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CARBON DIOXIDE BINDING IN THE HETEROGENEOUS SYSTEMS FORMED BY COMBUSTION OF OIL SHALE

1. CARBON DIOXIDE BINDING AT OIL SHALE ASH DEPOSITS

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About 95 % of electricity production in the Republic of Estonia is based on the firing of pulverised local solid fuel – oil shale. This process is accompanied by high CO_2 emission and followed by the formation of waste alkali ash in huge quantities. Ash is removed by hydrotransport and deposited in wet dumps.

A complex of laboratory and field experiments with different ashes and their transport water samples from operating power plant (1610 MW) was carried out. Low activity and low carbonation rate of dry ash were determined as well as a much higher activity of previously hydrated ash, which in favourable conditions could be completely carbonated. An up to 50–60 % degree of carbonation of transport waters by binding CO_2 from the air in the production cycle was established.

The total amount of CO_2 bound from the air and calculated basing on the results of laboratory model experiments reached 37 kg per 1 ton of oil shale burnt in the electricity production. The relative part of bound CO_2 formed 20.5 % and 4.2 % from the carbonate and total emission of CO_2, respectively. The recommendations to increase the amount of CO_2 bound at natural field conditions have been worked out. By implementing them, the amount of bound CO_2 could be essentially increased – up to 35–36 and 6–7 %, respectively.

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Introduction

According to the Kyoto Climate Conference Protocol adopted in 1997 it is necessary to reduce, by the year 2008–2012, the amount of greenhouse gases emitted into the atmosphere in Estonia by 8 % as compared to the level of 1990 [1] when emission of CO_2 was 25.2 ton per person in Estonia. This index was approximately 6 times higher than the world average [2], being 1.5–2 times higher as compared to our nearer neighbours in the Baltic Sea region [3] (Table 1).

Table 1. **Comparison of Estonia and its Neighbouring Countries in Production of Primary Energy and Emission of CO_2**

Indices	Estonia		Latvia	Lithuania	Finland	Sweden
	1990	1995	1995			
Primary energy:						
Total, PJ/year	416.6	225	208	417	936	1499
Per person, GJ/year	265	150	80	113	182	170
Emission of CO_2 in energy production:						
Total, Mt/year	39.6	21	14	19	51	54
Per unit of energy, Mt/PJ	0.095	0.092	0.069	0.046	0.054	0.036
Per person, t/year	25.2	13.5	5.3	5.2	9.9	6.1
Per km^2, t/year	875	464	216	223	151	120

The main part of total CO_2 emitted in Estonia results from electricity production, which is mainly based on the combustion of a local fuel – Estonian oil shale. The carbon emission factor (CEF) of oil shale is 29.1 t C/TJ calculated according to Equation (1) [4, 5], and that of coal and natural gas is 25.8 and 15.2 t C/TJ, respectively. The CEF for Estonian oil shale is higher due to its higher content of mineral CO_2 as compared to other fuels.

$$\mathrm{CEF}_{oil\ shale} = 10 \times [C_t^r + K_{\mathrm{CO_2}} \times (\mathrm{CO_2})^r_M \times 12/44]/Q_i^r \ (\mathrm{t\ C/TJ}) \qquad (1)$$

where C_t^r is the content of carbon in oil shale, %;

$(CO_2)^r_M$ is the content of mineral carbon dioxide in oil shale, %;

K_{CO_2} is the decomposition degree of the carbonate part of oil shale by pulverized firing of it ($K_{CO_2} = 0.95–1.0$);

Q_i^r is net calorific value for oil shale, MJ/kg.

It follows from this equation that on the average 18–20 % of CO_2 formed by combustion of oil shale is derived from its mineral part, mainly from carbonates. The decomposition products of carbonates are free Ca and Mg oxides, their content in ashes reaches up to 30 % [6, 7]. In certain conditions they could bind acidic gases (SO_2, CO_2, etc.) [8, 9]. So, it is assumed that oil shale ash formed in huge amounts (42–48 % of oil shale mass burned) could be carbonated by binding CO_2 from the air during ash hydroseparation and deposition in wet dumps (in ash fields) decreasing so the amount of CO_2 emitted. Therefore, the maximum amount of CO_2 bound by free Ca and Mg oxides is theoretically proportional to the content of mineral CO_2 in oil shale. Actually, it is lower due to the participation of free oxides in the formation of clinker minerals and binding of SO_2 in the furnace.

