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Conceptual design of the iodine–sulfur process flowsheet with more than 50% thermal efficiency for hydrogen production

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ABSTRACT

A conceptual design of a practical large-scale plant of the thermochemical water splitting the iodine–sulfur (IS) process flowsheet was carried out as a heat application of Japan Atomic Energy Agency's commercial Gas Turbine High Temperature Reactor 300 MW for Cogeneration (GTHTR300C) plant design. Innovative techniques proposed by JAEA were applied for improvement of hydrogen production thermal efficiency; depressurized flash concentration of H_2SO_4 using waste heat from Bunsen reaction, prevention of H_2SO_4 vaporization from a H_2SO_4 distillation column by introduction of H_2SO_4 solution from the 2nd flash bottom, and I_2 condensation heat recovery by direct contact heat exchange in an HI distillation column. A simulation of material and heat balance was made using PRO/II, a commercial chemical process simulator. The result demonstrated that hydrogen of about 31,900 Nm^3/h would be produced by 170 MW heat from the GTHTR300C. A process thermal efficiency of 50.2% would be achievable with incorporation of the innovative techniques and the following high performance components expected in future R&D; an electro-electrodialysis cell stack, a reverse osmosis membrane, a HI decomposition reactor incorporated with a H_2 permselective membrane, and heat exchangers.

1. Introduction

High Temperature Gas-cooled Reactors (HTGR) are expected for many applications taking advantage of their high temperature helium coolant of maximum 950 °C. Electricity generation, hydrogen production, process steam supply, and waste heat utilization are anticipated utilizing heat of various temperature ranges. Fig. 1 illustrates examples of heat applications. Japan Atomic Energy Agency (JAEA) has carried out R&D on such heat applications as electricity generation (Sato et al., 2014), hydrogen ironmaking (Kasahara et al., 2014), desalination (Kamiji et al., 2014), and district heating (Kasahara et al., 2016).

Hydrogen production is one of the most intensively studied applications in JAEA. The iodine–sulfur (IS) process is selected as the candidate hydrogen production method considering its potential to produce hydrogen with high thermal efficiency from water, which does not emit CO_2 from neither material source nor heat source. The IS process consists of the following three chemical reactions. H_2O is decomposed thermally into H_2 and O_2 in total; iodine (I) and sulfur (S) compounds cycle within the process.

$$I_2 + SO_2 + 2H_2O \rightarrow H_2SO_4 + 2HI \text{ (Bunsen reaction)}$$

$$H_2SO_4 \rightarrow H_2O + SO_2 + 0.5O_2$$

$$2HI \rightarrow H_2 + I_2$$

The highest temperature requirement in the process is 800–900 °C for H_2SO_4 decomposition. The demand matches the maximum temperature of the HGTR helium coolant of 950 °C. At present, a test facility made of industrial materials has been constructed, which is designed as 100 NL/h scale hydrogen production. An 8-h continuous operation tests are successfully demonstrated in February 2016 (Noguchi et al., 2016; Tanaka et al., 2016). Preparation of a longer-term test is now under way. After the success of the test, demonstrations of heat supply from the High Temperature Test Reactor (HTTR), a test reactor of HTGR in JAEA, to a helium gas turbine (GT) and the IS process is planned. Designing of a HTTR-GT/H2 plant for the plan is in progress (Yan et al., 2016). Technology obtained in the test will be transferred to private companies for commercialization.

A flowsheet analysis of the IS process using heat from a HTGR was performed. For the H_2SO_4 section, 3 stages flash drums and a direct contact heat exchanger (DCHX) H_2SO_4 concentrator were used. Electro-electrodialysis (EED) cell stacks and reverse osmosis (RO) membranes were applied for HI concentration. Removal of I_2 from HI decomposition field by reaction with Co was then utilized to enhance HI conversion ratio. Total heat input was standardized to 170.0 MW_t, the heat supply from Gas Turbine High Temperature Reactor 300 MW for

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Nomenclature

n_{H2}	hydrogen production rate, mol/s
Q	total heat input to the IS process, MW
t_+	H^+ transport number, molar ratio of H^+ permeation rate through a cation exchange membrane in the cell to electron reacted at electrodes, dimensionless
W	total electricity input to the IS process, MW
β	electroosmosis coefficient, molar ratio of H_2O permeation rate to H^+ permeation rate, dimensionless
ΔH_{H2}	higher heating value of H_2, (=0.2858 MJ/mol)
η	hydrogen production thermal efficiency, %
$\eta_{el.}$	net electricity generation efficiency, %

