Nitrogen Expander Cycles for Large Capacity Liquefaction of Natural Gas

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Abstract. Thermodynamic study is performed on nitrogen expander cycles for large capacity liquefaction of natural gas. In order to substantially increase the capacity, a Brayton refrigeration cycle with nitrogen expander was recently added to the cold end of the reputable propane pre-cooled mixed-refrigerant (C3-MR) process. Similar modifications with a nitrogen expander cycle are extensively investigated on a variety of cycle configurations. The existing and modified cycles are simulated with commercial process software (Aspen HYSYS) based on selected specifications. The results are compared in terms of thermodynamic efficiency, liquefaction capacity, and estimated size of heat exchangers. The combination of C3-MR with partial regeneration and pre-cooling of nitrogen expander cycle is recommended to have a great potential for high efficiency and large capacity.

Keywords: LNG, Liquefaction, Mixed Refrigerant, Nitrogen, Expander
PACS: 07.20.Mc

INTRODUCTION

A variety of thermodynamic cycles have been developed for the liquefaction process of natural gas [1,2] in order to achieve high efficiency and large capacity. The most popular liquefaction process under operation is based on propane pre-cooled mixed refrigerant (C3-MR) cycle by the Air Products and Chemicals Inc. (APCI). The natural gas feed is pre-cooled from ambient temperature to approximately 240 K by a multi-stage propane (C3) JT (Joule-Thomson) cycle, and then condensed and sub-cooled to LNG temperature by a mixed refrigerant (MR) JT cycle, as schematically shown in Fig. 1(a). The entropy generation due to temperature differences in the heat exchangers can be minimized with an optimized composition of MR, attaining high thermodynamic efficiency with a small number of individual components. The maximum capacity of C3-MR process is roughly 5 MTPA (million tons per annum) due to the technical limits on propane compressor and heat exchanger sizes. The dual mixed refrigerant (DMR) process is another efficient cycle, where a separate MR cycle is used for pre-cooling, as shown in Fig. 1(b). The DMR process has an even smaller number of individual components than the C3-MR process, but has a comparable level of thermodynamic efficiency when two MR compositions are optimized as described in [2].

In an attempt to impart a substantial increase in liquefaction capacity, the C3-MR process has been modified lately by the APCI [3,4]. The modification is the addition of a nitrogen (N2) expansion (i.e. reversed-Brayton) cycle for sub-cooling at the cold end, as shown in Fig. 2(a). This combination of three cycles (C3-MR-N2) is called AP-X Process. With the aid of N2 sub-cooling, the AP-X process can achieve the largest liquefaction capacity (up to 8 MTPA) with existing equipment at reasonable production cost. On the other hand, the thermodynamic efficiency may be smaller in comparison with the C3-MR or DMR processes.

This thermodynamic study intends to investigate the combination of the N2 expander cycle with the C3-MR and DMR processes, simultaneously aiming at high efficiency and large capacity. These existing and modified cycles are simulated with commercial software (Aspen HYSYS) for comparison in terms of thermodynamic efficiency, liquefaction capacity, and system size. Similar studies have been performed for various combined cycles of C3-JT, C2-JT, and N2-Brayton cycles by the present authors [5], and the same approach is utilized in this work.

This study is part of ongoing efforts [5-8] under a Korean governmental project supported by the LNG Plant R&D Center, which was established in 2008 with an eight-year program to prepare the competition for worldwide LNG plant market in the near future. Many of newly proposed LNG cycles are patented domestically or internationally, and two reduced scale test beds are under operation or construction at the Incheon LNG site of the Korea Gas Corporation.

Advances in Cryogenic Engineering
AIP Conf. Proc. 1573, 1652-1657 (2014); doi: 10.1063/1.4860905

MODIFIED CYCLES AND SIMULATION BASIS

Four different combinations of a nitrogen expander cycle with C3-MR processes are schematically shown in Fig. 2. Figure 2(a) is the existing AP-X process, where the N2 expander cycle is independently regenerative. In C3-MR-N2(I) cycle, the compressed nitrogen is pre-cooled by C3 and MR cycles in series before expanding through a turbine, as shown in Fig. 2(b). Alternatively, the compressed nitrogen may be pre-cooled by a C3 cycle and regenerated in an MR heat exchanger in C3-MR-N2(II) cycle, as shown in Fig.2(c), or may be regenerated in C3 and MR heat exchangers in C3-MR-N2(III) cycle, as shown in Fig.2(d). Figure 2(e) is the combination of nitrogen expander cycle with DMR process, denoted as DMR-N2.

