Possible options for the geological storage of carbon dioxide in Hungary

Éva Hartai[1]

Storage of anthropogenic carbon dioxide in deep geological formations is a possibility for the mitigation of climate change. CO_2 *can be stored in deep saline aquifers, depleted oil and gas fields and unmineable coal seams. Due to the tectonic setting and the evolution of the Pannonian Basin, in Hungary all storage options are available. Saline aquifers of the Pannonian Szolnok Formation and Újfalu Formation seem to be the best* CO_2 *reservoirs but depleted gas fields in the Great Hungarian Plain also have significant capacity. As for the coal fields, the Lower Jurassic Mecsek Coal Formation and the deep lignite seams of the Pannonian Újfalu Formation are potential storage places.*

Key words: *carbon capture and storage, saline aquifers, depleted gas fields, unmineable coal seams*

Introduction

It is widely accepted that global climate is influenced by the anthropogenic emission of large quantities of greenhouse gases, including carbon dioxide, into the atmosphere. There are several options for reducing CO_2 emission, like energy efficiency improvements, switch to less carbon-intensive fuels, use of renewable energy sources, or enhancement of biological sinks. Beside these options, CO_2 capture and geological storage (CCS, CO_2 sequestration) is also a possibility to achieve significant emission reduction and can be applied to large industrial CO_2 sources (IPCC, 2005).

CCS technology is a succession of processes in which CO_2 is captured at the emission sites (primarily coal and hydrocarbon power plants), purified if necessary, compressed and transported to a suitable injection place; then injected into a geological reservoir where it is stored safely and permanently (Oelkers & Cole, 2008). During the last years, there has been a gradually increasing research interest focusing on CCS as part of climate change mitigation solutions. Research has shown that it has a potential to be a safe and effective way to rapidly decrease anthropogenic CO_2 emissions. The study of natural analogues has also received a growing interest as it can provide useful information for performing CO_2 sequestration. CCS is currently in a transition state between pilot and demonstration phase, with a commercial deployment projected around 2020.

CO_2 geological storage conditions

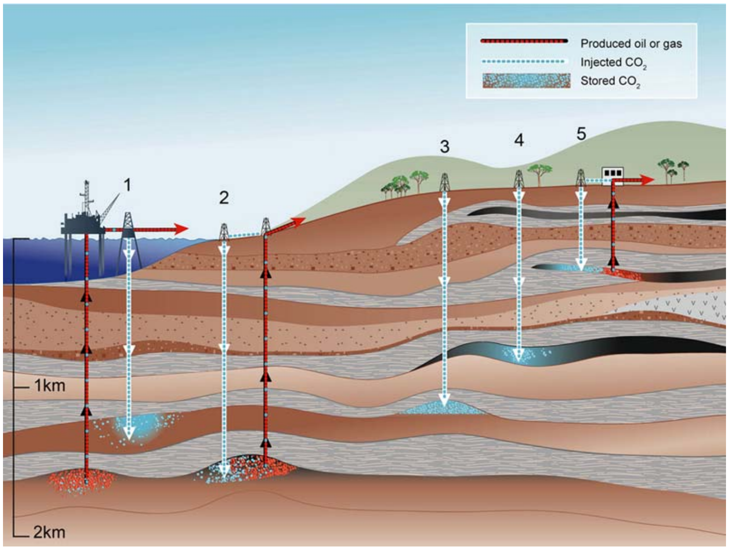

Fig. 1. Options for CO_2 *geological storage. 1, 3: deep saline aquifers (1: offshore, 3: onshore), 2: depleted hydrocarbon fields with possible enhanced oil recovery (EOR), 4: unmineable coal seams, 5: unmineable coal seams with enhanced coalbed methane recovery (ECBM) (after* CO_2*CRC).*

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[1] *Assoc. prof. Éva Hartai, PhD,* University of Miskolc, Faculty of Earth Science and Engineering, Institute of Mineralogy and Geology, H 3515 Miskolc-Egyetemváros, Hungary

There are four major options for the geological storage of CO_2 (IPCC, 2005) (Fig.1):

- injection into deep saline aquifers,
- injection into depleted gas fields,
- injection into oil fields with enhanced oil recovery (EOR),
- injection into unmineable coal seams, possibly with enhanced coalbed methane recovery (ECBM).

