The Hydrogen Isotope Separation System

Introduction

The Laboratory for Laser Energetics (LLE) routinely fields 860-μm-diam, thin-walled plastic shells filled with deuterium–tritium (DT) gas or DT ice layered on the inner surface of the shells to study inertial confinement physics. The multistep filling operation involves desorbing the DT fuel from uranium storage beds, measuring the tritium inventory, and removing decay ^{3}He from the DT fuel prior to filling the evacuated shells.

In the case of targets filled with gas, the DT fuel is gradually compressed to pressures in the range of 10 to 30 bar in the presence of the shells. DT permeates into the empty shells until the pressure within the shell comes close to equilibrium with the pressure outside the shell. Pressurizing the container holding the shells too quickly can buckle the thin-walled plastic spheres. An aluminum coating on the outside of the shells extends the permeation time constant into the tens-of-hours range. At the end of the pressurizing cycle, the DT is returned to the uranium storage beds. The filled targets are transferred to cold storage to further suppress permeation losses from the shells and are kept there until required for experiments.

In the case of cryogenic targets, the DT fuel is assayed, stripped of decay ^{3}He, and then transferred to a compressor where the gas pressure is first boosted from 1 bar to ~120 bar and subsequently to pressures in the range of 500 to 700 bar, depending on the final ice thickness required. At the end of the pressurization stage, the targets are gradually cooled to 15 K so that the DT gas that has permeated into the shells can condense on the inner surface of the plastic spheres. At ~30 K, gas surrounding the shells is returned to the storage beds. At this temperature, gas permeating from the plastic shells is negligible. From this moment on these targets are maintained at temperatures between 17 and 40 K until they are used to study implosion of DT ice driven by the laser system.

These manipulations and time have caused, over the past few years, the tritium-to-deuterium ratio to depart from the preferred 50:50 ratio for two reasons: (1) tritium decays at ~0.46% per month; (2) beta-induced isotopic exchange with hydrogen in the plastic wall and with hydrogen bound in water adsorbed on the inner walls of the process system increases the protium (H) content in the fuel at ~0.3% per year. The first effect makes the fuel deuterium rich. The second effect causes protium to concentrate inside the target and interfere with the implosion kinetics. Both degrade the fusion neutron yield.

Motivation

The fuel supply has gradually degraded to a tritium/deuterium/protium (T/D/H) ratio of 38/59/3. Additionally, during the commissioning of the high-pressure systems, LLE used 48 TBq (1300 Ci) of tritium to make up gas mixtures ranging from 0.1% T/D to 10% T/D. This gas is not useful as a fuel and has been stored on a separate uranium storage bed that is not connected to any of the existing gas-handling systems. Finally, LLE has installed emission-reduction equipment based on getter technology that will be discussed shortly. Elemental tritium can be extracted from a helium purge stream with a very high efficiency[1] using getters. While these getters can be regenerated to recover elemental tritium with negligible release to the environment, the gas will be strongly contaminated with protium, rendering it useless for inertial confinement physics studies.

LLE operates under a 1.5-g tritium inventory limit. The majority of this gas is required for the operations discussed in the previous section. LLE does not have the inventory headroom to replace downgraded tritium with fresh tritium without first disposing of the downgraded tritium off-site. For example, an accidental release of 30% of LLE's tritium inventory and its subsequent recovery as downgraded tritium would preclude LLE from undertaking any further inertial confinement fusion studies until the downgraded tritium was either disposed of as waste or treated to recover the tritium. Disposing of elemental tritium gas is prohibitively expensive and not a viable approach; isotopic separation is more attractive. Furthermore, outfitting LLE with an Isotope Separation System (ISS) expands the Laboratory's capabilities to explore fusion-reaction physics, in a controlled manner, over a very broad range of T/D ratios.

A review of the hydrogen isotope separation technologies suggested that a scaled-down version of the "thermal cycling absorption process" (TCAP)[2] developed at Savannah River National Laboratory would be ideally suited to meet LLE's needs. The system in its most-recent reincarnation offers several unique advantages: the system has no moving parts with the exception of one automated valve, and gases are shuffled within the system by heating or cooling storage beds or columns. These features increase the system's robustness against accidental release of tritium.

