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DEVELOPMENT OF ACCELERATOR DRIVEN ADVANCED NUCLEAR ENERGY (ADANES) AND NUCLEAR FUEL RECYCLE*

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Abstract

The proposed Accelerator Driven Advanced Nuclear Energy System (ADANES) is a new closed nuclear energy cycle system, aimed at improving the utilization rate of nuclear fuels, enhancing nuclear safety, reducing the nuclear proliferation, and at becoming a sustainable and low-carbon energy supply for thousands of years. The ADANES system consists of fuel burner like the Accelerator Driven System (ADS), used nuclear fuel processing and recycle system. Several prototypes have been made as the development of key technology R&D. The 25 MeV proton superconducting linac (CAFe) has been built to demonstrate the several mA continues-wave proton beam at low energy section. The material named SIMP (Steel designed by Institute of Modern Physics and Institute of Metal Research, CAS) is presented. The performance of resistance to oxidation, LBE corrosion, irradiation and the yield strength, tensile strength, ductility at temperature of 200°C till to 700°C have been shown. The nuclear fuel recycle approach with the help of the external neutron source is proposed. The rest spent nuclear fuel after removing the part fission products will be refabricated to new nuclear fuel and burned. The investigation about the separation of some fission products and fabrication of ceramic nuclear fuel microspheres were achieved in the laboratory.

INTRODUCTION

The global climate change and green-house-gas emission are the common issues around the world. To reduce the ration of fossil energy in the primary energy, nuclear energy is employed to play the important role of clean energy. It is an inevitable strategy to meet the demand on energy in China. The number of nuclear power plants keeps increasing despite effect of Fukushima-accident. Up to May 2019, there are 46 nuclear reactors are in operation, the 3rd of the world [1]. But the nuclear power electricity is only 4.22% of the total produced, the fossil energy is still the main part in the primary energy resources [1]. At present, 11 nuclear reactors are constructing in China, and it is the 1st of the world [1].

However, with the development of nuclear power, more and more spent nuclear fuel will be produced and accumulated. The nuclear waste and the limit mineral resources of Uranium become the key issues on the way of the sustainable development of fission nuclear. The accelerator driven sub-critical reactor (ADS) and the fast reactor system (FR) were proposed to transmute the minor actinides. The partition-transmutation strategy was known as the most efficient way to reduce the radiotoxicity of nuclear spent fuel (NSF). It was reported by NEA/OECD in 2002 that the ADS is advantage that it can burn pure minor actinides while avoiding a deterioration of the core safety characteristics [2]. In Europe, an ADS project MYRRHA as in Multi-purpose hYbrid Research Reactor for High-tech Applications was proposed in 1998. It is the prototype of a nuclear reactor driven by a particle accelerator under European Framework. The planning of the project is in 4 phases till 2033. The first stage started in 2017 and will complete the construction and start operating a 100 MeV practical accelerator in 2026 [3]. In China, a long-term development strategy started in 2011. "Future Advanced Fission Energy Program – ADS Transmutation System" strive for the series key technology from test facility to demonstrate facility, aiming at sustainable development of nuclear fission energy to contribute the national energy supply [4].

CONCEPT AND ROADMAP OF ADANES

With the development of nuclear power, more and more spent nuclear fuel will be produced and accumulated. Reprocessing of these spent nuclear fuels became the key issues for the environmentally friendly and sustainable development of nuclear energy. Nowadays the concept of a closed nuclear fuel cycle is highly discussed as a possible, which will make an optimized use of natural resource and minimize the amount of nuclear wastes. The Accelerator Driven Advanced Nuclear Energy System (ADANES) proposed by CAS is a novel closed nuclear energy cycle system, aimed at improving the utilization rate of nuclear fuels, enhancing nuclear safety, reducing the nuclear proliferation, and at becoming a sustainable and low-carbon energy supply for thousands of years. It consists of a fuel burner like the Accelerator Driven System (ADS), and a used nuclear fuel processing and recycle system. Closing fuel cycle can be achieved through partitioning of uranium and minor actinides and transmuting of long-lived radioactive nuclides in ADS. The new reprocessing of spent nuclear fuel only remove the part fission products including some volatile fission products and some neutron poisons (lanthanides), which is different from the traditional partitioning and transmutation (P-T) strategy. The rest spent nuclear

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fuel after removing the part fission products will be re-fabricated into new nuclear fuel which can be burned in ADS burner.