The following assumptions are made for simulation, as presented in [2].

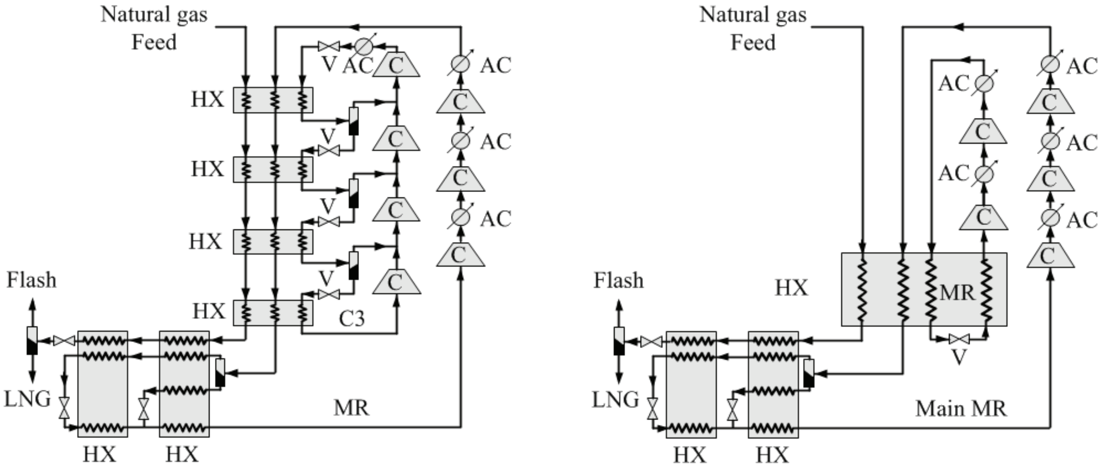
- The pressure drop in all heat exchangers is zero.
- The composition of natural gas is 4.0% N2, 87.5% C1, 5.5% C2, 2.1% C3, 0.5% nC4, 0.3% iC4, and 0.1% iC5 on mole basis.
- The natural gas feed at 65 bars is cooled from 300 K to 113 K for flash expansion to LNG storage.
- The exit temperature of after-cooler is 315 K for C3, 305 K for MR, and 300 K for N2.
- The number of compression stages is 4 for C3 and 3 for MR and N2.
- The minimum temperature approach between the hot and cold streams is 3 K in all cold heat exchangers.
- The evaporating refrigerant is fully evaporated in cold heat exchangers.
- The adiabatic efficiency is 80% for all compressors and expanders.
- The minimum temperature of pre-cooling refrigerant is 240 K.
- The maximum pressure of N2 is 80 bars.

A general purpose process simulator, Aspen HYSYS Version 7.3, is used in this study. The intermediate pressures for multi-stage compression are determined so as to minimize the total compressor work. The after-coolers are applied only where the exit temperature of compressor is higher than the ambient temperature.

The thermodynamic performance of a liquefaction system is evaluated in terms of the figure of merit (*FOM*) [9], defined as the ratio of minimum liquefaction work to actual work.

$$FOM = \frac{\dot{W}_{\min}}{\dot{W}} = \frac{\dot{m}_F\left[\left(h_{LNG} - h_0\right) - T_0\left(s_{LNG} - s_0\right)\right]}{\sum \dot{W}_{\mathrm{comp}} - \dot{W}_{\exp}} \qquad (1)$$

