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# Innovative thermodynamic underground coal gasification model for coupled synthesis gas quality and tar production analyses

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## KEYWORDS:

Underground Coal Gasification, Thermodynamic model, Synthesis gas quality, Tar production,  
Environmental impacts

## **ABSTRACT**

Underground Coal Gasification (UCG) technology is steadily improving due to high scientific and industrial efforts in currently over 14 countries worldwide. A fundamental UCG objective refers to syngas production for multiple end-uses, accompanied by environmental impact mitigation focusing contaminant reduction. In terms of this topic, the control of groundwater quality endangering tars has been a key problem rarely addressed in UCG publications so far. Considering UCG main sub-processes, operating parameters and tar spectrum knowledge grounded upon established thermodynamic equilibrium principles, an innovative and flexible model approach for coupled gas quality-tar production balances is presented here. The model is validated against literature data of the Hanna-I and Centralia-Partial Seam CRIP (PSC) field trials. For both trials good matching results were found. Main gas compounds and Lower Heating Values (LHVs) results are close to reported data partly reaching less than 10% deviation (relative error range for main compounds 4.32–18.6%, LHV 6.60–21.7%). Tar literature trend-modeling comparisons down to the single pollutant scale are addressed for the first time considering published data. Results here successfully reflect main qualitative tar tendencies, while current quantitative prognoses are on a satisfactory level, and expected to be further improved with the availability of more comprehensive in-situ data.

## **1. INTRODUCTION**

Underground Coal Gasification provides a promising technical and economic potential to solve current long-term power supply conflicts worldwide by controlled in-situ conversion of otherwise non-mineable coal reserves [1–7]. While the technique itself is a rather simple concept in theory, experience from major international field trials over the last decades shows its control

in practice is much more complicated. Various factors during the in-situ UCG reactor operation often hamper a continuously stable process performance (e.g. water ingress, heat losses), in consequence partially leading to lower synthesis gas qualities and undesired hazardous by-products [1,3,8–14]. Pollutant charged gas leakage from the UCG reactor due to roof rock failure as a thermo-mechanical response of in-situ pressure and temperature conditions recently marks one of the most severe potential environmental impacts [1,6,15–19]. With focus on the growing public debate on gas leakage affiliated with organic groundwater pollution risks as well as general tar plugging problems, a key future UCG by-product challenge thus lies in better tar production control and aligned organic pollutant minimization in UCG gases [20–23]. Principle UCG tar related organic pollutant groups, primarily consisting of BTEX, Phenols, PAHs and heterocyclic compounds have been well documented in diverse preceding publications [10,11,21,24–26]. Besides environmental motives, the tar control topic may also be deeper analyzed from an economic valuable tar perspective, regarding tar yield and spectrum optimization for marketable products [27]. Whether addressing environmental or economic targets, in terms of subsurface tar control factors and quantitative tar effects to date, overall few hard fact knowledge exists and rare literature data are available covering these issues. UCG chemical in-situ processes are of major relevance in terms of tar build up and transformation. In this context, previous studies indicate that UCG mixed synthesis gas tars can be interpreted as a reduction/partial oxidation induced lighter- and lower molecular weight/boiling point-fraction of heavier primary pyrolysis tars, whereby pyrolysis is consistently reported as main source process for tar build up. Tar spectrum and related quantities can depend on several factors, including gasification stage, inner-system variables such as coal type chemistry, in-situ gas temperature, as well as outer ones, e.g., choice of oxidant [10,26,28–30].

Due to complex reaction routes involved in tar chemistry and the numerous factors that contribute to the final UCG tar product, so far tars have seldom been explicitly focused in UCG modeling. Whenever considered in former compositional gas models, they were commonly treated as pseudo species [31–34]. In awareness of the recent debate on potential gas leakage and compound related water cycle restrictions, however a deeper tar modeling scale down to the specific single pollutant level becomes mandatory to deduce corresponding quantitative data on single compounds and optimization potentials regaining trust in UCG’s environmental performance.

## **2. Materials and methods**

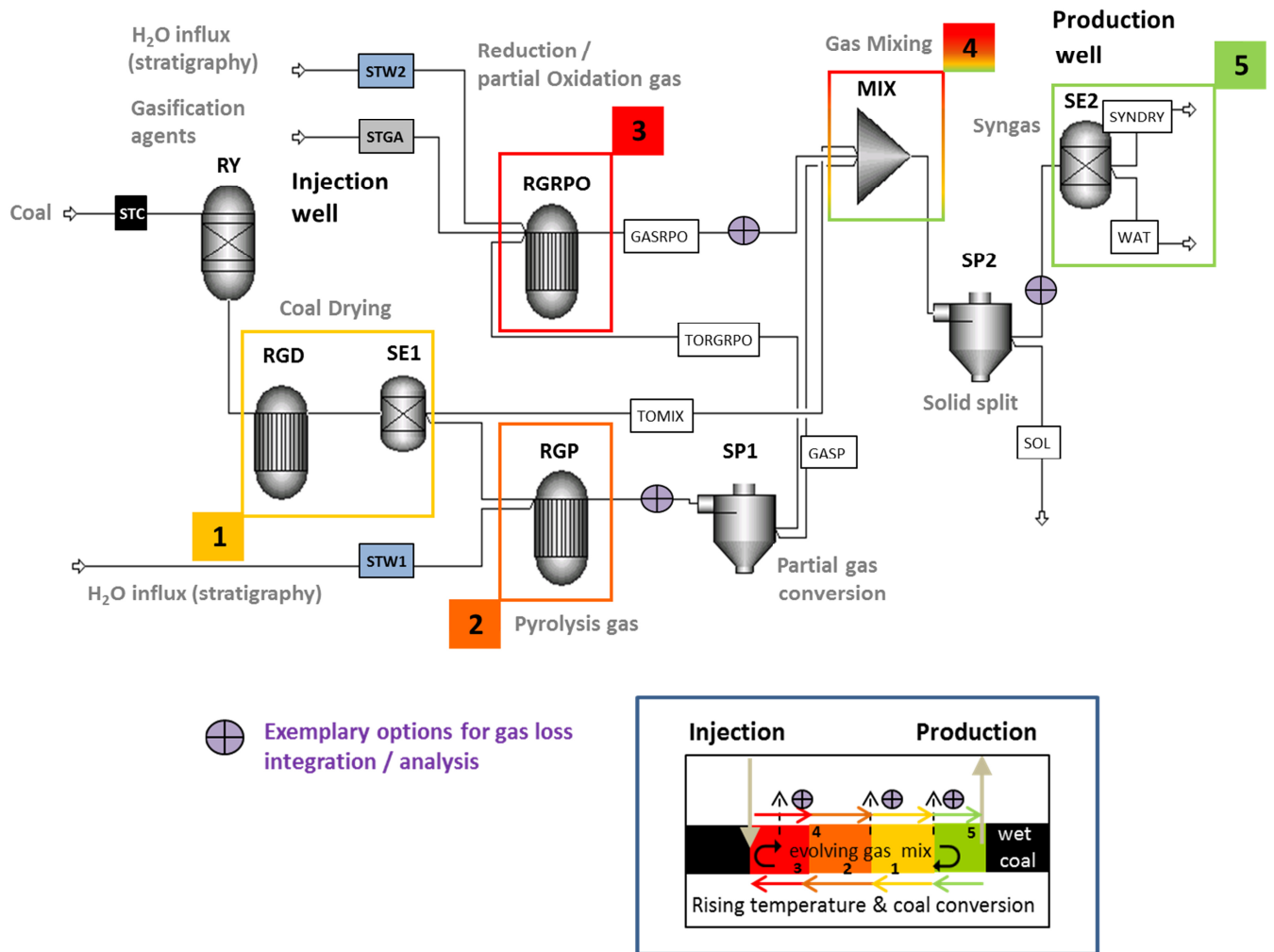
Using the established commercial simulator Aspen Plus®, we introduce a new UCG synthesis gas quality related tar model, which considers main chemical in-situ sub-processes of drying, pyrolysis, reduction/partial oxidation, related mixing of gas fractions, water influx, optional gas losses and respectively a selected model tar set including typical single pollutants [1,3,8,10, 24–25]. The model is based on chemical and phase equilibrium balances under minimization of Gibbs energy referring to the Peng Robinson Equation of state [35,36]. Equilibrium modeling has successfully been conducted within diverse previous UCG studies among other focusing basic test operating alternatives as well as economic analyses [37–39]. Considering these former modeling equilibrium approaches, we provide an innovative coupled main gas- and detailed tar compound accounting model. In this context we respectively chose the equilibrium balancing option faced to the fact that it marks an established approximation strategy, which is independent of reaction pathways [3,40], helping to overcome uncertainties and reduce complexity in terms of more than 500 potentially participating tar reaction routes [41]. While UCG may be performed with different technical setups [1,42] the focus here is on a general coal model establishment,

