

The relation between the lithotypes of Pliocene coals from Oltenia and their main quality characteristics

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Abstract. The main Pliocene lignite lithotypes from Oltenia (detrital coal, xylite, xylite coal, weakly xylite coal, clayey coal) were formed within various coal facies named after the dominant paleophytocoenoses: the forest with *Sequoia*, the grassy swamp (*Carex* ssp.), the deciduous forest swamp, the forest swamp with *Glyptostrobus*, the marsh with *Phragmites* and the aquatic plants prairie. In each of these coal facies 1-3 lignite lithotypes were formed. Due to the fact that the chemistry of the parental vegetal material was sensibly different with regard each community and that this parental vegetal material was accumulated within different environmental conditions, the resulted lithotypes present specific properties. In this paper are the following parameters are approached: ash, hygroscopic moisture, volatile matter, calorific value, combustible sulfur. Moreover, the main characteristics of the xylite lithotype are analysed. Xylite, by its properties (primary carbonification products, benzene-alcohol extract, pore structure, carbon content, etc.) represents a valuable lithotype as a prime matter source.

Key words: Pliocene, coal seams, xylite, lithotypes, Oltenia, Romania.

Introduction

The first attempt to study the relation between physico-chemical properties of Pliocene lignite lithotypes from Oltenia was made in 1989 (Ticleanu et al., 1988) and continued in 1992 (Ticleanu, 1992 and Ticleanu et al., 1992).

Recently, Ticleanu and Diaconita (1997) have analyzed the main coal facies and lithotypes of the Pliocene coal basin from Oltenia.

1. General geological setting

The coal basin of Oltenia, situated in the south-western part of Romania, contains almost all Pliocene deposits between Olt and Danube Valley (Fig. 1).

According to Andreescu et al. (1985) the sequence of Pliocene coal deposits consists of three lithostratigraphic units:

- the Berbesti Formation (Lower Dacian), mainly psammitic, having in its upper half clayey interlayers and six coal seams (A, B and I-IV), representing the Valea Visenilor coal complex.;
- the Jiu-Motru Formation (Upper Dacian-Middle Romanian), pelitic- psammitic, having nine coal seams (V-XIII);
- the Cendesti Formation (Romanian-Lower Pleistocene), mostly psammitic-psephitic, having in the central part the Balcesti coal complex (seams XIV-XVIII).

Structurally, the coal basin overlies the South Carpathian Foredeep, especially on the internal side of the

foredeep. Here the greatest thickness occurs made of the 22 coal seams (N. Ticleanu et al. 1988, N. Ticleanu - I. Andreescu, 1988). On the external side of the foredeep, the frequency and the thickness of the coal seams decrease. Within the southern part of external side, in the Upper Neogene sequence, the coal seams occur as local pockets with thicknesses exceeding 2 m, due to the compaction subsidence.

The Pericarpathian fault separates the foredeep in two compartments (M. Sandulescu, 1984). Coal deposits from the internal compartment are a little folded and rarely faulted. In the external compartment the coal seams are continuous and their slopes are among 3 and 7 degree. The coal seams overlaying the Moesian Platform are thinner (up to 7 meters) and horizontal.

Palaeogeographically, the coal deposits from Oltenia were accumulated in the western side of the Dacian Basin, in a huge eutrophic swamp. Due to the depositional evolution through time, there are four depositional systems in these successions: fluvial, deltaic, alluvial (I. Andreescu, unpublished) and telmatic (N. Ticleanu, 1995).

2. General petrographical description

According to the recommendations of the International Commission for Coal Petrography (Lexique, 1971) a lithotype is a homogeneous coal with a thickness greater than 5 centimetres. Using this recommendation, Ticleanu, Bitoiaru (1988), N. Ticleanu et al. (1988) and Ticleanu, Diaconita (1997) have separated for the coal deposits from Oltenia the following lithotypes: detrital coal, xylite, weak xylitic coal, xylitic coal, fibroid coal and clayey coal.