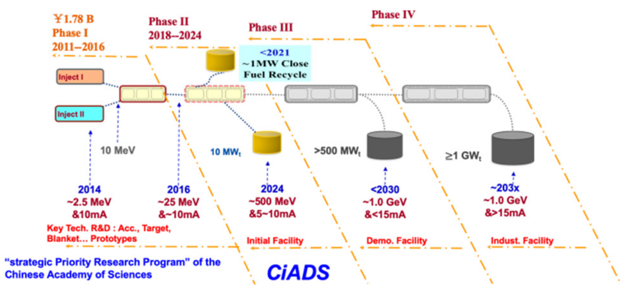

Figure 1: Roadmap of ADS/ADANES in China.

A four-phase developing roadmap is planned by CAS. It is shown in Fig. 1. The first stage started in 2011 till 2017, named Strategic Priority Research Program on "Future advanced fission nuclear - Accelerator Driven System". In the stage, the key technologies on superconducting linear accelerator, high power spallation target, LBE fast reactor, and materials have been developed. The second stage name Chinese initiative Accelerator Driven System (CiADS) located in Huizhou was approved in Dec. 2015, and ground broken in August 2018. It consists of a 500 MeV superconducting linac and a 7.5 MWt LBE fast reactor. It will be completed and operated in 2025. Meanwhile, a program of Accelerator Driven Recycle for Used Fuel (ADRUF) is in parallel. The third stage is a demo facility of ADS with a 15-MW lianc and a 500-MWt reactor. The goal is to build an industrial scale facility in 2030's.

CHINESE INITIATIVE ACCELERATOR DRIVEN SYSTEM (CIADS)

The ADS takes the spallation neutrons produced by the high energy proton bombard heavy metal target to drive the sub-critical fast reactor system. With the external wide-spectra, and high flux neutrons, the system can transmute the long-life radiation nucleus and output extra energy. It has inherent safety due to sub-critical mode. The Chinese initiative Accelerator Driven System (CiADS) as the second stage of roadmap started earthwork in 2018. The layout of CiADS is shown in Fig. 2. The top-level specifications list in Table 1.

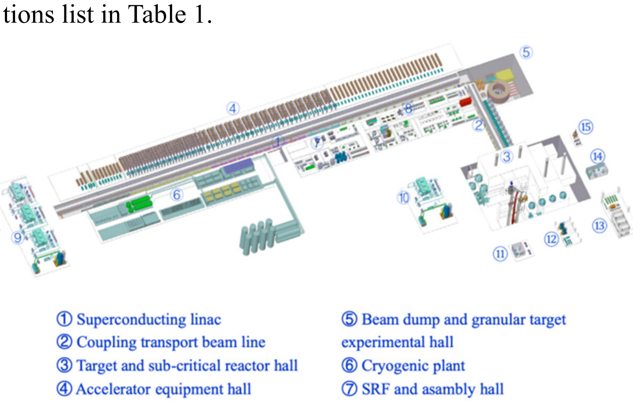

Figure 2: Layout of CiADS.

The driven accelerator is a superconducting linac around 200 meter long. The superconducting resonators are just after the room temperature RFQ. The exit energy of RFQ is 2.1 MeV. Five families of superconducting cavities are employed to accelerate particle to 500 MeV. 100-meter spare space is reserved for upgrading in the future. The output energy and numbers of cavity of each session of superconducting linac are shown in Fig. 3.

Table 1: Top-level Specifications of CiADS

Parameter	Value
Driven accelerator type	Superconducting linac
Particle	proton
Energy [MeV]	500
Current [mA]	5
Beam power [MW]	2.5
Target type	Granular flow
Target material	Tungsten alloy
Target power [MW]	2.5
Reactor type	Fast, sub-critical
Reactor power [MW]	7.5
Fuel	UO_2
Coolant	LBE
Keff	~0.75

The key parameters of each superconducting sessions list in Table 2. The failure compensation is both local and global modes. In the routine operation mode, the electrical field of cavities is 70% to 80% of the maximum.