In order to get the optimal fluid conditions, CO_2 should be in supercritical stage. For this reason, the sequestration can be applied to rock bodies at a depth of about 800 m. Taking the world-wide average geothermal gradient, this depth corresponds the pressure and temperature values for the critical point of CO_2 (31,1 ºC temperature, 7,3 MPa pressure). At these values the density of CO_2 is 50-80 % of that of the water. The CO_2 is partly dissolved in the formation water, partly fills the open pore spaces and due to the buoyancy effect it is enriched in the upward arching structures. In some formations it would slowly react with cations to form carbonates, which would lock up the CO_2 essentially permanently (Davidson et al., 2001).

CO_2 emission in Hungary

In the last few decades the population of Hungary has been around 10 million. The highest emission period in Hungary was between 1975 and 1985 (about 8 t per capita / year). In the late 1980s there was a sharp decrease, which can be explained by the introducing of the nuclear power plant in Paks and the lower rate of solid fossil fuels in the energy sector (Fig. 2) (http://cdiac.ornl.gov).

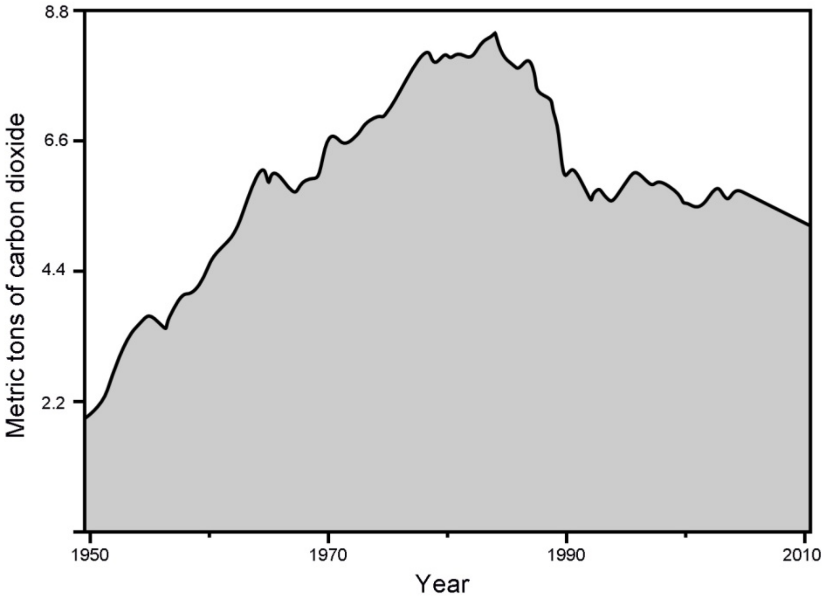

Fig. 2. The per capita CO2 emission in Hungary since 1950 (after http://cdiac.ornl.gov).

In 2008 the per capita CO_2 emission from the fossil fuels and cement manufacture was 5.5 t. About 75 % of the total emission comes from the energy sector (burning of fossil fuels), and the rest is produced by the other sectors, mostly the cement industry and oil refinery (Fig. 3) (http://cdiac.ornl.gov).

