17* International Mining Congress and Exhibition of Turkey- IMCET2001, ©2001, ISBN 975-395-417-4 Recent Developments and Outlook for Clean Energy from Coal without Combustion

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ABSTRACT: Development of new technologies and improvements on existing technologies are providing opportunities for coal *to* maintain its strong standing as a major competitive energy source for many centuries into the future. These technologies provide means for development of sustainable methods of production of hydrogen from coal without combustion and subsequent production of clean energy using fuel cell or turbine technology. In this paper, a review of past and current studies in this area is presented, concepts leading to design of zero emission power production are presented, and the impact of these developments on future energy and mining industries, contribution to improving environmental quality are examined. Finally, a preliminary estimate of power cost is provided.

1 INTRODUCTION

The release of greenhouse gases, primarily CO2 due to combustion of fossil fuels is a major global concern. Although, coal is not the only fossil fuel, which by combustion contributes to the continuously increasing CO2 content of the atmosphere, it is the major contributor to It per unit of energy produced. In the United States 84% of energy consumed, (81.557 10^{15} Btu, or 23.89 10^{12} kWh) comes from various fossil fuels. Specifically, coal, natural gas, and petroleum represent 23, 23, and 39% of energy consumption {DOE EIA 2000a) and contribute 549.3, 649.7, and 311.8 million metric tons/year of carbon emissions respectively (DOE EIA 2000b).

It is desirable to develop processes, which will allow the use of vast coal resources without the environmental consequences due to emission of greenhouse gases, SO2, NOx, and particulate matter. The traditional method of producing power by coal combustion and steam generation, in spite of many improvements, is losing ground simply because of the fact that conventional combustion itself leads to inefficient energy production and release of environmentally undesirable byproducts. In the following, we will examine the problems associated with production of energy by coal combustion, proposed methods for removing CO2 from the atmosphere, and review newly emerging technologies, which may provide economical and sustainable answers to these problems.

2 C0, DISPOSAL PROBLEM

Depending on the availability of local resources each country has a different mix of fossil fuel and renewable energy to meet its demand. Regardless of its impact on the global warming, CO2 produced by combustion of fossil fuels constitutes a serious problem in the long run. Since the beginning of the 19^{.h} century CO2 content of the atmosphere has risen by about 30%, from 280 ppm to 360 ppm (Siegenthaler & Oescher 1987; Keeling et al. 1995). The increase recorded during the last 40 years (from 315 ppm to 360 ppm) accounts for more than 50% of the total increase during the last two centuries. Considering the current and projected future fossil carbon consumption and the available fossil carbon resources, it is conceivable that in the distant future CO2 levels would reach intolerable levels (Yegulalp et al. 2000).

Safe and permanent disposal of CO_2 resulting from the combustion process is the key to sustainability of energy production from fossil fuels. Since coal is the most carbon intensive fossil fuel, this problem impacts the coal combustion more than other fossil fuels. In order to prevent CO2 accumulations in the atmosphere we need to collect and dispose of the combustion products. Unfortunately, as we burn coal, we produce a mixture of gases including CO, CO2, SO2, and NO_x along with remaining oxygen and nitrogen as well as particulate matter composed of carbon and ash. Separation of CO2 from this mixture for disposal is a formidable task. which makes this option technically difficult and economically undesirable (Yegulalp et al. 2000).

Here, we are not concerned with the approach of avoiding CO2 production by using other forms of energy. Although these methods can and will contribute to the reduction of CO2 emissions but energy conservation and energy efficiency will not be sufficient to meet the ever growing global demand. Alternative forms of energy that don't produce CO_2 are still far too expensive to compete. Without a major and unexpected technological breakthrough for an economically viable and sustainable carbon free energy resource, it is not realistic to expect a growing world energy market without a major contribution from coal.

Sequestration technologies can prevent the accumulation of CO2 in the air without limiting the use of fossil fuels. Sequestration can be accomplished In a variety of ways. CO2 can be collected at the point of combustion or later taken from the air. CO2 can be stored In gaseous form or could be chemically transformed before it is disposed as waste. It has been suggested that some of it can be recycled back into the economy.

