and related single tar pollutant production in pyrolysis-, reduction/partial oxidation- and syngas fractions by using a new self-developed thermodynamic model approach. Furthermore selected p/T dependent organic pollutant-water solubility scenarios were calculated in a separate second model approach. The main focus of this second approach was set on potential future UCG depth conditions to support the parameterization of pollutant transport models with regard to effective environmental management decisions for future UCG sites.

**Keywords:** UCG, IGCC, CTL, CO<sub>2</sub>-storage, Tar, Organic pollutants, BTEX, PAHs, heterocyclic compounds, CO2SINUS, Tar pollutant production, Tar pollutant - water solubility, Thermodynamic model



## 1 Introduction

In view of the world's long-term power supply debate global coal deposits could serve as a future energy substitute as they still offer abundant and widely distributed reserves, alongside with better price conditions compared to oil and gas (RODDY & YOUNGER 2010; SINGH & SINGH 2012). Recent statistics by British Petrol (BP) indicate a world coal fossil reserve-to-production (R/P) ratio of 112 years, surpassing oil and natural gas R/P's to more than 50 years (BP 2012). Comparing the potential of overall global coal reserves in terms of mineable reserves, however modern coal technology challenges especially lie in the field of technical feasibility, economic-efficiency and environmental sustainability, the latter respectively associated with the need to implement successful carbon capture strategies (MIT 2007; HOPF et al. 2011). The coupled process of Underground coal gasification (UCG) with combined Carbon Capture and Storage (CCS) thus offers a positive potential to meet these challenges of the future energy markets (KEMPKA et al. 2009, 2011; NAKATEN et al. 2011). Accordingly the number of active UCG demonstration sites is currently increasing in many parts of the world. To date main UCG spots can be found in Australia, New Zealand, South Africa, China, USA, Eastern Europe, India Indonesia, Vietnam, Pakistan and the UK (UCG ASSOCIATION 2013).

### 1.1 UCG process and tar related organic groundwater pollution issues

The basic setting for UCG involves a setup of two boreholes, injection and production well, which are connected by different linking procedures in order to provide a coal-to-gas flow path connection (c. Fig. 1).

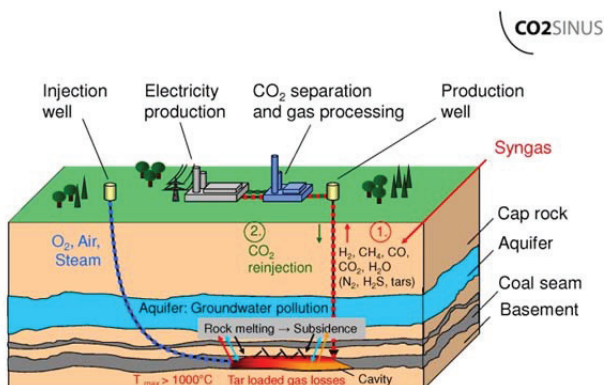


Abb. 1: Prinzip des UTV Prozesses mit angegliederter CO<sub>2</sub> Speicherung und potenziellen Umweltrisiken, geändert nach KLEBINGAT et al. (2009)

Fig. 1: UCG- (CCS) process principle and potential environmental hazards modified from KLEBINGAT et al. (2009)

The process starts with partial oxidation oriented substoichiometric input of oxidants in the injection well (oxygen/steam or air/water steam mixtures), which triggers a controlled self-ignition of the coal seam. With steadily increasing temperatures the coal is gradually dried out, pyrolyzed and finally converted to a medium-high calorific syngas which can be used for various industrial applications (e.g. electricity generation in the frame of the Integrated

Gasification Combined Cycle - IGCC Concept; or Coal-To-liquid - CTL procedures like Fischer-Tropsch synthesis). Apart from these promising technological perspectives to date, future UCG operations have to find new ways to overcome the gaps in process control and minimize the process-inherent environmental impacts of subsidence and groundwater pollution (COUCH 2009). With regard to the latter the understanding of tar production and related organic pollutants in UCG process gases and the nearby water cycle is a key issue for future UCG operations (FRIEDMAN et al. 2009). Several past UCG projects have thus been confronted with organic groundwater pollution issues in the form of enhanced BTEX, PAHs and heterocyclic compound concentrations due the condensation of tar loaded gas-losses (e.g. SURY et al. 2004; BURTON et al. 2006; SHAVIROVICH & VARMA 2009; YOUNGER 2011). More recently the actuality of this problem was demonstrated by the shutdown of the Australian UCG site Kingaroy, where enhanced benzene and toluene concentrations were detected in the local groundwater regime (LEWIS 2010; MC CARTHY 2012).

