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Coal-Based MEG Production by Ube Process

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Abstract

This review presents a technoeconomic evaluation of a newly commercialized monoethylene glycol (MEG) production process developed by Ube Industries Ltd. of Japan. The route and process, if they succeed commercially and achieve the desired level of product purity and catalyst stability on a sustained basis, could have a huge impact on the MEG industry with the possibilities of switching its production from the current ethylene-based source to a natural gas or coal-based source. The technology is divided into two principal steps: 1) the dimethyl oxalate (DMO) production process, which was refined to a commercial level by Ube; 2) the monoethylene glycol production process, which was originally developed by Ube but process updating and refining was done jointly by HighChem Company, Ltd. and East China Engineering Science & Technology Company, Ltd., based on operating data gathered from their pilot plant.

The process consists of several stages of reaction and separation of the resulting products. In the initial step, CO and methyl nitrite (CH₃ONO) react in a gaseous phase in the presence of a proprietary palladium catalyst producing a mixture of dimethyl oxalate (CH₃COOCOOCH₃) and nitric oxide (NO). The two reaction products are separated by condensing the dimethyl oxalate in a methanol-scrubbed column. Nitric oxide, along with unconverted CO and other associated light gases, leaves the scrubber column from the top in gaseous form.

In the second step, nitric oxide is reconverted back to methyl nitrite by reacting with methanol and oxygen. Water is also formed in this reaction. The reaction takes place in a countercurrent gas-liquid column with nitric oxide and oxygen entering from the bottom and methanol sprayed down the column from the top. The light gases (mainly unconverted NO, CO, CO₂, and N₂) leaving from the top of the column are cooled and, after purging a small portion, recycled to the first stage dimethyl oxalate reactor along with regenerated methyl nitrite. An aqueous solution of methanol is removed from the bottom of the regeneration column. Small amounts of higher oxides of nitrogen are also formed in the reaction, resulting in the loss of NO. Hence, a means is provided in the process to produce nitric oxide to make up for that loss.

The third reaction step consists of dimethyl oxalate conversion to ethylene glycol (EG) in a hydrogenation step in the presence of excess H₂. Methanol is regenerated in this reaction. This is a vapor-phase process in which a proprietary Cu-Cr-Ba (or Cu-Cr-Zn)-based catalyst is used. Some impurities/by-products such as methyl glycolate, dimethyl carbonate, methyl formate, 1,2-butanediol, etc., are also produced in small amounts. While most of the impurities are removed from the EG without a problem, 1,2-butanediol (BDO) separation possibly poses a difficulty in producing a fiber-grade EG due to the closeness of their boiling points. Product separation/refining is done through a series of distillation towers.

Process economics for standalone and integrated (with coal gasification/syngas) EG plants are presented in this review. Comparative economics with the Fujian process are also given (Tables 11–14).
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