considering the previously named main chemical in-situ processes as key elements of all UCG schemes. The model can be applied for environmental risk assessments and critical in-situ data estimation down to the sub-process level as important support for future laboratory and field setups. It may also be used as valuable support tool for tar yield optimization and composition analyses regarding surface gas processing plant design. In more general terms, it allows deeper insight into in-situ out-of-sight processes and respectively preferred operating modes to achieve coupled clean as well as high quality synthesis gas production solutions.

Corresponding to the process model development, standard Aspen Plus® software modules were governed as depicted in Fig. 1. An overview of selected model components, including the principal tar pollutant set balanced within the model is outlined in Table 1. Relevant information on parameterization for specific modules is provided in Table 2 [43–58]. In view of essential model validation test data only few precise background data on former UCG field trials are available to the public. As a starting point, here we used data related to the early North American trials Hanna I and Centralia PSC [43–47,53–57].

In deeper view of the basic flowsheet build-up procedure, the integration of main sections (1-5) was oriented at the in-situ reactor zone distribution known from principal previous UCG field experiments and related excavation experiences that have been well documented in several preceding UCG publications [1,3,8,59,60]. The basic reactor distribution is thus characterized by development of an oxidant impacted high-temperature reduction/partial zone near the injection well (cf. Fig. 1, section 3), in direction of the production well gradually followed by a mid-temperature pyrolysis section (cf. Fig. 1, section 2) and a low temperature drying section (cf. Fig. 1, section 1). In consequence of gas mixing between the previous named zones (cf. Fig. 1, section 4), finally a mixed synthesis gas stream forms (cf. Fig. 1, section 5).

Referring to the detailed process modules individual coal type chemistry is first defined using a basic material stream (cf. Fig. 1, STC). The coal chemistry information is subsequently transferred to an elementary C-H-O-N-S yield distribution in a RYield reactor (module RY) with a calculator block added for dry to wet basis transformation. The C-H-O-N-S yield distribution is a general essential data form for RGibbs balance calculations that we tested here in combination with a moisture separator unit to model coal drying (low temperatures up to 200-300 °C [1,3,61]; cf. Fig. 1, Section 1 RGibbs-RGD reactor and SE1 unit). Coal inherent moisture set free during the drying stage is thus separated in the SE1 separator unit and later re-added to the mixing stage of the different gas fractions (cf. Fig. 1, stream TOMIX) to establish idealized dried coal conditions in the subsequent pyrolysis process stage (Section 2 RGibbs-RGP reactor). Pyrolysis was modeled using a medium reactor temperature of 500 °C [1,3,61]. From a deeper conceptual perspective, the model assumes that the related pyrolysis gas will be partly consumed by the high temperature reduction/partial oxidation gas (Section 3), finally forming a mixed synthesis gas (Sections 4–5) during the UCG process. The percentage of the initially formed pyrolysis gas consumed in the subsequent process step of reduction/partial oxidation (cf. Fig. 1, splitter unit SP1 to stream TORGRPO and RGibbs RGRPO reactor) is thus found iteratively by analyzing the best-match between mixed model- and literature-based synthesis gas compositions. With priority to matching final synthesis gas compositions, we also considered average water influx, integrated by exemplary total sum split into water influx streams STW1 and STW2.



**Fig. 1.** Basic UCG model flow sheet with focus on main in-situ process sections 1–5.

Note: RY = RYield reactor, coal chemistry yield transformation to elemental C-H-O-N-S distribution essential for Gibbs reactor balancing, RGD = Gibbs equilibrium reactor for drying section (1), product balancing by minimization of Gibbs energy, RGP = Gibbs equilibrium reactor for pyrolysis section (2) RGRPO = Gibbs equilibrium reactor for reduction/partial oxidation section (3), (balancing cf. RGD), MIX = mixer module, mixing of section 1–3 related gas types (4), SE1 = coal moisture separator unit, SE2 separator unit for dry syngas balancing (5). SP1 = pyrolysis gas splitter unit for partial pyrolysis gas conversion, SP2 = splitter unit for solid separation from gas phase via material stream SOL, GASP = material stream residual pyrolysis gas to mixing section, GASRPO = reduction/partial oxidation gas stream to mixing section, STC = material stream coal/coal chemistry definition, STGA = material stream for gasification agent definition, STW1/STW2 material streams for water influx definition to pyrolysis- and reduction/partial oxidation sections, SYNDRY = material stream dry syngas, TOMIX = material stream coal moisture re-added to mixing section, (balancing cf. RGD), TORGRPO = material stream partial pyrolysis gas consumed in reduction/partial oxidation gas section, WAT = material stream residual water.



