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Dipl.-Ing. Roland Waitz, Linn High Therm GmbH (Germany)

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Editor: Dipl.-Ing. Stephan Schalm, Tel. +49 (0) 201/82002-12, E-Mail: s.schalm@vulkan-verlag.de

Muffle furnaces for temperatures from 200-1200°C with controlled atmospheres and vacuum

Roland Waitz

The "Hot Wall" heating technology for furnace operation under controlled atmosphere and vacuum up to 1200°C is briefly described on the basis of KS-S, a small to medium sized furnace line with chamber volumes from 80 dm^3 to 2 m^3. Realizable heating- and cooling rates as well as the influence of forced convection and multi zone heating on temperature distribution are discussed. A comparison between the residual oxygen content by fore-vacuum or flushing gas method and brief description of safety devices for operation with explosive gases are also part of this paper.

Thermal treatment of materials in protective gas or vacuum is a commonly used process in material science. Besides its function to prevent oxidation, gas and mixtures of gases can be used to modify material surface, e.g. nitrating or carbonising. In general, two different designs of furnaces for thermal treatment can be distinguished: A "cold-wall" design and a "hot-wall" design.

In "cold-wall" furnaces the heater and the insulation are installed inside a vacuum or gas tight chamber. In a "hot-wall" system a vacuum and/or gastight retort, so-called muffle, is heated from the outside. The design of the KS-S furnace line is based on the "hot-wall" principle.

The heating system a of KS-S furnace consists of spirals of Kanthal A1© wire or APM© wire. These wires are placed in notches of the inner insulation, which is usually made of ceramic light bricks. Micro porous material on the backside complete the insulation. This whole unit is encaged in a metal frame. A gap between the frame and the outside housing, filled with air, leads to an additional insulation effect. This design limits the maximum surface temperature of the furnace to 50°C above room temperature.

In the standard configuration the furnace is heated from three sides and also from the bottom. Additional heaters can be installed on ceramic tubes at the attic of the furnace. A furnace with this improved heating power allows an operation at elevated temperatures up to 1200°C or a faster heating cycle with heavy loads.

Kanthal A1 (APM) wires are produced from ferritic (iron/chromium/aluminium alloys doped with rare earth elements like lanthanium). Rare earth elements stabilize the grain boundaries and give protection against inner oxidation, while Cr acts as protection against corrosion. Aluminium, with its high affinity to oxygen, forms a gastight layer of Al_2O_3 on the heater's surface which gives further protection against oxidation. The maximum temperature for A1-wires is 1400°C, for APM-wires 1425°C, which is far higher than the maximum temperature of the furnace (1200°C). These temperatures allow a safe operation of the wires in the furnace.

The heaters are connected to three phase line voltage (400 V). The installed power vary from 15 kW (for 70 dm^3 useable volume in the furnace) to 51 kW (for 460 dm^3 useable volume), equivalent to 200-100 W/dm^3. This yields a heating up time of app. 1.5 h from room temperature to 1000°C (**Fig. 1**).

Muffle

The muffle is a welded box or cylinder made out of heat resistant steel, a water cooled frame for the door sealing, and a door with a combined insulation of

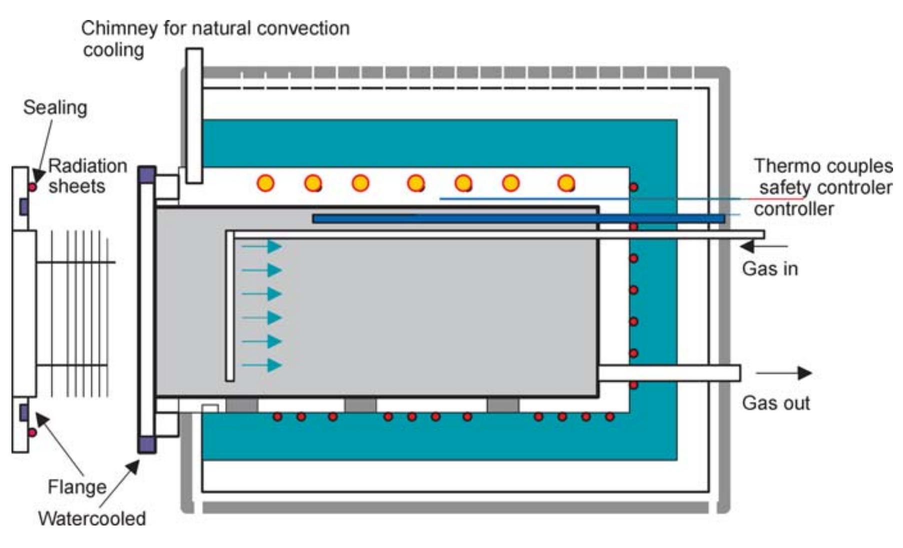

Fig. 1: Scheme of a KS-S Furnace

radiation sheets and encapsulated ceramic fibre. The wall thickness and the material depend on size, maximum temperature, and further process parameters.