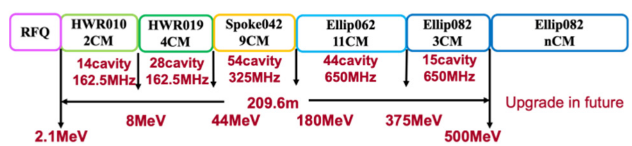

Figure 3: Structure of superconducting lianc.

Table 2: Key Parameters of Superconducting Sessions

	Unit	HWR010	HWR019	SPOKE042	Ellip062	Ellip082
f	MHz	162.5	162.5	325	650	650
type	-	Sqreezed	Taper	Double	5 cells	5 cells
β_{opt}	-	0.10	0.19	0.42	0.62	0.82
Vmax	MV	1.0	2.5	6.5	13	20
Ep norm.	MV/m	20	25	28	28	28
Ep max	MV/m	28	32	35	35	35
Q0	E09	5.00	5.00	6.00	10.0	10.0
Cav./CM	-	7	7	6	4	5
MT/CM	-	7	4	2	2	2
CM L	m	5.5	7.3	8.2	6.5	8.1
CM Num	-	2	4	9	11	3

The tungsten granular target is the basic option of CiADS [5]. The principle is shown in Fig. 4. The small granular with the size of 1 mm diameter flow down driven by gravity. The granular stays in the interaction area for around 1 second and heated by proton beam. The hot granular exchanges heat with oil and lifted by elevator to the top of target. The granular speed is controlled by the size of exit port. The merits are no stress due to beam trips and

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not thermal shock effect due to beam pulse, but the drawback is the dusty due to abrasion.

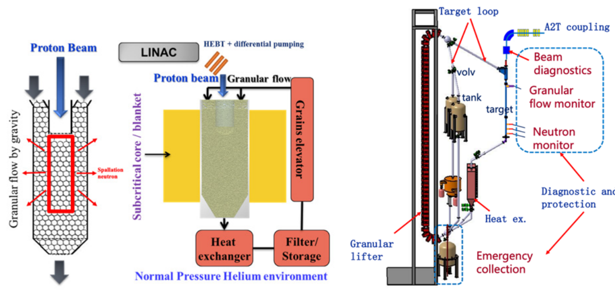

Figure 4： The principle and setup of granular target.

The granular target system has been designed and layout is shown in Fig. 4. The target pipe goes through the reactor and down to the heat exchanger. An electro-magnetic elevator is employed to lift the granular to the top of reactor. The emergent collecting tank is at the very end of whole system to store all the granular when earth quack or other emergent cases happen. The inlet temperature of target is 250℃ and average temperature at exit is less than 330℃.

Table 3: Parameters of Sub-critical Fast Reactor

item	Value
Type	Sub-critical fast reactor
Power [MWt]	10 (incl. beam)
Fuel and enrichment	UO_2 (19.75%)
Keff	~0.75
Coolant	LBE
Main coolant loop	Pool-loop mix
Main coolant driven mode	forced
Main loop pressure	normal
Main coolant temp. [℃]	280-380℃
Main coolant exchanger	Main ex. ×2
	Auxiliary ex.×4
Main bumps	Mechanical bump ×2
Secondary loop coolant	Pressure water
Second. loop pressure [Mpa]	1.4
Second. Loop temp. [℃]	225-260

The parameters of sub-critical fast reactor list in Table 3. It is a deep sub-critical fast reactor with design lifetime of 1095 EFPD. The fuel is ceramic UO_2 with enrichment of 19.75%. The core has 30 bundles with 60 rods in each and 78 dumbs. As the first demo for ADS, the power of sub-critical core is only tuned by the accelerator beam. There are not additional control rods for tuning or stopping the reactor. The external neutron source is ~2.87E16 /s and average neutron flux in the reactor is 4.86E13 n/cm2/s.

PROGRESS OF SUPERCONDUCTING DEMO LINAC FOR ADS

The Chinese ADS Front-end superconducting demo linac (CAFe) has been completed as the prototype of ADS accelerator in 2017 [6, 7]. It is shown in Fig. 5 which consists of ECR ion source, RFQ, and four cryomodules. It accelerates proton to 25 MeV. The parameters list in Table 4.