**Table 1** Overview of selected model components including full model tar set (1).

Name	Formula	CAS-RN	Name	Formula	CAS-RN
Carbon monoxide	CO	630-08-0	ne_Propylbenzene	C <sub>9</sub> H <sub>12</sub>	103-65-1
Carbon dioxide	CO <sub>2</sub>	124-38-9	ne_1,3,5-TMB	C <sub>9</sub> H <sub>12</sub>	108-67-8
Methane	CH <sub>4</sub>	74-82-8	ne_Naphthalene	C <sub>10</sub> H <sub>8</sub>	91-20-3
Hydrogen	H <sub>2</sub>	1333-74-0	ne_1-Benzothiophene	C <sub>8</sub> H <sub>6</sub> S	95-15-8
Water	H <sub>2</sub> O	7732-18-5	ne_1-Methylnaphthalene	C <sub>11</sub> H <sub>10</sub>	90-12-0
Nitrogen	N <sub>2</sub>	7727-37-9	ne_Biphenyl	C <sub>12</sub> H <sub>10</sub>	92-52-4
ac_Phenol	C <sub>6</sub> H <sub>6</sub> O	108-95-2	ne_Acenaphthalene	C <sub>12</sub> H <sub>8</sub>	208-96-8
ac_o-Cresol	C <sub>7</sub> H <sub>8</sub> O	95-48-7	ne_Dibenzofuran	C <sub>12</sub> H <sub>8</sub> O	132-64-9
ac_p-Cresol	C <sub>7</sub> H <sub>8</sub> O	106-44-5	ne_Fluorene	C <sub>13</sub> H <sub>10</sub>	86-73-7
ac_m-Cresol	C <sub>7</sub> H <sub>8</sub> O	108-39-4	ne_Phenanthrene	C <sub>14</sub> H <sub>10</sub>	85-01-8
ac_Benzoic acid	C <sub>7</sub> H <sub>6</sub> O <sub>2</sub>	65-85-0	ne_Anthracene	C <sub>14</sub> H <sub>10</sub>	120-12-7
ne_Benzene	C <sub>6</sub> H <sub>6</sub>	71-43-2	ne_Pyrene	C <sub>16</sub> H <sub>10</sub>	129-00-0
ne_Toluene	C <sub>7</sub> H <sub>8</sub>	108-88-3	ne_Chrysene	C <sub>18</sub> H <sub>12</sub>	218-01-9
ne_Ethylbenzene	C <sub>8</sub> H <sub>10</sub>	100-41-4	ba_Pyridine	C <sub>5</sub> H <sub>5</sub> N	110-86-1
ne_o-Xylene	C <sub>8</sub> H <sub>10</sub>	95-47-6	ba_Quinoline	C <sub>9</sub> H <sub>7</sub> N	91-22-5
ne_Cumene	C <sub>9</sub> H <sub>12</sub>	98-82-8	ba_Carbazole	C <sub>12</sub> H <sub>9</sub> N	86-74-8

Note: (1) full model tar set consists of 26 single compounds related to chemical main groups  
ac = acidic; ne = neutral and ba = basic tar fraction.

We use this concept as a flexible option to account for principal water influx effects to different stages during UCG operation, and hereby experienced enhanced quality of the syngas prognoses.

Equilibrium yields of reduction/partial oxidation processes (Section 3) are calculated using the RGibbs reactor RGRPO. This reactor represents the high temperature UCG section commonly characterized by temperatures in excess of 700 °C to partly > 1100 °C [1,3,61]. The section is located close to the injection well, where direct reaction with gasification agents occurs (cf. Fig. 1, stream STGA). As mentioned above, we also integrated an option for water influx consideration into the model (Fig. 1, stream STW2). Contrary to the fixed p/T approximation of the RGibbs-drying and pyrolysis reactors (RGD & RGP), we generally chose the pressure/heat duty input option for the RGRPO reactor to allow a better temperature approximation in this section. In special focus of heat duty, this parameter in the RGRPO reactor is assumed to be zero as first choice due to limited in-situ data availability.

**Table 2a** Parameterization basic setup and coal chemistry.

Cases	Basic setup data [43–46]				Coal chemistry information / calculation (stream STC) [46–52]													
	Base Est. 1	Base Est. 2	Ambient pressure	Ambient temperature	Operating pressure	Proximate analysis db				Ultimate analysis db					Sulfunal analysis db <sup>e</sup>			Heat of <sup>a</sup> combustion
	CSD (m)	COD (kg/m <sup>3</sup> )	COS (MPa)	GwT (K)		FC (wt%)	VM (wt%)	ASH (wt%)	ASH (wt%)	C (wt%)	H (wt%)	N (wt%)	S (wt%)	O (wt%)	PY (wt%)	SU (wt%)	OR (wt%)	
Han-I	122	2650	3.17	287	0.48	37.67	36.07	26.26	26.26	54.81	4.45	1.43	0.75	12.3	0.325	0.1	0.325	23.2
C-PSC	63	2100	1.30	285	0.43	33.7	41	25.3	25.3	54	4	1	1	14.7 <sup>e</sup>	0.45	0.1	0.45	22.1

Note: Est. = estimate, CSD = coal seam depth (for COS & GwT approximation); COD = coal overburden density (for COS approximation); COS = approximate/coal overburden stress = (CSD\*COD\*9.81 m/s<sup>2</sup>)/10<sup>6</sup>; GwT = approximate depth related normal groundwater temperature assuming average geothermal gradient of 3°C/100 m and near surface standard 10° C; STC = stream coal; db= dry basis; FC = fixed carbon; VM = volatile matter; PY = pyritic; SU = sulfunal; OR = Organic; e = estimated.  
<sup>a</sup> Boie correlation – higher heating value dry basis (final conversion btu/lb to MJ/kg).

**Table 2b** Parameterization feed streams.

Cases	STC <sup>a</sup> [53,54]			STW1 <sup>b</sup> [44, 45,47,54]			STW2 <sup>b</sup> [44, 45,47,54]			STGA <sup>c</sup> [45,54–56]					
	P (MPa)	T (K)	F (t/day)	P (MPa)	T (K)	F (m <sup>3</sup> /hr)	P (MPa)	T (K)	F (m <sup>3</sup> /hr)	P (MPa)	T (K)	F (mol/s)	O <sub>2</sub> (mole fraction)	N <sub>2</sub>	H <sub>2</sub> O
Han-I	3.17	287	13.1	0.62	287	0.030	0.62	287	0.525	0.48	522 <sup>e</sup>	12.4	0.21	0.79	0
C-PSC	1.30	285	67.7	0.63	285	0.155	0.63	285	0.915	0.43	400 <sup>e</sup>	30	0.29	0.06	0.65

Note: Stream STC = stream coal; STW1/STW2 = water influx streams 1 and 2; STGA Gasification agent stream; P = pressure, T = temperature; F= average stream flow rate; e = estimated. <sup>a</sup> Ambient temperature and pressure orientation. <sup>b</sup> Total outer water influx rate estimate fitted to syngas. <sup>c</sup> P operating pressure orientation, T estimated values to evolving syngas.