2.5 Biomass Sequestration

Biomass generation has been considered as a method of sequestration. This however is a means of collecting energy, which ultimately will be wasted. It İs not feasible to store the perpetual accumulation of carbon as biomass. A mature forest will lose about as much biomass as it generates. Since biomass collection rates are very small (Ranney & Cushman 1992), one needs to dedicate unrealistic amounts of land or ocean to use this option as the sole means of sequestration of CO2 (Lackner et al. 1998; Sedjo & Solomon 1989). It İs also shown that the annual collection of carbon on an acre (0.4 ha) of land at best compensates for a couple of minutes worth of CO2 released from a one GW coal-fired power plant (Ranney & Cushman 1992).

2.6 Underground Injection

Another option could be to inject CO2 into some suitable geological formation for permanent storage. This idea is already being practiced at a limited extent for various purposes such as to dispose of the CO2 stripped from natural gas in Norway. Because of high carbon tax in Norway (\$55/t CO₂) CO₂ it is feasible to inject CO2 stripped from natural gas into an aquifer 1000 m under the sea floor in the North Sea (Kaarstad & Audus 1997). In crude oil and natural gas production, CO₂ is injected to increase production rates. A recent study (Akihiro et al. 2000) forecasts that separation, liquefaction and injection of CO2 from a coal-fired power plant in Alberta

Canada would cost from C\$56 to C\$64 per ton of CO2 disposed into an aquifer. Some CO₂ can be injected to recover methane from deep coal seams (Gunter et al. 1997) as a combinatioeisf disposal and methane recovery system. v' v'

2.70cean disposal

There are various forms of ocean disposal, which differ in how and where CO; İs introduced into the ocean (Herzog et al. 1997). CO2 can be transported in an undersea pipeline from the shore, or it can be introduced from a ship that carries it to a deep part of the ocean. It can be introduced as a compressed gas at great depth, injected as a water clathrate. it can be introduced as dry ice or bubbled into intermediate depth where it dissolves in the water. Very deep storage has the advantage that the $C0, \ensuremath{\mathsf{becomes}}$ denser than water and forms a layer on the bottom of the ocean, which only gradually dissolves into the surrounding water (Herzog et al. 2000). However, although ocean circulation guarantees that over time the highly soluble CO2 is mixed into the ocean the allowable change in pH will limit how much can be stored İn the ocean. Approximately 1000-Gt of carbon added as bicarbonate ions to the ocean would change the overall pH by 0.3 (Yegulalp et al. 2000).

2.8 Carbonate Disposal

With the exception of biomass generation, all sequestration methods propose disposing of CO2 in gas form. A new technology originally suggested by Seifritz (1990) suggests that it is possible to dispose of CO; in the form of carbonates (Lackner, et al. 1998). The technology is based on the well-known reaction of CO2 with common mineral oxides to form carbonates like magnesite or calcite. The resulting product is an environmentally safe carbonate and it is thermodynamically stable.

In nature, however, calcium and magnesium are rarely available as binary oxides. They are found typically as calcium and magnesium silicates. The carbonation reaction is exothermic for common calcium and magnesium bearing minerals. As an example, consider the following carbonation reactions of forsterite and serpentine;

$$\frac{1}{2}Mg_2S_1O_4 + CO_2 \to MgCO_3 + \frac{1}{2}S_1O_2 + 95kJ / mole$$
(1)

$$\frac{1}{3}Mg_{3}Si_{2}O_{5}(OH)_{4} + CO_{2}$$

$$\rightarrow MgCO_{3} + \frac{2}{3}SiO_{2} + \frac{2}{3}H_{2}O + 64kJ \ (mole$$
(2)

Both of these reactions are favored at low temperatures. Technologies being developed involve the use of abundant natural silicates such as serpentine and accelerate the CO2 acceptance process at an industrial scale (Yegulalp et al. 2000; Lackner et ai. 1997). At present, this process is still at an early research phase. However, recent reports indicate that significant progress has been made during the last two years (O'Connor et al. 1999; O'Connor et al. 2000).

3 HYDROGEN FROM COAL

One approach to use coal as a clean energy source is to convert carbon In the coal to hydrogen as the first step to zero emission energy production. This is based on the well-known CO2 acceptor process, which has been pioneered by Consolidation Coal Company earlier (McCoy et al. 1976; Fink et al. 1977). The basic idea Is to assist the reforming shift reactions, which make hydrogen from water by turning carbon into CO2 and use CaO to remove the CO2 from the reaction products.