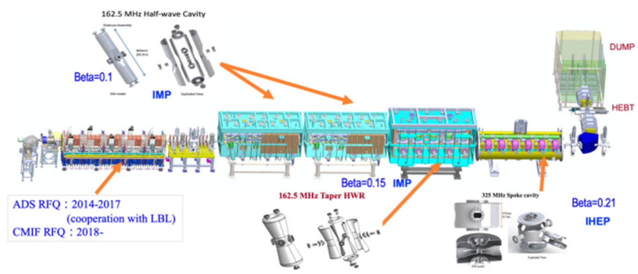

Figure 5: Layout of CAFe.

Table 4: Parameters of CAFe

	RFQ	CM1	CM2	CM3	CM4
frequency	162.5 MHz	162.5 MHz	162.5 MHz	162.5 MHz	325 MHz
output energy	2.1 MeV	5 MeV	10 MeV	18.5 MeV	25 MeV
cavity type	4-vane	HWR010	HWR010	HWR015	Spoke021
cavity number	1	6	6	5	6
field	65 kV	25 MV/m	25 MV/m	32 MV/m	32 MV/m

The RFQ was designed by Lawrence Buckley National Laboratory (LBL) and cooperated to fabricate in workshop of IMP. It operated in CW power around 6000 hours from 2014 to 2018. The MTBF was 11.3 hours at the first 3264 hours of operation with the Tetrode amplifier and 24.4 hours at the second 2638 hours with solid state amplifier.

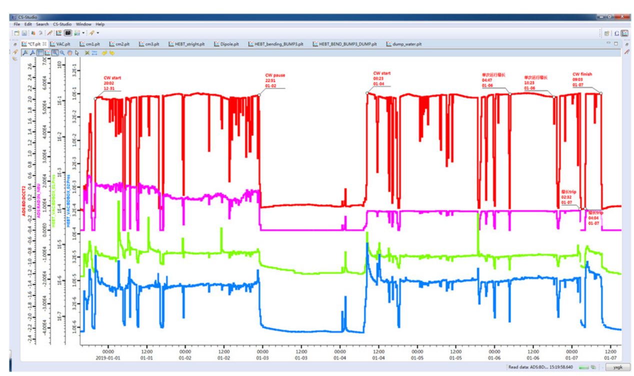

Figure 6: Operation record of high-power operation.

A high-power operation test campaign was done in Jan. 2019. The beam is 15.8 ~16.4 MeV @ 2.0 ~2.1 mA. Total operation time is more than 110 hours. The availability is 89%. The MTBF is 90.7 min and MTTR is 11.1 min in the first stage. The MTBF is 113. 7 min and MTTR is 14.6 min in the second stage. The maximum power is 45 kW at 17.5 MeV and 2.55 mA (see Fig. 6).

MATERIAL -- SIMP STEEL

Regarding to the excellent properties on swelling resistance, low activation and high thermal conductivity, reduced activation ferritic-martensitic (RAFM) steels are identified as the primary candidate structural materials for

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GEN IV fission reactors. However, LBE corrosion of materials is one of the key issues must be considered in the reactor using LBE as coolant.

In order to fulfil the properties mentioned above, we have developed a novel material named as SIMP steel (Steel designed by Institute of Modern Physics and Institute of Metal Research, CAS). In the past 8-years, systematic works on the design, fabrication, processing and evaluation of SIMP steel have been done. The routine performances, such as high temperature properties and resistances to intense ion irradiation and LBE corrosion, have been tested.

For the design of SIMP steel, low activation elements are selected (chemical composition design) at the first, control of element content, phase segregation, impurity elements as well as the resistance to oxidation and LBE corrosion are taken into account. Then optimization of smelting, casting with phase control of SIMP steel ingots were gradually prepared till to pilot 5 tons' ingots fabricated in a company and mechanical processing of SIMP steel has also been done (see Fig. 7).

