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ANALYSIS OF GREENHOUSE GAS EMISSIONS FROM ESTONIAN OIL SHALE BASED ENERGY PRODUCTION PROCESSES. LIFE CYCLE ENERGY ANALYSIS PERSPECTIVE

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Abstract. *The objective of the paper is to define CO_2 intensity for Estonian oil shale used for energy generation. Life cycle analysis (LCA) is selected as a modelling tool to reach the objective. The model analysis energy requirements and CO_2 emissions are associated with mining and transportation of shale rocks and production of shale oil. Since the oil shale retorting process works as a 100% self-compensation energy technology (heat and gas derived from retorting of oil shale are reused within the on-going and next retorting), the paper lays emphasis on analysis of the co-products life cycle in elimination of CO_2 emissions. Preliminary results of the research show that life cycle CO_2 emission factors of oil shale processing mainly depend on the technology used for production of final raw products as shale oil, semi-coke gas and generator gas. Also, boundaries of the researched system must be taken into account due to avoided emissions from co-products production instead of using universal CO_2 emission factors and assumptions. Future work will be focused on 1) material based life cycle analysis to investigate whether the energy produced from oil shale resources can compete with other fuels in terms of life cycle emissions, and 2) LCA impact assessment of the system.*

Keywords: *life cycle analysis, CO_2 emission factor, oil shale, retorting, solid heat carrier, gaseous heat carrier.*

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1. Introduction

Oil shale can be defined as a sedimentary rock which in its mineral matrix contains a solid, combustible organic matter commonly called “kerogen”. It is widely distributed around the world – some 600 deposits are known, with resources of the associated shale oil totalling almost 410 billion tonnes, or approximately 3.0 trillion barrels [1].

Kerogen is a kind of organic high molecular polymer, insoluble in common organic matter. Oil shale can be burned in boilers as fuel or used as a raw material in shale oil production. It has to be noted that the production of oil from oil shale requires chemical destruction of shale organic matter (kerogen). When oil shale is heated in absence of air or oxygen to 400–500 ^{o}C, known as retorting of oil shale, the kerogen is pyrolyzed to produce shale oil, gaseous product, solid residue and a small quantity of decomposition water. The inevitability of the chemical decomposition (i.e. pyrolysis) of oil shale organic matter in the production of shale oil differentiates oil shale from conventional crude oil sources. Oil shale is also distinct from oil sands. Oil sands are composed of sand, sandstone, limestone or other sedimentary rocks, with the surface covered by heavy oil; the heavy oil can be attracted and separated by hot alkaline solution and is soluble in organic solvent (unlike the organic polymer kerogen) [2]. The atomic ratio of hydrogen to carbon in oil shale is about 1.2 to 1.8 times lower than in crude oil [3].

Due to the decrease of world oil resources and intensive use of oil and oil products, the demand for additional alternative hydrocarbon sources will be increasing in the future. There is already a lot of experience in oil production from oil shale [4–9]. Today, considerable numbers of oil shale thermal processing factories are operating in Estonia, Russia, China and Brazil. Estonia’s oil shale industry is currently the most developed in the world, although China has the highest production volumes of shale oil.

Due to climate and environmental considerations, a set of European Union regulations [10, 11, 12], also known as the EU climate and energy package, focuses on the reduction of GHG emissions of liquid transportation fuels during their life cycle. Since in Europe shale oil is produced in Estonia only, there is scarce data on shale oil production technology and environmental performance for setting justified environmental limitations on or benchmarks to oil shale based production.

The overall objective of this paper is to analyse GHG emissions generated while producing shale oil in two technological processes – gaseous heat carrier and solid heat carrier, and to define the modelled system’s emission factors of these technologies per tonne of shale oil produced.