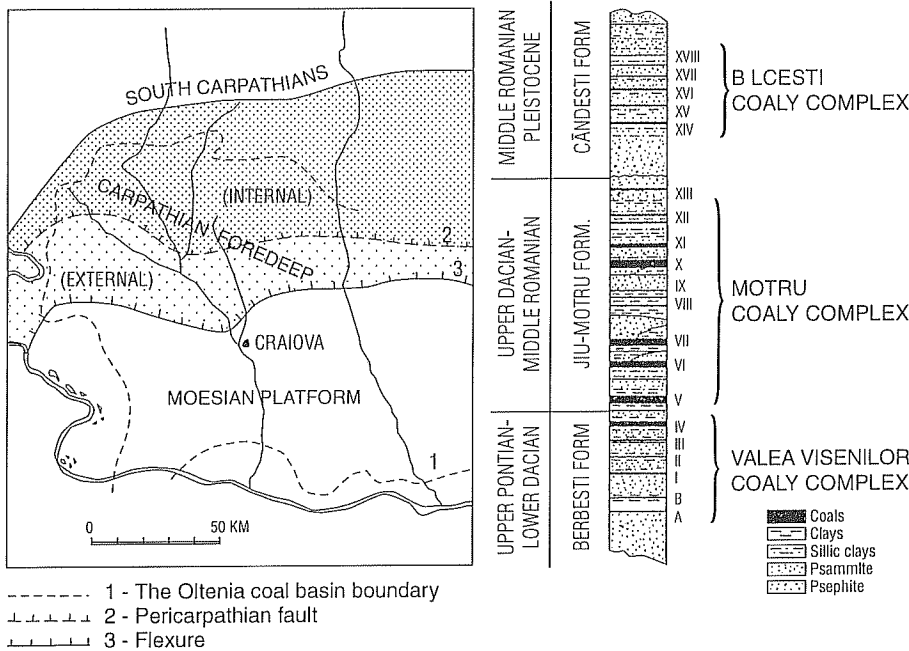


Fig. 1. The Oltenia coal basin occurrence and simplified lithographic log for the coal bearing deposits.

Beside the lithotypes, the coal seams have coaly clay, clay and rarely clayey silts and silts.

Detrital coal (DC) is the most frequent lithotype within the coal basins of Oltenia. The DC has a humic groundmass of carbonified vegetal fragments with fine detritic texture, more or less layered. In fresh state the DC is without luster, with colours between yellow-brown and brown-black. DC changes its physical and chemical properties in contact with the air, after a period longer than six months (Panaitescu, 1990).

In the coal seams lithotype DC is present like bank and lens with a thickness between 5 centimetres and few tens of centimetres; rarely, the DC has a thickness greater than

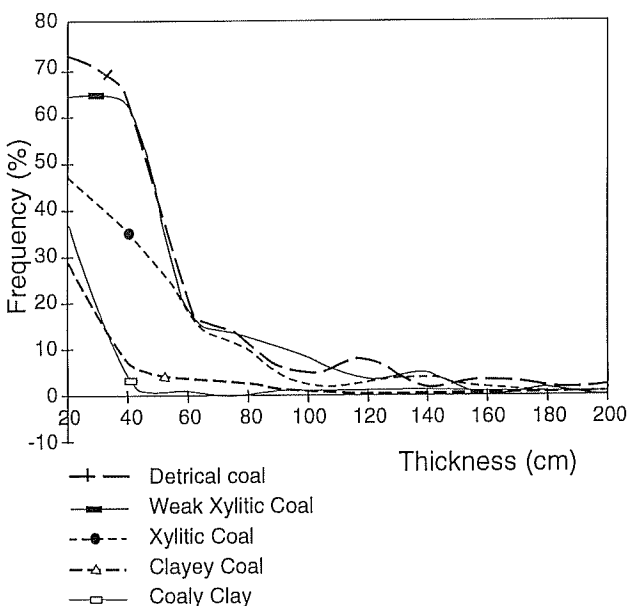


Fig. 2. Frequency polygons for the thickness classes distribution of banks and lenses of the main lithotypes.

1 m up to 3 m. The beds of DC with thickness of 20-40 cm are frequent (80 %) in coal seams. In 20 % cases, beds of DC have a thickness lower than 20 cm (Fig. 2).

Xylite (XL) is present in coal seams from Oltenia in percentages up to 25 %. The XL lithotype is composed of carbonified wood (roots, trunks and boughs) from Pliocene eutrophic swamps.

More than 80 % of xylite is composed of fragments of *Glyptostroboxylon tenerum* and other trees like *Taxodyoxylon toxodii*, a relict in Pliocene swamps (N. Ticleanu, unpublished).

