

*Notes on Gasification of Powdered
Coal in Suspension
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I. Gasification in suspension.

The theory of gasifying in suspension given by Dr. Gumz is essentially based on the rules of heat transfer and the complete analogy of heat transfer and the exchange of materia. But it is not quite sure that all those laws are valid in the case of a gas producer for powdered fuel, because two conditions are not regular in the sense of aerodynamics: The size of the particles is nearly the same as the thickness of the boundary layer and on the surface of the particles a heterogeneous reaction is going on, producing new materia in the gas phase, whereby the exchange of heat and materia is influenced.

The following is to outline a method of calculating the time required for gasification which is based on simpler propositions and leads to different results.

Generally speaking a chemical reaction may be influenced by several conditions, which are sometimes independent from each other. In such a case the speed of the process is limited by the condition requiring the longer time. In the special case of the heat consuming water gas reaction two conditions must be mentioned 1.) The difficulties of the exchange of materia through diffusion and convective exchange, 2.) the condition that the water gas reaction can only take place as long as a surface temperature of considerable hight is maintained.

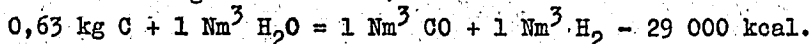
The readiness of carbon and steam to perform the water gas reaction is given by the concentration factor

$$K_{P_w} = \frac{CO \cdot H_2}{H_2O} \quad (1)$$

This factor marks a sharp edge with the abszissa about 800°C and therefore the mechanism of the heat absorbing reaction on the surface of a particle suspended in hot steam may be as follows: The particle is heated till the water gas process begins and the heat transferred to the surface is absorbed this way. Between 500 and 800°C the surface temperature can be raised by intensive heat transfer for the readiness of the substances is evidently not very great as could be seen from fig. (1) but at about 800°C the willingness for the reaction increases so sharply that the surface temperature can not be increased, even by most intensive

heat transfer, as long as enough reacting materia is at the disposal. If this is supposed a very simple theory can be derived:

The water gas reaction:



The decrease of weight W of a particle in differential time dt:

$$-\frac{dW}{dt} = \frac{0,63}{29000} \cdot q$$

where q means the quantity of heat transferred to the surface of the particle in unit time and consists of the two components: The heat transfer through radiation q_R and the heat transfer through convection q_C . Giving a special example, the temperature of the steam and the whole reactor is say 1200°C, the surface temperature of the particle is 800°C, q_R would be:

$$q_R = \pi \cdot d^2 \cdot \frac{10^{-6} \cdot 3,9 \cdot 0,5 \cdot 0,25 \left[\left(\frac{1200+273}{100} \right)^4 - \left(\frac{200+273}{100} \right)^4 \right]}{3 \cdot 600}$$

d [mm], q_R [kcal/sec]

The radiation is calculated as 50 % of the radiation of a "luminous flame" of 3 m diameter, for the inner of a gas producer is radiating less than a luminous flame and the reacting particle is partially surrounded by CO, absorbing heat radiation too. These factors cannot be given exactly but the problem is governed by the predominating influence of the Stefan-Boltzmann-link, containing T^4 , therefore a wrong estimation of a constant factor is of no great influence in this special case. The other part of the heat transfer is

$$q_C = -\frac{dW}{dt} \cdot \frac{1200 - 800}{0,63}$$

This formula will say that only this part of the surrounding medium is contributing to the convection heat transfer, which is taking part in the reaction itself, for all other medium is isolated from the surface of the particle by the reaction products H_2 and CO, which are of the same low temperature as the surface and cannot contribute to the heat surface therefore.

Substituting this formula for q and putting

$$W = \int \frac{\pi}{6} d \cdot 3 \cdot 10^{-9} \text{ and } -\frac{dW}{dt} = -\int \frac{\pi}{2} \cdot 10^{-9} \cdot d^2 \cdot \frac{dd}{dt}$$

a linear relation between dd and dt is found and on integration

$$Z_{\text{sec}} = d_0 \cdot 7,32 \cdot \frac{2900 - (i_{T_m} - i_{T_R})}{\left(\frac{T_m}{100}\right)^4 - \left(\frac{T_R}{100}\right)^4}$$

is obtained, where d_0 is the diameter of the particle at the beginning of the reaction given in [mm], T_m is the absolute temperature of the steam and T_R is the absolute temperature of the reacting surface [°Kelvin].

