The Production of Ultra-pure Hydrogen

QUALITY CONTROL OF PALLADIUM ALLOY DIFFUSION UNITS

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The successful development of Johnson Matthey Metals palladium alloy diffusion units for the production of ultra-high purity hydrogen is largely dependent on comprehensive testing of all units before they are released to customers. This article describes the test procedures and the equipment used in testing.

Ultra-pure hydrogen is being used today in an increasing number of industrial applications as diverse as gas chromatography and the furnace atmospheres required for the sintering and annealing of refractory metals. Some recent unpublished work even suggests that a dramatic reduction of wear in wire-drawing dies can be achieved by annealing wires in very pure hydrogen to prevent the formation of abrasive oxide

Dimensions and Recommended Operating Conditions of J.M.M. Diffusion Units

	Model	Physical Dimensions, mm W	D	H	Recommended Operating Temp. °C	Recommended Feed Gas Pressure lbf/in²	kgf/cm²	Pure H_2 Output ft³/h	l/h
Laboratory Range Units	H 28/1	203.2	203.2	558.8	300	300	21.0	1	28
	H 140/5	203.2	203.2	558.8	300	300	21.0	5	140
	H 480/17	203.2	203.2	558.8	300	300	21.0	17	480
	H 1260/45	203.2	203.2	558.8	300	300	21.0	45	1,260
Integral Units	HK 7	531	577	1399	300	150	10.5	250	7,000
	HK 14	531	577	1399	300	150	10.5	500	14,000
Large Units with Separate Console and Pressure Frame	HK 21	console 531	577	1399	300	150	10.5	750	21,000
		pressure frame 1371	686	1015					
	HK 28	console 531	577	1399	300	150	10.5	1,000	28,000
		pressure frame 1371	686	1015					

W = width   D = depth   H = height

Fig. 1 The rear of the pressure frame of an HK 28 diffusion unit, showing the compact arrangement of four diffusion modules and the ease of access for servicing

surface films. However, the major usage continues to be in the manufacture of semiconductor devices where the highest standards of purity and operating safety are required.

Johnson Matthey Metals caters for all these applications with its ranges of H and HK diffusion units (1). The table shows the principal dimensions and operating characteristics of all standard JMM units.

Performance of these diffusion units has been recently still further improved. A few early models were found to have a tendency to leak where the tubular alloy membranes were joined to the manifold or header plate but a new and advanced brazing technique has now virtually eliminated this source of leakage. The method consists of forming a controlled fillet of braze metal on each side of the joint while preventing the brazing alloy from undercutting into the thin-walled palladium tubes and thus causing pinhole perforations. This development has made a significant contribution to improved reliability and service life, particularly under conditions of thermal stress cycling.

Intrinsic Safety

All Johnson Matthey Metals diffusion cells are built to the British Standard Code of Practice CP.1003 Parts I and II, relating to the safety of electrical appliances in the presence of explosive or flammable atmospheres. Diffusion cells must be intrinsically safe, particularly the larger units such as the HK 28 shown in Fig. 1 since such a unit may be producing pure hydrogen at more than 28,000 l/h (1,000 ft^3/h). Johnson Matthey Metals engineers are continually seeking to improve the efficiency and safety of their diffusion units, and considerable care goes into the design of operating and safety features. For example, current HK units are now operated by illuminated push buttons linked to an interlocked sequential

