Effect of Temperature on Chemical Composition of Oil from Direct Liquefaction of Mukah Balingian Coal in Tetralin-Glycerol Mixed Solvent System

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ABSTRACT

Coal-derived liquids or oils from coal liquefaction are an attractive commodity as they could be a source of transportation fuels and feedstocks for the chemical industry. Analysis of the chemical composition is essential to determine the suitable usage of the liquids from the coal liquefaction process. The common instruments used for this purpose are GC-MS, FT-IR and ^{1}H NMR. In this study, the chemical composition of oils produced from direct liquefaction of Mukah Balingian coal in a tetralin-glycerol solvent system was carried out using GC-MS. This work aims to investigate the effect of liquefaction temperature on the qualitative and quantitative aspects of the oils produced. Although all of the oils contain similar trends of chemical composition, the per cent areas of these compounds showed a notable change due to different temperatures used during the liquefaction process. The liquefaction condition is of low pressure and high temperature, so the aromatisation reaction is favoured. As a result, the aromatic components in oils were increased as the temperature was raised due to the predominate reaction of dehydrogenation, thus producing more aromatics of hydrogen-depleted compounds.

Keywords: *Coal, liquefaction, coal-derived liquid, oil, temperature, GC-MS*

INTRODUCTION

The crisis of petroleum depletion has called for many efforts to develop substitutes. Coal is a recognised substitute due to its abundance and wide availability worldwide. Coal liquefaction has always been considered a cleaner method of converting solid fuel into liquids to produce fuel and other chemicals for various uses. Obtaining liquid fuel from coal through coal liquefaction can be performed by two routes - direct liquefaction (DCL), in which solid coal is directly converted to liquid fuel, while indirect liquefaction (ICL) first converts solid coal to gas before turning it further to liquid form [1]. By direct liquefaction (DCL), most of the coal structures initially present were retained.

In contrast, indirect liquefaction (ICL) destroyed the organic structures to finally produce a mixture of gases containing CO and H_2 [2]. Due to the issue of emission of concentrated carbon dioxide from ICL plants, more preferred technology to be used for coal liquefaction is DCL [3]. DCL is a process that involves coal cracking, often carried out at high temperatures and pressure [4]. Following DCL, three types of products will be obtained: oil, asphaltene and preasphaltene. The high energy density owned by liquid hydrocarbons such as coal-derived liquids makes them suitable as transportation fuels and the chemical industry's feedstock [5].

Coal consists of the primarily naphtalenic structure. Due to that, most oil components produced contain a high amount of aromatic content, majorly from naphthalene and benzene. Naphthalene is an abundant component readily present in the primary tar. In addition, its formation does not require reactions involving hydrocarbon growth [6]. As consecutive reactions proceeded, indene and toluene were produced, with the final and relatively stable product obtained as benzene. Other than naphthalene and benzene, another compound found in the oil component is a phenol [7].

Additionally, several reactions caused these molecules to polymerise, thus producing polycyclic aromatic hydrocarbons (PAHs). Ziegler and Robbat Jr [8] cited that PAHs are readily found in fossil fuels and their by-products. Aside from being the primary source of phenol, coal tar also serves as the primary source of nitrogenous compounds (indole, carbazole, pyridine and quinoline) in the chemical industry [9].

Due to the low boiling point of the sample, the preferable instrumentation to be used to identify the components in coal-derived liquid or oils is Gas Chromatograph-Mass Spectrometer (GC-MS). GC-MS has been used to evaluate the selective hydrogenation of aromatics in coal-derived liquids over NiW and NiMo carbide catalysts [10]. GC-MS was paired with ^{1}H-NMR analysis in another work to demonstrate the hydrogen addition mechanism in oil+gas products from the co-liquefaction of Elbistan lignite and manure [11]. Pyrolysis-GCMS (Py-GCMS) has been employed for studying Indian coals and their solvent-extracted products [12] and to investigate the compositions of volatiles from pyrolysis of different extracts of low-temperature coal tar in six-stage processes [13].