There are two types of xylite (Abel, 1965): fasser xylite, the most frequent and the bruch xylite which is very rare.

Xylitic coal (XC) is frequent enough: together with xylite, it represents 26.6 % in the studied coal seams. XC has a matrix of detrital coal with fragments of xylite. The beds and lenses of xylite and detrital coal are up to 5 cm thick. XC is a transitional lithotype between detrital coal and xylite lithotypes.

The beds of XC have a thickness of 20 cm (more than 45 %), rarely of 60-80 cm and very rarely (1-2 %) 100-200 cm (Fig. 2).

Weak xylitic coal (WXC) is a lithotype of transition among DC and XC. SX consists in beds of detrital coal and xylite with thickness up to 5 cm. The most frequent thickness of SX is 20-40 cm and very rarely 200 cm.

Charcoal (CH) is very rare, mostly lens-like, with thickness greater than 5 cm. CH is frequently present like discontinuous beds with thickness up to 1 cm in DC, WXC and XC. Pieces of charcoal with a variety of different size occur frequently in the detrital or xylitic coal. These show the presence of frequent natural fires in the forest swamp.

Clayey coal (CC) is present in the upper coal seams like beds with thickness up to 10-20 cm and sometime, rarely, 30 or 70 cm.

Coaly clay (AC) have frequently thickness of 5-10 cm and sometimes 15, 20-40 or, very rarely 55 cm (Fig. 2).

The preliminary researches indicate that in the Pliocene coal seams from Oltenia the frequencies of the lithotypes are: DC- 32.21 %, WXC-25.58 %, XC+XL- 23.01 %, AR-11.33 %, CC-5.17 %, AC-2.73 % and CH up to 1 %. (Fig. 2). The coal with this distribution of lithotypes is a lignite.

The xylite is frequent within the basal seams (I-IV,) being especially frequent within the seams V-VII. The frequency of coaly xylitic facies increases from the border

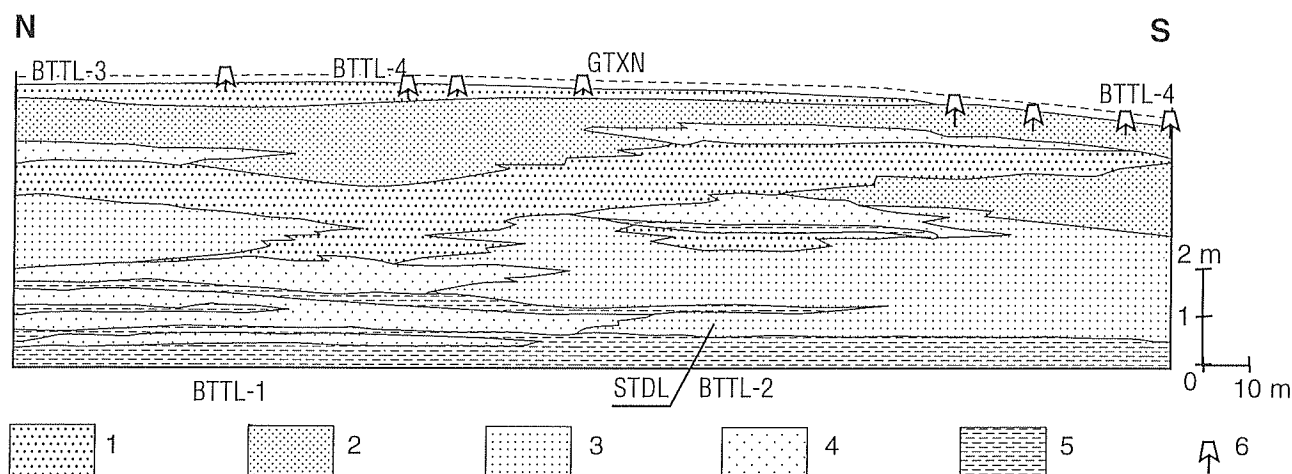


Fig. 3. Macropetrographical and paleobotanical section trough of coal seam no. VI from Lupoia Quarry (the central zone of the exploitation area in summer 1988).

1 - xylite and syllitic coal, 2 - detrital coal with centimeter level of xylite interlayers, 3 - weakly detrital coal, 4 - detrital coal (the increase of the xylite content is emphasized by an increasing of observation points density, 5 - clays, 6 - roots in growth position.