The main electricity producing enterprises in the Republic of Estonia are Baltic and Estonian Thermal Power Plants (TPP) located near the town of Narva. The waste ash is removed from boilers using hydrotransport and deposited in wet dumps at so-called ash fields having a surface area of about 2,000 ha. The amount of ash deposited so far is about 282 million tons (01.01.2000), and 5–6 million tons will be added every year. Thus, the CO_2 binding capacity of this material is essential.

The carbonation of lime is well known [10], but data about specific characteristics of the carbonation process of free lime in oil shale ash and of its reactivity towards CO_2 are lacking. The latter depends on the chemical composition and physical properties of the ash as well as on the conditions of heterogeneous interaction of ash with CO_2 (temperature, time, concentration of components in gaseous and water medium, etc.).

The first attempt for estimation of the amount of CO_2 bound by oil shale ash in nature was done by Punning [11]. It proceeded from theoretical considerations and was not connected with any practical measurements. Therefore, at present we have no data dealing with the character, rate and amount of CO_2 bound in nature. However, knowledge about the regularities of binding processes enables to determine the amount of CO_2 bound by ash during its transporting and storing under technological conditions actually used and to find out the ways for intensifying the binding process. These results would be the basis for specifying the CO_2 emission caused by power engineering and establishing respective pollution taxes. Fulfilling this problem was the main task of the present work.

Methods

Preliminary experiments were carried out using cyclone ashes formed in the boilers TP-17 and TP-67 at Baltic TPP. Content of total CaO, free CaO, SO_3, R_2O_3, CO_2 and moisture in the ash from TP-17 was 49.4, 20.0, 3.9, 4.0 and 0.1 %, respectively. The aim was to explain the CO_2 binding ability of cyclone ash under the best conditions – the samples of dry or hydrated ash were stored as a 1-mm thin layer in the open air in laboratory conditions, mixing and moistening them once a day. The room temperature was 19–20 °C, relative moisture of the air 65–68 %.

Model experiments, whose aim was the observation of the carbonation process and its progress into the depth of ash samples depending on the time and natural conditions of storing, were carried out. The model ash used was a mixture of ashes formed at Estonian TPP mixed at the mass ratio furnace ash : superheater ash : cyclone ash : electrostatic precipitator ash = = 15 : 15 : 60 : 10.

The content of free CaO and CO_2 in the model ash (on dry basis) was 19.9 and 1.9 %, respectively. The dry ash was then hydrated with an amount of water exceeding the stoichiometric ratio for 5 times considering the content of CaO_{free} in the ash. The samples of hydrated ash were set on special pans (sample holders) divided into sections of 4 cm × 4 cm × 6 cm size (sub-samples). One sample holder was stored in winter conditions in the open air under the cover (I series) moistening the ash once a week, the other one – fully in the atmospheric conditions (II series).

So, the temperature conditions corresponded to the outdoor temperature during this period for both series as well as the moistening conditions for II series. For analyses once a week one sub-sample from both series was divided into six layers with thickness of 1 cm. The content of moisture, CaO_{free} and CO_2 was determined separately in each layer and the binding degree of CO_2 by CaO_{free} with formation of $CaCO_3$ was calculated using the changes in the content of CO_2 as well as of CaO_{free} in these layers.

The previous treatment and storing of the samples of the III and IV series was analogous to those of the I and II series, but the samples of 12 cm × × 12 cm × 6 cm size were carefully mixed once a week to explain the influence of mixing in the case of storing the samples in a thick layer. Samples of the V and VI series were stored under analogous conditions as those of the I and III series, but in summer.

To investigate the carbonation process in wet ash, the sample of ash from the II section of ash field (WA) of Estonian TPP was used. The content of mois-ture, free CaO and CO_2 in the sample was 18.2, 9.6 and 1.9 %, respectively. The sample with a size of 25 cm × 25 cm × 2.5 cm was divided into three parts. The first part was stored without moistering, the second one was moistened with distilled water and the third one – with transport water once a week. The content of moisture in the samples was kept at 30 %.