2. General characterisation of oil shale and shale oil

Estonian oil shale dry matter consists of three components: organic, carbonate and sandy clay. The chemical and mineral composition of all these separate components is relatively constant, being independent of deposit location. However, the proportions of different components in the composition of oil shale as received fuel depend upon mining technology, enrichment rate, and other factors [4, 5]. Based on monitoring data, compositional differences of oil shale used for production of shale oil in Estonian shale oil processing plants (in 1990–2010) are given in Table 1.

Table 1. Characteristics of oil shale used for shale oil production in 1990–2010

Component	Technology applied	
	Solid heat carrier	Gaseous heat carrier
Moisture of oil shale as received, %	11.43–11.72	14.04–14.46
Amount of oil shale ash as received, %	44.93–45.68	39.73–40.49
Content of oil shale mineral CO_2 as received, %	17.37–17.96	10.55–11.27
Sandy-clay part in oil shale as received, %	23.79–23.82	25.88–26.89
Organic part of oil shale as received, %	26.47–27.52	35.18–37.15
Carbon content of oil shale as received, %	19.31–20.27	23.00–27.84

The composition of oil shale is experimentally determined by the contents of moisture, ash and mineral (carbonate) carbon dioxide (CO_2) in the sample (proximate analysis). The main components in the carbonate part of oil shale are limestone and dolostone. In the sandy-clay part silica-, aluminum- and potassium-containing minerals dominate. The portion of free quartz in oil shale mineral matter is about 40 m-%. Marcasite is also included in the composition of the sandy-clay part.

Thus, the average characteristics of Estonian [3] oil shale are as follows:

- lower calorific value Q^r_L = 1,992.2 kcal/kg (8.3 MJ/kg);
- moisture content W^r = 10.7%;
- carbonate content $(CO_2)^r_M$ = 18.9%;
- ash content A^r = 45.3%.

On the basis of the results of technical analysis of oil shale [4, 5], and depending on its calorific value, the following mathematical formulas for determining the contents of ash, carbonate and humidity of oil shale are deduced:

$$A^d = 0.052 \times (Q^d_L)^2 - 2.3049 \times Q^d_L + 68.929 \tag{1}$$

$$(\mathrm{CO}_2)^d_c = -0{,}112 \times (Q^d_L)^2 + 1.0723 \times Q^d_L + 20.323 \tag{2}$$

$$W^r = 0.6695 \times Q^d_L + 5.9141\,, \tag{3}$$

where A^d is the ash content of dry oil shale, %; Q_L^d is the lower calorific value of dry oil shale, MJ/kg; $(CO_2)_c^d$ is the carbon content in dry oil shale, %; and W^r is the moisture content of dry oil shale, %. The relationship between the amounts of laboratory ash, mineral CO_2 and moisture, and the lower calorific value (LHV) of oil shale is studied and the results are presented in Figure 1.

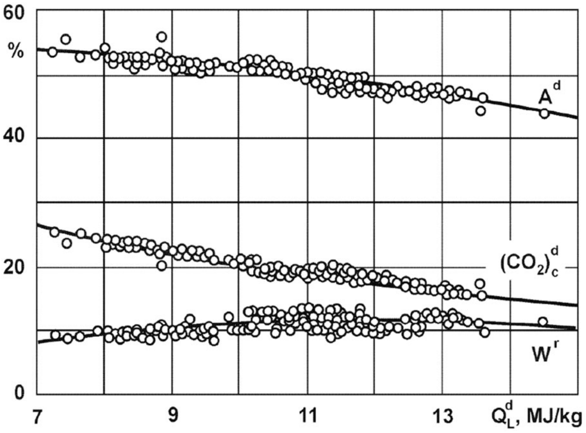

Fig. 1. Relationship between the lower calorific value and ash, mineral carbon dioxide and moisture contents of oil shale [4, 5].

As a result of long-term studies [13], a correlation between the laboratory oil yield T (m-%) (Fischer Assay) and the calorific value of dry oil shale Q_L^d (MJ/kg) has been found and is presented in Formula 4:

$$T = 1.78 \times Q_L^d. \tag{4}$$

3. Methodology of the research

The current research represents an energy based analysis, but inventory of the materials used in construction and decommissioning of the mining, retorting, condensation and distillation plants is not taken into account.