### 1.2 Selected UCG tar production and fate investigations within the CO<sub>2</sub>SINUS project

With focus on potential tar reducing strategies the present CO<sub>2</sub>SINUS study investigated the influence of selected single process parameters on tar related organic pollutant production by using a new self-developed thermodynamic model approach according to the principal chemical processes within a fixed bed reactor (c. Fig. 2).

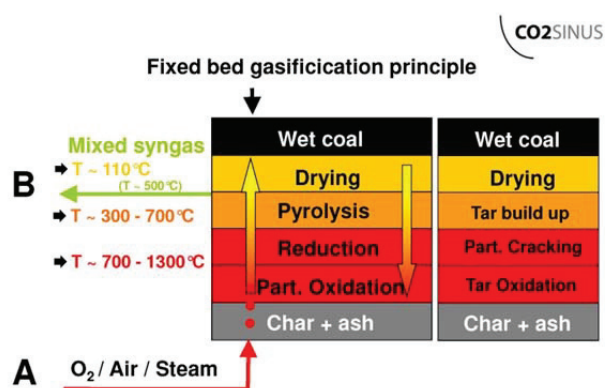


Abb. 2: Übersicht der wesentlichen chemischen UTV Prozesse und des hiermit assoziierten Teer-Verhaltens während der Festbettvergasung

Fig. 2: Main UCG chemical processes and related tar behaviour during fixed bed gasification

This conceptual UCG approach was chosen in agreement with similar younger UCG process descriptions by Min & Edgar (1987), who stated that fixed modeling can be a first step when evaluating basic controlling mechanisms. Furthermore selected tar pollutant-water solubility scenarios were investigated in a separate second model approach. The main focus of this second model approach was set on a basic prognosis for high p/T tar pollutant-water partitioning with regard to environmental hazard assessment for potential deep future European UGC sites.

## 2 Methodology

### 2.1 UCG - Tar pollutant production model development

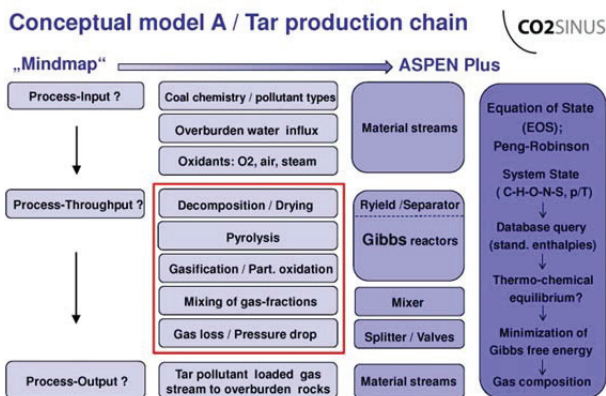


Abb. 3: Übersicht der konzeptionellen Teilschritte und Berechnungsgrundlagen im Rahmen der Entwicklung und Rechnung eines thermodynamischen UTV Modells zur Teer-Schadstoff Quantifizierung

Fig. 3: Overview of conceptual steps and mathematical foundations in the frame of thermodynamic UCG tar pollutant production model - development and - calculation

In view of the main tar controlling processes and flow regimes within fixed bed gasification (c. Fig 2) a thermo-chemical model of these processes for UCG tar production was set up using the software ASPEN PLUS (c. conceptual overview Fig. 3). In this context selected software units like material streams, Mixer- Splitter-, reactor- and valve types were chosen to represent the main tar altering processes (red box: respectively pyrolysis, reduction /partial oxidation and gas mixing to syngas) as well as significant other UCG principal boundary conditions (e.g. gasification agent flux/composition, pyrolysis temperature). The hierarchical order of these units was governed in a separate model flowsheet. Quantitative tar pollutant yield evaluation for the abovementioned main processes was carried out due to the principle of Gibbs free energy minimization by solving the Peng-Robinson Equation of State (PENG & ROBINSON 1976).

