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METHOD FOR REDUCING THE PERMEABILITY OF ALLOYS BY HYDROGEN

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5 Claims. (Cl. 148—16.7)

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This invention relates to metal containers for holding hydrogen at elevated temperatures and to a method for treating metals to reduce the permeability thereof by hydrogen at elevated temperatures.

It is well known that metals, such as alloy steels, Nichrome, etc., commonly used for hydrogenation equipment and other hydrogen containers, laboratory equipment, etc. are permeable by hydrogen at elevated temperatures resulting in large loss of hydrogen from such equipment. Because of such losses, the determinations of the dissociation temperatures, etc. of many compounds which evolve hydrogen at elevated temperatures have varied greatly and have been unreliable or only approximate.

I have discovered that alloys comprising iron, nickel and chromium, such as stainless steel, etc., when heated at an elevated temperature in contact with hydrogen for a prolonged period of time become gradually less permeable by hydrogen and after prolonged heating in contact with hydrogen for several days their permeability by hydrogen may be reduced one hundredfold or more. The temperature used should not be substantially less than that at which the gas is fully mobile with the metal being treated. In general, this temperature is about 600° C. In the treatment of such alloys I have obtained good results by heating at a temperature between about 600° C. and 900° C. but I prefer to employ a temperature of about 800° C. A higher temperature may be used but usually is not necessary. I have further discovered that the advantageous results thus obtained can be destroyed to a large extent if the heated metal is exposed to air at room temperature and to a greater extent if the treated metal is exposed to air while heated at an elevated temperature. Therefore, in order to preserve the imparted property of low permeability by hydrogen, the treated metal or the treated fabricated article or at least one surface thereof should be kept in an environment free of oxygen, such for example as hydrogen, argon or other monatomic gas.

The invention is illustrated further by the following specific examples showing the diffusion rate of hydrogen through specific iron, nickel and chromium alloys. In each of these examples the diffusion rate was measured with a mercury slug flow meter which was connected by a hermetically sealed connection to one end of the metal tube being tested. A thermocouple well of silica was inserted through the other end and the latter was hermetically sealed. The tube being tested was placed in an electrically heated thermostatically controlled tube furnace. In each example, hydrogen at atmospheric pressure was used. The materials used were Nichrome, 304-Alloy stainless steel, 316-Alloy stainless steel and 347-Alloy stainless having the composition in per cent indicated below.

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Element	304-Alloy	316-Alloy	347-Alloy	Nichrome
Carbon	0.08 max	0.10 max	0.10 max	0.10
Manganese	2.00 max	2.00 max	2.00 max	
Silicon	1.00 max	1.00 max	1.00 max	
Phosphorus	0.04 max	0.04 max	0.04 max	
Sulfur	0.04 max	0.04 max	0.04 max	
Chromium	18–20	16–18	17–19	16
Nickel	8–10	10–14	9–12	60
Molybdenum		1.75–2.50		
Columbium			1.00 max	
Iron	bal	bal	bal	24

Example 1

In this example the material treated was a 304-Alloy stainless steel tube having a wall thickness of 0.9 mm. (0.036''). The hydrogen loss as indicated is cubic centimeters of hydrogen at room temperature per square centimeter of surface per hour.

Hours heated	H-loss cc./cm.²/hr.	Temperature, ° C.
6	0.195	800
27	0.108	800
51	0.0369	800
78	0.0324	800

Example 2

In this example, after heating the 304-Alloy stainless steel tube for 78 hours, the tube was cooled to room temperature, the hydrogen pumped out and air admitted for thirty minutes. It was then evacuated, filled with hydrogen and heated at 800° C. The diffusion rate now was 0.0434 cc./cm.²/hr. The hydrogen in the tube then was replaced by air and the tube heated about 50 minutes at 800° C. The air in the tube then was replaced by hydrogen and the tube heated at 800° C. for about twenty hours and the diffusion rate was 0.0650 cc./cm.²/hr.

Example 3

In this example, a 316-Alloy stainless steel tube was treated. The hydrogen loss is given as cubic centimeters of hydrogen at room tempera-

ture per square centimeter of surface per hour per millimeter of thickness.

Hours heated	H-loss cc./cm.²/mm./hr.	Temperature, ° C.
24	0.15	650
240	0.03	650
504	0.02	650

Example 4

In this example, a 347-Alloy stainless steel tube was treated. The hydrogen loss is given in the same terms indicated in Example 3.

Hours heated	H-loss cc./cm.²/mm./hr.	Temperature, ° C.
96	0.1	650
384	0.01	800

Example 5

In this example, a Nichrome tube was treated. The hydrogen loss is given in the same terms indicated in Examples 3 and 4.

Hours heated	H-loss cc./cm.²/mm./hr.	Temperature, ° C.
24	0.12	650
120	0.06	650
240	0.002	650

I claim:

1. The method for reducing the permeability of an alloy by hydrogen which comprises heating the alloy in an atmosphere consisting essentially of hydrogen at a temperature not less than about 600° C. for a period of time not less than about 24 hours, said alloy comprising iron, nickel and chromium.

2. The method for reducing the permeability of stainless steel by hydrogen which comprises heating the steel in an atmosphere consisting essentially of hydrogen at a temperature not less than about 600° C. for a period of time not less than about 24 hours.

3. The method for reducing the permeability of an alloy by hydrogen which comprises heating the alloy in an atmosphere consisting essentially of hydrogen at a temperature not less than about 600° C. for a period of time not less than 24 hours, said alloy comprising iron, nickel, chromium and not more than about 0.10 per cent carbon.

4. The method for reducing the permeability of an alloy by hydrogen which comprises heating the alloy in an atmosphere consisting essentially of hydrogen at a temperature not less than about 600° C. for a period of time not less than 24 hours, said alloy comprising iron, nickel, chromium and not more than about one per cent columbium.

5. The method for reducing the permeability of an alloy by hydrogen which comprises heating the alloy in an atmosphere consisting essentially of hydrogen at a temperature not less than about 600° C. for a period of time not less than 24 hours, said alloy comprising iron, nickel, chromium and between about 1.75 to 2.50 per cent molybdenum.

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