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Experimental characterization of a high sulfur Hungarian brown coal for its potential industrial applications

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ABSTRACT

Differently from the conventional combustion and entrained flow gasification technologies, fixed-bed up-draft gasification process tested in pilot scale appears very suitable for the "multipurpose transformation" of Hungarian low-rank and high sulfur brown coal (for the production of electrical energy, heat and eventually gaseous or liquid fuels). This paper presents the positive results of the wide-range analysis (led jointly by Sotacarbo in Italy and by Ormosszén in Hungary) on the possibility to use the Hungarian coal from the North-East basins for energy purposes. At this stage, there is no scientific literature on this subject (the energetic use of this fuel, which is very complex in conventional power generation plants), whereas there are several studies on the geological properties of the Hungarian brown coal and on the energetic exploitation of the Mecsek basin's high-rank coal.

In addition to the conventional characterization (proximate, ultimate and thermal analyses), a series of bench-scale experimental tests were carried out to preliminary assess the combustion performance of Hungarian brown coal and to set a pilot-scale experimentation in the Sotacarbo gasification pilot plant. As results from the experimental tests, the air-blown up-draft gasification of about 11 kg/h of Hungarian brown coal allows to produce about 24–25 kg/h of raw syngas, characterized by a lower heating value of 3.55 MJ/kg.

The high sulfur content in primary fuel is one of the main problems when that coal is used. As a matter of fact, conventional cold gas desulfurization processes are typically not sufficient for an efficient syngas desulfurization. But a very high H_2S and COS removal efficiency has been experimentally obtained treating syngas with a zinc oxide-based hot gas desulfurization system. This process allowed to obtain a final H_2S and COS global concentration lower than 20 ppm (by volume), operating at about 400 °C.

1. Introduction

Coal has the largest reservoir in the world compared to the other fossil sources (oil and natural gas) [1,2] and it currently represents one of the most important energy sources with its 27.3% share (corresponding to about 3472 Mtoe — tons of oil equivalent) of world primary energy [3]. Just it happens for other fossil fuels, a small number of nations control the vast majority of the world's coal reserves [4]: at the end of 2012 the so-called "Big Six" (United States, the former Soviet Union – mainly Russian Federation, Ukraine and Kazakhstan – China, India, Australia and South Africa) controls together the 86.3% of the world's coal reserves [1].

About one-half of the overall coal reserves are constituted by low-rank coal (i.e. sub-bituminous coal and lignite), which is typically more widely distributed worldwide in comparison with hard coal, oil and natural gas. Currently, low rank coal sees little use (mainly for power generation) in spite of its large reserves and low price, because of its high moisture content and its low heating value [5,6]. Many studies [4,7–12] indicate that coal, and low rank coal in particular, will be widely used for power generation for several decades, at least until 2100.

Hungary's coal reserves amount to about 1660 Mt; 99.2% are constituted by low rank coal, with a production of 1.9 Mtoe [1]. Brown coal was widely mined throughout recent decades, supplying a significant amount of Hungary's energy needs. A simplified map of the main Hungarian coal field was proposed by Landis et al., 2003 [13] and here adapted in Fig. 1 (well explored coalfields are shown in dark gray; poorly explored coalfields are shown in light gray). Hungary's principal bituminous (high rank) coal basin is located in the Mecsek mountains, in South Hungary; coal is characterized by a higher heating value between 18.5 and 30.0 MJ/kg [14]. But other very interesting coalfields are the sub-bituminous/lignite coalfields of the mountains in Trans-Danubian and North-East Hungary [15].

Abbreviations: AFT, ash fusion temperature; AT, ash sintering temperature; DTA, differential thermal analysis; DTG, derivative thermogravimetric profile; ESP, electrostatic precipitator; FT, ash fluid temperature; HT, ash hemispherical temperature; HT, high temperature; ICE, internal combustion engine; IT, ash initial deformation temperature; LT, low temperature; MDEA, methyldiethanolamine; MEA, monoethanolamine; PSA, pressure swing adsorption; ST, ash softening temperature; TG, thermogravimetric analysis; toe, ton of oil equivalent (41.868GJ); WGS, water-gas shift.

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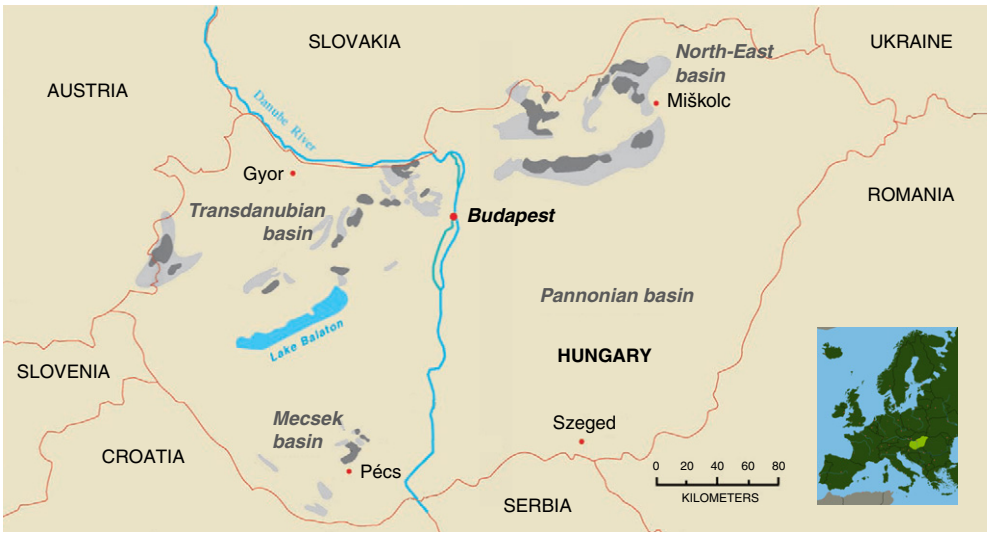

Fig. 1. Simplified map of Hungarian coal basins (adapted from [13]).

In particular, the brown coal basin located in North-East Hungary (Fig. 2, adapted from Vass et al. 2005 [16]) is characterized by a very high sulfur and moisture content and a very low heating value. Therefore, this fuel is very complex to be used in conventional power generation plants. Whereas several studies [13,14,16] investigate the properties of this coal from the geological point of view, no recent studies have been found in the scientific literature regarding the use of this fuel for energetic purposes.

During the recent years, the role of lignite has increased significantly in the primary energy sector in Hungary and the North-East brown coal basin could be a very important resource from the strategic point of view.

In this scenario, Ormosszén Ltd. and Sotacarbo S.p.A. have kicked off a strong cooperation to investigate the potential "multipurpose transformations" of Hungarian brown coal for a sustainable production of electrical energy, clean gaseous and liquid fuels and heat. This paper reports the first results of this cooperation: a wide-range characterization of the high sulfur Hungarian brown coal for synthesis gas (syngas) production through gasification. In particular, coal was preliminarily characterized through the conventional proximate, ultimate and thermal analyses. Then, coal combustion was analyzed through a thermogravimetric study and bench scale gasification tests were also performed. Finally, a series of gasification tests were carried out in the Sotacarbo pilot platform in order to verify the suitability of the Hungarian coal for fixed-bed up-draft gasification.

Fig. 2. Map of Edelény and Vadna coal basin in North-East Hungary (adapted from [16]).