### 2.2 Tar pollutant – water solubility model development

Analogue to the procedure for the first model approach, a second model approach considering UCG tar pollutant – water solubility was developed under use of principal Aspen Plus process units (Flash reactor and material streams, c. Fig. 4). This approach was simplified under the assumption of an already cooled UCG gasifier, focusing on the post-phase liquid-liquid tar pollutant-water partitioning behavior.

In this context a main development step implied the regression of specific single pollutant KLL- Liquid-Liquid partitioning coefficients to enhance the quality of the tar pollutant –water partitioning results of the Aspen Plus Flash reactor unit. Referring to these regression results selected high pressure / temperature tar pollutant – water solubility prognoses were calculated under use of the Peng-Robinson Equation of State (PENG & ROBINSON 1976).

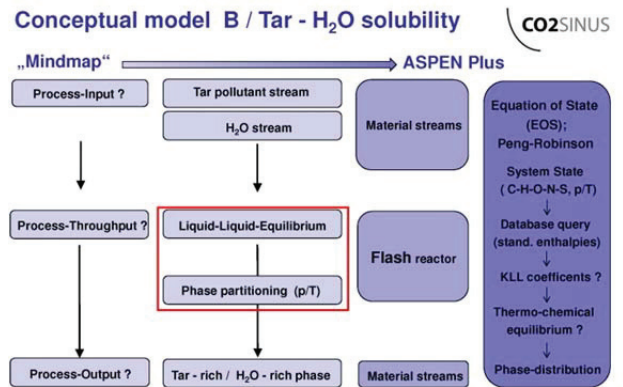


Abb. 4: Übersicht der konzeptionellen Teilschritte und Berechnungsgrundlagen im Rahmen der Entwicklung und Rechnung eines thermodynamischen UTV Modells zur Teer-Schadstoff Wasserlöslichkeit

Fig. 4: Overview of conceptual steps and mathematical foundations in the frame of thermodynamic UCG tar pollutant water model - development and - calculation

### 2.3 Selected model parameter studies

Based upon technical data reports from subbituminous and bituminous coals of the former US-UCG trials at Hanna-, Centralia and Pricetown (amongst others CAMPBELL et al. 1974; BARTKE & GUNN 1983; HILL et al. 1983, 1984; Moskowschuk 1997) more than 30 single scenarios were calculated within model A (c. 2.1) referring to the main influences of principal process parameters on total and related single tar pollutant yields in UCG gas fractions. An overview of the main investigated process parameters is provided in Fig. 5.

#### 6 key model parameter studies: Tar pollutant range in UCG gas fractions

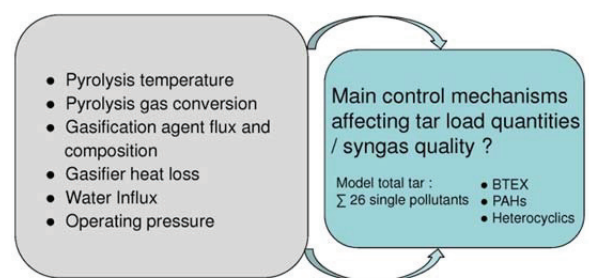
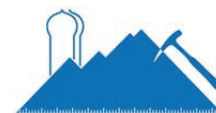


Abb. 5: Übersicht der untersuchten Hauptparameter im Rahmen der Szenarienrechnungen zum UTV Teer-Schadstoff Modell im Hinblick auf die Evaluierung zentraler Teer-Einflussmechanismen

Fig. 5: Overview of key model parameters investigated in UCG tar pollutant production model scenarios with focus on main tar control mechanisms

Each of these parameters was investigated with regard to its quantitative tar-load altering effect upon the main gas types of UCG (Pyrolysis-gas, Reduction/partial oxidation gas and mixed syngas fractions). Furthermore technical interrelations between different types of parameters were examined. Under consideration of model A results (c. 2.1) principal parameter studies for model B (2.2) focused the coupled effects of different p/T regimes on individual tar pollutant – water solubility systems (amongst others evaluation of the



tar pollutant – water solubility system Benzene-H<sub>2</sub>O, presented here). Basic scenarios for these systems included low, as well as high coal deposit-depth calculations, up to a maximum depth of 5 km against the background of future applications for European UCG sites.