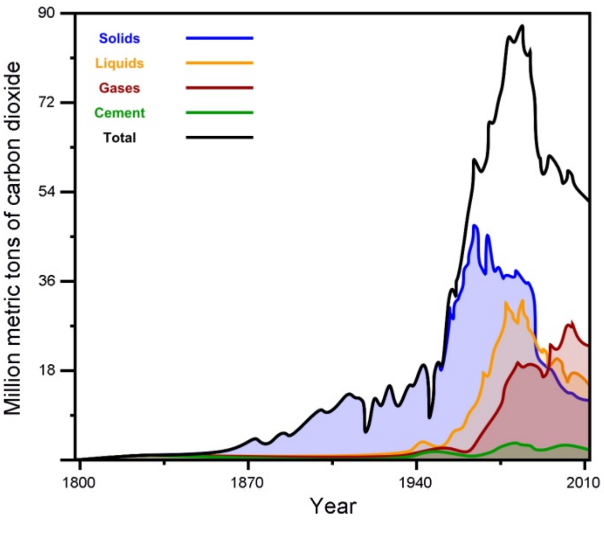

Fig. 3. CO2 emissions in Hungary since 1800 by different sources (after http://cdiac.ornl.gov).

Geological sketch of Hungary

The tectonic setting and geology of Hungary have been a result of a complex structural development. The area was in the collision zone of the African and European plates, and this caused the fragmentation of the marginal parts of the plates. In the Alpine orogeny, the fragmented micro-plates were sheared and rearranged due to the multiply folding and overthrusting. At the end of the Miocene a mantle diapir caused the formation of the large basins of the Pannonian Basin system, which determined the recent structural setting of the country. Contemporaneously with the formation of the basin system the Alpine-Carpathian mountain system emerged.