**Table 2c** Parameterization coal conversion related modules.

Cases	RY <sup>a</sup>		RGD <sup>b</sup> [45,57,58]		RGP <sup>c</sup> [45, 46]		RGRPO <sup>c</sup> [45,46]		SE1 <sup>c</sup> [45,46]		SE2 <sup>c</sup> [45,46]		SP1 <sup>c</sup>		SP2 <sup>c</sup>		Mix <sup>d</sup> [45,46]	
	P (MPa)	T (K)	P (MPa)	T (K)	P (MPa)	T (K)	P (MPa)	Heat duty (Watt)	P (MPa)	H <sub>2</sub> O TOMIX (fraction)	P (MPa)	H <sub>2</sub> O WAT (fraction)	Fluid Split TORGRPO (fraction)	Fluid Split GASP (fraction)	Solid Split SOL (fraction)	P (MPa)	T-estimate (K)	
Han-I	3.17	287	0.62	434 <sup>e</sup>	0.48	773	0.48	0 <sup>e</sup>	0.48	1	0.48	1	0.4	0.6	1	0.48	873	
C-PSC	1.30	285	0.63	435 <sup>e</sup>	0.43	773	0.43	0 <sup>e</sup>	0.43	1	0.43	1	0.3	0.7	1	0.43	903	

Note: RY= RYield reactor; RGD = RGibbs reactor drying; RGP = RGibbs reactor pyrolysis; RGRPO = RGibbs reactor Reduction/partial Oxidation; SE1/SE2 = Separator units, SP1/SP2=Splitter units; P = pressure; T = temperature; F= average stream flow rate; e = estimated. <sup>a</sup> Ambient temperature and pressure orientation. <sup>b</sup> P hydrostatic head orientation. <sup>c</sup> Average operating pressure orientation; heat duty/temperatures and section specific fractions cf. text explanations. <sup>d</sup> Average of pyrolysis and reduction/partial oxidation gas temperature pre-tests.

Downstream mixing of coal moisture derived steam, residual pyrolysis-, reduction/partial oxidation gas (cf. Fig. 1, Section 4, streams TOMIX, GASP, GASRPO into module MIX) and accompanied synthesis gas evolution (Section 5) represent the last key UCG process parts considered in our model. Under field perspective, synthesis gas reaches its final composition at the production well, where it is produced to the surface for end use processing (cf. Fig. 1) [1,3,11]. Residual solid carbon remaining underground in real operations is split from the main gas stream by units SP2 and stream SOL before entering the synthesis gas section. With regard to literature gas comparisons on a dry basis, the unit SE2 is furthermore added, allowing for water splitting (cf. Fig. 1, stream WAT) and particularly balancing of final dry synthesis gas (stream SYNDRY).

In a deeper sense of all sections described above regarding further model development, Fig. 1 also illustrates exemplary options for gas loss integration in the model, e.g. by splitting the main gas streams, which may be of interest for real-site gas loss scenarios in terms of their different tar loads of section specific gases (cf. Fig. 1, purple circle symbol marker). Addressing the basic model cases focused in the present paper, however we first assumed an idealized closed UCG in-situ process without gas losses to analyze the principal maximum in-situ pollutant production under the expected geological settings as well as a safe and stable UCG operation [1,10,11].

### **3. RESULTS AND DISCUSSION**

An overview of the simulation results, including a comparison against literature data yielded under the prescribed conditions of Table 2 is outlined in Figs. 2–4. Figs. 2 and 3 compare the dry synthesis gas composition, its lower heating value, and total tar production regarding the main source process of pyrolysis and expected mixed synthesis gas yields [26,47,53–55]. In context of the syngas heating value analysis here, we used the standard internal Aspen Plus® software

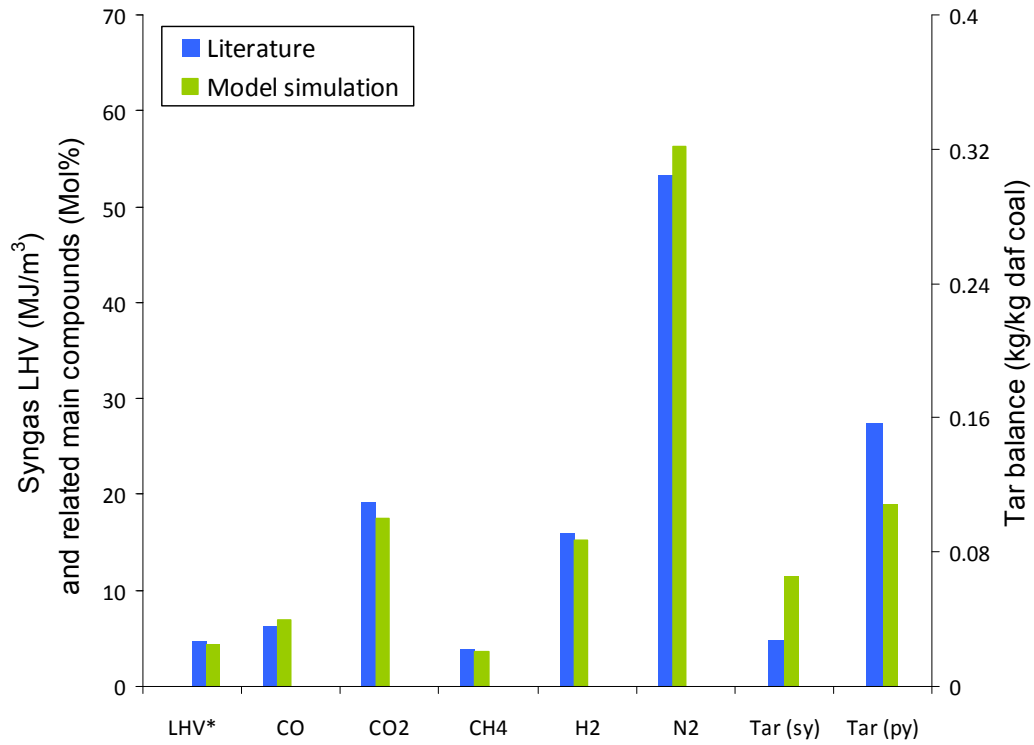
subroutine (Refsys correlation manager, LHV-0). With regard to deeper single tar pollutant production comparisons, only very scarce literature data is available by now. Due to this background, Fig. 4 outlines selected single tar pollutant product line data of the Centralia PSC trial [26], used here for a trend analysis of selected simulation results.