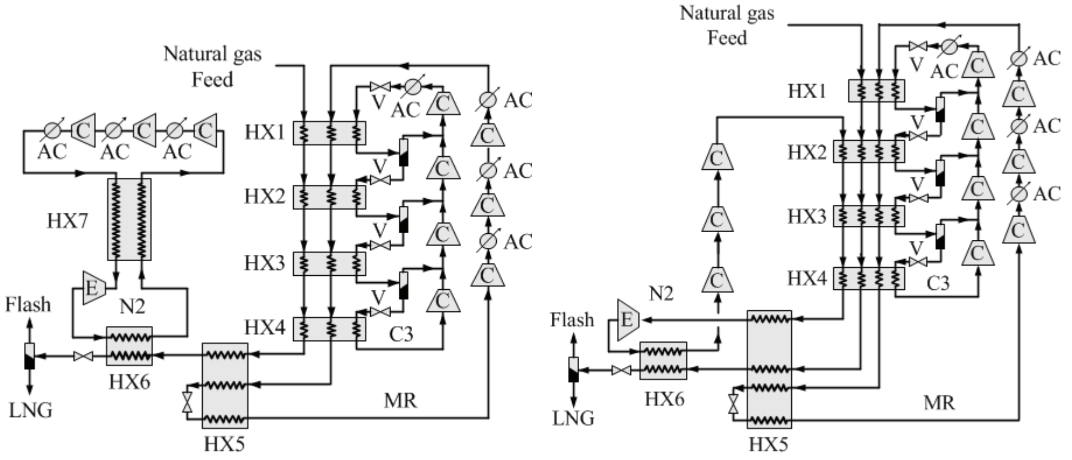
where h and s are the specific enthalpy and entropy of natural gas feed, respectively, and the subscripts *LNG* and *0* denote LNG (113 K) and ambient temperatures (300 K), respectively. The *FOM* is called thermodynamic efficiency [10] (based on second law) or fraction of useful effect [2].



(a) Propane pre-cooled MR (C3-MR) process

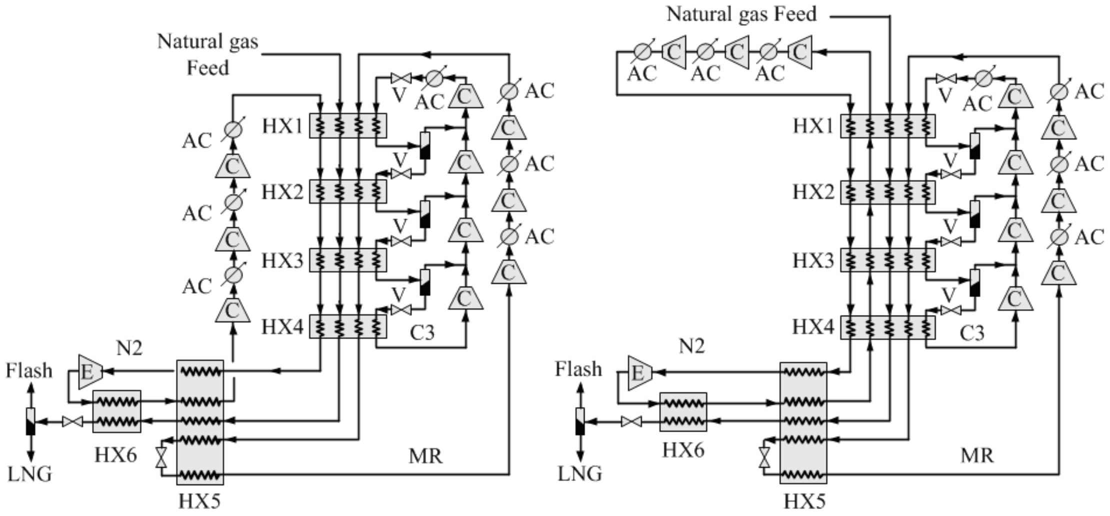
(b) Dual MR (DMR) process

FIGURE 1. Existing mixed refrigerant (MR) cycles (HX: heat exchanger, V: valve, C: compressor, AC: after-cooler).



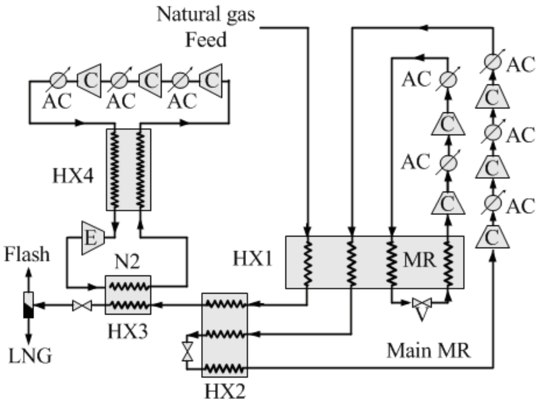
(a) AP-X process

(b) C3-MR-N2 (I)



(c) C3-MR-N2 (II)

(d) C3-MR-N2 (III)



(e) DMR-N2

(E: expander, HX: heat exchanger, V: valve, C: compressor, AC: after-cooler)

FIGURE 2. Existing AP-X process and four modified cycles with nitrogen expander cycle.