The system is compact. Atomic sorting of the hydrogen species by mass occurs in the presence of palladium on kieselguhr (Pd/k). Molecular hydrogen isotopologue sorting by mass occurs on a cold molecular sieve (MS). Both processes are complimentary and reinforce the movement of the heaviest species to the front end of the Pd/k column, while the lightest species prefer to accumulate at the exhaust end of the MS column.

Emission from the system is controlled by adjusting the operating conditions of the ISS and the number of times the gas is shuffled between the two columns. Additionally, ISS raffinate effluents with measurable activity can be directed to and concentrated in the Glovebox Cleanup System as hydrides and subsequently returned to the ISS for tritium recovery.

Description of LLE's ISS

1. Overview

An overview of LLE's ISS is provided in Fig. 144.1. The complete system is made up of five subsystems: Gas Handling, Core, LN_2 Management, Vacuum, and Glovebox Cleanup. The Gas Handling System feeds isotopically diluted tritium to the Core System and provides a temporary storage capability for purified tritium. The Core System decomposes the mixed hydrogen isotopes and separates them to isolate pure tritium. The LN_2 System provides a high cooling capability for the Pd/k and MS columns. The Vacuum System supports all evacuation steps required in handling tritium within the Gas Handling and Core Systems. The Glovebox Cleanup System provides secondary containment for any tritium releases from the Gas Handling and Core Systems. Only the Gas Handling System and the core are housed inside the glovebox.

2. Gas Handling System

The Gas Handling System comprises a uranium storage bed, a palladium storage bed, and a valve tree for diagnostics and for transferring gas into and out of the Core System. Swagelok BNBW Series bellows valves fitted with copper stem tips are used throughout. The valve tree is an integrated, welded assembly with a global leak rate below 1×10^{-9} atm-cm^3/s. The valves are rated to operate from vacuum to 3.55 MPa (500 psig), spanning a temperature range from 10°C to 204°C. Swagelok destructively tested a set of seven BN valves at their factory in support of this project. The burst or fail pressure of these valves is 90 MPa (13,070 psig) when closed and 32.5 MPa (4700 psig) when open. The stem bellows twist and relieve the pressure when the valve is open.

MKS 870 series Baratron pressure transducers were deployed in both the Gas Handling and Core Systems. The burst pressure of these transducers was measured to be 132 MPa (19,140 psig).

The uranium storage bed features a flow-through configuration to permit circulation over 32 g of depleted uranium powder

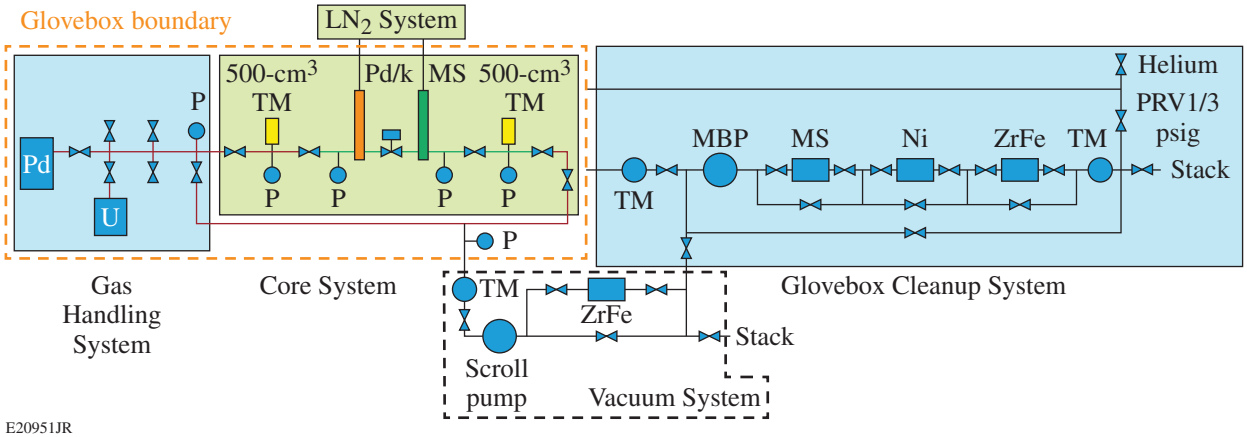

Figure 144.1
Overview of LLE's Isotope Separation System (ISS). P: pressure transducer; TM: tritium monitor; MS: molecular sieve; MBP: metal bellows pump; PRV: pressure-relief valve.