Most works related to coal liquefaction focus on the effect of temperature on conversion and the types of components in oils. In contrast, only several works have reported on the effect of liquefaction temperature on the composition of oils produced from DCL. In general, products from DCL are rich in PAHs and heteroatoms [14]. As liquefaction temperatures being varied, the components in oils also being affected. Rahman et al. [15] reported that coal liquid from Canadian Bienfait lignite is progressively becoming hydrogen-depleted as liquefaction temperature increases as polycondensation reaction dominate the reaction, suppressing active thermolysis reaction. For oxygenic compounds, lower temperature promotes the formation of ketones rather than carboxylic acids [16]. It was reported that high-temperature usage allows more oxygenic compounds to be concentrated in oils due to a decarboxylation reaction that removes oxygenic compounds from the coal structure. Nonetheless, increasing liquefaction temperature causes a reduction in the acid content of the oils due to degradation [17].

In this study, GC-MS was used to investigate the effect of liquefaction temperature on the chemical composition of oil from DCL of Mukah Balingian coal using a tetralin glycerol mixed solvent system. Previous work [18] has delineated that tetralin-glycerol solvent produced oils with a combination of polar and non-polar components. This study is an extension work which further explores the effect of using this co-solvent at different temperatures on the oil composition.

EXPERIMENTAL

Materials

Low-rank coal used in this study was Mukah Balingian (MB) Malaysian coal. The coal was pulverised to less than 212 µm and dried in a vacuum oven at 100 °C overnight to remove inherent moisture. Table 1 shows the characteristics of MB coal.

Table 1: Characteristics of MB coal sample

Ultimate analysis (wt. % daf)		Proximate analysis (wt. %)	
Carbon	60.8	Volatile matter	42.72
Hydrogen	4.9	Fixed carbon	48.77
Nitrogen	1.6	Ash content	4.02
Sulphur	0.2	Moisture	3.08
Oxygen*	32.5	Fuel ratio[#]	1.14
H/C ratio	0.95	Calorific value	21.6 MJ/kg

*calculated by difference #fixed carbon/volatile matter

Liquefaction experiment and product separation

The liquefaction experiments were carried out in a 300 mL batchwise reactor system. In each run, 10 g of dried MB coal with 100 mL of the tetralin-glycerol co-solvent system in 70:30 (% v/v) were charged in the vessel with a steel ball added. Figure 1 shows a schematic diagram of the reactor system used in this study.
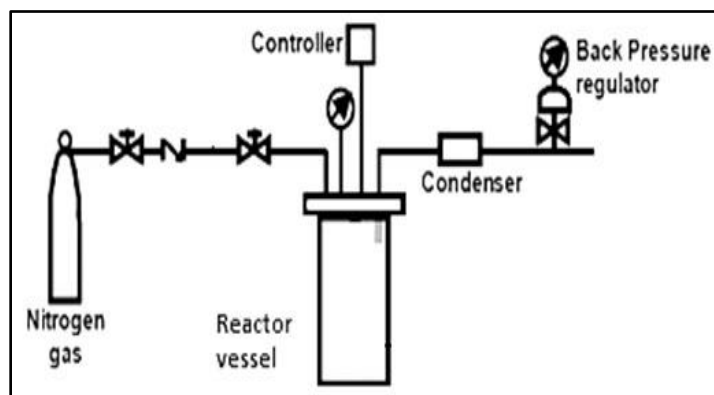
Figure 1: Schematic diagram of batchwise reactor system

The liquefaction works were carried out at temperatures of 380, 400, and 420 °C with a constant pressure of 4 MPa. Liquefaction experiment started with the leak-check test, followed by introducing nitrogen into the system and heating to the desired temperature. After 30 minutes of reaction, the reactor vessel was rapidly cooled to room temperature in an ice bath. The reactor vessel was then depressurised to ambient pressure. The products in the vessel and condenser were collected, washed with tetrahydrofuran (THF) and filtered to obtain crude tar with coal liquefaction residue (CLR). The crude tar was then separated into oil (hexane-soluble), asphaltene (toluene-soluble) and preasphaltene (THF-soluble) by sequential Soxhlet extraction method. The oil is the product of interest for analysing chemical composition using GC-MS.