The most important challenge for the muffle design is to avoid thermal stress. Especially during fast heating and cooling cycles thermal stress can damage the muffle. Rounded corners reduce thermal stress. In addition, the temperature gradient along the section of the water cooled door sealing must be minimized. The design of the door with several radiation sheets diminish the thermal stress inside the muffle mouth because the temperature gradient is distributed over a longer distance.

The maximum temperature for continuous operation of the silicone door sealing is 220°C. Therefore the flat face of the rectangular frame must be water cooled. With additional cooling in the door, the sealing is located in a cold labyrinth passage and perfectly protected.

The water cooled part of the muffle acts as heat sink. If the furnace is used for heat treatment by steam or with high flow of bubbler wetted gases it is recommended to use oil as coolant rather than water. Oil cooling allows operation temperatures up to 140°C, which is beyond the boiling temperature of water and therefore avoids condensation inside the door frame.

The standard material in furnace design for temperature up to 1050°C is steel 1.4841 (X15CrNiSi25-20). Depending on the operating conditions and the process, other materials may be used, e.g. Nickel alloy 2.4851 (Inc 601) NiCr23Fe for temperatures up to 1150°C or 2.4633 (Inc 602CA) NiCr25FeAlY which allows temperatures up to 1200°C.

Control Unit

The design of the Control Unit, particularly the positioning of the thermocouples in a hot wall furnaces, requires some preparatory thoughts which consider the temperature – time regime of the process.

One practicable solution is to place the thermocouple of the controller into a welded-in tube in the protective gas chamber. Thereby it can be easily changed from the outside and is not exposed to the possibly stressful furnace atmosphere. This is important for the KS-S furnace line designed for temperature over 1050°C using Pt-Rh thermocouples Type S. Exposed to hydrogen these elements are "poisoned" and the thermoelectricity changes.

A safety controller supervises the temperature in the room between the heater and the protective gas chamber. This assures that the protective gas muffle remains undamaged during fast heating – up cycles and/or heavy charges.

In addition, a flexible dragging thermocouple with a display und recording temperature inside the chamber enhances the features of the furnace. As it measures and records the real temperature in/on the charge it guaranties the quality and safety of the heat treatment.

Nevertheless, it still may come to overheating at fast heating-up cycles and/or during dwell times at lower temperatures (<400°C). The thermocouple in the tube, in combination with the controller, may react with delay e.g. in the beginning of a cycle.

This effect can be avoided by choosing a slow heating rate in the beginning and suitable PID control parameter. By positioning the thermocouple outside of the gastight chamber a linear heating without overheating can be accomplished. Still the difference in temperature between the furnace and the chamber for each temperature range can only be measured empirically.

With the help of modern difference controller, which uses the inner elements as guidance and the outside element as actuating variable with an adjusted offset, this problem can be solved.

Temperature distribution

The temperature distribution or uniformity in a furnace is primarily influenced by the geometry of the furnace and the distribution of the heating elements inside and the insulation.

For a "hot-wall" design a deep furnace is preferred. A very effective and uniform heating is a heating system from 5 sides (4-sided heating system shows only small temperature deviations which might also be tolerated). The temperature uniformity in a KS-S furnace is approximately +/-15 K to +/-20 K, depending on overall temperature and size.

The general factors which determine heat transmission and temperature uniformity are convection and radiation. Thermal conductivity of the gas has only a marginal influence. Two options for improved heating are offered by Linn High Therm: A 3-zone heating and a forced convection.

In a 3-zone heating each zone of the furnace can be heated independently of the others. Thus possible local heat loads or local heat sinks can be adjusted also locally. This design improves the over-all temperature uniformity inside the furnace.

Secondly, forced convection is offered. The heat is transported inside the furnace by an enforced gas circulation. Normally a fan integrated in the furnace door provokes this gas flow. A normal gas circulation works up to a maximum temperature of 850°C. In processes up to 1050°C with gas circulation it is necessary to slow down the fan at high temperatures. However, a minimum revolution, e.g. 2 Hz, must always be maintained to avoid damage to the axle. Otherwise the axle warps.