So far, the use of the LCA method for analysis of GHGs from the Estonian oil shale energy sector has been described in a joint project of Eesti Energia and the Finnish Environment Institute SYKE for the investigation of the life cycle of oil shale-based electricity production [14]. In his publications Brandt [15, 16] describes GHG emissions from producing liquid fuels from Green River oil shale with the Alberta Taciuk Process (ATP), and includes analysis of emissions from thermal retorting, fuel combustion, upgrading and refining and other processes. In their study, Aarna and Lauringson [17] present the LCA model for the "Enefit280" technology with a focus on co-products market and CO_2 credits.

An algorithm worked out in this study for definition of GHG emissions from Estonian oil shale-based energy production processes is given in Figure 2.

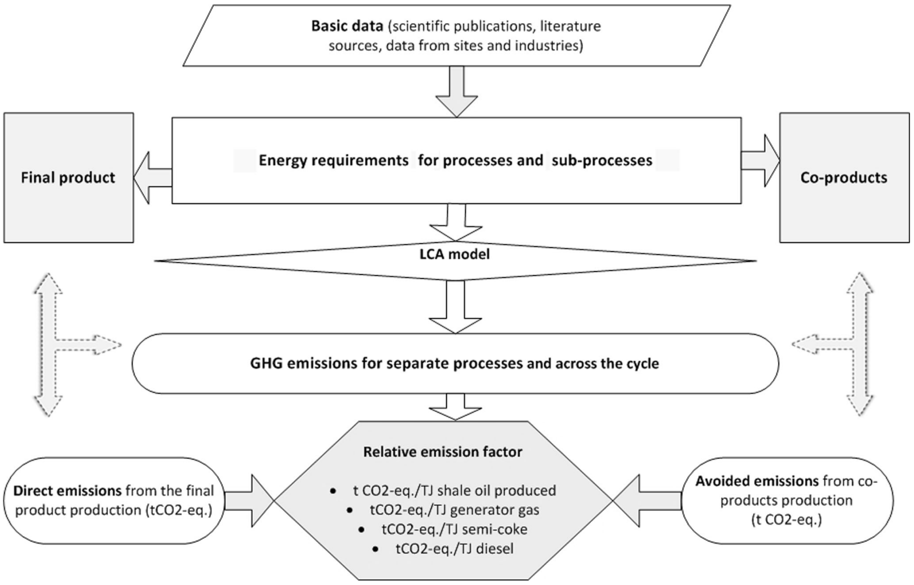

Fig. 2. Algorithm for definition of GHG emissions from Estonian oil shale-based energy production processes.

The net emission factor (e_{net} in tonne CO_2-eq./TJ final product) is used to demonstrate the full cycle efficiency of the modelled system (Fig. 3), taking into account emissions generated from production of final products (here shale oil, generator gas, semi-coke and diesel), called in this paper direct emissions (E_{direct} in tonnes CO_2), and co-products (products generated besides the final products), called in this work avoided emissions ($E_{avoided}$ in tonnes CO_2).

The amount of GHG emissions (CO_2, SOx, CO, CH_4, NOx, etc.) are recalculated to CO_2 equivalents (CO_2-eq.). The final product emission factor across the modelled life cycle is calculated according to Formula 5:

$$e_{net} = \frac{\Sigma E_{direct} - \Sigma E_{avoided}}{S} = \frac{(E_m + E_r + E_{cd} + E_{ref} + E_u + E_{bp})}{S}, \qquad (5)$$

where S is the shale oil produced, TJ; E_m, E_r, E_{cd}, E_{ref}, E_u and E_{bp} are respectively emissions generated during mining, retorting, condensation/distillation, refining, upgrading and co-products production processes.

Analysis and also calculation of CO_2 emission factors are performed for two oil shale retorting technologies – gaseous heat carrier and solid heat carrier.