GC-MS analysis of oil component

GC-MS used in this study was from Agilent Technologies, Model 6890N and MS Model of 5973Network. GC Capillary column used was of DN-5MS with dimensions of 30 m x 0.25 mm x 0.25 µm, with split or splitless type of auto-injector. Prior to analysis, the oven temperature was set at 60 °C to 270 °C with a heating rate of 5 °C/min. The carrier gas used was helium gas at a flow rate of 20 mL/min, and the injector type used was split mode, with a 1:30 ratio. For

chromatogram analysis, obtained peaks were identified using the National Institute of Standards and Technology (NIST) 05 Library software.

RESULTS AND DISCUSSION

Effect of liquefaction temperature on peaks distribution

Figure 2 shows a chromatogram of oils from the liquefaction of MB coal at different temperatures. At 380 °C, few peaks were observed in the chromatogram, and as the temperature was raised to 400 °C, almost double the number of peaks were observed in the oil produced, with few apparent peaks observed at around 24 to 32 minutes. As the temperature was further increased to 420°C, the oil produced contained more compounds. Peaks around 24 to 32 minutes of retention time were not observed, while more peaks were eluted at around 35 to 40 minutes.

From these chromatograms, it was deduced that increasing liquefaction temperature caused the oil chromatogram to contain a higher number of peaks, indicating oil obtained has a more significant number of compounds. This reflects that higher thermal treatment promotes cracking [15]; hence, more compounds from the coal are released and contained in the oil. Peaks with higher intensity were eluted at longer retention times, indicating that the oils contained heavier materials as liquefaction temperature increased.

Effect of liquefaction temperature on the chemical composition of oil

Researchers have analysed coal liquids to evaluate the component produced following the coal liquefaction process. The analysis allows the coal liquid's chemical component to be identified, which can serve as a foundation for turning the products into other uses, such as liquid fuel and raw chemical materials [19]. Compared to petroleum products, products of DCL contain a higher number of cyclic alkanes, less linear alkanes and relatively high concentrations of aromatic and naphtheno-aromatics [20].