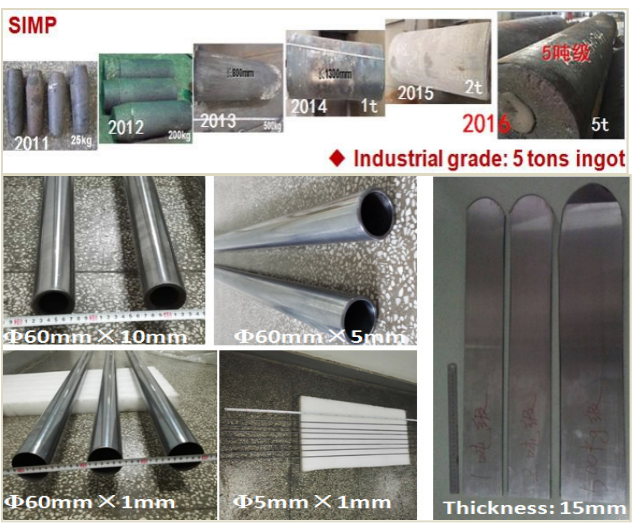

Figure 7: Progress of smelting, casting and mechanical processing of SIMP steel.

It was found that the obtained SIMP steel are of whole martensite phase and with excellent thermal stability. The yield strength, tensile strength, ductility of SIMP steel at 200°C till to 700°C are higher than that of T91 steel. More, the creep at 200°C till to 500℃are also higher than that of T91 steel.

Figure 8 shows the comparison of LBE corrosion between SIMP and T91 steels under the same experimental conditions (LBE with saturated oxygen at 450℃). It is clear that the thickness of the corrosion layers increases with increasing corrosion time and the corrosion rate decreases gradually. Moreover, double structural corrosion layers are clearly observed at the specimen exposing to LBE for 2000 hours and longer, the thicknesses of outer and inner corrosion layers are slightly different.

Figure 9 shows the comparison of cavity formation in SIMP, T91 and one home-made RAFM steel specimens at the peak damage regions induced by the same ion beam irradiation. For the irradiation performed at 450℃, the cavity size distributions of T91 and home-made RAFM steel specimens are of two peaks. The exist of larger size cavities leads to larger cavity swelling. Smallest average cavity size, lowest cavity number density and thus smallest cavity swelling values are observed in SIMP steel. For the case of specimens irradiated at 550℃, the cavity size distributions are of one peak with smaller average cavity size and lower cavity number density than that at 450℃, which means that the irradiation resistance of these steels is better than that at 450℃. More， smaller average cavity size, lower cavity density and thus smaller cavity swelling values in SIMP steel than that in the home-made RAFM steel specimen.

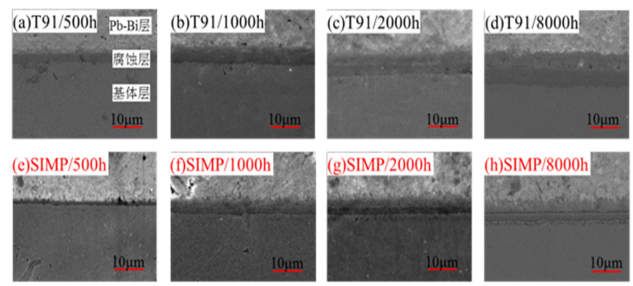

Figure 8: SEM micrograph of cross-sections of SIMP and T91 steels after exposing to LBE with saturated oxygen at 450℃ for different time.

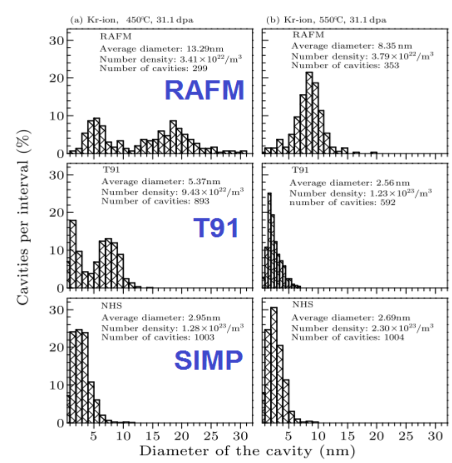

Figure 9: Comparison of cavity formation in different steels under same ion beam irradiation.