Expanding on this idea, Ziock and collaborators are developing an anaerobic process for hydrogen production from coal (Lackner et al. 1999b; Yegulalp et al. 2000) in which coal, water and lime are used to form hydrogen and limestone as an intermediary. The process of producing hydrogen consists of the following four steps:

Step 1: Carbon in coal is reacted with hydrogen to produce methane, i.e.

$$C+2H_2 \rightarrow CH_4 + 75 \, kJ$$
 (3)

Step 2: Methane is reacted with water and lime to produce hydrogen and CaCOî, i.e.

$$CH_4 + CaO + 2H_2O \rightarrow CaCO_3 + 4H_2 - 75 \, kJ \tag{4}$$

Step 3: CaO is recovered by calcining CaCOî and producing a pure stream of $C0_2$, i.e.

$$CaCO_3 + 178 \ 8 \ k J \rightarrow CaO + CO_2 \tag{5}$$

Step 4: Electrical power is generated using a fuel cell or heat from combustion by combining hydrogen and oxygen, i.e.

$$2H_2 + O_2 \rightarrow 2H_2O + 571 \ 7kJ \tag{6}$$

The energy released m the fourth step is partly used for the heat needed in the third step for calcining $CaCO_3$. The remaining energy (571.7 - 178.8 = 392.9 kJ) is approximately the same amount of energy released by combustion of carbon m the coal.

$$C + O_2 \rightarrow CO_2 + 3935 \text{ M}$$

The hydrogen needed in step 1 ls produced in step 2 by recycling half of the hydrogen output in step 2. The purpose of producing intermediate methane in the first step is to keep the lime used for carbonation isolated from the impurities present in the coal. For example, the sulfur is separated between steps one and two so that methane entering into reaction with CaO is sulfur free. The net result of this four-step process is clean energy from coal with easily manageable pure waste products such as gypsum from sulfur, ash separated in the first step, and pure CO2 ready for an efficient sequestration process. Several detailed discussions of this process are available elsewhere (Lackner et al. 1999; Saunders, 2000; Yegulalp et al. 2000).

(7)

A preliminary cost estimation has been carried out as a basis for more detailed and design-based cost study (Saunders 2000). This study concluded that the cost of producing electric power using solid oxide fuel cell technology and hydrogen from coal based on the methodology outlined above could be as low as \$0.05/kWh without CO2 sequestration.

4 POWER GENERATION

Hydrogen, a clean carrier of energy, can be used in a variety of ways to generate power Conventional technologies based on combustion and subsequent steam generation as a basis for power generation are inefficient and are giving way to more efficient turbine technology or fuel cell technology to produce electric power at efficiencies in the order of 60 - 70%. It is clear that new 'energy generation complexes will have the opportunity and economic incentive to employ cleaner generation systems with higher efficiencies.

4.1 Hydrogen fueled combustion turbines

The US DOE Office of Fossil Fuels and Office of Industrial Technology are partners in a program called Advanced Turbine Systems. The program has invested in research to produce high efficiency, low cost, low emission gas turbine systems that will be commercially available in 2002. The technical goals for utility systems are: efficiency greater than 60%, emissions less than 9 ppm NO_x and 20 ppm CO2 and unburned hydrocarbons, and cost 10% below environmentally equivalent 1992 turbine systems (Layne & Zeh 1999). General Electric and Westinghouse are developing systems rated at 400MW and 420MW respectively. Hydrogen fueled turbines are considered part of the long-term plan of the *next* generation systems with anticipated employment in

2010 (Layne 1999). Utilization of hydrogen combustion turbines is associated with implementation of the Vision 21 Program (Ruth 2000) containing research and development plans for 2000 through 2008. This technology will enable Vision 21 to reach the goals of 60-70% electrical efficiency and zero emissions while producing market rate electricity by 2015. Hydrogen combustion technology is also being developed through Japanese government funded research.

The New Energy and Industrial Technology Development Organization through its World Energy Network Program is developing hydrogen energy systems. This program consists of multiple phases. Phase I (1993-1998) included the development of an optimum hydrogen combustion turbine system. The goal of the program is to reach greater than 70.9% thermal efficiency (LHV) without CO_2 , NO₄, and SO* emissions. The system must also be as reliable, available, and maintainable as current natural gas combined cycles (Bannister et al. 1997). Three corporations are involved: Westinghouse, Mitsubishi, and Toshiba.