CO_2 binding degree (BD_{CO_2}) was calculated by the change in the content of CO_2 in the samples:

$$BD_{CO_2} = CO_2/CO_{2\,max} \times 100\ (\%) \tag{2}$$

where CO_2 is analytically determined CO_2 content of the sample in current time, %;

$CO_{2\,max}$ is maximum possible CO_2 content of the sample calculated on the basis of CaO_{free} in the initial sample, %, and was calculated as:

$$CO_{2\max} = \frac{CaO^{i}_{free} * \mathbf{M}_{CO_2}/\mathbf{M}_{CaO} + CO^{i}_{2}}{100 + (CaO^{i}_{free} * \mathbf{M}_{CO_2}/\mathbf{M}_{CaO})} * 100\ (\%) \tag{3}$$

where CaO^{i}_{free} and CO^{i}_{2} are the content (%) of free CaO and CO_2 in the initial sample, respectively.

All values used are given on dry basis.

Different samples of circulating water from different points of ash removal system – inlet (initial pulp) and outlet (pond I) points of ash field, outlet point of transport-water sedimentation pond (pond II) and outlet point of refined water back-flow channel before entering into the new cycle (channel) were studied. Main characteristics of these samples are presented in Table 2.

Table 2. **Main Characteristics of Ash-Transport Water Samples**

Sample	Content				pH	Alkalinity, mmole/l	
	mg/l	mmole/l				Ap	At
	Ca^{+2}	OH^-	CO_3^{-2}	HCO_3^-			
Collected on 03.05.1999							
1. Initial pulp	1242	79.9	2.4	0	13.2	81.1	82.3
2. Pond I	972	77.8	2.6	0	13.2	80.4	77.8
3. Pond II	926	76.2	2.4	0	13.2	77.4	78.6
4. Channel	692	62.4	3.0	0	13.1	63.9	65.5
Collected on 09.06.1999							
5. Initial pulp	831	81.6	4.0	0	13.0	83.6	85.7
6. Channel	480	63.4	4.0	0	13.0	65.5	67.5
Collected on 03.11.1999							
7. Initial pulp	775	80.2	4.4	0	12.7	82.4	84.6
8. Pond II	686	81.2	4.0	0	12.7	83.2	85.2
9. Channel	309	61.4	4.0	0	12.6	63.4	65.5

Carbonation of ash-transport water samples was carried out by leading the air into the water through a porous plate. The height of water column above the plate was 5 or 10 cm and the volume of water – 100 or 200 cm^3. For simulating flue gases the air–CO_2 mixture was used, the P_{CO_2} in the gas mixture was 115 mm Hg.

Results

Preliminary experiments (Fig. 1) indicated that dry oil shale ash was carbonated (bound with CO_2 from the air) slowly – during 1 month – by approximately 11 %. However, free CaO present in hydrated ash when stored in thin layer and mixed periodically could be carbonated during the same period practically completely.

Therefore, 1 ton of ash containing 18 % of CaO_{free} could bind up to 0.14 t of CO_2 from the air.