In addition, heteroatom-containing compounds such as oxygen- and nitrogen-containing compounds are more dominant than sulfur-containing species in DCL products [21]. In this section, an analysis of oil from MB coal liquefaction using GC-MS was carried out to investigate the effect of liquefaction temperature on the qualitative and quantitative aspects. For a more straightforward analysis, the components in oil were characterised into different categories, namely aliphatics, aromatics, oxygenic compounds, nitrogenous compounds and sulfuric compounds. Table 2 presents the detailed characterisation of oils with several different types of compositions, and their respective per cent are at different temperatures.
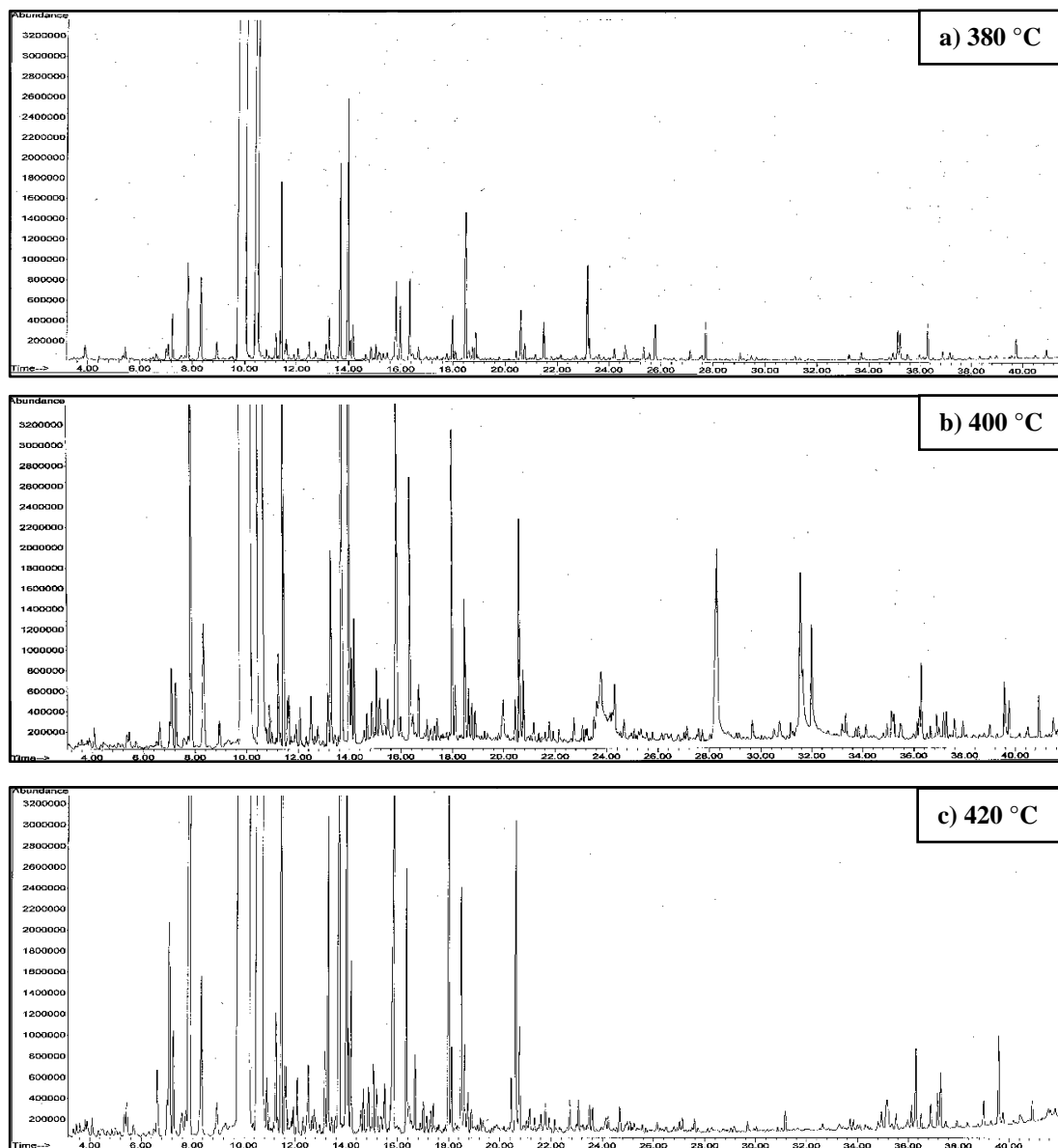
Figure 2*:* Chromatograms of oils from liquefaction of MB coal using 70:30 (% vol/vol)
the tetralin-glycerol solvent system at different temperatures

Table 2: Categories of Components and Percent Area in Oils Obtained from Liquefaction[1] of MB Coal Using 70:30 (% vol/vol) Mixed Tetralin-Glycerol Solvent System at Different Liquefaction Temperature

| Categories | Compositions | Liquefaction temperature (° C) | | | | | |
| | | 380 | | 400 | | 420 | |
		Types count	Per cent area (%)	Types count	Per cent area (%)	Types count	Per cent area (%)
Aliphatics	Alkanes	-	-	2	0.18	3	0.24
	Alkenes	1	0.16	1	0.05	2	0.08
Aromatics	Benzene	6	8.30	18	7.10	26	12.75
	Naphthalene	24	75.65	27	70.27	26	72.17
	Indene	6	7.39	6	1.52	5	4.39
	Indane	1	0.26	-	-	1	0.44
	Fluorene	1	0.33	1	0.52	2	0.52
	Anthracene	-	-	-	-	1	0.16
	Phenanthrene	-	-	3	0.48	5	0.56
	Pyrene	-	-	4	0.41	3	0.23
	Perylene	1	0.13	1	0.08	1	0.25
	Binaphthalene	2	1.14	4	1.56	5	2.04
Oxygenic compounds	Alcohols	1	0.78	8	1.71	7	1.10
	Phenols	4	3.65	12	2.36	14	3.24
	Aldehyde, ketone	2	1.07	6	0.90	6	0.76
	Acid, ester	-	-	11	11.89	2	0.14
	Pyran	1	0.42	-	-	-	-
	Oxirane	-	-	-	-	1	0.14
	Oxirene	-	-	-	-	1	0.26
	Oxepin	-	-	1	0.08	1	0.07
Nitrogenous compounds	Amine	1	0.26	2	0.23	2	0.18
	Pyrazole	-	-	1	0.10	-	-
	Imidazole	-	-	1	0.10	-	-
	Nitrile	1	0.46	2	0.46	2	0.28
TOTAL		**52**	**100**	**111**	**100**	**116**	**100**