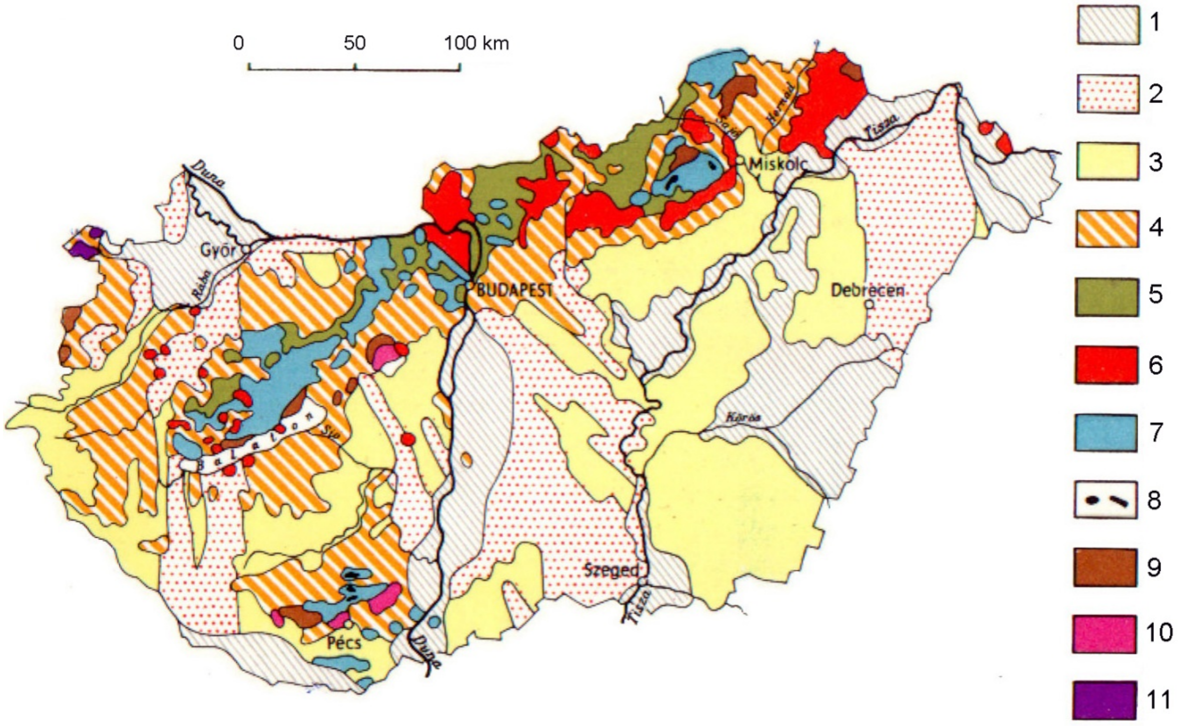

Fig. 4. Geological sketch of Hungary. 1. Holocene alluvium, 2: Pleistocene pebble, sand, 3: Pleistocene loess, 4: Neogene sedimentary rocks, 5: Paleogene sedimentary rocks, 6: Tertiary volcanic rocks, 7: Mesozoic sedimentary rocks, 8: Mesozoic volcanic rocks, 9: Paleozoic sedimentary rocks, 10: Carboniferous granite, 11: Paleozoic schist, gneiss (Haas et al., 2001).

The Paleozoic-Mesozoic basement outcrops in a relatively narrow NE-SW striking mountain range, and in smaller blocks in the western and south-western part of the country (Fig. 4). Because of the intense blocking subsidence in the Neogene tectonic evolution, in the plain areas of the country the basement rocks can be found at a depth of several thousands of meters. From the Late Miocene, in the Pannonian Basin System even 6-7 km deep basins have been formed (Haas et al., 2001).

The tectonic setting, and the character and thickness of the geological formations of Hungary provide a significant CCS potential. The conditions for all the four storage options are given but the most significant storage capacity is available in the deep saline aquifers.

Storage possibilities in deep saline aquifers

A saline aquifer is a porous and permeable sedimentary rock body (usually sandstone) saturated with non-potable water, from which the water can be drawn, and into which fluids can be injected to be stored for a longer period of time. The water of saline aquifers has usually high dissolved material content (a few percent to tens of percent). Saline aquifers are believed to have the greatest potential to store CO_2 (Oelkers & Cole, 2008).

In order to get the safe and appropriate storage options the potential CO_2 reservoir has to fulfil the following requirements (Solomon, 2007):

- large horizontal and vertical extent for the storage capacity,
- high porosity and permeability,
- impermeable caprock above the storing unit,
- the geological structure restrains CO2 from escape,

- tectonically stable (no joints, faults, folds as potential paths for escape),
- isolation from fresh water bodies.

The significant depth of the Neogene basins and the large thickness of sedimentary sequences indicate remarkable storage capacity. Regarding the horizontal and vertical extent, especially the Pannonian (Upper Miocene) formations meet the volumetric requirements. From the surrounding Alps and Carpathians a large amount of clastic sediments arrived into the Hungarian part of the Pannonian Basin. In the Transdanubian area the transport came from west and north-west while in the Great Hungarian Plain from north-west and north-east (Juhász, 1994).

The rate of subsidence was different in the different parts of the Pannonian Basin. However, the Pannonian lithostraigraphic units are relatively easy to follow in the basin system because the characters of the sedimentary formations are similar in the Transdanubian area and on the Great Hungarian Plain (Juhász, 1994).

Based on the stratigraphical and sedimentological examinations, Juhász (1998) described eight sedimentary formations in the Pannonian sequence. From this eight, there are two formations which meet the conditions for the CO_2 geological storage, both for their lithologic character and the horizontal and vertical extent: the Szolnok Formation and the Újfalu Formation.

The Szolnok Formation is a turbidite system with the connecting sedimentary formations which fill the deeper parts of the Pannonian Basin (Fig. 5). Its maximum thickess is around 1000 m. Lithologically it consist of mostly fine-grained sandstones with interlayered silt and marl. The thicker sandstone layers are built up by smaller rhythms. In the lower parts of the Szolnok Formation the pelitic sediments are more frequent, while in the upper zones the sandstone layers become dominant and their thickness is more significant. The Szolnok Formation is covered by the Algyő Formation which has dominantly a pelitic character, in a thickness of 200-100 meters (Juhász, 1998). This overlying formation can form a good seal above the sandstone layers of the Szolnok Formation.