3.1. System boundary, functional unit and assumptions

From a methodological point of view the model consists of two parts (see Fig. 3):

- direct LCA system is based on monitoring and calculated data collected from Estonian shale oil processing plants [13, 14] and employs an exclusive approach – emissions generated from production of co-products are excluded from total emissions;
- indirect LCA system is based on the results (indicators) obtained in other studies [14–17], which are redirected to serve as a continuation of the direct LCA model.

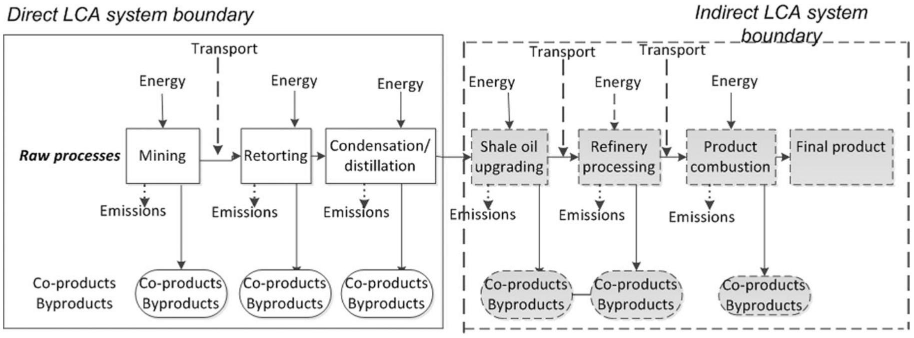

Fig. 3. Model system boundary.

The functional unit of the analysed system is the amount of emissions (in tonnes CO_2-eq.) generated per 1 TJ of shale oil produced from mining to retorting.

The time frame of the model is 2011: in the research, the corresponding data for 2011 from shale oil and oil shale production plants is used. However, few data coming from life-cycle databases (for example, energy consumption in mining process and transportation of oil shale) apply to the period 2006–2010.

A scheme of the direct LCA system boundary of the researched model as well as the carbon intensity values calculated for shale oil include the processing and transport steps "from cradle to gate": from mining to condensation and distillation processes (direct LCA) and then from upgrading to diesel production (indirect LCA).

Despite the findings of previous studies [15, 16, 17], the full cycle LCA approach needs to take into account that production of crude shale oil gives a wide range of high value chemicals as well as some unique fuel oils, and thus the generated emissions are eliminated from related industries. Additionally, it has to be acknowledged that depending on the retorting technology used, the production of crude shale oil affords a significant amount of a co-product – waste gas. The waste gas is used as fuel for producing electricity and heat in cogeneration plants (CHP) related to the shale oil industry. The produced heat and electricity fully cover the self-consumption needs of shale oil production and the rest is sold out (shale oil production results in the net export of heat and electricity). Depending on the technology, the remaining physical heat of the waste gas and ash from the pyrolysis process can be used for producing steam in utilisation boilers and heat exchangers; this steam can be used for producing electricity. The waste ash from shale oil production contains large amounts of clinker minerals. While research of using the ash as a clinker additive in cement production is on-going, the construction materials industry already uses ash as raw material. Thus, the assessment of LCA for shale oil production and production of motor fuels from it needs to use the LCA to co-products displacement approach.

The presented LCA model is based on the following assumptions/ limitations:

- processes related to the maintenance of the mining site after the extraction process are not taken into account;
- any processes related to abnormal working conditions (for example, interruption of the mining process due to weather conditions) are not taken into account;
- transportation distance from the mining site to the retorting site is shorter than 20 km;
- condensation and distillation processes take place at the retorting plant, thus the transportation distance is 0 km;
- quality of co-products satisfies the average quality requirements of Estonian oil shale and shale oil industries;
- quality of shale rocks does not change during the transportation process.

3.2. Inventory analysis

The inventory analysis of life cycle processes has been carried out using a mathematical model developed in VBA. Long-term monitoring data collected from Eesti Energia Oil Industry and VKG Oil AS was used to calculate fuel and energy inflows and outflows. Ecoinvent database data is used for collection of missing data (mainly on transport and mining).