RESULTS AND DISCUSSION

Figure 3 compares the simulated *FOM* of the five different cycles combined with nitrogen expander cycle. The *FOM* of existing AP-X process is 42.6%, which is compared with 50.8% for C3-MR and 48.3% for DMR [2]. The main reason for the difference is the large irreversibility associated with the cryogenic expander in the nitrogen cycle. Among the modified cycles, *FOM* is the highest (46.1%) for C3-MR-N2(II) and the lowest (41.4%) for DMR-N2.

For more detailed analysis, it is useful to examine the exergy (or availability) balance [10] in each cycle. By combining the first and second laws of thermodynamics,

$$\dot{W} = \dot{m}_F \left[\left(h_{LNG} - h_0 \right) - T_0 \left(s_{LNG} - s_0 \right) \right] + T_0 \sum \dot{S}_{gen} \tag{2}$$

where the left-handed side is the net power input for liquefaction, and terms inside the bracket represent the useful effect or the flow availability of LNG. The fraction of useful effect is *FOM*, as defined in Equation (1). The last term is called the thermodynamic irreversibility, which is the entropy generation rate multiplied by the ambient temperature. The contribution of entropy generation can be itemized for all components in the cycle. Figure 4 shows the composition of useful effect and irreversibility by components for the AP-X process and four modified cycles. The irreversibility is subdivided into five groups of components: JT valves and mixing devices (V+M), expander (E), compressors (C), heat exchangers (HX), and after-coolers (AC). It can be noted that the major reason for the highest efficiency of C3-MR-N2(II) is the small irreversibility (i.e. the small temperature difference) in HX and AC.

The liquefaction capacity and size of the heat exchangers is also examined for the five simulated cycles. As mentioned above, the reputable C3-MR process has a technical limit in liquefaction capacity, mainly because of the compressor capacity of C3 and the heat exchanger size. In order to compare the potential regarding these technical limits, two parameters are plotted as column graphs in Fig. 5 from the simulation results. Figure 5(a) shows the mass flow rate per unit mass of feed gas, which is closely related with the compressor capacity. All five cycles are nearly the same in terms of compressor capacity of N2, but C3-MR-N2(III) and DMR-N2 are slightly superior to the existing AP-X in terms of compressor capacity of C3 or pre-cooling MR. Figure 5(b) plots the sum of heat exchange rate multiplied by the number of streams, as an index of the heat exchanger size. The total stacked column is shortest for the existing AP-X, which means that all other modified cycles could require a larger size of heat exchanger for the same liquefaction. However, it should be noted that the N2 heat exchanger plays a more important role in actual size, because the heat transfer coefficient of N2 gas is typically smaller than that of two-phase C3 or MR.

The relative significance of thermodynamic efficiency vs. liquefaction capacity is one of the most important factors in selecting liquefaction cycles. Out of all the modified cycle examined, the C3-MR-N2(II) cycle would be an obvious choice, when both efficiency and capacity are given equal consideration. The simulated FOM is 46.1%, which is notably greater than 42.6% of AP-X, even though somewhat larger N2 compressors and larger heat exchangers may be required. For the C3-MR-N2(II) cycle, the temperature profile in heat exchangers as well as temperature-entropy diagram are plotted in Fig. 6(a) and 6(b). It is noticeable that the temperature difference is significantly reduced near the cold end by the N2 expander cycle, resulting in an improved efficiency.