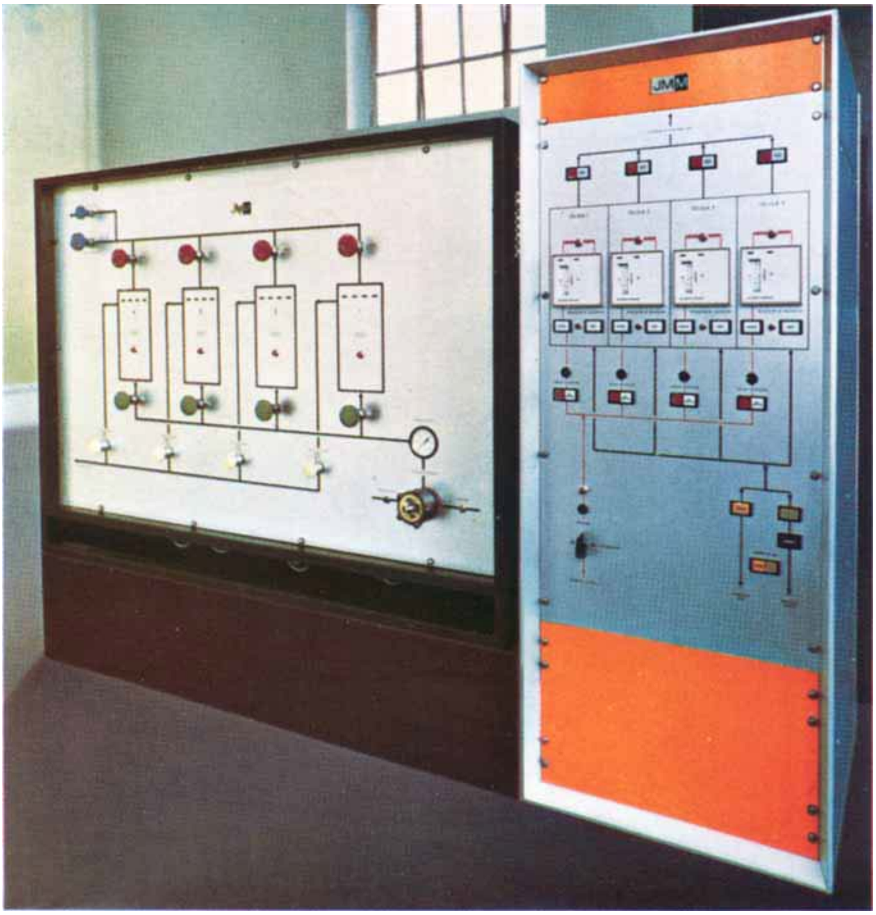

Fig. 2 An HK 28 diffusion unit viewed from the front, showing the ergonomic layout of the controls on the pressure frame (left) and on the control console (right)

control system, making the units virtually foolproof in use. The push button control system can be seen on the front of an HK 28 unit in Fig. 2. Other safety devices include both strategically placed thermal fuses and automatic purging with nitrogen to ensure that the units fail safe in the event of fire or of the failure of gas or power supplies. Solid state temperature controllers are used to regulate the operating temperature (normally 300°C) of the units and these controllers incorporate over-temperature detection devices.

In theory palladium alloy diffusion membranes are capable of supplying hydrogen of 100 per cent purity. In practice very high purity can only be achieved by skilful design and by stringent quality control procedures during fabrication to minimise the occurrence of leaks in components and assemblies. Each H or HK diffusion unit consists of assemblies of silver-palladium tubes, all of which are eddy current-tested for flaws or for manufacturing defects before closure at one end to form diffusion membranes. Each tubular membrane is then pressure-tested to 6,900 kN/m^2 (1,000 lb/in^2) to detect leaks at the closure weld. Membranes passing this test are assembled into a manifold or header plate to form a diffusion module and the

Fig. 3 A typical diffusion module being evacuated in preparation for leak testing on the helium mass spectrometer. Only modules with leak rates less than 2×10^{-11}cm³/sec at 1×10^{-5}torr are passed for installation in H and HK units

tubes are sealed into the manifold by the method described on page 125. This stage of assembly having been completed, the modules are given their most searching test by evacuation and leak testing on a helium mass spectrometer. A module on test is shown in Fig. 3. Only modules with a leak rate of less than 2×10^{-11} cm³/sec at an operating pressure of 1×10^{-5} torr can be passed for installation into H and HK units.

Testing of Assembled Units

Completed units are subjected to a thorough testing and inspection before leaving the works. After testing to ensure correct functioning of the complex electrical circuits, each unit is coupled to a suitable gas supply and is run up to its normal operating temperature and pressure on a special flow test rig. Figure 4 shows both H and HK units under test connected to this rig. The output of pure hydrogen is checked there against the design requirements and during this latter test the pure hydrogen being produced is sampled and checked for its moisture content, using a Shaw dew-point meter. The dew-point of the gas produced must be less than −60°C (equivalent to a moisture content of 10 p.p.m. of water vapour), although experience has shown that the dryness and purity of the gas improves steadily with time, and after three hours operation the dew-point can be expected to approach −80°C (1 p.p.m. water vapour). The relationship between water vapour content and cell running time is shown in Fig. 5. Further improvement is only limited by the overall leak rate of the system in which the unit is operating. On completion of flow and purity tests, each complete unit is returned to the mass spectrometer and is rechecked to the same high standard of leak tightness.