Below each of the above steps of producing motor fuel from shale oil will be described and the GHG footprint values of those steps given; they are significantly different from the production of motor fuel from conventional crude oil.

3.2.1. Mining and transport of shale rock to site

Oil shale as raw material is extracted from open casts and underground mines. The same mines also supply power plants running on oil shale with the rock. Inventory data on the mining process and transportation of shale rock is given in Table 2.

Table 2. Inventory data on shale rock mining process and transportation

	Process description or assumptions	
	Gaseous heat carrier	Solid heat carrier
Energy requirements	Electricity consumption for extraction process[a] – 2.3 kWh/t of shale rocks Emission factor of electricity[b] – 1.04 t CO_2/MWh_e	
Quality of shale rocks[c]		
Lower heating value of oil shale as received	11.35 MJ/kg	8.17 MJ/kg
Moisture of oil shale as received	13.52%	11.38%
Amount of oil shale ash as received	41.52%	46.56%
Content of oil shale mineral CO_2 as received	13.68%	18.61%
Sandy-clay part in oil shale as received	24.88%	23.91%
Organic part of oil shale as received	32.84%	24.98%
Carbon content of oil shale as received	24.81 %	18.40%
Transportation of rocks to retorting plant		
Distance[c]	< 25 km	12.5 km
Type of transportation[c]	railway (one way-full, back-empty)	conveyor belt (one way-full, back-empty)
Load[d]	100%	100%
Emission factor for transportation[c]	1.7 kg CO_2/t shale oil	1.56 kg CO_2/t shale oil

[a, c] – Calculated based on monitoring data given in Table 1; [b] – calculated based on Ecoinvent data base; [d] – assumption.

Oil shale transport for the retorting industry in VKG Oil AS is provided by railway and a 12.5 km belt conveyor was installed in 2011. Thus, the LCA of CO_2 from oil shale mining and transport results in different values depending on the location of the retorting plant.

Avoided emissions at this stage may come from the use of mining waste in road construction – today up to 25% of the waste is utilised in this industry [14]. However, the use of mining waste and its later positive environmental effect on total process emission factor were not taken into account in calculations.

3.2.2. Retorting, condensation and distillation

The thermal processing of oil shale to shale oil has quite a long history and various facilities and technologies have been used. In principle, there are two ways of thermal processing:

- low-temperature processing by heating oil shale up to about 500 °C – semi-coking or retorting;
- high-temperature processing by heating oil shale up to 1000–1200 °C – coking.

The coking of oil shale for production of shale gas was used in Estonia in the 1950s–1960s. Nowadays, the oil shale industry employs only low-temperature processing for shale gas production.

In Estonia, only ex-situ technologies are used for shale oil production employing internal heating retorts with solid or gaseous heat carrier. Depending on technology, the retorting process (pyrolysis) takes place without the use of external fuels.

According to heat consumption pyrolysis of oil shale can be divided into the following stages:

1. drying the source oil shale, i.e. a drying stage;
2. heating the dry oil shale to the process temperature;
3. thermal decomposition of oil shale, i.e. the pyrolysis process;
4. vaporising the oils resulting from the pyrolysis process;
5. decomposition of carbonates.

The heat necessary for the process is gained in the equipment from either the retort gas (waste gas in case of gaseous heat carrier technology) or solid residue (in case of solid heat carrier technology). In Estonia, both technologies are applied to shale oil production [3–8].