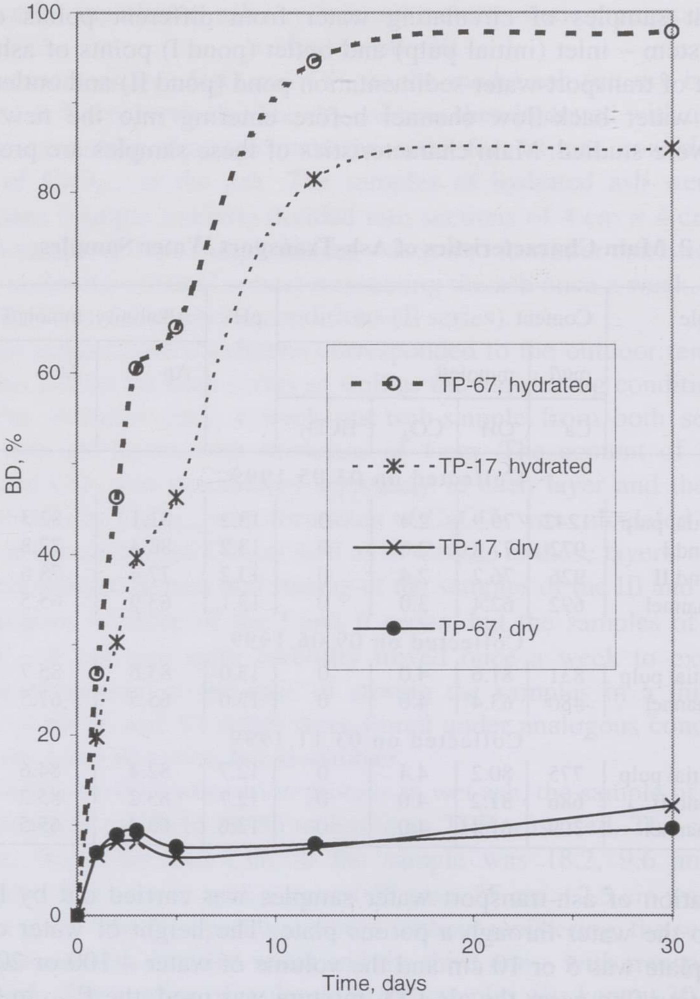

Fig. 1. Dependence of CO_2 binding degree (BD) of oil shale ashes from TP-67 and TP-17 boilers on the storing (in the open air in laboratory conditions as 1-mm thin layers with periodical mixing of samples) time

Investigations with the model ash indicated that the samples previously hydrated and deposited in winter conditions carbonated intensively only in the depth of the upper layer (1 cm) – during a seven-weeks storing the content of CO_2 increased from 2.8 to 12.2 %, or the BD_{CO_2} reached up to 80 %. After the same period this value for the 6th layer (5-6 cm) was only 10–30 %, depending on the natural conditions of storing – temperature and moistening regimes. For example, the content of CO_2 in the 1st and 2nd layers of sub-samples of series II increased essentially between the fifth and seventh weeks of experiment as there was more precipitation (rain) comparing with the previous period (moisture content of the sample increased from 14.3 % to 25.0 %) (Fig. 2).

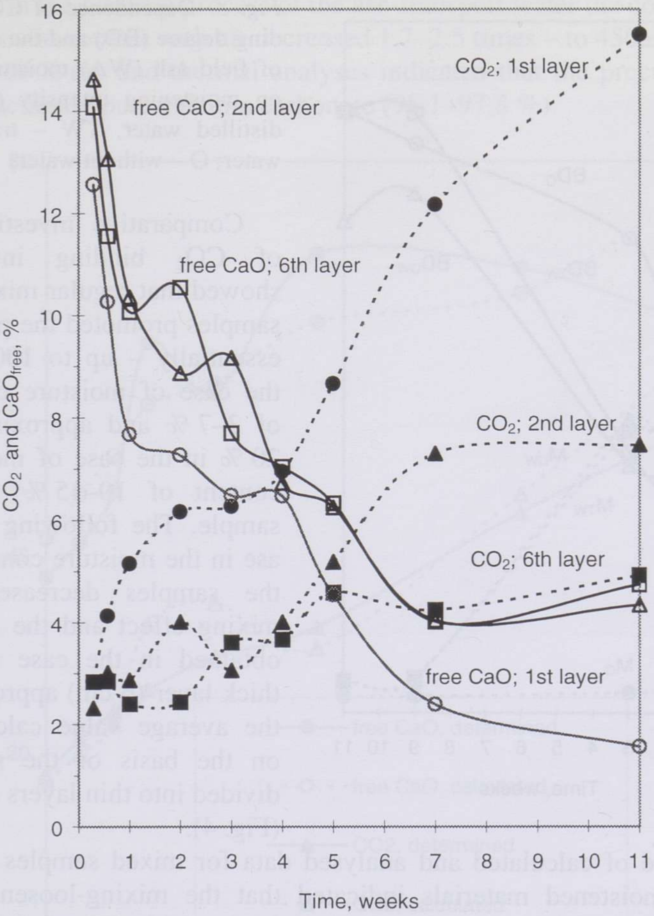

Fig. 2. Changes in the content of CO_2 and free CaO in the exposure time by simulating the carbonisation of model ash in the field conditions (II series)

Investigations carried out with the samples from the ash field (WA) confirmed the importance of moistening conditions (regime) in the carbonation of ashes laying in static conditions. Excess moisture (approximately 35–40 % of sample mass, paste consistence) hindered the CO_2 diffusion into the solid matter, and the process proceeded more slowly compared with samples without moistening (Fig. 3). After the third week the intensity of moistening was diminished (down to 5–7 % of the sample mass), accompanied by an incre-ase in the CO_2 content. After nine weeks of storing no practical differences in the content of CO_2 were observed in the samples kept with or without moistening – in all cases the content of CO_2 was 7–7.6 % and BD_{CO_2} about 80 %.