Natural convection caused by a temperature gradient inside the furnace provokes a gas flow inside the furnace. It is the dominant factor for heat transport up to approximately 200°C, depending on surface and emission factor of the

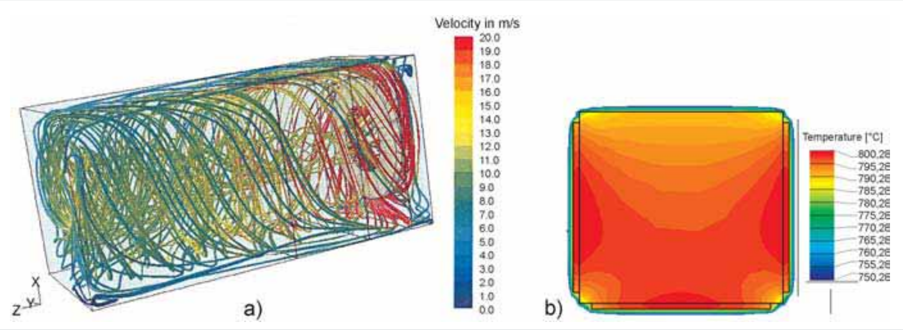

Fig. 2: a) Path lines and gas velocity inside KS80 S furnace with forced convection, b) Calculation of middle cross section of KS 80 S

used materials. At higher temperatures the thermal transfer by radiation prevails. Forced convection by means of a blower extends the range of dominance up to approximately 400°C and stays effective up to 800°C. The efficiency of forced convection increases from Argon, Nitrogen to Helium and Hydrogen. Beyond 800°C the thermal transfer and tem-perature is primarily controlled by radiation.

In a furnaces with appropriate controlled atmosphere where metal has an oxide free surface with low emission factors down to less than 0,1 the heat transfer coefficient in influenced by forced convection even up to more than 1000°C (**Fig. 2**).

If an exact temperature distribution up to 400°C over a large volume is required a gas recirculation should be used for heat treatment. Over 800°C furnace temperature a 3-zone heating is essential. Between 400°C and 800°C both methods should be combined.

Forced convection additionally improves the heat transmission. This is important when metals are heat treated at low temperatures. The main function of a protective gas furnace (or a vacuum furnace) is to prevent oxidation. The emission factor for non oxidised metals is comparable low, generally values 0.05 to 0.2. By forced convection the heating time is reduced by more than 50%.

Regarding a multi-zone heating, in most cases it is put into practice with 2 or 3 control paths which are allocated over the length of the furnace (door area, middle part, rear side). In such an arrangement, temperature deviates between +/-3 K and +/-7 K over the total furnace volume. For special applications even better uniformity can be guaranteed by a 6-zone heating. However, instead of building a 6-zone heating in most cases it is sufficient to reduce the used volume inside the furnace to achieve the same effect but at lower costs.

Fast cooling devices

The natural cooling rate of a furnace can be described roughly by the function $T=T_{,}e^{-kt}$. For KS-S furnaces the natural cooling rate is around 8 K/min at 1000°C, 6 K/min at 800°C, 3 K/min at 500°C, descending to 1 K/min at 250°C.

For oxygen sensitive materials the furnace temperature should fall below 150°C before opening the doors. This leads to extremely long cool-down cycles. A fast cooling device can reduce the cooling-off time significantly. Fresh cold air, blown by fans into the space between gas chamber and the heaters, reduces the total cooling-off time. Used hot air leaves the furnace through an exhaust on the upper side (and must be eliminated). In order to protect the inner muffle from damage, a fast cooling device should operate only at temperatures below 500°C. Starting the blowers at 500°C, the cooling rate will increase to 10-12 K/min and remains effective (>2 K/min) even at 200°C. A frequency controlled blower motor raises the starting temperature for forced convection to 1000°C because it allows to reduced the rotation speed of the blowers at high temperatures. This design achieves an the average cooling rate of 10K/min above 500°C.

Further improvement for cooling above 500°C can be obtained by a recirculation of the working gas over a heat exchanger. This device reduces the cooling time to room temperature to app. 50 min.

Table 1: Evacuation time for 240 l volume 16 m³/h rotary vane pump

Pressure [mbar]	10	1	10^{-1}	$3x10^{-2}$
S_{eff} [m³/h]	15,96m³/h	15,64	12,4	4
evacuation time	4 min 10 sec	6 min 35 sec	9 min	26 min

Vacuum operation

Furnaces for vacuum operation contain a reinforced muffle because of the pressure from outside. At temperatures higher than 500°C the used alloys loose their mechanical stability. The strength of the Ni based alloys (INCONEL) is higher than Fe based alloys. The maximum temperature for a rectangular muffle (depending on its volume) is limited to 600-700°C. With a cylindrical form the maximum working temperature under vacuum increase to 800-1000°C.