BTTL - *Bytneriophyllum tiliaerfolium*, GTXN - *Glyptostroboxylon tenerum*, STDL - *Stratiotes dacicus*.

Frequency indexes: 1 - very rare, 2 - rare, 3 - frequent, 4 - very frequent.

to the centre of the basin. The percentage of IC increases from the basalmost coal seams to the upward layers. In the Lupoia quarry the IC represents 4.34 % in the seam V-VII and 16.39 % in the coal seam X. Similar to IC, the proportion of CC increases from 1.98-7.07 % in the basal seams, to the 14.63 % in the coal seam X (in the quarries Rosiuta and Jilt Sud).

For the all coal seams from the basin of Oltenia, the proportion of xylitic lithotype increases toward the superior part of the seam (Fig. 2).

3. The origin of lithotypes

Each lithotype has its origin in one to three specific coal facies. The coal facies are defined by the main palaeophytocoenoses (Ticleanu, Bitoianu, 1989 and Ticleanu, Diaconita, 1977).

These paleophytocoenoses represented ecological assemblage series grading from borders towards the centre of the coal generating swamps. Influenced by the tectonic, climatic, depositional and paleogeographic factors, the paleophytocoenoses changed and so the coal facies within space and time. In this way, inside the same coal bed, entirely the same complete genetic series rarely occurs. Short-term deposition gaps occurred, being caused by catastrophic floods or by substantial, long-term changes.

Each cited lithotype was formed within one or three coal facies (Ticleanu, Bitoianu, 1989; Ticleanu - Diaconita, 1997), being determined by the changes that occurred within the hydrologic regime of the swamp (the subsidence increasing or decreasing). This phenomenon can be caused by the subsidence rate variation (tectonic regime) or by the compaction subsidence due to the clayey rocks of the substrate. In this way, within the vertical direction of the coal seams several coal facies changes can be recorded.

Each coal facies was formed within a distinct paleobiotope that was characterised by typical environmental conditions, such as the flooding, pH, Eh and microbial regime, etc. These conditions determined the occurrence of typical phytocoenoses having ecological affinities for the various parameters.

Following coal facies and the resulted lithotypes were recorded:

- Sequoia forest, with XC, XL lithotypes;
- grassy marsh with *Carex* ssp., with lithotype DC. This coal facies, evolves to a forest coal facies;
- swamp deciduous forest, with DC, WXC and rare XL lithotypes;
- swamp with *Glyptostrobus*, with XC, XL, WXC lithotypes;
- reed swamp (2 m depth), with DC, CC lithotypes;
- aquatic macrophyte prairie with CC, DC.

In this way, the parental vegetal material of each coal facies was chemically individualised (e.g. the grass facies contained almost exclusively cellulose whereas the swamp forest facies contained lignine, cellulose, etc.). Also, the primary mineral content was very low compared to the vegetal material coming from the forests (under 1 %), whereas within the grass facies (*Carex*, *Phragmites*, *Typha*, etc.), the primary mineral substance could reach up to 6 %, inducing a residual accumulation between 2.4 % and respectively 8 %. To these levels it can evidently be added the tertiary mineral substance, being the main ash generator, grading from a paleobiotope to another (Ticleanu & Diaconita, 1997).

To conclude, each coal facies was characterised by typical accumulation conditions of the parental vegetal material and by a typical chemical composition. Within each coal facies 1-2 (rarely 3) lithotypes were formed, that, as it will be emphasised, present physical and chemical differences.

4. Ash content as a lithotype indicator

The main aim of the statistical analysis of ash content is to find out a parameter to discriminate among the six coal lithotypes.

The first step has been an analysis of the observed distribution of ash content for each lithotype. Except of the xylite (with lognormal distribution of values of ash contents), for all lithotypes the values of ash contents have Gaussian distributions.

In the second step of analysis, the means of ash content for the six lithotypes were evaluated (using 555 values) (table 1).

Table 1.