Szamosfalvi et al. (2011) calculated the CO_2 storage capacity for the Szolnok Formation in the area of the Great Hungarian Plain using three different methods. In the calculation they did not consider the solution and mineralization of the CO_2. By the equations method worked out by József Pápay, which is based on the compressibility of the formation and the highest applicable overpressure, 650-750 million ton storage capacity was estimated. Using the GeoCapacity-TNO model, which is the “standard” formula in Europe, 500-650 million ton was determined. By the application of the CSLF (Carbon Sequestration Leadership Forum) formula1800-3750 million ton was calculated. As for the whole area of Hungary, they estimated 1500-3500 million ton CO_2 storage capacity in the Szolnok Formation.

The Újfalu Formation is deposited above the Algyő Formation and is built up by sandstone (Fig. 5). Its thickness is generally 200-300 meters but at certain places there are extreme increases in thickness, e.g. 1400 meters at Etyek. It represents a protruding delta facies with upward increasing grain size. The Újfalu Formation is covered by the Zagyva Formation, which is dominantly pelitic, but also has sandstone and lignite layers (Juhász, 1998). Kubus (2009) calculated 424 million ton CO_2 storage capacity of the Újfalu Formation. He used the Pápay’s equations method.

It has to be noted note that the CO_2 storage capacity of saline aquifers might be overestimated because of the lower CO_2 density due to the high geothermal gradient in Hungary and the high temperatures at storage depths of the specific basins.

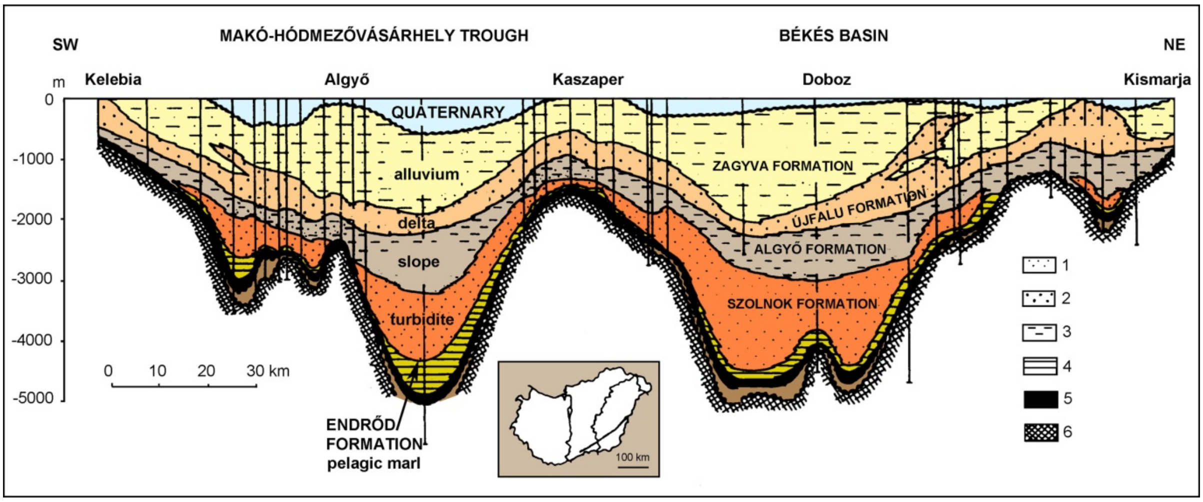

Fig. 5. Pannonian formations in the southern area of the Great Hungarian Plain showing the situation of the Szolnok Formation and Újfalu Formation (after Juhász, 1998).

Storage possibilities in depleted gas fields

Considering natural hydrocarbon fields, it is clear that geological structures can hold crude oil and natural gas (including carbon-dioxide) for millions of years. In the case of depleted gas fields, an important factor is the ability of injected CO_2 to reoccupy the pore space formerly containing natural gas. Depleted gas fields are well known in terms of the lithology of the reservoir rock, the geology of the reservoir and the production, which helps to estimate the CO_2 storage capacity. When selecting a storage site, the capacity, the injectivity and the containment have to be considered.