Both Westinghouse and Mitsubishi Heavy Industries analyzed hydrogen combustion turbine systems employing current components or easily modified existing technology. The near term Westinghouse plant model had a net efficiency of 65.2% (LHV) while the optimal Mitsubishi plant design had 72.8% (LHV). However, the Mitsubishi data is from an interim report while the Westinghouse data is from the final report and refinements may change the outcome. The Westinghouse near term model plant efficiency was reported to be as high as 73.5% (LHV) İn an interim report (Bannister et al. 1997). Both systems will be considered, it appears Mitsubishi Heavy Industries has been selected for researching hydrogen fuel utilization in Phase II (1999-2003) of the WE-NET program (Saunders 2000).

4.2 Fuel cell technology

Fuel cells generate electricity and heat electrochemically like batteries. A fuel at the anode (natural gas or hydrogen or CO) and an oxidant such as oxygen or just air at the cathode is supplied. Depending on the electrolyte used we consider three basic types of fuel cells: Phosphoric acid, molten carbonate (lithium or potassium), and solid oxide (stabilized zirconia). Most of the recent research and development effort has been focused on the solid Fuel Cell technology. Currently in the U.S. research is being funded by the Department of Energy (DOE), the Electric Power Research Institute, and the Gas Research Institute. Under the DOE Vision 21 Program conceptual plant designs that would provide market rate electricity from fossil fuels with zero emissions are being developed with the target of commercialization by 2015. The goal is to produce fuel cell/gas turbine hybrids with 60% efficiency by 2003 and 70% efficiency by 2010. Additionally, 2P' century fuel cells using natural gas or coal-dertved fuels are targeted for 70% efficiency in 2010 and 80% efficiency in 2015 (Ruth 2000). The first two groups of projects were selected for Vision 21 in March and August 2000. Central to the success of meeting zero emissions criteria is the development of fuel cells.

5 A COMPLETE SYSTEM

A complete system for near-zero emission power production using coal as its raw material can be implemented in the near future as new technologies for coal processing, hydrogen production, power generation, and CO₂ sequestration become technically and economically feasible. Such a system will consist of the following components (Figure 1): (*i*) one or more coal mines, (*ii*) a coal processing and hydrogen production plant, (*Hii*) a power plant using hydrogen as fuel, (*iv*) "a CO2 processing plant producing MgCOî, (*v*) a surface or underground mine to supply raw material for CO2 processing and as a waste disposal site

J. / Coal mines

There is essentially little difference from the conventional coal mining operation. The only significant difference is that coal quality would have minimal restrictions. Since there will not be a conventional coal combustion in the subsequent stages, sulfur is not a quality factor. The amount of sulfur would however affect the consumption of limestone at the subsequent stages. It is foreseen that essentially all sulfur is converted into marketable gypsum. The ash content of ROM coal would affect the efficiency and material throughput at the hydrogen plant. Therefore, coal washing at the mine site will be the same as before.

Transportation of coal from mine site to the hydrogen plant could be in various ways. Since it is desirable to have fine coal (e.g. <-1 mm) in the hydrogen plant, it would be feasible to crush all coal and transport it as slurry by a pipeline. Otherwise crushing will take place at the hydrogen plant.



Figure 1. An example of a complete power system with CO: sequestration.

5.2 Hydrogen Plant

The plant will consist of three serially connected fluidized bed reactors (Figure 2).

5.2.1. Reactor One (Gasifier)

In the first reactor, hydrogen and carbon are reacted to form methane as described in equation (3). Since this reaction is exothermic, it is expected that some water will be added to control the temperature and to cany the excess heat energy to the second reactor where it is needed. This water could automatically be introduced since coal input stream would be moist and in the form of slurry. The primary product of this part would be methane. There will be also steam, sulfur compounds, ash, minimal CO and CO2, since there is no air introduced into the input stream. Solids are separated from gases and gases are processed through a scrubber to separate sulfur from the gas stream.