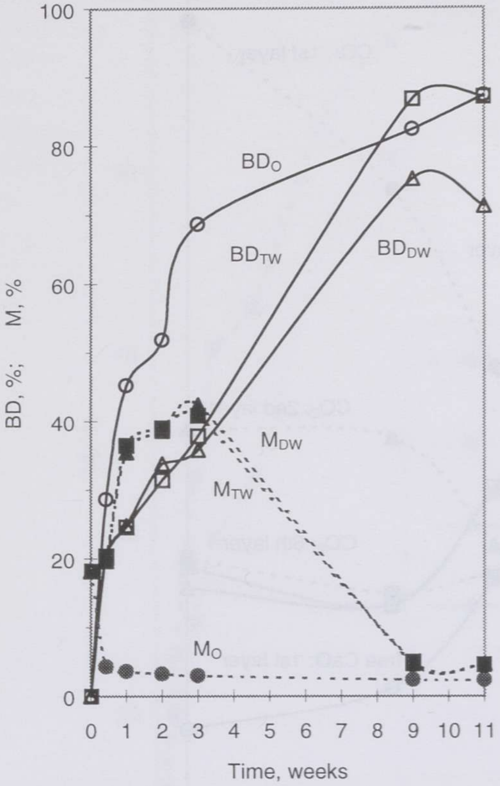

Fig. 3. Dependence of CO_2 binding degree (BD) and the content of field ash (WA) moisture (M) on moistening intensity (DW – distilled water, TW – transport water; O – without water)

Comparative investigation of CO_2 binding intensity showed that regular mixing of samples promoted the process essentially – up to 100 % in the case of moisture content of 3–7 % and approximately 20 % in the case of moisture content of 10–15 % in the sample. The following increase in the moisture content of the samples decreased the mixing effect and the results obtained in the case of the thick layer (6 cm) approached the average value calculated on the basis of the sample divided into thin layers (1 cm) (Fig. 4).

Coincidence of calculated and analysed data for mixed samples in the case of overmoistened materials indicated that the mixing-loosening of material would be effective at a comparatively dry (elder) part of the ash field.

The formation of different calcium compounds besides calcium carbonate explains why the values of BD_{CO_2} calculated on the content of CaO_{free} and CO_2 in the sample differ from each other (Fig. 4). Therefore, the CO_2 binding capacity of the samples must be calculated on the basis of changes in CO_2 content of the ash. Results of X-ray analyses of the samples indicated that the main calcium-containing compounds in dry initial ashes were lime (CaO) and anhydrite ($CaSO_4$), but in carbonated ash – calcite ($CaCO_3$), portlandite ($Ca(OH)_2$) and some amount of calciumsilicates. In the hydrated ash (WA) both in the initial and in the carbonated ash the main compounds were portlandite, calcite, acermanite (Ca_2MgSiO_7) and larnite (Ca_2SiO_4) – in different proportions.

The samples of circulating water from the inlet point of the ash removal system (initial pulp) contained 770–1240 mg Ca^{2+}-ions/l and all the samples used were deeply alkaline – their pH index was 13.2–12.6 (Table 2). Hydroxide ion (OH^-) prevailed (60–80 mmole /l), the concentration of carbonate ion (CO_3^{-2}) was 2.4–4.4 mmole/l, hydrogen carbonate ion (HCO_3^-) was absent. In the clearing process of the ash-transport water the concentration of Ca^{2+}-ions in contact with air decreased 1.7–2.5 times – to 450±100 mg/l. The results of X-ray and thermal analyses indicated that the precipitate formed was practically pure calcium carbonate (95.1–97.8 %).