The use of depleted gas field for CO_2 storage has a special advantage that the injection system could be linked directly to the existing model of the natural gas production system. However, it can be a risk as well because carbon-dioxide can leak through abandoned wells. The first large-scale storage in a gas reservoir was the In Salah project in Algeria, where 1 million ton CO_2 has been injected annually into the depleted parts of the the Krechba sandstone gas reservoir. The estimated total stored CO_2 is 17 million ton (IPCC, 2005).

In Hungary a comprehensive study by the MOL and the Eötvös Loránd Geophysical Institute examined 180 hydrocarbon reservoirs for their storage capacity. The theoretical storage capacity of depleted oil and gas fields was calculated 430 million ton. These storage sites can be found in the area of the Great Hungarian Plain and are grouped into three larger blocks. It was also pointed out that 155 million ton capacity would be available within 10 years and 16 million ton for the following 25 years (Kubus, 2009).

Underground gas storage sites can also be used as CO_2 reservoirs. The CO_2 storage capacity of these storage sites is 47 million ton. However, after using an underground gas storage site as a CO_2 reservoir, it cannot be used as a natural hydrocarbon gas storage site anymore because of the impurities. That is why CO_2 storage is not planned in these sites (Kubus, 2009).

Storage possibilities by enhanced oil recovery

The technology of Enhanced Oil Recovery (EOR) has been known for decades. By the injection of CO_2 into the reservoir, the recovery of crude oil can significantly be increased. The rate of oil displacement depends on the reservoir temperature, the pressure and the composition of crude oil. About half to two-third part of the injected carbon dioxide returns with the exploited oil. This CO_2 is separated and re-injected to the reservoir.

The most well-known EOR project was established at the Weyburn Oil Field, Saskatchevan, Canada. The injection started in 2000 and about 18 million ton CO_2 is expected to inject in the lifetime of the project. That means an additional 21 million m^3 oil recovery (IPCC, 2005).

In Hungary the application of EOR technology started in the early 1970s in the Pannonian sandstone reservoirs of the Budafa and Lovászi oil fields (SW Hungary). The industrial-scale technology began to be applied in 1980 in the Nagylengyel oil field (SW Hungary), where the reservoir is karstic Triassic dolomite and Upper Cretaceous limestone. The gas for the injection was piped from the Budafa field, where the CO_2 content of the natural gas was 81 mol%. In four years, 96 million m^3 was injected and 40 600 m^3 oil was exploited (Bíró et al., 1999).

In the Szank oil and gas field (Great Hungarian Plain) the EOR technology has been applied since the 1990s. The reservoir is built up by Middle Miocene clastic sediments and the fractured upper zone of the Proterozoic crystalline basement. The CO_2 is separated from the natural gas produced from the same field. By the injection of CO_2, the oil recovery has been increase by 5-14% depending on the type of reservoir and the technology applied (Kubus, 2009).

Storage possibilities in unmineable coal fields with potential ECBM

The porosity and the consequent high adsorption capacity of coal are widely recognized. Coal seams even contain fractures that give some permeability to the system. During the coalification, volatiles like methane, carbon dioxide and nitrogen are produced by the transformation of the plant material. The amount of gas increases with increasing coalification. By the stage of black coal the methane production can be as high as100-200 m^3 per ton of coal. During the further transformation of black coal, at the anthracite stage the volatiles are driven off the coal seams (Fodor, 2006).

The carbon dioxide is preferably adsorbed onto the pore of coal to methane: for each molecule of methane two or three molecules of carbon dioxide can be adsorbed. About 90% of the methane is not physically adsorbed, rather stored in the internal coal structure. Tests have shown that carbon dioxide can be efficient elution gas to mobilize the methane. It was also pointed out that physical impact might provide the necessary energy to expel the methane from the internal coal structure (Némedi-Varga et al, 2006).