[1]Pressure = 4 MPa nitrogen; Reaction time = 30 minutes; Coal-to-solvent ratio = 1:10

In general, as liquefaction temperature increased from 380 – 400 °C, aromatic content decreased while oxygenic content increased. Seeing that aliphatics and nitrogenous compounds also slightly increased, this observation probed the possibility that the aromatic components were thermally broken down to form other categories of a compound due to the usage of higher temperatures. As MB coal is low-rank coal with a high presence of oxygen in its structure and thermal effect, more oxygenic components were observed in oil at this temperature. An almost similar trend in categories of compounds found in the oil was observed as the temperature was further increased to 420 °C. However, at 420 °C, aromatics dominated the oil, with oxygenic compounds drastically reduced as the second highest compound, followed by nitrogenous compounds, while aliphatics were produced at the smallest amount.

Despite the similar trend in categories of the compound produced, detailed characterisation of the oils showed that the number of compounds increased as the liquefaction temperature increased. According to the NIST 05 Software, lower temperatures produced 52 compounds, which doubled as temperature increased to 400 °C. Additional liquefaction temperature to 420 °C produced oil with a small increment of compounds to 116. This reflects that high temperature allowed more minor compounds with lower molecular weight to be obtained in liquid yield through thermal and hydrocracking reactions [15].

All the oils produced following liquefaction at three different temperatures were mainly aromatics. Mzinyati [22] states that low pressure and high temperature promoted aromatisation via two pathways. Firstly, by dehydrocyclisation of *n*-paraffins to give aromatic compounds (an endothermic and slow process) and secondly, by dehydrogenation that occurred as a result of reversibility of the hydrogenation reaction of aromatics. This high aromaticity content of oil also could be explained using thermodynamic investigation. It was revealed that high partial pressure of hydrogen and low temperature favours hydrogenation, while low hydrogen partial pressure with high-temperature favours dehydrogenation. Dehydrogenation is an endothermic and fast reaction. As the oil produced at all temperatures tested was shown to contain very high aromatic content, it was expected that dehydrogenation predominates the reaction in liquefaction of MB coal using 70:30 (% vol/vol) tetralin-glycerol solvent system at 30 minutes of reaction time at all temperatures investigated.

GC-MS analysis of the oil showed that increasing temperature caused aromatic content to increase. Rahman et al. [15] also observed that increasing temperature caused an increment in aromatic content in coal liquids producing in liquefaction of Canadian Bienfait lignite using an industrial hydrotreated solvent. This phenomenon could be explained by understanding different hydrogen consumption at different temperatures. At low temperatures, minor hydrogen consumption occurred with the internal reorganisation of hydrogen predominates. During this temperature, hydrogenation is compensated for by the hydrogen liberated in condensations. As the temperature is raised, the net consumption of hydrogen increases due to bond cleavage reactions and aromatisation reactions becoming more pronounced [23], thereby increasing the aromaticity of the oil produced.

The primary compound, naphthalene, showed an almost comparable per cent area in all analysed oils. However, as the temperature increased, a slight reduction of naphthalene with increasing benzene content was observed. This indicated that the higher temperature interrupted the stability of naphthalene's structure, causing minor degradation. Benzene, otherwise, is a final and stable product. Due to this, it was observed that the per cent area of benzene in oil increased with different temperatures. Indane originated from the cleavage of covalent C-C bonds interconnecting bicyclic aromatic rings rather than the result of condensation reactions of aromatic rings occurring at higher temperatures [17]. This could explain the reasoning behind the appearance of indane in oil as the liquefaction temperature increased to 420 °C.

Other than naphthalene, benzene and indane, these oils also contained PAHs of different rings. Materials such as petroleum and various tars commonly consist of PAH as their component. Coal conversion processes such as gasification, carbonisation, combustion and liquefaction released PAHs [24]. Either naturally occurring in fossil fuels or as by-products following fuel processing, PAH existed in mixtures. Among the standard components in PAH mixtures are anthracene and phenanthrene. These two components are isomers with a chemical formula of $C_{14}H_{10}$. Anthracene, however, was less observed in oils from coal liquefaction due to its volatile characteristics [25].