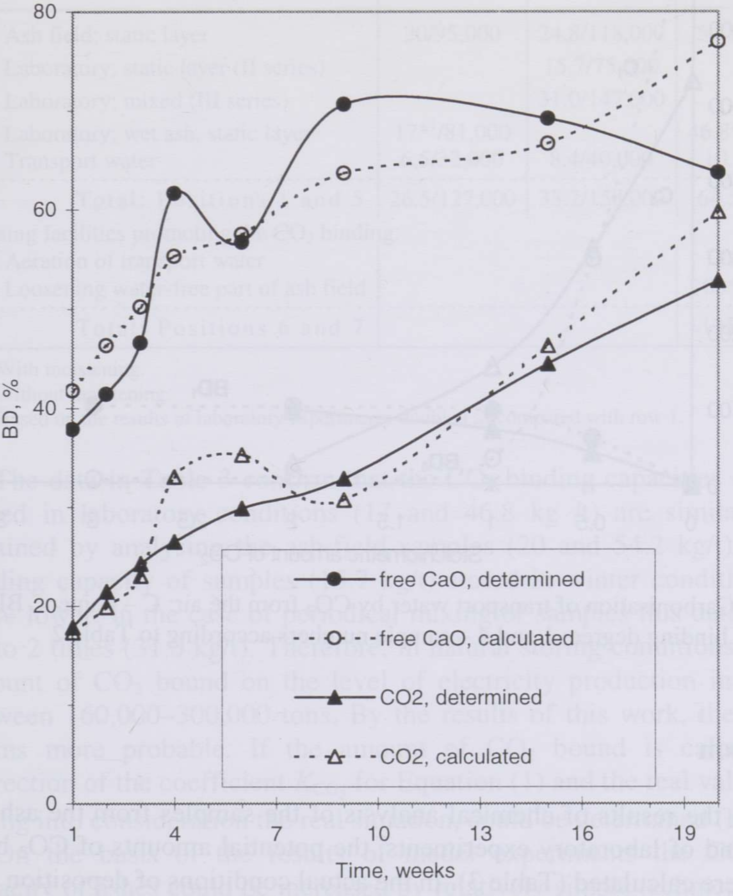

Fig. 4. Comparison of CO_2 binding degrees (BD) calculated as average and determined on the basis of the content of CO_2 and free CaO

The results of aeration of the ash-transport waters confirmed that they intensively bound CO_2 from the air – the content of Ca^{2+}-ion decreased to 10–30 mg /l and pH of the solution to 10.2 as well as the alkaline indexes Ap and At – more than 2 times. Carbonation was possible by using initial water (pulp-water) as well as by using back-flow channel water. In summer conditions initial water having a mole ratio of $CO_2/CaO_{free} = 1.0$ carbonated up to 81–97 %, the channel water up to 71–86 %, in winter conditions both of them up to 65–75 % (Fig. 5). Storing these samples hermetically closed increased the carbonation degree up to 94–95 % during seven days, the content of Ca^{2+}-ion decreased to 20–40 mg/l. Similar results were obtained by using for aeration CO_2–air mixture.

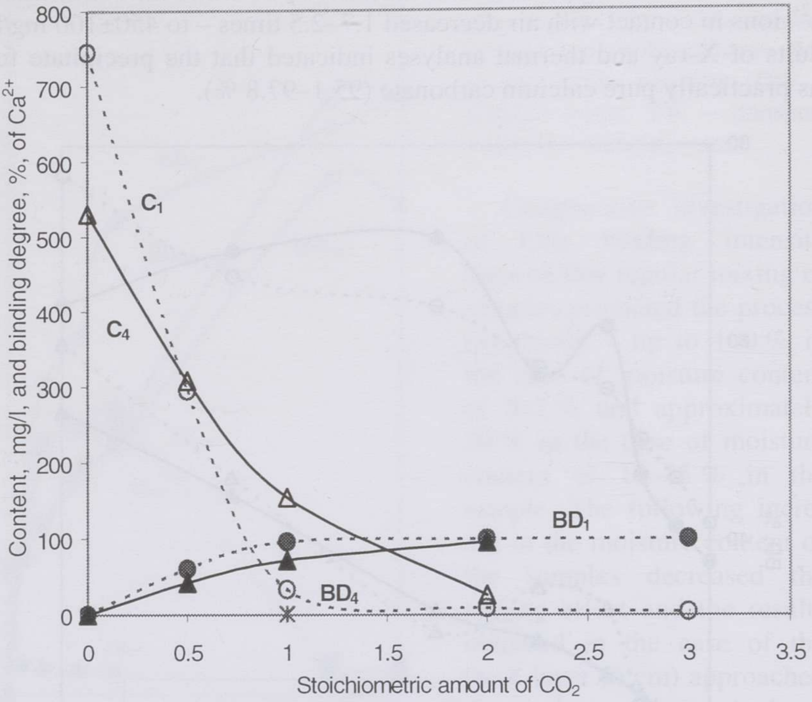

Fig. 5. Carbonisation of transport water by CO_2 from the air: C – content, BD – binding degree; 1 and 4 – sample numbers according to Table 2