and is doubly contained to reduce heat and tritium losses to the glovebox when the bed is heated. The bed was designed and built to ASME VIII Div 1 and is registered as a pressure vessel. The maximum-allowable working pressure is 1.48 MPa (200 psig) at 430°C for the primary vessel and 0.45 MPa (50 psig) at 120°C for the secondary vessel. The hydrogen storage capacity is 4.5 sL.

An illustration of the cross section of the uranium storage bed is provided in Fig. 144.2. In purge mode, gas enters via the upright valve to flow into the uranium powder that is trapped between two 2-μm, 1.6-mm-thick stainless-steel frits, exits via the lower filter, passes through channels cut into the inside of the primary vessel wall, and flows along an annular region of the central tube before exiting the bed via the valve positioned at 90° relative to the central tube. The central tube incorporates a formed bellows to relieve stress between the primary and secondary containers when the bed is heated to 430°C. The annular region between the primary and secondary containers is kept under vacuum for most operations involving the bed.

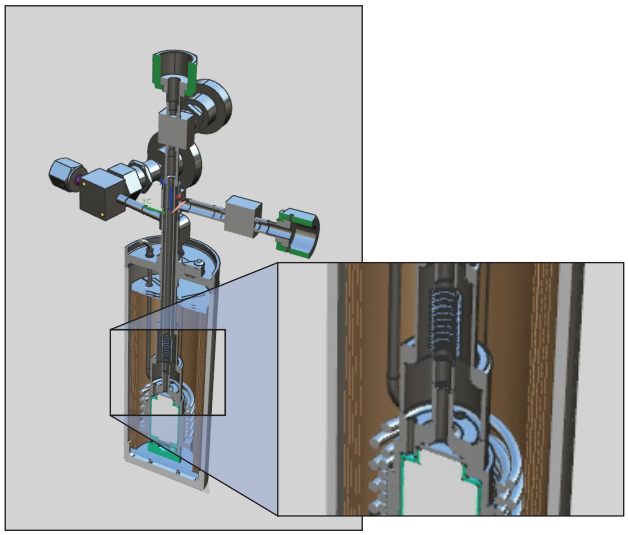
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Figure 144.2
Cross section of the flow-through uranium storage bed.

The design of the palladium storage bed design parallels that of the uranium bed. It also features a flow-through design and utilizes double containment. The maximum-allowable working pressure is 1.48 MPa (200 psig) at 210°C. The storage capacity of the Pd bed is 5.7 sL. The Pd bed typically operates between 150 K and 150°C. To achieve subzero temperatures, an acoustic Stirling (pulse-tube) cryocooler[3] is thermally coupled to the Pd bed via a cold-finger joint that separates at elevated temperatures. The pressure-wave generator and the compliance tank with an inertance tube coil reside outside the glovebox while the primary containment vessel of the Pd bed is located inside the glovebox. The configuration is shown in Fig. 144.3.

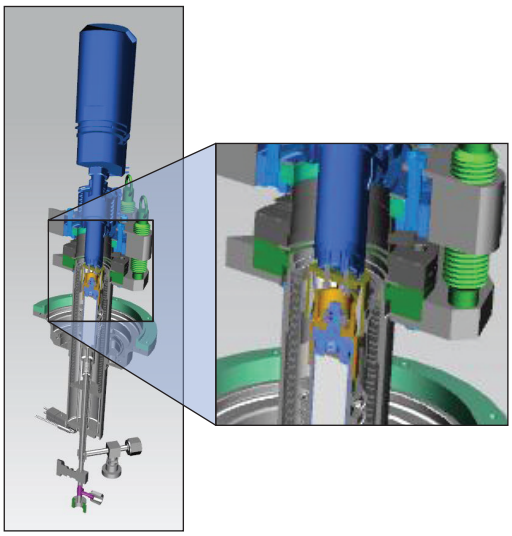
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Figure 144.3
Cross section of the flow-through Pd storage bed. The flange welded to the Pd bed's secondary containment seals against the glovebox ceiling to isolate the cryocooler from the glovebox atmosphere.