Subscript

e	electricity
t	heat

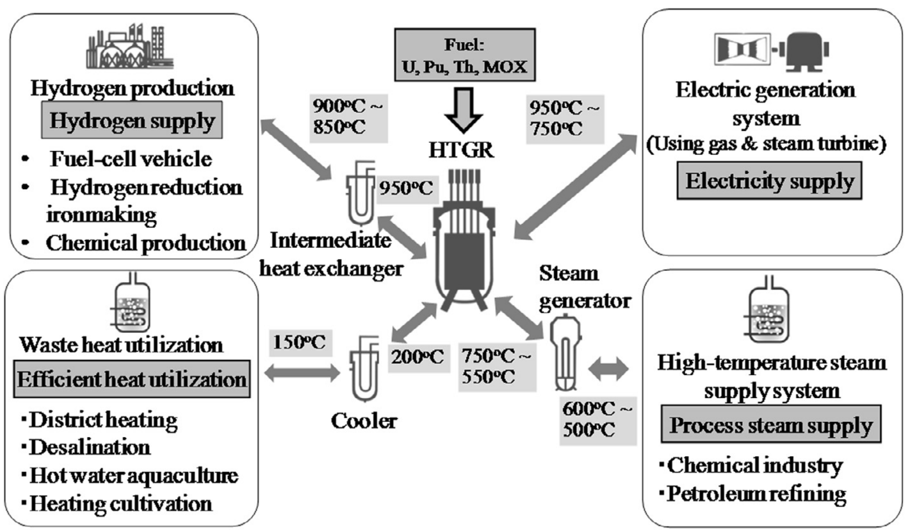

Fig. 1. HTGR heat applications.

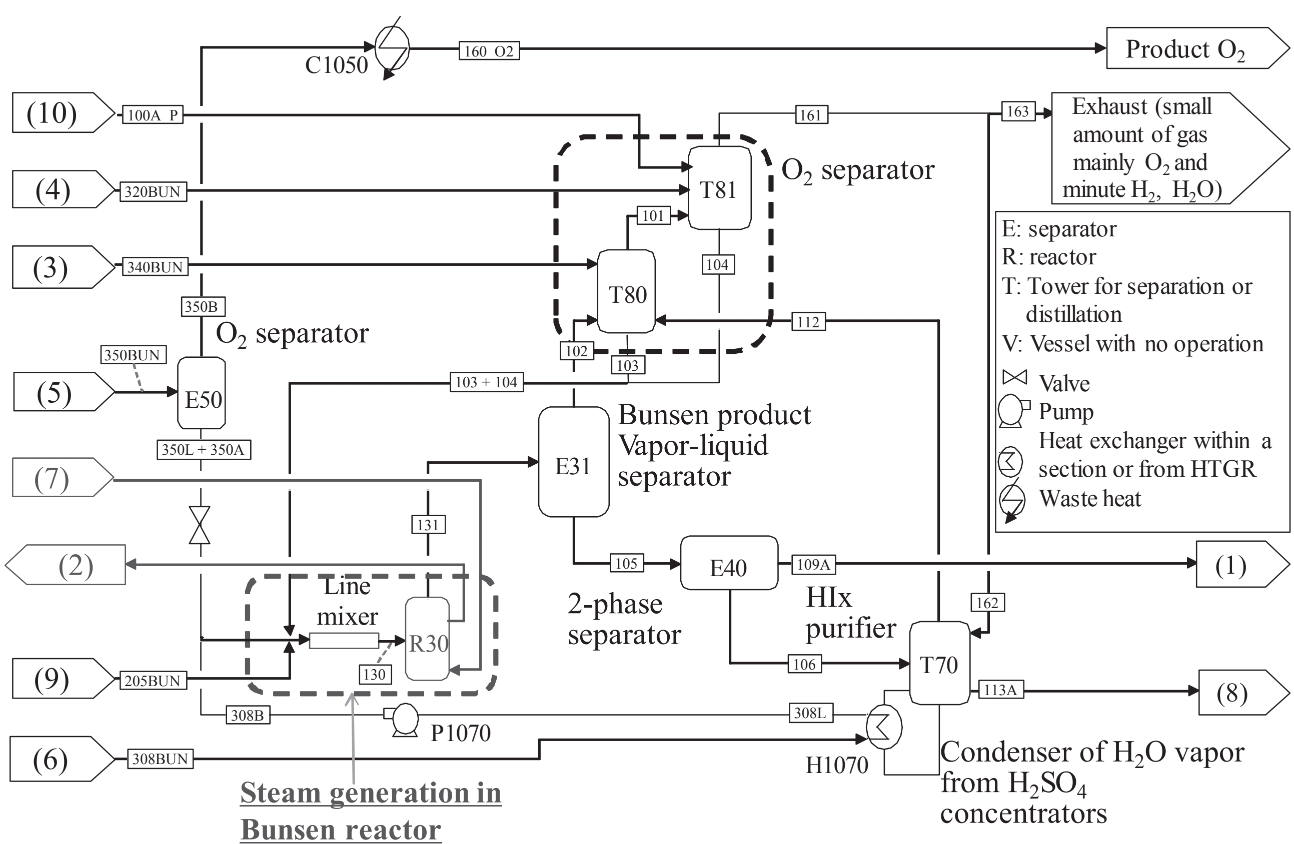

Fig. 2. Bunsen section flowsheet. The component with underline is a newly added one in this study. Numbers with parentheses mean stream connection to Figs. 3 and 4.