3.2.3. Gaseous heat carrier (GHC)

Gaseous heat carrier (Kiviter technology) retorts require oil shale feed as a raw material with a particle size of 25–125 mm and a minimal heating value of 10 MJ/kg. Oil shale with such specifications constitutes only part (maximum about 40%) of the oil shale resource extracted. The enrichment and fractionation of total extracted material is justified by the relatively high oil yield of approximately 17% for Kiviter technology retorts. The heat for the process is gained in the equipment from retort gas. The gaseous heat carrier (formed by recycled retort gas combustion) has a temperature of 750–950 °C. Besides crude shale oil, the co- and byproducts of the Kiviter process are retort gas and phenolic water. The retort gas co-product yield per ton of processed oil shale is about 400–500 $\mathrm{Nm^3/t}$, while the retort gas has a minimal lower calorific value of about 2.1 $\mathrm{MJ/Nm^3}$. The retort gas is used as a waste gas fuel in combined heat and power plants (CHP). Phenolic water, the byproduct of the Kiviter process, is used as a raw material for refined chemicals production. The gaseous heat carrier process as system boundary is given in Figure 4, the energy requirements for retorting are summarised in Table 3.

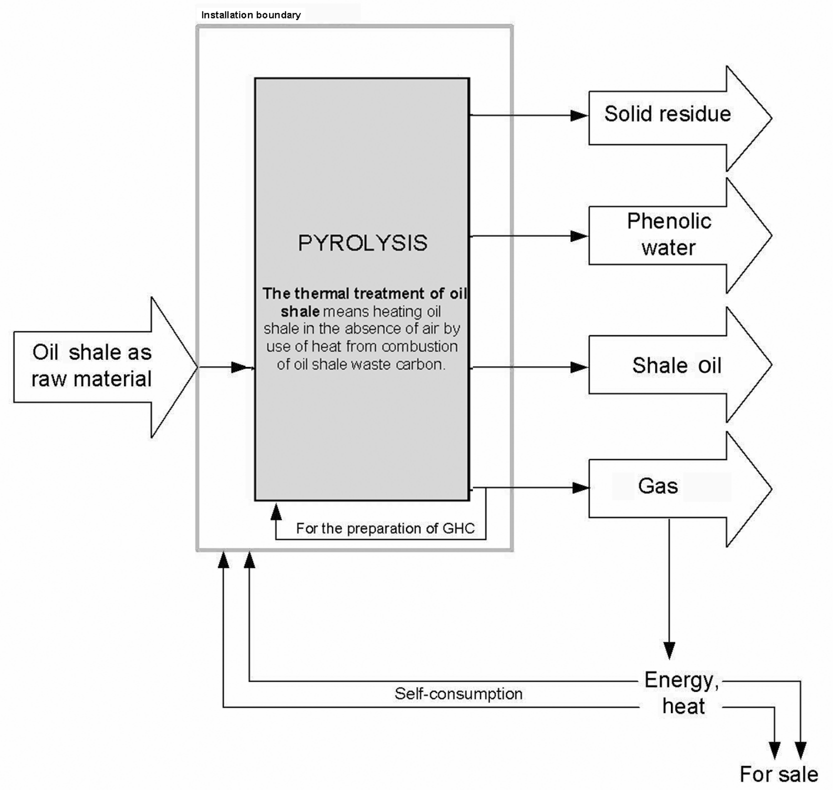

Fig. 4. Gaseous heat carrier pyrolysis process for shale oil production (solid residues and phenolic water are not included in the LCA study).

3.2.4. Solid heat carrier (SHC)

Solid heat carrier retorts process oil shale feed with a particle size of 0–25 mm and a calorific value of about 8.5 MJ/kg. In case the extracted and enriched shale oil is not sufficiently fractionated, the raw oil shale has to be crushed to guarantee the required specification of raw material for the retorts. Due to the lower calorific value of oil shale used in SHC technology retorts, oil yield (ca 12.5%) is lower than in Kiviter retorts. The heat necessary for the process is gained in the equipment from waste carbon in the solid residue. The solid heat carrier is recycled back to the retorts at a temperature of approximately 800 °C and after mixing it with raw material, the pyrolysis process takes place in the reactor at about 500 °C. The relatively small fraction of oil shale used in the process enables an efficient transfer of heat from the heat carrier to raw oil shale, thus the residence time of oil shale in the pyrolysis process is shorter. Besides crude shale oil as a co-product, the SHC retorting process also affords retort gas as a co-product and phenolic water as a byproduct. The retort gas yield per ton of processed oil shale is about 35 Nm^3/t, its lower calorific value is around 35 MJ/Nm^3. The retort gas is used as a waste gas fuel in CHP. Phenolic water, the byproduct of the SHC process, can also be used as a raw material for refined chemicals production. The SHC process as system boundary is given in Figure 5.