When coal adsorbs carbon dioxide, it swells, which reduces the permeability. Furthermore, coal seams have low matrix permeability, and gas can flow in the meso- and macropore cleat system, which is difficult to control.

Although there are many ECBM projects, especially in the United States, the risk of leakage cannot be clearly determined (IPCC, 2005).

In Hungary the coal fields were formed in the period of Early Jurassic to Late Miocene-Pliocene (Pannonian) (Fig. 6). Recently active mining is carried out only on the Pannonian lignite, although there are minor activities in one of the mines of the Eocene coal deposits.

Coal fields above the depth of 800 m are not taken into account as at least this depth is needed to the supercritical stage of carbon dioxide and the gas sorption capacity of coal is very low above 400 m depth because the gas migrates et shallow depths. Consequently, the Upper Cretaceous, Eocene and Miocene coal basins are not considered as CO_2 reservoirs. The Mecsek Coal Formation displays the best parameters with the potential of enhanced coalbed methane recovery. However, the deeper units of the Pannonian lignite in the Újfalu Formation could also be used for CO_2 storage (Hámor-Vidó, 2008).

The Lower Jurassic Mecsek Coal Formation is the only black coal occurrence and the largest hard coal complex in Hungary. Several underground and open pit mines were operated in the coal field from 1782 with combined annual production rate of 2,5-4 Mt. The explored but undeveloped coal resource is about 980 Mt. The horizontal extent of the coal field is 350-400 km^2 (Fodor, 2006). The coal-bearing sequence forms a complex faulted syncline-anticline fold structure, between Triassic footwall and Middle-Jurassic hanging wall sedimentary formations, hosting several Cretaceous alkaline basalt sills and dikes. The coal-bearing series is 120 to 1000 m thick, with 36 coal seams in three seam-groups (Némedi-Varga et al, 2006).

The gas content of the coal increases towards depth. Fodor (2006) estimated the coalbed methane resource of the Mecsek Coal Formation as 200 billion^3, and the amount of the exploitable methane as 28.5 million m^3. Hámor-Vidó (2008) characterized the producible gas reserves as 18.9-62.3 billion m^3, and the CO_2 storage capacity of the coal formation as 68-224 million ton.