### **3. 1 Case study Hanna I vs. model simulation**

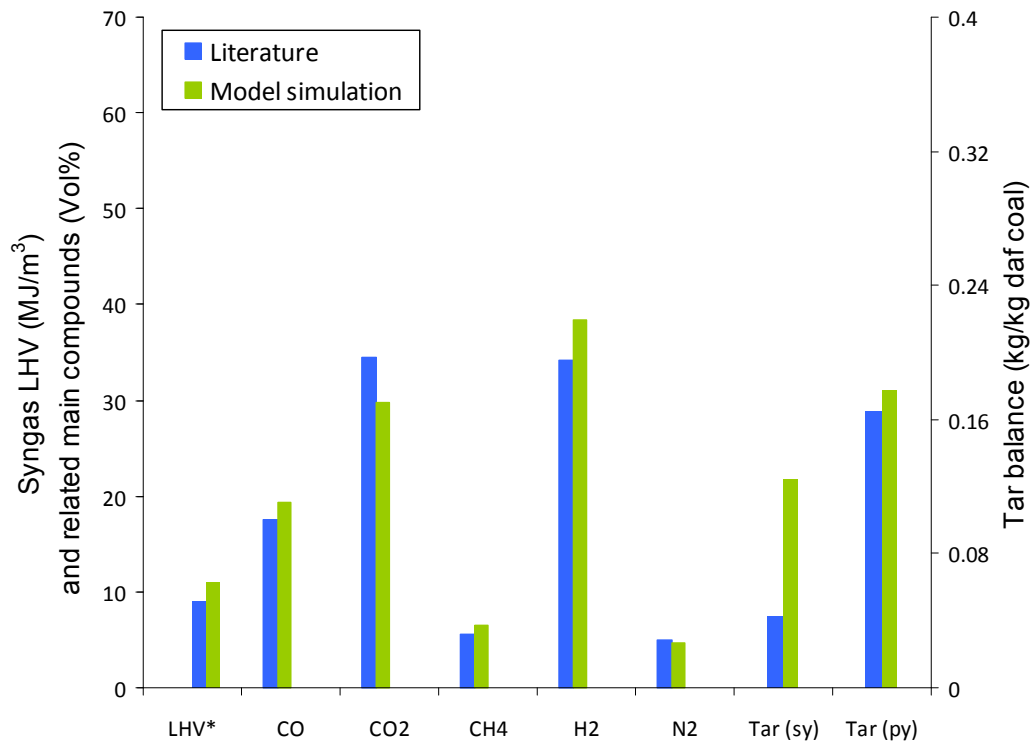
In terms of the Hanna I field trial literature synthesis gas composition (Fig. 2), the relative error range for the main gas single compounds ranges between 4.32 and 9.95% [53], overall indicating quite good agreement, keeping in mind that this is only a snapshot of a reported average gas composition. Alongside, the relative error for the synthesis gas LHV comparison was found to be relatively low (6.60%) [55], probably slightly overestimated as a result of the higher tar and H<sub>2</sub> production prognosis. Regarding the few available tar reference data available and discussed here, these data partly represent pyrolysis tests at laboratory conditions, and therefore should rather be seen as an average trend rather than representing in-situ data. Against this background, a direct percental error comparison is not drawn here. Overall pyrolysis model tar results were found to come quite close to the expected literature tar trend. The synthesis gas tar yield prognosis seems to be overestimated by the idealized model, however the simulation successfully confirms the Hanna I tar trend of lower synthesis gas tar yields, compared to pyrolysis as a matter of enhanced tar cracking [26]. In our model, the synthesis gas tar yield prognosis may still be improved by using more realistic input data for the pyrolysis gas conversion module SP1, which had to be estimated in lack of data. Based on more reliable data, future work should also consider tar condensation and gas residence time effects.

### 3.2 Case study Centralia PSC vs. model simulation

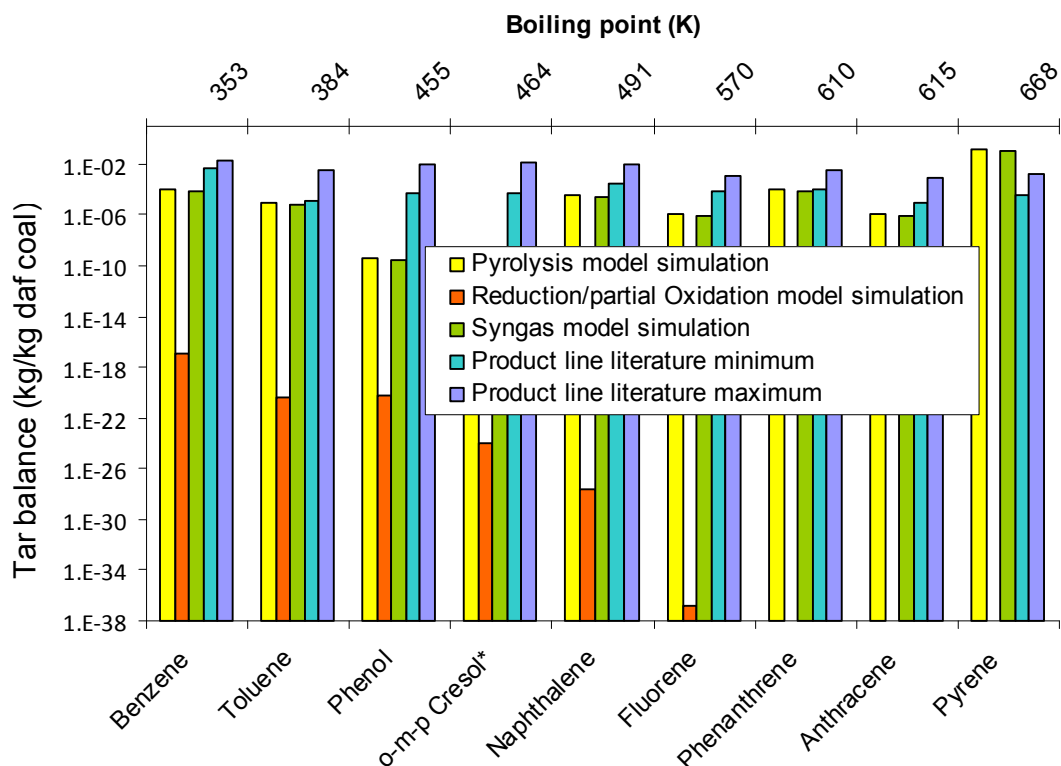
Regarding the Centralia PSC field trial literature syngas composition (Fig. 3), the relative error range for the main gas single compounds is between 5.88 and 18.6% [54]. In terms of the synthesis gas LHV comparison, slightly higher deviations are observed (relative error 21.7%) [47]. Compared to the Hanna I synthesis gas simulation, these enhanced deviations probably refer to the higher degree of uncertainty in terms of water influx background data, yet overall still mark a reasonable model prognosis. With regard to reported tar trends good qualitative simulation agreement was found [26]. As mentioned above synthesis gas tar prognoses may still be improved in future model revisions, especially by improving input data for the pyrolysis gas conversion module. The main literature tar trend with high tar loads from the pyrolysis source process and reduced yields in synthesis gas as a matter of cracking was again confirmed in the Centralia PSC model simulation. Focusing the rare available tar product line single pollutant literature data [26], our C-PSC case simulation results match in closest tendency with the reported minimum production of the depicted neutral compounds (Fig. 4). In this context, respectively the calculated lower boiling BTEX class compound toluene ( $C_7H_8$ ) and the higher boiling PAHs naphthalene ( $C_{10}H_8$ ) and phenanthrene ( $C_{14}H_{10}$ ) exhibit good agreement with literature data (Fig. 4). The overall literature trend reflects higher tar production rates of the lower boiling compounds, what is reasonable due to enhanced cracking at that stage. This trend is predominantly reproduced in the simulation with greater exceptions seen for phenol, cresol and pyrene. Deviation for the oxygen-associated phenol ( $C_6H_6O$ ) and cresol compounds ( $C_7H_8O$ ) thus likely refer to the abovementioned uncertainty in terms of water influx background data. The lower literature values for pyrene ( $C_{16}H_{10}$ ) actually indicate more cracking is taking place in-situ, than forecasted in the simulation. However, this issue can be improved by reverse modeling