5.2 2. Reactor Two (decarbonizer)

The second reactor will receive a gaseous input stream primarily methane and steam. Additional water or steam is introduced to complete the hydrogen balance (equations 3 and 4). CaO regenerated in the reactor three is introduced here to react with methane and steam. The resulting products will be hydrogen and CaCOj. Half the hydrogen generated is recycled back to reactor one, and the remaining is sent to the power plant after a final scrubbing and filtering process. CaCOî is sent to reactor three for regeneration of CaO by calcining and reuse.



Figure 2 Concept of hydrogen and power production from coal

5.2.3. Reactor Three (Calciner)

The third reactor is a typical calcining system. The necessary heat (178 8 kj/mole) is supplied from the power plant. The product CaO is sent to reactor two, and CO2 is sent to the sequestration plant for disposal. Since CO_2 generated at this stage is pure and concentrated, the subsequent sequestration process will be much more efficient than that of separating and processing of conventional combustion gases.

5.3 Power Plant

Any one of the systems described above (Section 4) can be utilized in the power plant. The success in the development of efficient and high capacity fuel cells would determine the near and long-term configuration of the power plant It can be also feasible to transport the hydrogen produced to another site where it is converted to electrical power. However, the heat needed in the third reactor would necessitate at least some of this hydrogen to be used locally.

5.4 Serpentine Mine

In this configuration a surface or underground mine is used to provide the necessary raw material (e.g. serpentine, forsterite) to be used in the sequestration process. It is economically advantageous to place the hydrogen plant where the raw material for this process is located. This will prevent large quantities of rock from being transported to the source of CO2 or large quantities of COi from being transported to the mine site. Furthermore, the final product of the carbonation process (MgC0₃), along with sihca and other solid waste could be economically disposed off at the mine site.

A typical bituminous coal has about 70% carbon. Complete combustion of a ton of coal will yield 2.6 tons of CO2. A typical serpentine would have approximately 40 % MgO (Goff & Lackner 1998) content, which can be used to accept CO_2 Thus for every ton of coal processed one would need approximately 6 tons of serpentine to be mined and processed.

Mining method and m..hods to be used for disposal of sequestration products would strongly depend on the local geology and structure. However, because of quantities involved, costs associated with mining and crushing serpentine would be comparable to those of large-scale bulk mining costs.

5.5 Carbonation Plant

The complete system will have a carbonation plant at the site. The plant will utilize one of the two promising processes for carbonation of serpentine. The first process involves chlorination leading to production of magnesium hydrochloride, which in turn will react with CO2 to yield MgCO₃. This process is recently put to use to produce MgCb for electro winning of Mg metal from asbestos waste in Canada (Watson et al. 2000). The second process involves direct reaction of CO2 with serpentine and water at favorable temperatures and pressures to produce MgC0₃ (O'Connor et al. 1999; O'Connor et al. 2000). Both findings are promising and are expected to lead to design and implementation of plants that can be employed for sequestering large quantities of concentrated CO2.

6 ECONOMICS

A preliminary cost estimation was carried out as a basis for more detailed and design-based cost study (Saunders 2000). This study concluded that the cost of producing electric power using solid oxide fuel cell technology and hydrogen from coal could be as low as \$0.05/kWh excluding CO2 sequestration costs. Another study was carried out to assess the product markets, the technology market and the effects of clean coal technology on the related industries (Knight 2000). This study focused on SO_2 , NOx, and CO2 abatement markets and status and future projections of power needs and costs. It was concluded that in the long-term, if for sustainable development all anthropogenic carbon emissions must equal anthropogenic carbon sequestration, a competitive environment for carbon sequestration technology will exist, and carbon sequestration technologies will become a part of the global energy infrastructure

Economics of zero emission power generation should not be assessed solely on the basis of power generation at a plant but should also take into account external costs and benefits to the society as a whole.

On the cost side, we need to include the cost of coal mining and transportation to the power plants, hydrogen generation, power production with a fuel cell or turbine system, waste disposal at the power plant, and CO2 sequestration. Except for the CO?

sequestration, these costs can be estimated with some extrapolation or interpolation of current plant construction and operating costs. Sequestration cost will depend on the method employed (see Section 2). In case of sequestration by carbonation, mining cost would be of the same order of magnitude of current large-scale mining costs. Processing of serpentine rock for CO2 removal would depend highly on the results of ongoing research and development efforts.