Lithotype	Number of values	Lower limit	Mean	Upper limit	a
XL_1	160	3.618	3.71	4.279	3 %
XL_2	72	3.582	3.71	4.249	3 %
XC	88	12.738	13.52	15.51	3 %
SX	83	18.203	19.81	23.098	3 %
DC	83	23.742	25.58	28.889	3 %
CA	44	40.247	42.21	44.214	3 %
AC	25	58.76	61.85	63.814	3 %

The differences between means are significant (Fig. 4) for a low level of the probability of making a type error ($\alpha=0.03$).

In this case the means of ash content are a good parameter to discriminate among the lithotypes of Pliocene coals from Oltenia.

5. Analysis of the main quality parameters of lithotypes

Within Table 2 there are presented the variability limits and the statistic averages for the main quality parameters of the lignite lithotypes, except the total moisture (W_t). The last one has a coal-field average of 41 % and an ash content (A) previously analysed.

The content of hygroscopic moisture (W^h) of lithotypes is very variable (4-8.5 %) and does not correspond to the carbonification for the lignite level, being more de-

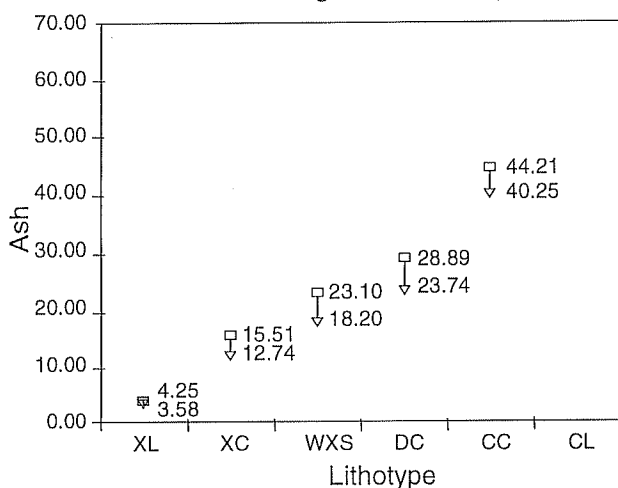


Fig. 4. Means ash content of the lithotypes of Pliocene coals from Oltenia.

Table 2. Main quantitative parameters of the Pliocene lignite lithotypes from Oltenia

Parameter 1	Lithotype 2	UM 3	Samples 4	Mean 5	Max 6	Min 7
W^h	XL	%	234	4.19	10.55	2.09
	CX	%	45	4.87	15.64	2.66
	WXC	%	32	7.16	17.85	3.16
	DC	%	53	5.5	19.15	2.45
Q_i^i	XL	kcal/kg	134	4250	5877	2365
	XC	kcal/kg	45	3521	5054	2076
	WXC	kcal/kg	32	3181	4984	1860
	DC	kcal/kg	57	2801	4984	1583
Q_i^{inc}	XL	kcal/kg	96	5952	6717	5217
	XC	kcal/kg	44	5819	6242	4946
	WXC	kcal/kg	32	5671	6406	4862
	DC	kcal/kg	52	5684	6977	4678
V^{inc}	XL	%	136	64.32	93.7	19.67
	XC	%	44	57.67	64.76	53.26
	WXC	%	32	56.69	62.31	43.62
	DC	%	57	58.6	79.71	37.78
S^{inc}_c	XL	%	99	2.3	7.62	0.9
	CX	%	44	2.92	6.78	0.95
	WXC	%	32	2.94	8.9	1.34
	DC	%	7	3.08	12.65	0.62

creased than the carbonification level. The average values for the Romanian lignites vary between 12.3-14.8 (Blum, Barca, 1966). The explanation of this phenomenon is linked probably to the pore structures of the lithotypes that make the lignites.

A very wide variability is presented by the volatile matter content for the xylite lithotype (19.67-93.7 %), with a high statistic average (64.32 %) that reflects the low carbonification degree. It is evident that this large variability is given both by the diversity of the parental vegetal material (angiosperms and gymnosperms) and by the occurrence conditions of the xylite within the three main coal facies.

Also, for understanding the extreme values, it has to be taken into consideration that these values can be referred to special conditions such as that the xylites come from frequent natural wood fires. Some have been the subject of a natural primary carbonisation, with a massive loss of volatile matter. An important part of the parental vegetal material fell into the swamp, after becoming dry and partially rotten from the trees.