Based on the increasing per cent area and ring sizes of PAHs as liquefaction temperature increases, it could be deduced that higher temperature positively affects the formation of PAHs. Furthermore, the complexity of aromatics was witnessed with the per cent area of higher-ring PAHs such as perylene and binaphthalene were improved as temperature increased. Dong et al. [24] observed a similar situation in which their experimental results of coal pyrolysis showed that increasing temperature had also caused the amount and large size PAHs to increase. They concluded that the formation of PAHs at low-temperature pyrolysis mainly from the aromatic structure within coal and thermolysis of coal macromolecular. As higher pyrolysis temperature is applied, PAHs could be formed by synthesising and polymerising smaller molecules.

The alcohols/ethers, mainly aromatic species, such as naphthalenol, were observed with the highest per cent area at the lowest temperature for oxygenic compounds. Like alcohols/ethers, the highest fraction of carbonyls were also observed at the lowest temperature. Cao et al. [16] inferred that lower temperature promotes the formation of ketones rather than carboxylic acids. This supports the GC-MS analysis, which showed no presence of acids/esters content in oils at 380 °C and began to be produced extensively as the temperature was raised to 400 °C. This reflects an increment of temperature, which promotes decarboxylation that effectively eliminates oxygenic compounds from the organic structure in the coal sample. As a result, these oxygen compounds were released and concentrated in the oil. Acids, however, decreased in type and per cent area as the temperature was increased to 420 °C, as they degrade at high temperatures. This trend was also observed in a previous study by Lievens et al. [17].

This phenomenon also probes the possibility that parts of the acid participated in acetal formation from the reaction of an aldehyde with alcohol. This was supported by the existence of

acetal oxygenic compounds such as oxirane, oxirene and oxepin, which were unique to oil of 420 °C. In addition, it was also observed that ketones were obtained at a higher per cent area than aldehyde. Cao et al. [16] also observed ketones in bio-oil produced when glycerol was used as a co-solvent in the hydrothermal liquefaction of rice straw. They hypothesised that glycerol addition in the solvent system facilitates the production of ketones. In this study, it was suspected that a similar role of glycerol as a co-solvent had occurred. Together with the involvement of more reactive aldehyde with other chemical compounds, ketones were observed at higher content in oils.

It was observed that amine and nitrile were produced in oils of all temperatures for nitrogenous compounds. Amines and nitriles were among the nitrogenous compounds commonly identified in products of coal pyrolysis, gasification and liquefaction [26]. Nevertheless, their content gradually decreased as liquefaction temperature increased. Seeing the per cent area of aliphatics increased, higher liquefaction temperature might have thermally cracked the bonds within the structure to produce more aliphatic compounds.

CONCLUSION

Oils from DCL of MB coal in the tetralin-glycerol solvent system under different liquefaction temperatures have shown an almost similar trend of compound categories. However, the effect of temperature was observed as the number of compounds in oils increased with an increase in temperature reflected in thermal and hydrocracking reactions. Regarding composition, aromatics are majorly produced in all temperatures investigated, but the per cent areas were revealed to be decreasing as the liquefaction temperature increased. PAHs also showed an increment of per cent areas due to polymerisation reaction occurring at higher temperatures. Oxygenic compounds such as ketones are more concentrated in oils produced using higher liquefaction temperatures, while the acids degrade under similar conditions. Nitrogenous compounds are present in all temperatures, with the amount gradually decreasing as liquefaction temperature increases due to more cracking reactions to produce more aliphatic compounds.

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AUTHOR'S CONTRIBUTION

Siti Nurlia Ali carried out the research and wrote and revised the article. Mohd Azlan Mohd Ishak and Khudzir Ismail designed the research, supervised the progress, and provided suggestions. Ali H. Jawad conceptualised the central research idea and provided the theoretical framework, while Nur Nasulhah Kasim assisted in the content and formatting of the article.

CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted without any self-benefits or commercial or financial conflicts and declare the absence of conflicting interests with the funders.

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