**Fig. 2.** Case Han-I literature [26,47,54] vs. model simulation comparison. (Note: sy = syngas, py = pyrolysis gas, \*LHV = Lower heating value on dry basis, daf = dry and ash free basis.)



**Fig. 3.** Case C-PSC literature [26,53,55] vs. model simulation comparison. (Note: sy = syngas, py = pyrolysis gas, \*LHV = Lower heating value on dry basis, daf = dry and ash free basis.)



**Fig. 4.** C-PSC literature single tar pollutant balances [26] vs. model simulation comparison. (Note: daf = dry and ash free basis, missing Reduction/partial Oxidation column indicates zero production during simulation, \*sum of o-, m-, and p-Cresol.)

and specifically matching improved data for the SP1 pyrolysis gas conversion module in combination with water influx. A further reasonable mechanism which could explain the differences for pyrene in-situ data to model results is fractionation and related condensation of high boiling compounds in the UCG product line due to gas cooling before entering the tar sampling unit. Although a likely mechanism focusing recent UCG experience [10], deeper trial information was not found here, consequently not allowing further analysis of the condensation issue [26].

Overall, our simulations mark a reasonable UCG process representation in terms of reproduced highest tar loads in pyrolysis (source process), reduced tar yields in reduction/partial oxidation

(cracking of higher boiling compounds, dominance of smaller compounds) and finally evolving mixed synthesis gas tar balances (pyrolysis- and reduction/partial oxidation gas fractions) [10,26,30,62], in our case the latter here strongest reflecting pyrolysis.

### **3.3 Conclusions**

In terms of existing UCG models our new approach first time considers coupled balances for main gas compounds and detailed tar production down to the single pollutant level. The recently yielded simulation results thus mark an excellent starting point for further investigation on the UCG process environmental impacts. The model can be used for various tasks, e.g., as a reverse modeling tool to analyze specific reactor performance problems, for optimization of tar yields in view of surface gas processing plant operation as well as support for laboratory studies. As mentioned previously, various model extensions may be implemented for additional process integration, e.g., gas losses. Against this background, our current modeling studies focus best fit analyses to optimize gas qualities and simultaneously reduce tar yields under given geological boundary conditions.

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## REFERENCES

- [1] Couch GR. Underground coal gasification. London, UK: IEA Clean Coal Centre; 2009.
- [2] Roddy DJ, Younger PL. Underground coal gasification with CCS: a pathway to decarbonising industry. *Energy Environ Sci* 2010;3:400–7.  
<http://dx.doi.org/10.1039/b921197g>.
- [3] Bhutto AW, Bazmi AA, Zahedi G. Underground coal gasification: from fundamentals to applications. *Prog Energy Combust Sci* 2013;39:189–214.  
<http://dx.doi.org/10.1016/j.pecs.2012.09.004>.
- [4] Durucan S, Korre A, Shi J, Idiens M, Stan'czyk K, Kapusta K, et al. TOPS: technology options for coupled underground coal gasification and CO<sub>2</sub> capture and storage. *Energy Proc* 2014;63:5827–35. <http://dx.doi.org/10.1016/j.egypro.2014.11.616>.
- [5] Nakaten N, Schlüter R, Azzam R, Kempka T. Development of a techno-economic model for dynamic calculation of cost of electricity, energy demand and CO<sub>2</sub> emissions of an integrated UCG–CCS process. *Energy* 2014;66:779–90.  
<http://dx.doi.org/10.1016/j.energy.2014.01.014>.
- [6] Nakaten N, Azzam R, Kempka T. Sensitivity analysis on UCG–CCS economics. *Int J Greenhouse Gas Control* 2014;26:51–60. <http://dx.doi.org/10.1016/j.ijggc.2014.04.005>.
- [7] Nakaten N, Islam R, Kempka T. Underground coal gasification with extended CO<sub>2</sub> utilization – an economic and carbon neutral approach to tackle energy and fertilizer supply shortages in Bangladesh. *Energy Proc* 2014;63:8036–43.  
<http://dx.doi.org/10.1016/j.egypro.2014.11.840>.
- [8] Gregg DW, Edgar TF. Underground coal gasification. *AIChE J* 1978;24:753–81.  
<http://dx.doi.org/10.1002/aic.690240502>.
- [9] Krantz WB, Gunn RD, editors. Underground coal gasification – the state of the art. New York, USA: American Institute of Chemical Engineers; 1983.
- [10] Sury M, White M, Kirton J, Carr P, Woodbridge R, Mostade M, et al. Review of environmental issues of underground coal gasification; 2004. Report COAL R272 DTI/Pub URN 04/1880. Birmingham, UK: Department of Trade and Industry: WS Atkins Consultants Ltd.; 2004.
- [11] Burton E, Friedmann J, Upadheye R. Best practices in underground coal gasification; report draft contract No. W-7405-Eng-48. Livermore, California, USA: Lawrence Livermore National Laboratory; 2006.
- [12] Shafirovich E, Varma A. Underground coal gasification: a brief review of current status. *Ind Eng Chem Res* 2009;48:7865–75. <http://dx.doi.org/10.1021/ie801569r>.
- [13] Yang D, Sheng Y, Green M. UCG: where in the world? *TCE – Chem Eng* 2014;872:38–41.
- [14] Yang D, Koukouzas N, Green M, Sheng Y. Recent development on underground coal gasification and subsequent CO<sub>2</sub> storage. *J Energy Inst* 2015.  
<http://dx.doi.org/10.1016/j.joei.2015.05.004>.
- [15] Biezen ENJ. Modelling underground coal gasification Ph.D. Dissertation. Delft, Netherlands: University of Technology; 1996.