Fig. 2 is showing the time Z for various steam temperatures, when the temperature of the reacting surface is estimated with 800°C in accordance with the above described mechanism of the water gas process. (The objection could be made, that the estimation of the temperature of 800°C is a very rough one, but as may be seen from fig. 1 the real temperature cannot differ very much from this estimation and the difference to the general result is very small in consequence of T_m^4). (Fig. 5)

Fig. 3 shows the time required for gasification according to the calculation of Dr. Gumz, mentioning the difficulties of diffusion only without respect to the temperature of the reacting surface. The diagram shows a very small difference between the behaviour of coal dust from 900° to 1500°C . This is not likely to be proved true by experience and I consider the theory leading to such results not as wrong, but as incomplete and the worst is that it makes too optimistic promises especially at lower temperatures suggesting this way the possibility of a dry process.

Fig. 4 shows the results of the calculation by the two methods for the temperature of 1200°C . Line 1 marks the time required under the supposition of a reaction temperature of 800°C on the surface of the particles and line 2 shows the time required mentioning the difficulties of diffusion only. Fig. 4 says that between 0 and 0,4 mm diameter of particles the short times for gasification promised by line 2 cannot be verified in a real process, for it would sink the temperature of the surface considerably under 800°C , where the water gas reaction would be stopped for lack of temperature.

Quite the other way with particles bigger than 0,4 mm where the bottleneck is not the temperature but the exchange of materia. - In any case both limits are valid, but for practical consequences that limit must be considered, which causes the

time. In fig. 4 the times required are marked by the dotted line.

For gas producers heated with fuel powder of the same quality as it used for boiler firing most particles are below 0,1 mm ϕ , therefore the process in gas producers will be ruled by the relations shown in Fig. 2, for the diffusion limit given by Dr. Gumz will intersect these lines only at bigger diameters and very high temperatures.

In order to avoid the objection that the constant surface temperature of 800°C is estimated too high, fig. 5 shows the smallness of the influence of this estimation at a gas temperature of 1200°C, when the estimated reaction temperature is varying between 600 and 800°C.

Influence of mineral constituents.

A particle of say 0,05 mm diameter, which came from a particle of 1 mm original size will not behave exactly like a particle of 0,05 mm, which comes directly from the coal mill. The reason of the different behaviour is the different contents of mineral constituents. Mathematically this contents may be expressed as a function of the ratio of the diameter d at the moment when observed and the original diameter d_0 at the beginning of the process. It would be very difficult to find out this function by experimental methods, and the results could not be generalised, because there is always a catalytic influence, especially when bituminous coal is used, but it may easily be estimated what this function must look like (fig. 6). If we put the differential equation of the decrease of weight

$$-\frac{dW}{dt} = k \cdot q \cdot f(A)$$

then we can assume that the obstruction influence of the ash ~~contents is considerable only, when the ratio d/d_0 is very small,~~ that is towards the end of the process. If such a function of the ash influence is to be considered, the integration of the above written differential equation must be made graphically and would lead to infinite time required for complete gasification. ~~This is in accordance with the practical experience that in every process a rest of carbon remains unused, but the trouble with the infinite time can be avoided this way, that the time is searched,~~ where about 95 % of the carbon is consumed.

For practical application of the results in design of gas producers for synthesis gas with oxygen the following facts must be mentioned: The heat absorbing process begins at the moment, when all free oxygen is consumed. The temperature t_m is an average one between the temperature of the oxydation zone and the end of the gas way.

II. The "falling space theory"
and why it is not applicable.

Very small particles sink extremely slowly in the medium they are suspended in and there could be suggested that the watergas reaction on the surface of such a small particle might be an extremely slow one by the same reason.

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II. The "falling space theory" and why it is not applicable.