This cooling arrangement provides fine temperature control and simplifies the liquid nitrogen cooling circuit inside the glovebox. Heat loss to the secondary containment determines the Pd bed cooldown rate as well as the lowest-attainable temperature. As illustrated in Fig. 144.4, when the secondary

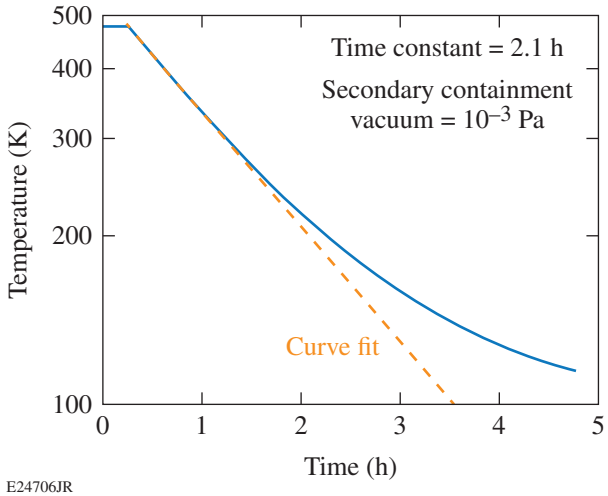

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Figure 144.4
The Pd bed's primary vessel cools down exponentially for the first hour when the secondary containment is evacuated to 10^{-3} Pa.

containment vacuum is below 10^{-3} Pa; the Pd bed temperature drops exponentially with a 2.1-h time constant for the first hour, and then at a slower rate to reach an operating temperature of 157 K in 3 h. The time required to reach 150 K from 20°C is 2 h.

3. Core System

The Core System uses two columns—palladium on kieselguhr (Pd/k) and molecular sieve (MS)—separated by a low-volume diaphragm valve. An exploded illustration of a column assembly is shown in Fig. 144.5. The column assembly is made by coiling the column comprising 6.4-mm-diam (1/4-in.) stainless-steel tubing filled with Pd/k and sandwiching the column between a coiled heater on one side and a 6.4-mm-diam LN_2 cooling coil on the opposite side. The three coils are brazed together to improve heat transport, while the column is thermally cycled using liquid nitrogen cooling and electrical heating. High-density insulation is inserted between the column assembly and the column end covers to reduce heat losses to the glovebox. The burst pressure of the column is 158 MPa (22,940 psig).

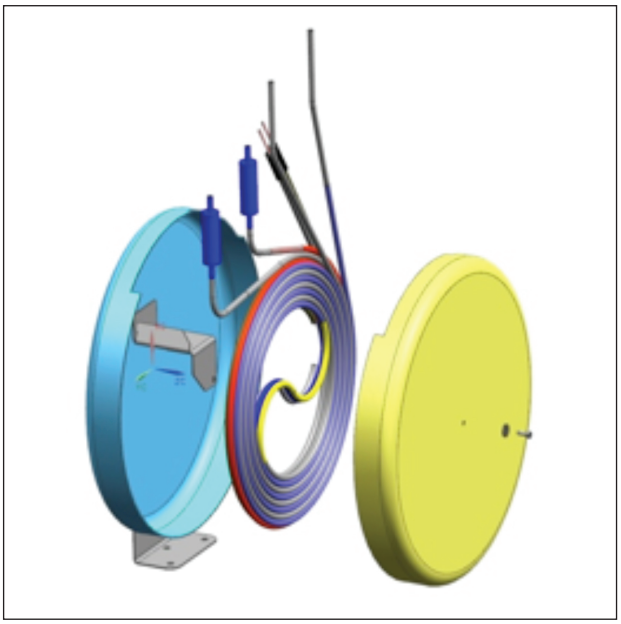

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Figure 144.5
Exploded view of a column.

A modified, all-metal, low-volume Swagelok 6LVV-DP series valve close couples the Pd/k and MS column assemblies. The burst pressure of this valve is 84.2 MPa (12,200 psig). The valve is oriented in the circuit to relieve gas pressure from the MS column into the Pd/k should the system inadvertently lose LN_2 cooling capability.