Discussion

Based on the results of chemical analysis of the samples from the ash field studied and of laboratory experiments, the potential amounts of CO_2 bound by ash were calculated (Table 3). In the actual conditions of deposition of oil shale ash in wet dumps, 33.2–64.5 kg of CO_2 would be bound by 1 ton of ash (on dry basis) at ash field, from which 8.4–10.3 kg would be bound by the transport water. The lower range was determined on the basis of CO_2 content in the sample taken from the fresh part of the ash field (paste consistence). In such conditions the binding of CO_2 is minimum. So, the upper index calculated on the basis of the samples from the elder part of the ash field is more realistic.

Table 3. **CO_2 binding Capacity of Ashes by Storing (kg CO_2/t, in numerator) and Amount of It per Year Calculated on the Basis of Data in 1998 (t; in denominator)**

Position	Minimum	Average	Maximum
1. Ash field; static layer	20/95,000	24.8/118,000	54.2/258,000
2. Laboratory; static layer (II series)		15.7/75,000	
3. Laboratory; mixed (III series)		31.0/147,000	
4. Laboratory; wet ash, static layer	17$*^1$/81,000		46.8$*^2$/222,000
5. Transport water	6,5/32,000	8.4/40,000	10.3/50,000
Total: Positions 1 and 5	26.5/127,000	33.2/158,000	64.5/308,000
Using facilities promoting the CO_2 binding:			
6. Aeration of transport water			17.0/81,000
7. Loosening water-free part of ash field			108$*^3$/513,000
Total: Positions 6 and 7			125/594,000

$*^1$ With moistening.
$*^2$ Without moistening.
$*^3$ Based on the results of laboratory experiments doubled as compared with row 1.

The data in Table 3 confirm that the CO_2 binding capacities of samples stored in laboratory conditions (17 and 46.8 kg /t) are similar to those obtained by analysing the ash-field samples (20 and 54.2 kg/t). The CO_2 binding capacity of samples (15.7 kg/t) stored in winter conditions is 10–20 % lower. In the case of periodical mixing of samples this data increased up to 2 times (31.0 kg/t). Therefore, in natural storing conditions of ash the amount of CO_2 bound on the level of electricity production in 1998 was between 160,000–300,000 tons. By the results of this work, the last value seems more probable. If the amount of CO_2 bound is calculated, the correction of the coefficient K_{CO_2} for Equation (1) and the real value of CEF, taking into consideration the real situation, could be determined (Fig. 6).

On the basis of the results of model experiments the CO_2 binding capacity of ashes could be increased by relatively simple technical methods. The deeper carbonation of transport water is possible by:

- Leading all the water from the ash field to the sedimentation pond through fountain sprayers.
- Aeration of water in the sedimentation pond. There are good possibilities for it in all the area of sedimentation pond and in back-flow channel. Consumption of air and energy needed for that depend on the deepness of carbonation and the content of Ca^{2+}-ion in the selected point and is lower in the back-flow channel.
- Aeration of transport water with flue gases. The exact spot for that would be selected considering the above-mentioned factors and the possibilities of pumping of flue gases.

For intensifying the carbonation process of ashes in the water-free part of the ash field the latter must be periodically loosened to the depth of 6–12 cm.