### 3 Selected preliminary results

Validated against the syngas data of the former shallow US Hanna 1 trial using a subbituminous coal, model A - total tar yields and BTEX-PAH-heterocyclic group-representative single pollutant distributions are illustrated with regard to exemplary effects of pyrolysis temperature and gasification agent influx / composition (c. 3.1-3.2, Fig. 6-10). In focus of first model B results, an exemplary p/T water solubility prognosis for the water-benzene system is discussed in chapter 3.3

#### 3.1 UCG - Tar pollutant production model / Tar effects of pyrolysis temperature

In view of the principal UCG chemical sub-processes (c. fig 2) the main source of tar production and related organic pollutant production depends on the performance of the pyrolysis sub-process (Sury et al. 2004). Due to the natural boundary conditions as well as hard operating control (e.g. water influxes from overburden aquifers, gas losses) the temperature profile of the pyrolysis process varies during UCG operation, which in turn triggers compositional tar-load changes within the different gas fractions. In the present case scenario (300-700 °C) considering respectively low to intermediate range pyrolysis (Guntermann 1989; Vargas 2006) as expected from the theoretical background total tar-mole fractions were found to be highest in the UCG pyrolysis gas, lowest in the reduction/partial oxidation gas and intermediate in the mixed syngas (c. Fig. 6-8). As pyrolysis reflects idealized pure thermal coal decomposition without participation of oxygen, higher temperatures did not significantly reduce the total tar mole fraction in pyrolysis gas (c. Fig. 6). More than 99% of the model-tar constituents in the pyrolysis gas consisted of the second heaviest model-tar single pollutant Pyrene (C<sub>16</sub>H<sub>10</sub>), while the yields of the lighter tar fraction with exception of benzene increased with increasing temperatures according to the single tar pollutant boiling points (bp), reflecting the principals of a distillation process (Pyrene C<sub>16</sub>H<sub>10</sub> bp ~ 394°C > Naphthalene C<sub>10</sub>H<sub>8</sub>, bp ~ 394°C, > Phenol C<sub>6</sub>H<sub>6</sub>O bp ~ 182°C, > Pyridine C<sub>5</sub>H<sub>5</sub>N bp ~ 115°C). In the oxidizing environment of the reduction partial/oxidation gas (Fig. 7) developing higher temperatures overall caused total tar yields to decrease, obviously as a result of more tar cracking towards higher temperatures. According to the cracking process, destruction of heavier components caused an increase in the lighter single tar pollutant fraction, resulting in an opposite trend to the pyrolysis tar-distribution. Pyrene was completely converted in the reduction/partial oxidation gas. Tar yields and distribution in the mixed syngas (c. Fig. 8; present case assuming 47% of pyrolysis gas conversion due to end-process syngas data fitting) overall reflect the dominating effect of the high tar-loaded pyrolysis gas compared to the minimal tar-loads in the reduction/partial oxidation gas. In this context slightly decreasing tar mole fractions were observed within the

mixed syngas, while the compositional tar spectrum and single pollutant trends remained the same as observed in the pyrolysis gas.