The pressure transducers are high-pressure MKS Series 870 Baratron sensors described in **Gas Handling System** (p. 172).

The expansion vessels double as tritium monitors. The design is based on the 1-L process monitor cited in Ref. 4. The cylindrical wall length was shortened to reduce the internal volume of the chamber to 500 cm^3. The end caps have been thickened to help support the floating BNC used to measure the ionization current. The monitor is registered under ASME VIII Div 1. With a burst pressure of 33 MPa (4800) psig, the tritium monitor represents the weakest link in the Core System. The two monitors operate from vacuum to 3.6 MPa (500 psig) over the temperature range of –10°C to 90°C. The internal surfaces were gold coated to reduce the memory effects of the monitor.[4]

Cross sections of the two expansion vessels are illustrated in Fig. 144.6. The collector within the raffinate monitor is a solid cylindrical configuration. The outer shell of the monitor is at zero potential. The ionization current induced by tritium decay within the detection region increases linearly with tritium activity in the range of 74 kBq/m^3 to 37 GBq/m^3 for flow rates below 40 sLPM and is practically independent of operating pressures above 9 kPa, as discussed in Ref. 4.

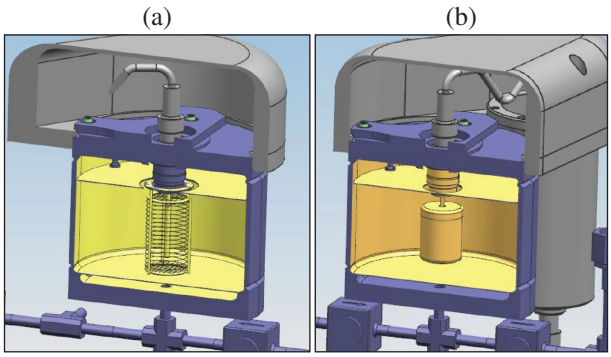

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Figure 144.6
Cross sections of the expansion vessel/tritium monitor on (a) the product side and (b) the raffinate side.

The collector in the product monitor is a wire cage housed at the center of the 500-cm^3 expansion vessel. The anode is ~3 mm in diameter. The detection volume is 20 cm^3. The minimum-detectable tritium activity in helium is 4 kBq/m^3 when this detector is coupled to a 1-fA transimpedance amplifier.

The saturation current's dependence on tritium concentration for fixed total pressures in the 20-cm^3 wire-cage detector increases linearly with tritium content over the 20- to 60-kPa

(150- to 450-Torr) pressure range as shown in Fig. 144.7. Figure 144.8, however, indicates that the increase in the saturation current with increasing pressure for different tritium activities in deuterium is less than linear for the 20-cm^3 detector. Evidently, electron–ion recombination in the gas becomes progressively more significant as the total pressure in the chamber increases.

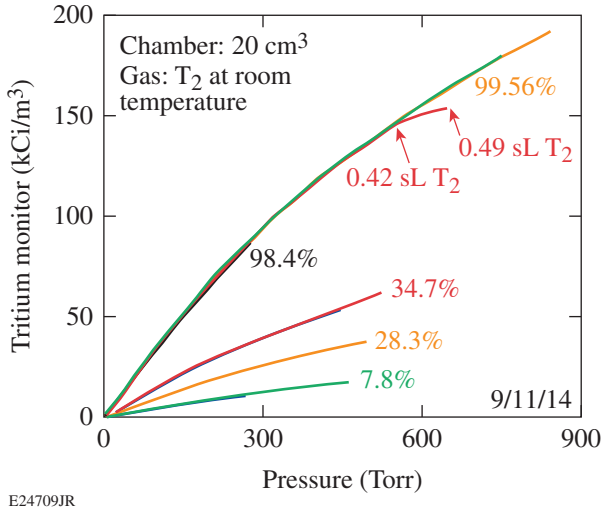

Figure 144.7
Response of the 20-cm^3 wire-cage ionization chamber to tritium activity at fixed total pressures.