- [16] Wolf KH, Bruining H. Modelling the interaction between underground coal fires and their roof rocks. *Fuel* 2007;86:2761–77. <http://dx.doi.org/10.1016/j.fuel.2007.03.009>.
- [17] Buscheck TA, Hao YA, Morris JP, Burton E. Thermal-hydrological sensitivity analysis of underground coal gasification. In: Proceedings of the 26<sup>th</sup> annual international Pittsburgh coal conference, Pittsburgh, PA, October 23–26th 2009. p. 704–11.
- [18] Otto C, Kempka T. Thermo-mechanical simulations confirm: temperature-dependent mudrock properties are nice to have in far-field environmental assessments of underground coal gasification. *Energy Proc* 2015;76:582–91. <http://dx.doi.org/10.1016/j.egypro.2015.07.875>.
- [19] Otto C, Kempka T. Thermo-mechanical simulations of rock behavior in underground coal gasification show negligible impact of temperature-dependent parameters on permeability changes. *Energies* 2015;8:5800–27. <http://dx.doi.org/10.3390/en8065800>.
- [20] Friedmann SJ, Upadhye R, Kong FM. Prospects for underground coal gasification in carbon-constrained world. *Energy Proc* 2009;1:4551–7. <http://dx.doi.org/10.1016/j.egypro.2009.02.274>.
- [21] Kapusta K, Stan'czyk K. Pollution of water during underground coal gasification of hard coal and lignite. *Fuel* 2011;90:1927–34. <http://dx.doi.org/10.1016/j.fuel.2010.11.025>.
- [22] Edwards R. Fears for toxic leaks under firths as Australian gas extraction firm faces contamination charges. *The National*; 2015. August 17th 2015 Available from: <<http://www.thenational.scot/news/fears-for-toxic-leaks-under-firths-as-australian-gas-extraction-firm-faces-contamination-charges.6437>> [last accessed 05.05.2016].
- [23] Klebingat S, Azzam R, Schulten M, Kempka T, Schlüter R, Fernández-Steeger TM. Modeling optimized UCG gas qualities and related tar pollutant production under different field boundary conditions. In: Lollino G, Giordan D, Thuro K, Carranza-Torres C, Wu F, Marinos P, et al., editors. *Engineering geology for society and territory*, vol. 6. Cham: Springer International Publishing; 2015. p. 1063–6. [http://dx.doi.org/10.1007/978-3-319-09060-3\\_193](http://dx.doi.org/10.1007/978-3-319-09060-3_193).
- [24] Stuermer DH, Ng DJ, Morris CJ. Organic contaminants in groundwater near an underground coal gasification site in northeastern Wyoming. *Environ Sci Technol* 1982;16:582–7. <http://dx.doi.org/10.1021/es00103a009>
- [25] Verma RP, Mandal R, Chaulya SK, Singh PK, Singh AK. Contamination of groundwater due to underground coal gasification. *Int J Water Res Environ Eng* 2014;6:303–11. <http://dx.doi.org/10.5897/IJWREE2014.0520>.
- [26] Stephens DR, Thorsness CB. Partial seam CRIP test tar results. In: Proceedings of the 11<sup>th</sup> annual underground coal gasification symposium, DOE/METC-85/6028, Denver, Colorado, August 11–14. p. 179–99.
- [27] Simbeck DR, Dickenson RL, Moll AJ. By-product considerations in underground coal gasification. In: Proceedings of the 7th underground coal conversion symposium, DOE/LLNL-81/CONF-810923, Fallen Leaf Lake, California, September 8–11. p. 572–9.
- [28] Barbour FA, Cummings RE. Comparison of coal tars generated by pyrolysis of Hanna coal and UCG Hanna IVB coal tars; report cooperative agreement No. DE-FC21-83FE60177, DOE/FE/60177-2427. Laramie, Wyoming, USA: U.S. Department of Energy, Western Research Institute; 1986.
- [29] Barbour FA, Covell JR. Rocky mountain 1 underground coal gasification test Hanna, wyoming – trace gas, product water and particulate characterisation for rocky mountain 1 UCG Project; Report WRI-89-R018. Laramie, Wyoming, USA: Western Research Institute; 1989.

- [30] Kapusta K, Stan'czyk K, Mocek P. Evolution of tar compounds in raw gas from a pilot-scale underground coal gasification (UCG) trial. In: Proceedings of the 7th international Freiberg conference on IGCC & XtL technologies, Huhhot, June 9th, 2015. Available from: <[http://tu-freiberg.de/sites/default/files/media/professur-fuer-energieverfahrenstechnik-und-thermische-rueckstands-behandlung-16460/publikationen/2015\\_20-3.pdf](http://tu-freiberg.de/sites/default/files/media/professur-fuer-energieverfahrenstechnik-und-thermische-rueckstands-behandlung-16460/publikationen/2015_20-3.pdf)> [last accessed 05.05.2016].
- [31] Thorsness C, Rosza R. Lawrence livermore laboratory in situ coal gasification program: model calculations and laboratory experiments. In: Proceedings of the 51th annual fall meeting of the society of petroleum engineers, New Orleans, LA, USA, 3rd October 1976; SPE 6182. New York, USA: American Institute of Mining, Metallurgical, and Petroleum: Engineers; 1976.
- [32] Perkins G, Sahajwalla V. A mathematical model for the chemical reaction of a semi-infinite block of coal in underground coal gasification. *Energy Fuels* 2005;19:1679–92. <http://dx.doi.org/10.1021/ef0496808>.
- [33] Perkins G, Sahajwalla V. Steady-state model for estimating gas production from underground coal gasification. *Energy Fuels* 2008;22:3902–14. <http://dx.doi.org/10.1021/ef8001444>.
- [34] Khan M, Mmbaga J, Shirazi A, Trivedi J, Liu Q, Gupta R. Modelling underground coal gasification—a review. *Energies* 2015;8:12603–68. <http://dx.doi.org/10.3390/en8112331>.
- [35] Andrianopoulos E, Korre A, Durucan S. Chemical process modelling of underground coal gasification and evaluation of produced gas quality for end use. *Energy Proc* 2015;76:444–53. <http://dx.doi.org/10.1016/j.egypro.2015.07.870>.
- [36] Peng D, Robinson DB. A new two-constant equation of state. *Ind Eng Chem Fund* 1976;15:59–64. <http://dx.doi.org/10.1021/i160057a011>.
- [37] Cena RJ. A simple UCG field performance and economic model. In: Proceedings of the 13<sup>th</sup> annual underground coal conversion symposium, DOE/METC/-88/6095 (CONF-8708106), Laramie, Wyoming, August 24–27. p. 391–403.
- [38] Dufaux A, Gaveau B, Létolle R, Mostade M, Noël M, Pirard PJ. Modelling of the underground coal gasification process at Thulin on the basis of thermodynamic equilibria and isotopic measurements. *Fuel* 1990;69:624–32. [http://dx.doi.org/10.1016/0016-2361\(90\)90149-K](http://dx.doi.org/10.1016/0016-2361(90)90149-K).
- [39] Kerinin EV, Shifrin EI. Mathematical model of coal combustion and gasification in a passage of an underground gas generator. *Combust Explos Shock Waves* 1993;29(2):148–54. <http://dx.doi.org/10.1007/BF00755871>.
- [40] Atkins PW, de Paula J. *Physikalische chemie*. 4th ed. Weinheim, Germany: Wiley-VCH; 2006.
- [41] Wang H, Frenklach M. A detailed kinetic modeling study of aromatics formation in laminar premixed acetylene and ethylene flames. *Combust Flame* 1997;110:173–221. [http://dx.doi.org/10.1016/S0010-2180\(97\)00068-0](http://dx.doi.org/10.1016/S0010-2180(97)00068-0).
- [42] Self SJ, Reddy BV, Rosen MA. Review of underground coal gasification technologies and carbon capture. *Int J Energy Environ Eng* 2012;3:1–8. <<http://link.springer.com/article/10.1186/2251-6832-3-16/fulltext.html>> [last accessed 23.06.2016].
- [43] Youngberg AD, Santoro RD. Hydrogeologic evaluation of the Hanna coal gasification site. In: Proceedings of the 7th underground coal conversion symposium, DOE/LLNL-81/CONF-810923, Fallen Leaf Lake, California, September 8–11. p. 659–74.