Pre-evacuation the muffle before operation is a possibility to eliminate oxygen in the chamber. Applications for this method are wire coils, bulk material or powder which are to be heat treated. Oxygen in the narrow gaps between the windings or particles can easily be removed by vacuum, but hardly rinsed only by scavenging gas. Normally, a final pressure of 10^{0}-10^{-1} mbar is low enough before purging with gas. Repeating this procedure improves the purity. However, it cannot remove adsorbed water and gas in acceptable time. For loads with large surfaces

Fig. 3:
KS240Vac for annealing of springs

and/or high sensitivity it may be helpful to repeatedly evacuate the muffle at a temperature >150°C.

A two stage rotary vane pump with a volume flow rate of some m^3/h generates a fine vacuum of 10^{-1} mbar, sufficiently low for most applications. In the process of the heat treatment the evacuation time is not critical. E.g. the evacuation time for a muffle volume of 240 litre connected to a pump with 16 m^3/h volume flow between $p = 10^3$ mbar and $P = 10^{-2}$ mbar with an estimated loss of pump efficiency by a leak rate of $L = 0.1$ mbar l/s = 0.36 mbar m^3/h. The connecting tubing should be wide and short enough to have no influence (**Table 1**).

Normally the leak rate is in range of 0.01-0.001 mbar l/s. However, this times can only achieved in a clean and dry furnace. A moist furnace at 20°C and 20 mbar evaporates mainly condensed water. Most of absorbed water and gases evaporate between 10^{-2} to 10^{-4} mbar. At pressure lower than 10^{-4} mbar desorption becomes the dominant factor. The inner surface of the vacuum muffle is comparatively small. Therefore in a cold state a final pressure in the range of 10^{-5} mbar is realizable with wide flange connection and strong turbo molecular pump or a diffusion pump. Under hot conditions still 10^{-4} mbar can be accomplished (**Fig. 3**).

Protective gas

Protective gases can be classified into three groups, which are oxidising gases, inert gases and reducing gases (**Table 2**). Examples are:

- oxidising gases: air, oxygen, water (at high temperature) and carbon dioxide.
- inert gases: noble gases, particularly argon and helium, and (in the lower temperature range) nitrogen.
- reducing gases: hydrogen, methane, ammoniac.

In general, reducing gases are burnable and mixtures of them with air are explosive. Only forming gases, a mixtures of hydrogen and neutral gases, with less than 5% H_2 in more than 95% neutral gas, is reducing but not burnable under normal conditions. Therefore, after loading a furnace it must be filled with scavenging gas to reduce the oxygen level (air) inside before any operation under a reducing atmosphere can start. Reducing atmospheres require a neutral scavenging gas. A flushing factor of 5 must be assured. A muffle with a volume of 10 dm^3 (including piping) needs a flow through of 50 dm^3 before the gas supply may be switched over to burnable gas. Theoretically, a purging factor of 2 is enough but blind holes and corners make a higher flushing factor very recommendable (**Table 3**).

During the heat treatment the required gas flow depends on the desired application. For "clean" processes like annealing the gas flow should be app. 1-2 times the furnaces volume per hour. For "dirty" processes like graphitisation the value rises to 10 times per hour. This may also necessary for oxygen sensitive materials like stainless steel in hydrogen atmosphere to keep the H_2/H_2O ratio high.

The gas feeding device is installed directly to the furnace and tightly connected with it. It provides two fittings and lines for gas with blocking valves and needle proportioning valves. These tubes are connected to one single gas supplying line. The flow meter with the needle valve is installed in this line. A throttle valve in the exhaust line keeps the furnace under a slight over pressure of 4-7 mbar. This prevents the penetration of oxygen in case of leaks. All tubes are made of copper or stainless steel.

Beside this standard gas supply a lot of customize solutions are available. For example installations for 3 to 8 different gases with mass flow controllers and accurate mixing of gases are available. For nitrating, carbonising and nitro-carbonising processes a complete system with sensors and control equipment can be offered. The use of inflammable gases is possible in combination with a burning-off device. Inflammable protective gases must be burned off before it gets into the air or exhaust lines. This is done by a small gas burner with flame supervision. The ignition by a spark can be started by a pressure switch or by the furnace controller. When the flame burn down, the magnetic valve in the protective gas line closes; at the same time the magnetic valve in the nitrogen line opens. The burn-off device is readily