The calorific power (Q^{daf}) presents the largest variability limits for the DC lithotype 4678-6977 kcal/kg), the statistic average being of 5684 kcal/kg. The maximum value of this parameter is given by the DC lithotype (6977 kcal/kg) which is due to the fact that in some sequences bituminous coals with Botryococcus occur (Em. Demetrescu, inedite). This bituminous occurrence increases the content in H^{inc} more than 6.68 %.

Finally, the combustible sulfur content (S_{comb}^{daf}) reflects very well the existence of different accumulation conditions of the parental vegetal material, the last one being maximal for the DC lithotype (3.08 %) due to predominant anoxic environments. The combustible sulfur content was formed in the same time with the syngenetic pyrite. XL has the lowest content (2.3 %) because the coal facies with Glyptostrobus is characterised by an aerobic

anaerobic environment not so favourable to the syn-genetic pyrite genesis.

6. Xylite: one of the most interesting lithotypes

Morphologically, the typical wood structured xylite occurs within bands, lenses or half-cone bodies, in function of the vegetal organ from which it came. The dimensions are either variable, due to the same reasons. In this way, the trunks fallen within the swamp have elliptical transversal shape, having lengths up to 10-15 m and thickness up to 0.5 m. The branches are within a millimetre scale, up to 15-20 cm thickness, with oval transversal shape. The roots within the coal mass are half cone shaped, 1-1.4 m wide and 1-3 m high.

According to I. Petrescu and N. Ticleanu (inedite) over 80 % of the xylite is generated by *Glyptostroboxylon tenerum* and secondary by *Taxodioxydon*, *Sequoioxylon* and other species.

The coal facies containing the greatest quantity of xylite belonged to the swamp with *Glyptostrobus europaeus*, within the almost permanently flooded areas having a larger development at the level of the coal beds no. I, IV, V and VI-VII. This fact can be explained by the development of the coal generating swamps occurring within a deltaic flood plain.

Taking into account the aspect and the mechanical character, the coals from Oltenia can be grouped within two main types of xylite:

Fibroid xylite, with evident wood structure, predominant yellow-brown, with elastic characters;

Breaking xylite, rare, with clear wood structure, olive, brown, black in colour, with conked, angular breaking.

While drying, both types of xylite become black, shiny, with conked breaking and the wood structure which is hardly distinguishable.

To the petrographic composition of the xylite participate almost exclusively the humotellinite subgroup macerals, represented by textinite (4.26-86.68 %), texto-ullminite (5.68-72.34 %) and eu-ullminite (4.35-46.9 %), within alternating bands.

The large variability limits are explained by the variability of the accumulation conditions of the parental vegetal material.

The humodetrinite is absent, occurring especially on small fractures and as a peel around fresh xylite fragments from the quarry. The humocollinite is absent or very rare, reaching exceptionally 10.33 %, usually being under 1 %.

From the point of view of visible xylite fragment thickness, the dominant dimensions occur within 20-50 mm. Frequently, within the quarries from the north of the coal-generating basin, the thicknesses surpassing 20 mm represent more than 50 % of the total xylite fragments and can reach 86 % (Fig. 5). This fact correctly reflects the dominance of the trunks compared to the branches as parental vegetal material.

The large variability limits for the dry ash, free volatile

Table 3. Tar and primary carbonisation gas (T_{PC}^{daf} , G_{PC}^{daf}), carbon (C^{daf}), humic acids (H_A^{daf}) and cellulose ($Cell^{daf}$) for the xylite lithotypes XL, XC, WXC.

Parameter	Litho-type	MU	Average	Maximal	Minimal	Number of analyses
T_{PC}^{daf}	XL	liters/ton	153.53	302.29	33.3	105
	XC	liters/ton	71.07	200.46	32.1	30
	WXC	liters/ton	55.44	89.84	38.6	13
G_{PC}^{daf}	XL	Nm ³ /ton	162.94	263	103	105
	XC	Nm ³ /ton	208.24	257.28	159.69	30
	WXC	Nm ³ /ton	223.62	297	180.37	13
C^{daf}	XL	%	63.94	75.8	58.05	54
B^{daf}	XL	%	7.6	33.39	1.24	62
H_A^{daf}	XL	%	9.38	33.52	1.42	29
$Cell^{daf}$	XL	%	17.96	49.69	4.07	53

matter, calorific value (see Table 2), carbon, extract with benzene alcohol, cellulose and humic acid (see Tab. 3) show the xylite provenance from different species of trees and the fluctuation of environmental conditions (Eh, pH, bacterial activity, etc.) within the coal facies in which this material has been accumulated (Ticleanu, Diaconita, 1997).