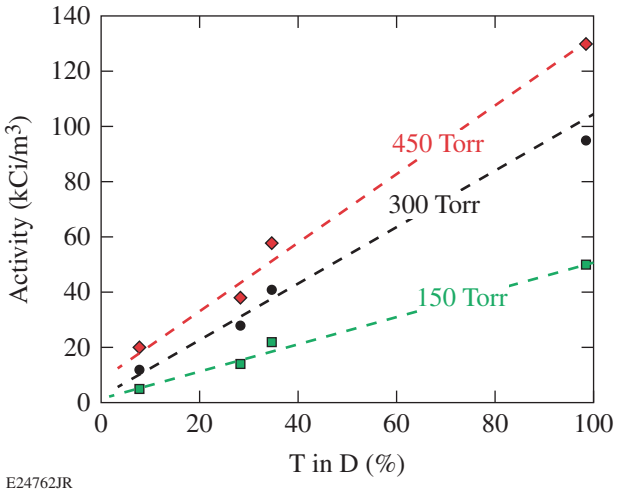

Figure 144.8
Response of the 20-cm^3 wire-cage ionization chamber to an increasing total pressure for different concentrations of tritium in deuterium.

4. LN_2 System

The thermal ramp of the columns in the Core System must be in the vicinity of 0.5 to 1 K/s for good separation. A liquid nitrogen cooling circuit was installed to achieve this precipitous temperature swing. A 350-L dewar is housed in a separate room that is ventilated directly to the room exhaust ventilation duct. Liquid nitrogen is transferred to the columns via vacuum-jacketed lines. Gaseous nitrogen is directed to the room exhaust via a second set of vacuum-jacketed lines. Several automated responses have been designed into the LN_2 System to address off-normal events. These include oxygen-deficiency monitors; system isolation if the cooling rate deviates from the expected trajectory; an independent, centralized monitoring station; and pagers to notify key personnel if parameters depart from design values.

5. Vacuum System

The Vacuum System is manually isolated from the Gas Handling System by two Swagelok 8B series valves fitted with copper stem tip valves to reduce the potential for inadvertent evacuation of high-activity gas from the process loop. A 1-L expansion vessel that also serves as a tritium detector separates the two manual valves. Vacuum is generated by a conventional turbomolecular/scroll pump combination.

The Gas Handling System is evacuated under stringent safety protocols to prevent the release of tritium into the environment. Safety protocols require operators to pre-evacuate the 1-L detector, isolate the detector from the Vacuum System, expand the contents of the Gas Handling System into the tritium monitor, assay the activity of the gas, and then direct the gas in one of three directions. The effluent can be (a) discharged directly to the room exhaust ventilation duct; (b) passed over a ST198 alloy to reduce the amount of effluent discharged to the room exhaust; or (c) injected into the Glovebox Cleanup System for additional treatment after a preliminary pass over the ST198 alloy.

6. Glovebox Cleanup System

The Glovebox Cleanup System is a stand-alone recovery system used to extract tritium from gloveboxes that use helium gas as the working atmosphere.[5–7] The system is built around ST198 alloy from SAES Getters, located in Milan, Italy. Elemental hydrogen forms a hydride with this alloy. The process is reversible. The alloy is periodically regenerated to recover tritium gas and the "emptied" bed is pressed back into service.

Typically the life expectancy of such a bed is several years before the capacity of the alloy is reduced to unacceptable levels by oxidation. Both water vapor and oxygen permeate into the glovebox from the atmosphere via rubber gloves. Both gases irreversibly consume the ST198 alloy.

The elements of the Glovebox Cleanup System are illustrated in Fig. 144.1. Gas is extracted from the glovebox with a Senior Flexonics' MB158 diaphragm pump through a tritium monitor and discharged into a train of three getters: a molecular sieve drier to remove water vapor, a nickel bed to remove oxygen, and the ST198 bed. Treated gas is returned to the glovebox via a second tritium monitor. The box pressure is adjusted to remain about 2 Pa below the ambient pressure. Treated helium can be discharged to the environment just ahead of a 2-kPa pressure relief valve (PRV) to reduce the box pressure or fresh helium can be added to the box downstream of the PRV to increase the box pressure. This system is currently operational and maintains the glovebox atmosphere at a dew point below −60°C and the oxygen and tritium concentrations below 10 ppm and 7 MBq/m^3, respectively.

ACKNOWLEDGMENT

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