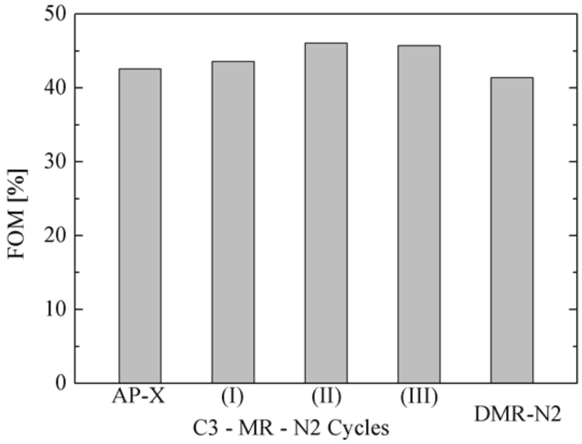
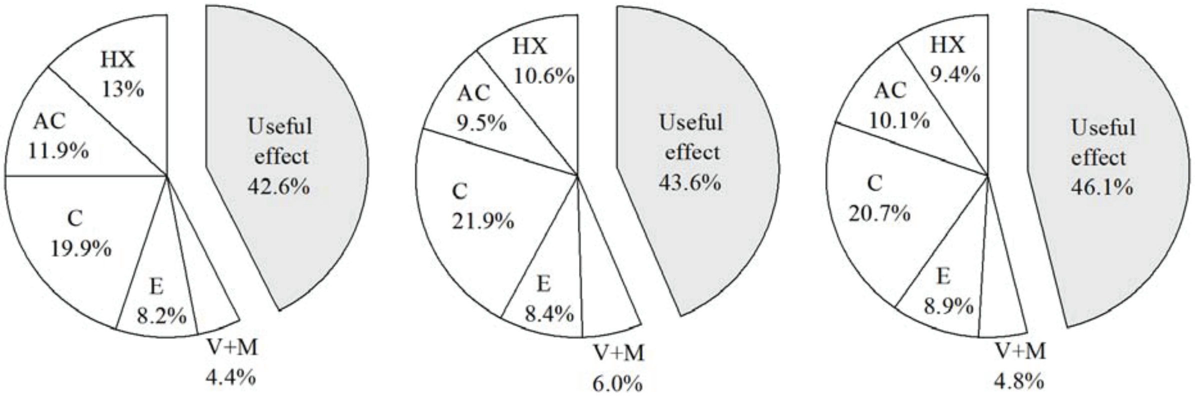
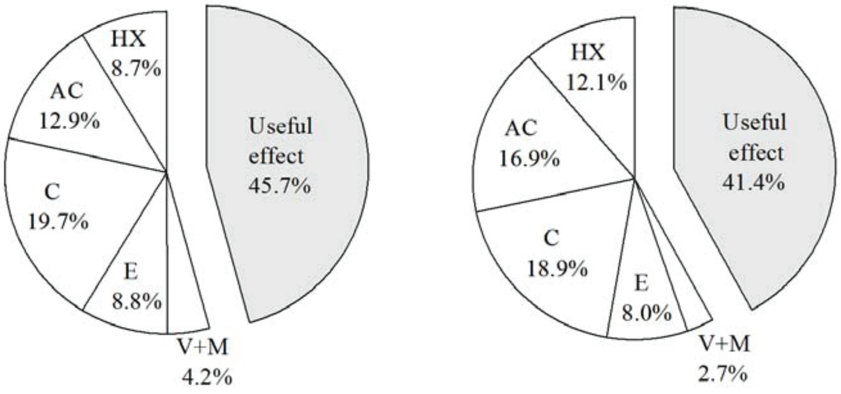


FIGURE 3. Simulated *FOM* values for existing AP-X process and four modified cycles.



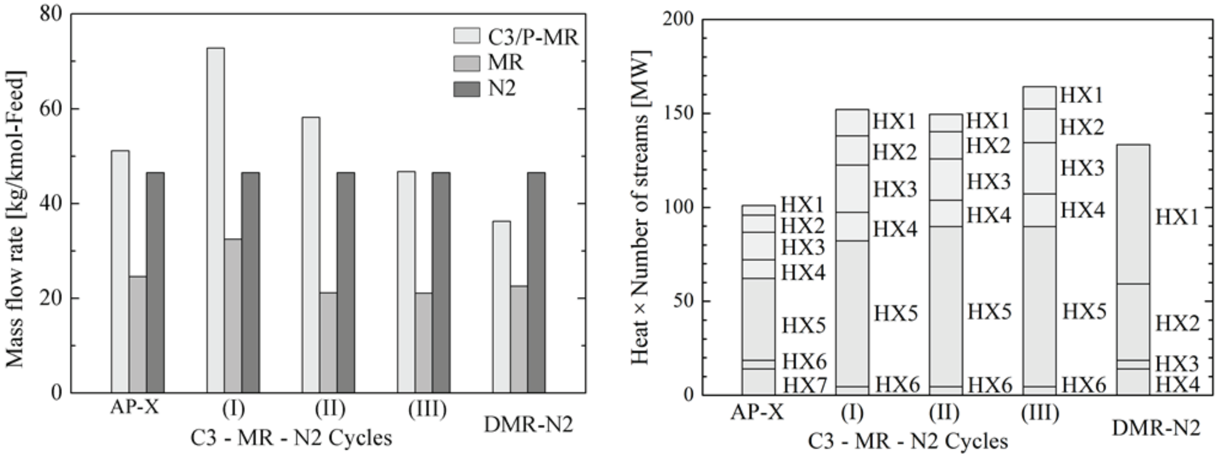
(a) AP-X process (b) C3-MR-N2 (I) (c) C3-MR-N2 (II)