Table 2: Maximum allowable concentration MAC

Gas	Chem. for mular	Spez. Weight Compared with air	Explosion limit in air (low) 20 °C, 1013 m bar	Explosion limit in air (high) 20 °C, 1013 m bar	Min Temp. for burning	Danger Explosive Poisonable Suffocation
Ammonia	NH_3	0,72 kg/m^3 (-)	15 %	27 %	690 °C	Ex, Po, Su
Hydrogene	H_2	0,084 kg/m^3 (-)	4 %	74 %	570 °C	Ex, Su
Methan	CH_4	0,671 kg/m^3 (-)	5 %	15 %	580 °C	Ex, Su
Carbonmonoxid	CO	1.17 kg/m^3 (-)	12.5	74 %	630 °C	Ex, Po, Su
Propan	C_3H_8	1.88 kg/m^3 (+)	2.2 %	9.5 %	480 °C	Ex, Su
Endogas 1 based OnC_3H_8	31 % H_2, 23 % CO 46 % N_2	0,89 kg/m^3 (-)	7 %	72 %	560 °C	Ex, Po, Su
Endogas 21 based OnCH_4	40 % H_2, 23 % CO	0,79 kg/m^3 (-)	7 %	12 %	560 °C	Ex, Po, Su
Exogas	14 % H_2, 7 % CO 5 % CO_2,	1.12 kg/m^3 (-)	17 %	72 %	560 °C	Ex, Po, Su
Cracked ammonia	25 % N_2, 75 % H_2,	0.38 kg/m^3 (-)	3 %	72 %	530 °C	Ex, Su

Table 3: Flushing factor and remaining oxygen

Flushing factor	0	1	2	3	4	5	6	7	8	9
Comparable Vacuum [mbar]	1031	378	137	45	18	6.9	2.4	1	0.3	0.1
Remaining oxygen [%]	20	7.7	2.8	1	0.38	0.14	0.05	0.019	0.007	0.0025

installed with electric pilot burner, flame supervision and electric warning horn.

An additionally safety package improves the protection against explosions.

It includes:

a. Supervision of the purging time and the flow rate of purging gas.

Flow rate times purging time = 5 times furnace volume.

b. Supervision of the gas stock, if bottles are used. Before the system can be started the available gas stock in the bottles must be 10 x the furnace volume.

c. Monitoring of the residual oxygen content inside furnace. If the oxygen content rises higher than 1 % the system is automatically flowed with neutral gas. This gives protection against leakage or increasing oxygen levels.

d. The flow rate of protective gas is additionally monitored. Too little flow of gas during cooling might create partial vacuum. Any vacuum inside a normal gastight chamber could lead to implosion or suck in of air. Explosive mixtures of gases are possible.

e. Blocking of the furnace door during operation with protective gas. For opening the door, the purging by neutral gas has to be finished.

For environment protection against organic waste (gases) a thermal or catalytic combustion systems can be installed (**Fig. 4**). These systems are used in furnaces for debinding of MIM and CIM (metal/ceramic injection moulding) or graphitisation. In catalytic systems the organic load must not exceed 12 g/m^3. Catalytic systems are available for gas flows from 10 m^3/h to more than 100 m^3/h.

Additional equipment

- Bubbler for active brazing and partial oxidation.

  In a bubbler, gas sparkles through a heated water volume. By the temperature of the water the dew point of the gas and with it the oxidising potential is determined. For the main applications hydrogen with a dew point between -40°C and +40°C is common.
- For the generation of lower dew points than room temperature the wet gas is mixed with dry gas by MFC`s.
- Inspection glasses and lead through for sensor installation.
- Racks for bulk material and other charging equipment.

The applications of the KS-S furnaces are widely spread in many different industries and research projects:

- Metal-working industry:
  carbonitrating, nitrating,
  brazing and soldering,
  debinding of MIM parts.
- Springs and wire:
  annealing of wire coils,
  annealing of springs.
- Precious metals:
  development and quality control of car catalysts, production of catalysts for chemistry, refining of precious metals.
- Diamond and hard metal cutting tools: pre-sintering and brazing of diamond and hard metal cutting tools.
- Chemistry and graphite:
  production of luminescent powder, production of ferro oxide, production of carbon brushes.
- Ceramics:
  debinding of CIM parts.
- Precious mechanics and optic:
  brazing of lenses in lens mounting, brazing of photomultipliers under vacuum.
- Testing and quality control:
  long – time testing on materials for fuel cells, quality control on sheets for cans and tube materials.

Fig. 4:
KS 80 S with safety package

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Dipl.-Ing. Roland Waitz

Linn High Therm GmbH
(Germany)

Tel. +49 (0)9665 / 9140
waitz@linn.de