- [44] Spratt B, Katz LJ. Post-burn high resolution seismic evaluation of Centralia, Washington underground coal gasification Site; Report to US Department of Energy Contract Number DE-AC20-83LC11009. Utah, USA: Utah Geophysical Inc.; 1984.
- [45] Britten JA, Thorsness CA. Mechanistic model for axisymmetric cavity growth during underground coal gasification. *Am Chem Soc Fuel Preprints* 1988;33:126–37.
- [46] Campbell GG, Brandenburg CF, Boyd RM. Preliminary evaluation of underground coal gasification at Hanna, Wyoming. Technical Progress Report 82. Pennsylvania, USA: US Department of the Interior, Bureau of Mines Coal Gasification Program; 1974.
- [47] Moskowtschuk W. Mechanismen der in-situ vergasung unter einbeziehen ukrainischer kohlen. Labor- und feldversuche Ph.D. Dissertation. Aachen, Germany: Rheinisch-Westfaelische Technische Hochschule – RWTH Aachen University; 1997.
- [48] SGS. Société Générale de Surveillance – Coal Calculations. <http://www.sgs.com/Mining/Analytical-Services/Coal-and-Coke/Coal-Calculations.aspx> [last accessed 05.05.2016].
- [49] Hamersma JW, Kraft ML. Coal Sulfur measurements – interagency energy/environment R & D program Report; Report EPA-600/7-79-150. NC, USA: United States Environmental Protection Agency, Industrial Environmental Research Laboratory; 1979.
- [50] Liu G, Peng Z, Yang P, Wang G. Sulfur in coal and its environmental impact from Yanzhou mining district, China. *Chin J Geochem* 2001;20:273–81. <http://dx.doi.org/10.1007/BF03166149>.
- [51] Mason DM, Gandhi K. Formulas for calculating the heating values of coal and coal char: development, tests and uses. *Am Chem Soc Fuel Preprints* 1980;25:235–45.
- [52] Thompson A, Taylor BN. Guide for the use of the international system of units (SI) – NIST special publication 811 2008 edition, appendix B9 factors for units listed by kind of quantity or field of science/heat. Gaithersburg, USA: National Institute of Standards & Technology; 2008.
- [53] Schriber LA, Jennings JW, Brandenburg CF, Fischer DF. An underground coal gasification experiment, Hanna, Wyoming. In: Proceedings of the 49th annual fall meeting SPE AIME, Houston, Texas, October 6–9. Society of Petroleum Engineers Preprints: SPE No. 4993; 1974. p. 25.
- [54] Hill RW, Thorsness CB, Cena RJ, Stephens DR. Results of the Centralia underground coal gasification field test. In: Proceedings of the 10th annual underground coal gasification symposium, DOE/METC-85/5, Williamsburg, VA, August 12–15 1984. p. 11–26.
- [55] Bartke TC, Gunn RD. The Hanna Wyoming underground coal gasification field test series. In: Krantz W, Gunn R, editors. *Underground coal gasification – the state of the art*. New York, USA: American Institute of Chemical Engineers; 1983. p. 4–14.
- [56] Bell GJ, Brandenburg CF, Bailey DW. Arco’s research and development efforts in underground coal gasification. In: Krantz W, Gunn R, editors. *Underground coal gasification – the state of the art*. New York, USA: American Institute of Chemical Engineers; 1983. p. 44–56.
- [57] Brandenburg CF, Fisher DD, Campbell GG, Boyd RM, Eastlack JK. The underground gasification of a subbituminous coal. *Am Chem Soc Fuel Preprints* 1975;20:3–10.
- [58] VDI. Verein Deutscher Ingenieure – VDI-Wärmeatlas: [Berechnungsunterlagen für Druckverlust, Wärme- und Stoffübertragung], 10, bearb. und erw. Aufl. Berlin, Heidelberg: Springer; 2006.
- [59] Gunn RD, Krantz WB. Underground coal gasification: development of theory, laboratory experimentation, interpretation, & correlation with the hanna field tests, Final Report No.

- DOE/LC/10442-2545. Morgantown, West Virginia, USA: U. S. Department of Energy; 1987.
- [60] Cena RJ, Britten JA, Thorsness CB. Excavation of the partial seam crip underground coal gasification test-site. In: Proceedings of the 13th annual underground coal conversion symposium, DOE/METC/-88/6095 (CONF-8708106), Laramie, Wyoming, August 24-27. p. 382-90.
- [61] Guntermann K. Experimentelle und theoretische Untersuchungen zur Untertagevergasung. Habilitationsschrift. Aachen, Germany: Rheinisch-Westfaelische Technische Hochschule – RWTH Aachen University; 1989.
- [62] King SB, Guffey FD, Gardner K. An investigation of aromatic fractions from coal tar produced by an underground coal gasification test. Am Chem Soc Fuel Preprints 1977;20:113-7.