(d) C3-MR-N2 (III) (e) DMR-N2

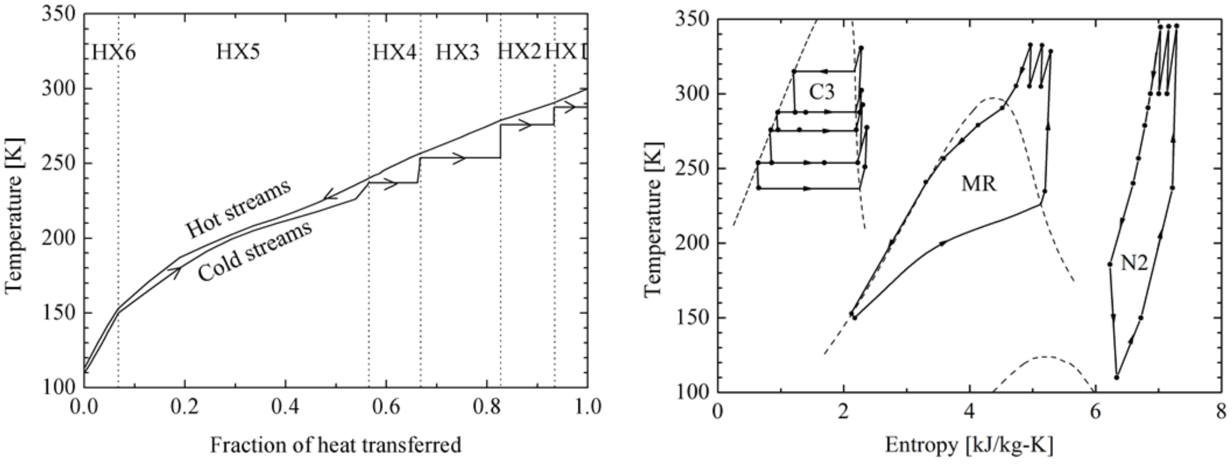
V+M (valves and mixing) E (expender) C(compressors) AC(after-coolers) HX(heat exchangers)

FIGURE 4. Useful effect and itemized irreversibility for existing AP-X process and four modified cycles.



(a) Mass flow rate (b) Product of heat and number of streams

FIGURE 5. Mass flow rate of refrigerants and product of heat and number of streams for existing AP-X process and four modified cycles.



(a) Temperature profile

(b) Temperature-entropy diagram

FIGURE 6. Temperature profile of hot and cold streams in heat exchangers and temperature-entropy diagram for C3-MR-N2 (II).

CONCLUSIONS

Five different combined systems of nitrogen (N2) expander cycle with mixed-refrigerant cycle are investigated for efficient and large-capacity liquefaction of natural gas. These cycles are simulated with commercial software (Aspen HYSYS) and compared in terms of the thermodynamic efficiency (*FOM*), required flow rate of refrigerants, and estimated size of heat exchangers. The existing AP-X process is a simple combination of independent N2-Brayton cycle with reputable C3-MR cycle, which is evaluated to be the most compact and suitable for large-capacity liquefaction. On the other hand, the thermodynamic efficiency may be considerably increased by modifying the cycle with partial regeneration and pre-cooling of N2 cycle in C3-MR cycle.

ACKNOWLEDGMENTS

This research is supported by a grant from the LNG Plant R&D Center, funded by the Ministry of Land, Transportation and Maritime Affairs (MLTM) of Korean government.

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