In most advanced SHC retorts (Petroter technology), heat recovery also generates steam and heat as byproducts suitable for district heating. A new Enefit-280 SHC shale oil factory (under development) will also produce electricity as a byproduct.

Calculated inventory data on energy requirements for the SHC and GHC retorting processes is given in Table 3. The table presents no data on energy self-consumption of both technologies since it is fully compensated for by the co-products of the retorting system (as heat and gas), which in this paper are assumed to be avoided emissions.

As seen from Table 3, the energy requirements for both technologies are the same. However, calculations show that in case of the SHC technology, an additional amount of CO_2 is produced during decomposition of carbonates, which accounts for 17% of total CO_2 emissions from the retorting stage.

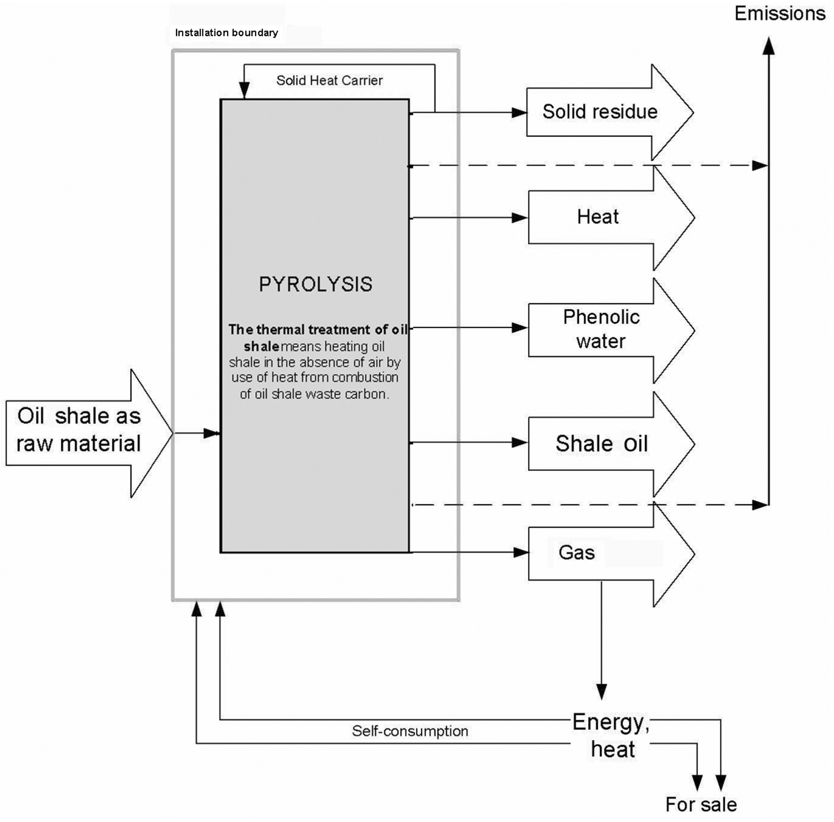

Fig. 5. Solid heat carrier process for shale oil production (solid residues and phenolic water are not included in the LCA study).

Table 3. Inventory data of the retorting process

Stage of the retorting process	Energy required, MJ/t of shale oil
1. Drying the source oil shale, i.e. a drying stage, heating water vapours to the process temperature	5,513.6
2. Heating dry oil shale to the process temperature	4,346.2
3. Thermal decomposition of oil shale, i.e. the pyrolysis process	850.3
4. Vaporising the oils resulting from the pyrolysis process	334.9

4. Results and discussions

Table 4 presents life cycle CO_2 emission factors (LCEF) per final products (in this paper, shale oil and generator gas) for the modelled system's life cycle stages.