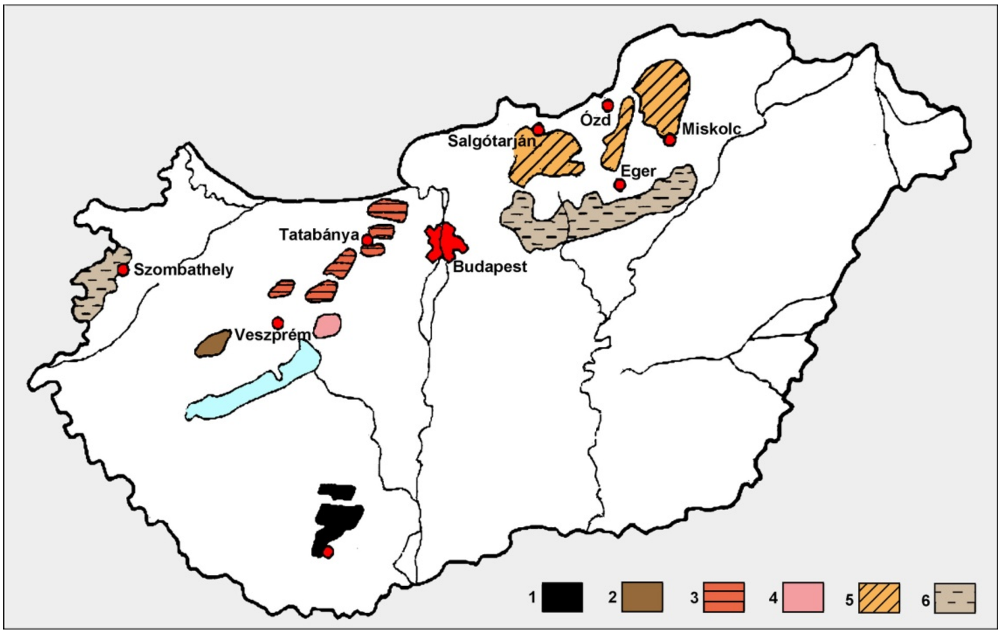

Fig. 6. Distribution of the coal fields in Hungary. 1: Lower Jurassic black coal, 2: Upper Cretaceous brown coal, 3: Eocene brown coal, 4: Miocene lignite, 5: Middle Miocene brown coal, 6: Upper Miocene-Pliocene (Pannonian) lignite (after Belláné in Fodor, 2006).

The Pannonian lignite is mined on surface in the southern foreground of the Mátra and Bükk Mountains. Because of the subsidence of the basin areas, lignite formations are in deeper positions and thicker below the Great Hungarian Plain and in the Dráva Basin. The Pannonian sequence is covered by thick Pleistocene sediments. The heteropic facies of the near-surface lignite formations is the Újfalu Formation, which has also lignite seams below the depth of 1000 m.

Methane sorption capacity of the lignite is 2 m^3/t in the deep basin areas. The lignite formations are 200-1000 km thick with several seams of lignite, of which 5-7 are economic. Lignite is generally has low methane content but can have 8-10 times higher sorption capacity for carbon dioxide than that for methane (Hámor-Vidó, 2008). According to these, Hámor-Vidó (2008) calculated 427 Mt CO_2 capacity for deep lignite formations in the area of the Great Hungarian Plain and the Dráva Basin.

Summary and conclusions

Due to the tectonic setting, the basin character and the related sedimentary processes, Hungary has the possibility for all kinds of geological storage of CO_2. As for the saline aquifers, the Pannonian (Upper Miocene-Pliocene) Szolnok Formation and Újfalu Formation meet the storage requirements (lateral and vertical extent, physical parameters). The storage capacity of these two sedimentary formations is 2000-4000 million ton carbon dioxide (Kubus 2009, Szamosfalvi et al., 2011).

The storage in depleted gas fields is also possible as the geological and volumetric data of the depleted fields are known and Hungary has gained experience in underground gas storage. The theoretical storage capacity of depleted hydrocarbon fields is estimated as 430 million ton CO_2 (Kubus, 2009).

The EOR technology has been applied in Hungary for decades, first in the SW part of the country, then in Szank, Great Hungarian Plain. However, the used CO_2 is not of industrial origin but separated from the natural gas exploited from the same field, which is much more cost-effective than using captured carbon-dioxide from power plants. Thus, the application of EOR in CCS is not planned in Hungary.

Unmineable coal seams also provide a possibility of carbon dioxide storage. The black coal seams of the Lower Jurassic Mecsek Coal Formation provide the opportunity for the ECBM technology as they contain relatively large amount of methane in the coal pores. The Upper Cretaceous, Eocene and Miocene coal fields are not suitable because of their higher position. The deep-seated, low-rank Pannonian coal formations (lignite) can also be taken into account as CO_2 reservoirs because although the lignite has relatively little amount of methane, its sorption capacity for carbon dioxide is much higher for methane. The carbon dioxide storage capacity of unmineable black coal seams and lignite formations in Hungary was calculated 500-650 million ton (Hámor-Vidó, 2008).

Summarizing all possible options, the total CO_2 storage capacity of Hungary is 3000-5000 million ton. However, this value is only theoretical. In order to achieve a more precise calculation and plan for future storage projects, detailed data on tectonics, closed reservoir structures, permeability and hydrogeology have to be collected.

Acknowledgement: *The work was carried out with the support of the TÁMOP-4.2.1.B-10/2/KONV-2010-0001 project in the framework of the New Hungarian Development Plan. The realization of this project is supported by the European Union, co-financed by the European Social Fund.*

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