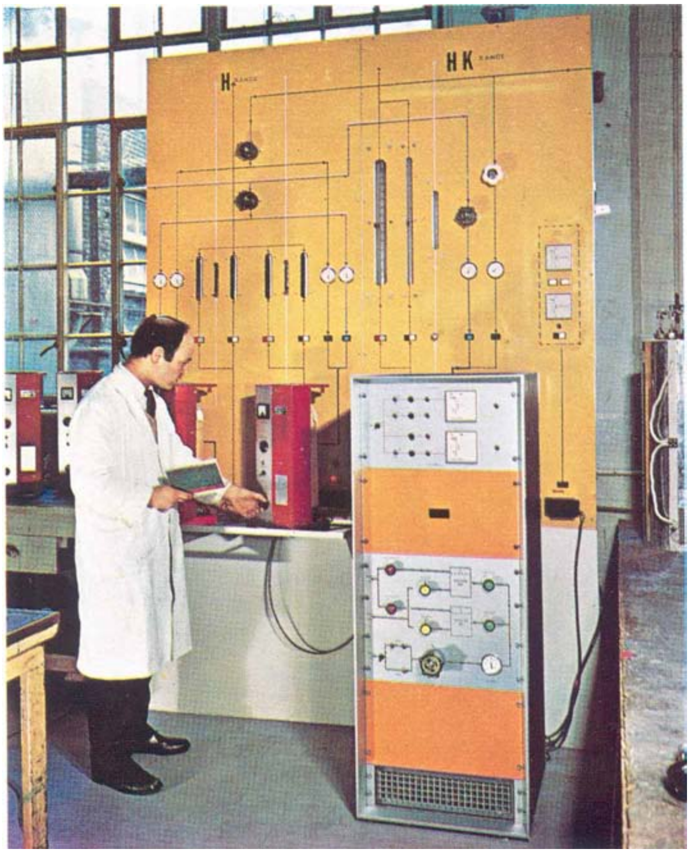

Fig. 4 Performance testing of H and HK diffusion units on a specially developed flow test rig. The operator is adjusting an H series unit while an HK series unit is on test on the right

The value of such thorough quality control can be seen in the remarkable performance achieved by Johnson Matthey Metals diffusion units, some of which have produced more than 42,500 m^3 (1.5×10^6 ft^3) of pure hydrogen in the course of their normal service life. JMM engineers have not only developed these highly reliable units but they also supplement them with a comprehensive range of advice and maintenance services to ensure continuous operation.

Reference

1 P. M. Roberts and D. A. Stiles, *Platinum Metals Rev.*, 1969, **13** (4), 141–145

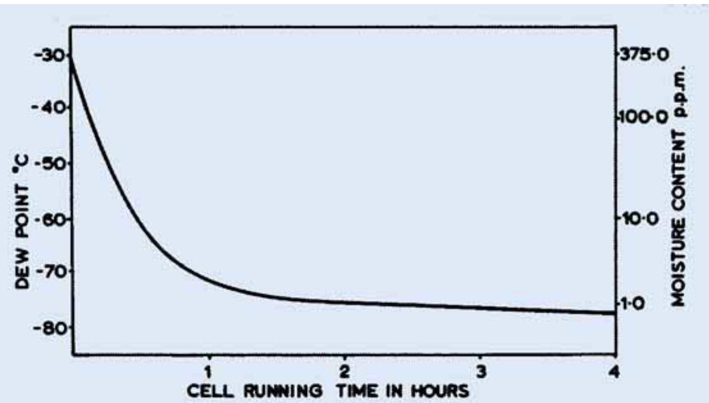

Fig. 5 Typical relationship between the dew-point of pure diffused hydrogen and the cell running time