On the benefit side, economic and societal benefits of production of power without pollution, elimination of penalties or limitations (SO2, NO x, particulate matter, fly ash disposal) for coal burning power plants, as well as elimination of CO2 emissions need to be accounted for. In addition, in case of sequestration by carbonation, products and byproducts of this process would yield marketable quantities of MgCO₃. SiOi, and iron oxides, and in case of certain serpentine types, some quantities of Cr could also be produced. For example chemical analyses of various peridotites show (Goff & Lackner 1998) that while MgO content varies from 42 to 50%, these rocks also contain 39 to 44% SiO₂ 7 to 9% FeO and FeA., 0.1 to 0.3 % NiO. 0.1 to 0.17% MnO, and 0.3 to 1.2 % Cr₃O₃ by weight.

Ultramafic rocks contain many mineral resources. Chrome, platinum group metais, nickel, cobalt, and diamonds come from various ultramafic rocks and their eroded products, whereas manganese copper, mercury, and other metals are sometimes obtained from within the bodies from enclosing rocks (Maddock 1964). Determination of the exact quantities or an average of the quantities of mineral byproducts that would be produced in the US or globally is beyond the scope of this paper. However, following an example of the Del Puerto, CA ultramafic rock formation discussed in literature (Goff & Lackner 1998), and considering the chemical composition of major elements found in a serpentinized peridotite sample from Del Puerto ultramafic body shown in Table], we can estimate that for each ton of CO2 sequestered 2.1 tons of rock needs to be processed. Table 2 shows the corresponding amount of products and byproducts of this process for each ton of sequestered C02

Table I. Chemical composition of Serpent inized Peridotite in Del Puerto Ultramafic Body".

			%	We	ight
SiO.			3	8.80)
MgO			4	2.50)
FeO				4.74	ŀ
Fe ₂ Oi				3.23	3
NiO				0.27	1
MnO				0.13	;
СЇЇОТ				0.60)
Other				9.73	;
Total			IO	0.00)
a .	~	CC 0			100

"Source Goff& lackner 1998

Table 2. Yield Per Ton of CO; Sequestered

	<u>Yield (fa)</u>
S103	815
MgCOî	1867
Fe	126
Ni	4
Mn	2
Cr	9
Other	204

Del Puerto rock body is reported to contain 33.6 Gt ultramafic rock with varying properties (Goff & Lackner 1998). With the above properties, it would sequester 16 Gt CO_2 . and yield 2 Gt iron, 70 Mt nickel, 28 Mt manganese, and 140 Mt chromium. It is clear that economic recovery of these byproducts will depend on the process utilized to treat serpentine for CO2 acception and separation of MgCOs from Fe, Cr, Ni and Mn compounds.

To set a scale for the future impact of such a development on mining and related industries, we may just look at the current coal usage for energy generation in the United States. In 1999, The United States consumed $(21.698 \times 10^{15} \text{ Btu})$ 6359 GWh energy generated at coal-fired power plants (DOE EIA 2000a). This in turn generated 549 Mt C0₂. If all of this were sequestered by mineral carbonation method using serpentine bodies similar to Del Puerto rock body as raw materia! sources, the annual stone production for this purpose would be 1153 Mt. This is in the same order of magnitude of the US crushed stone industry's annual output of 1560 Mt (USGS 2000). '

7 CONCLUSIONS

In this paper, we summarized the recent significant developments towards creation of sustainable clean energy supply systems based on coal. As these new technologies introduced at industrial scales new power generation capacities could be created using new and clean technology replacing the inefficient and polluting power plants. This is particularly significant for the developing countries, where serious power shortages need to be overcome without contributing to the production of greenhouse gases. Because of improved efficiencies on the order of 60 -70% instead of 33% at coal-fired power plants, coal mining needs would be reduced to almost half of what is needed today to meet the current energy output based on coal. This may be bad news for coal miners, but generation of a huge job and investment opportunities for mining industry at the sequestration side would easily counterbalance the anticipated loss. From resource conservation point of view, this development would extend the life of existing resources by mining less for the same energy needs,

and by allowing high-sulfur reserves to be included among the minable reserves. Implementation of hydrogen generation and hydrogen-based power plants can come to be a reality before the sequestration systems are in place. This is an interim solution for areas where atmospheric pollution from coal-fired power plants needs to be addressed urgently.

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