REPROCESSING AND FABRICATION FOR THE SPENT NUCLEAR FUEL

It is well known that spent nuclear fuel contains uranium oxide, fission products and transuranic (TRU) elements. As a rule of thumb, the spent nuclear fuel for a standard PWR with a burn-up of 33 GW/t consists of about 95.5% UO_2 matrix, 3.6% fission products and 0.9% trans-uranium elements. 6 A wide range of fission products are generated, representing more than 30 chemical elements, which also

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undergo further radioactive decay. Figure 10 shows the technical route of reprocessing and fabrication for the spent nuclear fuel.

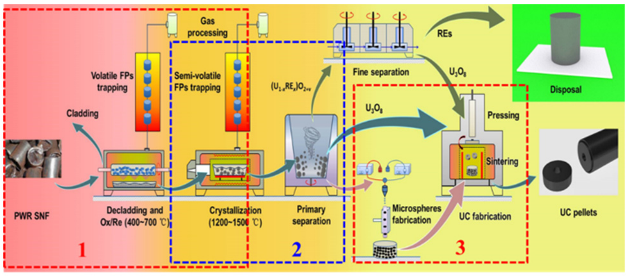

Figure 10: Technical route of reprocessing and fabrication for the spent nuclear fuel.

For those volatiles and semi-volatile fission products (^{3}He, ^{14}C, Kr, I, Xe, Mo, Tc, Ru, Te, et al.) would be easily removed by using a dry-processing technique known as OREOX (Oxidation and RE duction of OXide fuel) processes. In our laboratory, the studies of the pulverization about UO_2 pellet to U_3O_8 powder and the subsequent high temperature oxidation/reduction were carried out. Firstly, the UO_2 powder was pressed into simulated pellets and sintered at 1700℃ for 7 hours in an atmosphere of 4%H2/Ar. In oxidative process, the UO_2 simulated pellets all showed good pulverization results. The results of three cycles as oxidation at 450℃ in air or 50% O_2/Ar and reduction at 700℃ in 4% H_2/Ar showed that the particle size of two conditions are both less than 10 microns. The UO_2 real fuel pellets was also studied through 3 cycles of oxidation-reduction process. In addition, several of FP elements have been added into UO_2 powder to prepare the simulated PWR spent fuel in the work. The results showed that Mo, Ru, Te and Se can be effectively removed at 1200℃.

For lanthanides, which accounts for about 1/3 of the fission products, must be separated from minor actinides before next transmutation step because the high neutron captures cross sections of some lanthanides, named as neutron poisons, inhibit efficient transmutation. However, mutual separation of MA and Ln is very different due to their similar chemical properties. We describe herein an environmentally benign strategy for removing of fission products from spent nuclear fuel by the selective dissolution using carboxyl-functionalized ionic liquid. Water-statured can dissolve lanthanides oxide from simulated spent nuclear fuel with a dissolution ratio of 100% at 40℃. However, the dissolution of uranium is almost neglect (<1%) under the same conditions. This process of separation displays an outstanding performance to separate some key fission products oxide such as neutron poisons Ln_2O_3 efficiently and allows the recovery of actinides AnO_2 as a group in solid form. Less volume of high level radioactive liquid waste (HLW) was generated during this process and there was no need for dissolving spent nuclear fuel since only some fission products was dissolved selectively and separated. The dissolved Nd and U were recovered efficiently from the loaded ionic liquid phase by back-extraction with 1M HCl solution. The ionic liquid phase can be reused again for the dissolution of Nd_2O_3 after washing off the residual acid with water. The high Ln_2O_3 dissolution capability is elucidated by thermo-dynamical analysis on the microprocess of the solubilizing and the U/x value related to the lattice energy U for MxOy lattice disruption [8].

The preparation of ceramic nuclear microspheres was also performed using an improved microwave-assisted rapid internal gelation process. This improved microwave-assisted internal gelation process greatly simplifies the conventional sol-gel process and avoids secondary organic radioactive waste. It can provide an important guidance for the preparation of Pu and MAs carbides microspheres, especially (U+Pu)C, (U+Am)C, for nuclear reactors in the future [9].

CONCLUSION

The ADANES was proposed as a new closed fuel cycle system aiming at the sustainable nuclear energy. The prototype of front-end of CiADS has demonstrated 2.5 mA CW beam. The procedures of recycle SNF was proved with SIMFUEL in lab. CiADS as the burner of ADANES started constructing in 2018.

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