Table 4. Life cycle CO_2 emission factors of the modelled system

System stage	Gaseous heat carrier (Kiviter)		Solid heat carrier (Petroter)	
	LCEF, t CO_2/TJ shale oil produced	LCEF, t CO_2/TJ generator gas produced	LCEF, t CO_2/TJ shale oil produced	LCEF, t CO_2/TJ semi-coke gas produced
Mining	0.35	1.96	0.51	2.02
Transportation	0.25	1.39	0.33	1.32
Retorting, condensation and distillation	10.8	19.1	10.7	26.9

The calculated results of the energy LCA show that there are differences in LCEF for different retorting technologies and different final products: production of shale oil in the Kiviter technology generated almost the same amount of CO_2 emissions as in the Petroter technology. However, in the production of generator gas less CO_2 emissions are released than in the semi-coke gas production. Thus, the total summarized results (mining, transportation, retorting, condensation and distillation) for the combined relocated final product for the GHC and SHC are 30.95 t CO_2/TJ final product and 38.72 t CO_2/TJ final product. At the same time, as expected, the higher LCEF are related to the retorting, condensation and distillation stages – 96.61% in GHC and 97.13% in SHC.

Previous studies [15, 16, 17] report different emission indicators for the mining, transportation, retorting, condensation and distillation processes (so-called direct LCA system). The authors of this paper are of the opinion that the results of the referred studies are not comparable with their findings for the following reasons:

- lack of description of the methodology, boundaries and assumptions of Brandt's and Aarna's models;
- use of different inventory data and different case studies;
- allocation of the results to different final product/products;
- neglect of avoided emissions from co-products production;
- mixing of results of material- and energy-based LCAs.

To evaluate the final processing of shale oil products, an additional "indirect LCA system" (see Fig. 3) was developed. Estonian shale oil companies have an option to upgrade shale oil into final products by conventional refining. Due to its high specific gravity and high organic oxygen content, raw shale oil can be upgraded using refinery type hydrotreaters and catalyst. About 41–44 kg of hydrogen shale oil is needed for upgrading of 1 tonne of shale oil. It is known that during the process the weight of the end product (diesel) reduces by approximately 7% but the heating value increases by about 12% compared to the respective values of the input shale oil. Lack of actual upgrader refinery data makes it impossible to perform an LCA for the GHC for shale oil. For a preliminary estimation of the LCA of the shale oil upgrading process data on hydrogen production via natural gas steam reforming and crude oil refineries [17, 18] would be useful. These studies estimate life cycle emissions from shale oil upgrading to be 18.2 g CO_2/MJ diesel and from final refining, 5.1 g CO_2/MJ diesel.

5. Conclusions

1. The first inclusive LCA analysis case study for the Estonian oil shale processing is developed and a concept for calculation of avoided emissions from the process worked out and approbated with real processing plants data for two oil shale processing technologies – gaseous heat carrier (Kiviter technology) and solid heat carrier (Petroter technology).
2. The results of the LCA show that life cycle CO_2 emissions from shale oil processing are sensitive to the retorting technology used, the system's boundary and generated co-products. Kiviter technology gives a lower CO_2 emission factor, but the content of carbon in the landfilled ash will stay higher. Petroter technology gives a higher LCA emission factor (in tonnes CO_2-eq.), but provides a possibility of using the high calorific value gas as a fuel for upgrader.
3. Unlike previous studies this paper is based on actual data of shale oil plants and takes into account the self-consumption of heat and power, as well as uses the co-products displacement method for elimination of total CO2 emissions.
4. There is a great potential to reduce greenhouse gases generated in the shale oil processing through use of the waste ash in the cement

production. This concept needs to be evaluated and proved by the LCA results in the future.

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