With regard to the volatile matter, there can be recorded a decrease with the coal occurrence depth (the Hillt rule). So, at 60-70 m, under the X-th coal seam, within the V-th and the VI-th coal seam, the volatile matter content decreases by 2-3 %.

As it has been shown in the above chapter, the average statistic content for the xylite dry ash is 4.25 %, but this content refers to the xylite directly extracted from the quarry. Actually, the clean xylite, sorted and granulated has only 2.4 % ash for the 20 mm class.

The relation between some xylite properties and the xylite origin is shown also in Table 4, that has been made on the ground of identification of the xylite generating taxa and of their content in extracts with benzene-alcohol (B^{daf}) and cellulose. It is to be emphasised that the maximal values enter in the first category and probably come from the wood of a *Pinus* species, occurring within the Sequoia forest coal facies (Ticleanu, Diaconita, 1997). The extract with benzene-alcohol is represented in equal proportions by waxes and resins.

The same causes probably induced the large variability of the primary carbonisation products (temperature of about 500 degrees C), with their variability limits shown within Table 3. The variability limits for tar (T_{PC}^{daf}) are extremely large, 33.6 l/t - 302.29 l/t, with a statistic average of 153.33 l/t, in the same way as the primary carbonisation gases, having values between 103 Nm³/t and 263 Nm³/t.

The pore structure of the primary carbonisation coke is close (according to A. Georgescu, inedite) to the one of *Fagus silvatica*.

With its physical and chemical properties, the xylite is highly different from the other Pliocene coal lithotypes from Oltenia and it can make the prime matter for various industrial fields.

Table 4. Xylite distribution with regard to the bitumene and cellulose content

Xylite origin	Number of samples	B ^{daf} (%)	Number of analyses	Cellulose (%) (daf)	Number of analyses
Glyptosoboxylon tenerum	15	4.28-27.15 %	13	13.47-26.20	3
Taxodium (generally)	5	3.81-29.03	5	12.76-30.81	4
Conifers (not identifiable)	14	7.98-28.66	12	9.52-23.32	8
Various xylites, impossible to identify systematically	6	14.52-35.52	6	21.74-26.52	2

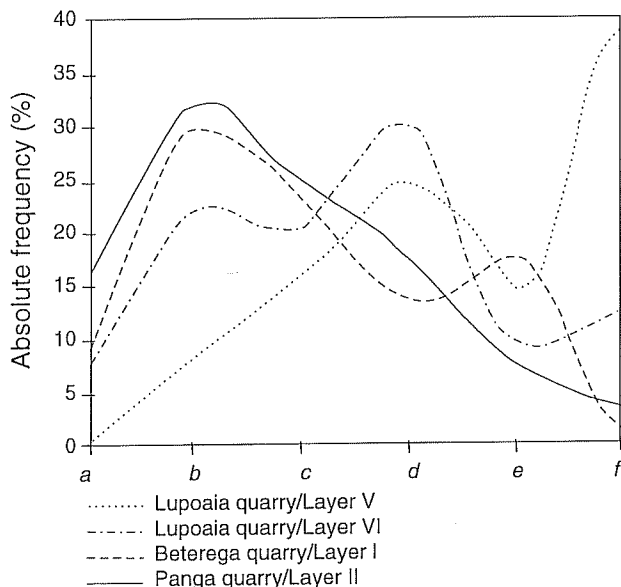


Fig. 5. Frequency polygons for the xylites occurring in various beds and coal quarries from Oltenia.

Thickness categories (in mm): a - (1-5), b - (5-10), c - (10-20), d - (20-50), e - (50-100), f - (more than 100).

7. Concluding remarks

The quality parameters (physical, chemical and technical) of the lithotypes present wide variability limits and they differ in each other due to lithotype genesis within distinct coal facies, of different vegetal parental material and of various facies.

Within the coal facies with forest swamps, a part of the wood parental material suffered rotting processes or natural half-carbonisation, before falling in the peat, fact that explains the wide variability limits for a series of parameters.

Due to its physical, chemical and technical properties, the xylite lithotype is very important taking into consideration that it can represent the prime matter for various products.

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