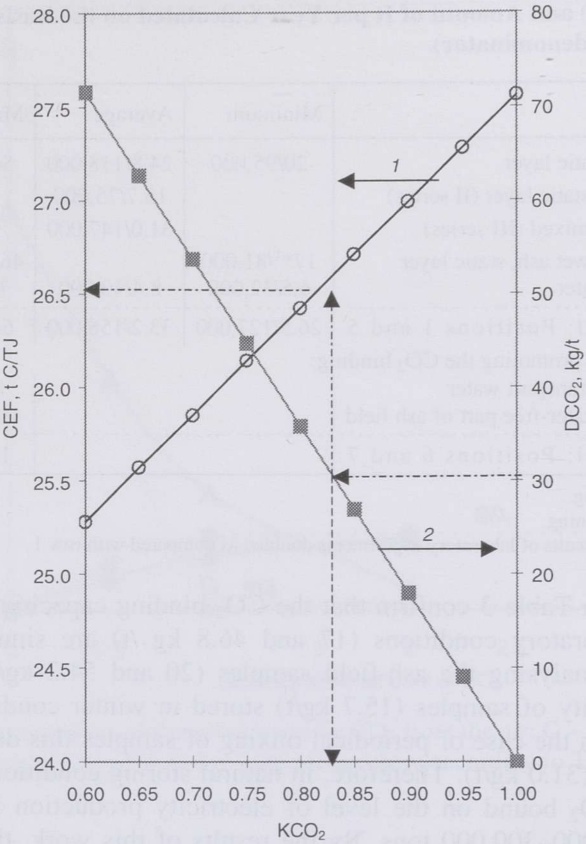

Fig. 6. Correction of coefficient K_{CO_2} by calculating of CEF on the basis of the amount of CO_2 bound (for CEF and K_{CO_2} see Equation (1) in the text; ΔCO_2 – amount of CO_2 bound per 1 ton of the initial oil shale; *1* – CEF = *f*(*K*); *2* – *K* = *f*(ΔCO_2))

It is simply realizable by using bulldozers (levelling the ash fields) with cultivators or other similar equipment for loosening the surface area and, which is especially important, the bulldozers footprints. The amount of CO_2 bound by using these methods could be increased approximately up to 600,000 tons per year (Table 3).

The amount of CO_2 bound by the fly ash during gas-solid interaction in the gas tract or by the ash deposited on the heating surfaces, or which was not emitted from the non-decomposed part of carbonates, was considered. Assuming that by using pulverized firing of oil shale the decomposition degree of carbonates is 1.0, the amount of CO_2 bound in boiler and in gas tract is 16.5 kg per 1 ton of ash or 7.9 kg per 1 ton of oil shale.

So, the total amount of CO_2 bound in the technological tract is 36.7 kg per 1 ton of oil shale, and the relative part of it, considering only the carbonate

CO_2 or total formation of CO_2 is 20.5 and 4.2 %, respectively. By using simple technological methods it is possible to increase this relative amount up to 35.6 % and 7.3 %. Therefore, in spite of incorrectness of assumptions in [11] the general assessment is quite realistic. Thus, the economy in pollution taxes for the producer of electricity in the case of taxes of 5 Estonian kroons per 1 ton CO_2 emitted is on the level of 1.5 –3.4 million Estonian kroons per year.

Conclusions

A complex of experiments for estimation of CO_2 binding ability of model samples compiled on the basis of dry oil shale ashes, wet ashes from ash field, and samples of ash-transport waters from Estonian TPP in laboratory as well as in field conditions was carried out. The hypothesis that it is possible to bind CO_2 from the air at a considerable speed on the basis of alkali components present in these samples by using oil shale ashes was confirmed.

It was indicated that dry oil shale ash at normal outdoor temperatures (20 °C) is inactive and is carbonated slowly – during one month of storing by 10–30 %. Hydrated ash is carbonated completely during the same storing period in the case of periodic moistening and mixing.

The experiments with model samples, hydrated samples and natural wet samples from the ash field indicated the formation of reaction front in the case of static storing (without mixing) of samples. The periodical mixing-loosening of the material promotes the carbonation process 1.2–2 times depending on the storing conditions.

The ash-transport waters were strongly alkaline (pH = 13.2–12.6), saturated by Ca^{2+}-ions – depending on the conditions – up to 80–140 %. They bound CO_2 from the air spontaneously, and as a result precipitation of $CaCO_3$ took place and the concentration of Ca^{2+}-ions in the solutions decreased 1.7–1.8 times. These waters were carbonated by the air as well as by flue gases.

The specific amount of CO_2 bound in the technological tract and in ash fields per 1 ton of dry oil shale ash (up to 64.5 kg from which up to 10.3 kg is bound by transport waters) and per 1 ton of oil shale (up to 28.8 kg totally and 4.6 kg by transport water, respectively) was calculated and a diagram for correcting the coefficient K_{CO_2} by calculating the value of CEF was proposed. The relative parts of carbonate CO_2 and total CO_2 bound were 20.5 and 4.2 %, respectively. By using simple technical methods the relative amount could be increased up to 35.6 and 7.3 %, respectively. On the basis of these data the economy from the decrease of pollutant taxes was calculated.

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