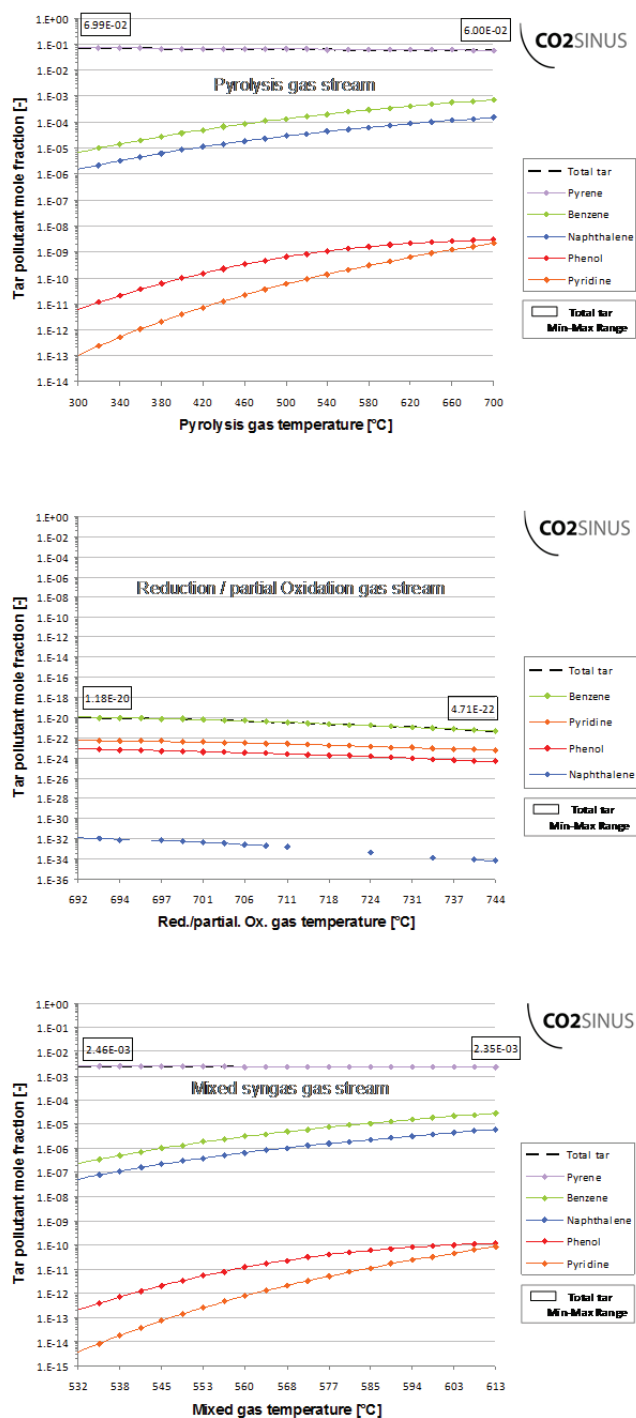


Abb. 6-8: Beispielhafte Gesamtter- und Einzelschadstoffproduktion in UTV-Prozessgasen bei Umsetzung einer Braunkohle mit geringem Schwelgrad als Funktion der Pyrolysetemperatur

Fig. 6-8: Exemplary total tar and single pollutant production in UCG process gases under conversion of a non-swelling subbituminous coal as a function of pyrolysis temperature

### 3.2 UCG - Tar pollutant production model /Tar effects of gasification agent flux and composition

To investigate principal tar-load effects of the parameters gasification flux and compositional changes a nominal air flux of 0.044 kmol/h (Bell et al. 1983 in ref. to Cena & Thorsness) was tested within an estimated range of +/- 20% to account for basic variability during Hanna 1 process operation. In a second step original pure air injection was replaced by different scenarios testing the effects of slightly enhanced oxygen contents within air with regard to the total and single pollutant tar-loads in principal gas fractions. With regard to the nominal gas flow of 0.044 kmol/h used for the Hanna 1 trial a decrease of the total tar loads and related single pollutants to zero in the reduction /partial oxidation gas was found for an oxygen-enriched air scenario of N<sub>2</sub> to O<sub>2</sub> ratio of 0.75 to 0.25 (c. Fig. 9,10). - As in the present basic model A-pyrolysis gas reactor an idealized non-disturbance by gasification agents was assumed with regard to the inert atmosphere classic definition of pyrolysis (Kabe et al. 2004) tar yields and related single pollutants in the pyrolysis gas were not affected. - In the mixed syngas fraction intermediate tar loads and related single pollutant distributions were found, basically reflecting the pyrolysis - reduction/partial oxidation gas mixing ratio. Overall tar yields and related single pollutant distributions in all three gas-fractions were found to behave analogue to the processes described under 3.1, respectively representing the fundamentals of a distillation process, sensitive to boiling points. The main effect of increased fluxes and compositional changes, whether air or oxygen-enriched air is used, was an observed increase in UCG system temperature due to higher oxygen fractions, reducing the total and single tar pollutant yields as a result of tar cracking.

### 3.3 Tar pollutant – water solubility model/ p/T dependent Benzene-water solubility

To date present simulations of selected case studies indicate average benzene production in model A may reach several grams per hour of UCG process operation. In case of undesired gas losses to the overburden cooling of UCG gases in higher strata levels may lead to condensation and tar-water partitioning of principal gaseous tar pollutants (Sury et al. 2004). In this context water solubility will be one of the decisive transfer mechanisms with regard to pollutant quantities that can enter the water-water cycle. Exemplary p/T dependent water solubility prognoses within model B simulations for the benzene-water system thus indicate temperature is the main controlling mechanism increasing water solubility, whereas pressure just has a subordinate effect. For maximum deep coal future scenarios up to 5 km an increase of average factor five to near surface benzene-water solubility is seen in present simulations (c. Fig. 11).

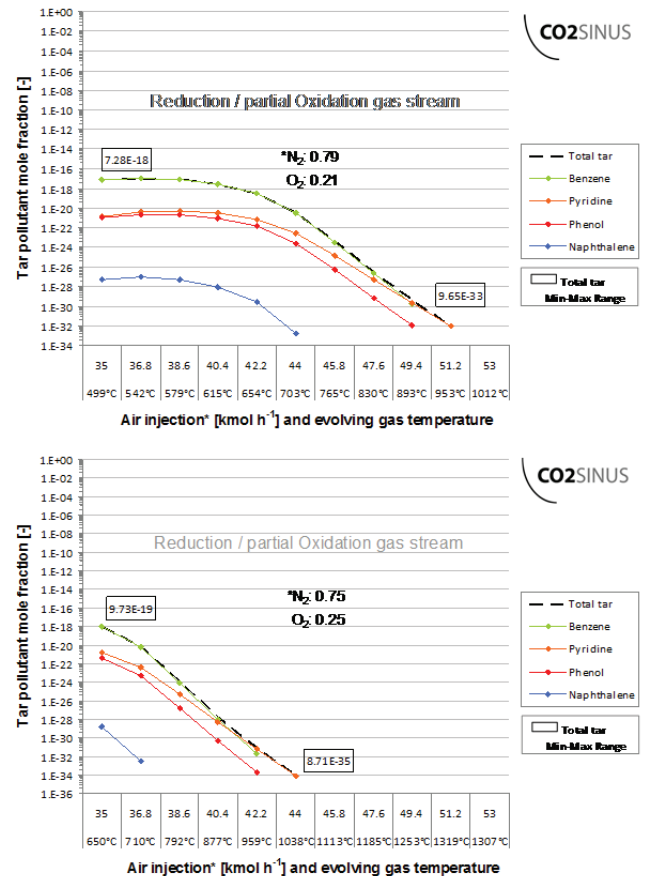


Abb. 9-10: Beispielhafte Gesamtter- und Einzelschadstoffproduktion in der UTV Gasfraktion des Reduktions-/partiellen Oxidationsgases bei Umsetzung einer Braunkohle mit geringem Schwelgrad als Funktion des Vergasungsmittelstroms unter Nutzung von reiner Luft (oben) und O<sub>2</sub> angereicherter Luft (unten)

Fig. 9-10: Exemplary total tar and single pollutant production in UCG reduction- /partial oxidation gas under conversion of a non-swelling subbituminous coal as a function of the gasification agent flux using pure air (top) and O<sub>2</sub> enriched air (bottom)

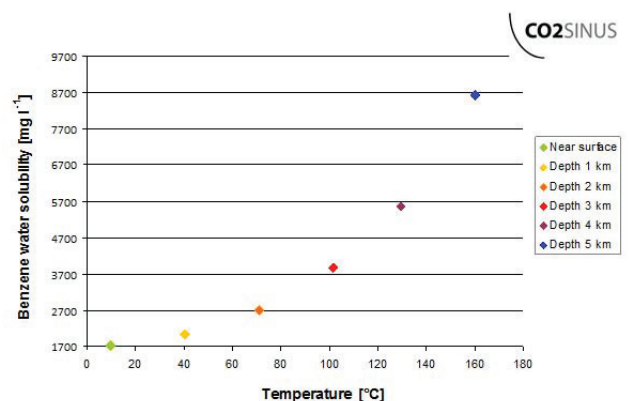


Abb. 11: p/T abhängige Benzol-Wasserlöslichkeitsprognose im Hinblick auf potenzielle europäischer UTV Kohleteufen bis 5km  
 Fig. 11: p/T dependent benzene-water solubility prognosis in view of potential european UCG coal depths up to 5km



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