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In order to bring this thought into a mathematical form we can assume that the surface is reacting with a quantity of steam only which the particle is able to touch physically. This quantity of reacting medium per unit time is contained in a cylinder of the volume

$$v = \frac{\pi}{2} \cdot d^2 \cdot u \text{ [mm}^3\text{/sec]}$$

wherein d is the diameter of the sinking particle in mm. and u is the sinking speed in mm/sec.

The sinking speed u of particles between 0 and 0,15 mm can be given with satisfying accurateness by the parabola

$$u = f(\rho) \cdot d^2$$

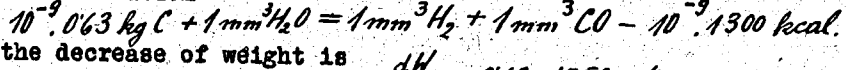
where f(\rho) depends from the toughness of the medium, its temperatur and the specific weight of the particle. Therefore

$$V = \frac{\pi}{2} \cdot d^2 \cdot f(\rho) \cdot d^2 = f(\rho) \cdot d^4$$

Assuming a temperatur \rho of 1200 °C the normal contents of the falling space would be

$$V = 1950 \cdot d^4 \text{ (mm}^3\text{/sec)}$$

Applying the equation for the water-gas reaction in an adequate transformation



the decrease of weight is $-\frac{dW}{dt} = 0,63 \cdot 1950 \cdot d^4$

Substituting the decrease of weight by the decrease of diameter

$$W = \rho \cdot \frac{\pi}{6} \cdot d^3 \quad \frac{dW}{dt} = \rho \cdot \frac{\pi}{2} \cdot d^2 \cdot \frac{dd}{dt}$$

the complete differential equation is

$$-\rho \cdot \frac{\pi}{2} \cdot d^2 \cdot \frac{dd}{dt} = 0,63 \cdot 1950 \cdot d^4$$

This equation can be solved by separation of the variables

$$\frac{dd}{d^2} = -\frac{2}{\pi \cdot \rho} \cdot 0,63 \cdot 1950 \cdot dt$$

and by integrating both sides

$$t = \left(\frac{1}{d_e} - \frac{1}{d_b} \right) \cdot \frac{\pi}{2} \cdot \rho \cdot \frac{1}{0,63 \cdot 1950}$$

where d_e is the diameter of the particle at the end of the time t and d_b is the diameter at the beginning of time t.

This formula means that a complete gasification requires infinite time in any case and in order to make the formula say anything about the difference between the gasification of big and small particles the assumption is made that a rest of 10% of the original weight consisting mainly of ash is tolerated. Then

$$d_b = d_e \cdot \sqrt[3]{10} = d_e \cdot 2,1544 \quad \frac{1}{d_e} - \frac{1}{d_b} = \frac{1,1544}{d_e}$$

and a very simple formula for t is acquired

$$t = \frac{1,1544}{d_b} \cdot \frac{\pi}{2} \cdot \rho \cdot \frac{1}{0,63 \cdot 1950}$$

representing a hyperbolic law. For a temperatur of 1200 °C:

$$t = \frac{1}{d_b} \cdot 1,85 \text{ (sec)}$$

For a diameter d=0,1 mm this would be 18,5 sec.

The result is that the time required for gasification increases rapidly with smaller diameters and that an increase of the medium temperature would cause an increase of time required. It is quite clear that this law would be applicable only below the diameters where the hyperbola intersects the line of the minimal surface temperatur (Fig.8) i.e. below 0,5 mm, but even than it would make the whole gasification of powdered coal impracticable, for the maximum of ordinary fuel dust is below 0,1 mm. But even in the realm of small diameters the theory of the falling space is not applicable because of the following reasons:

- 1.) It is true that the sinking ~~particls~~ speed decreases rapidly with the diameters of the particles, but at the same time the reacting surface per unit weight of fuel is tending to an infinite size and therefore even a small transport way of materia causes considerable results.
- 2.) If the sinking speed is very small the mechanism of diffusion will interfere and it is not necessary, that all reacting steam is touched physically by the sinking particle. The basic assumption of the contents of the "falling space" reacting only is too pessimistic therefore.
- 3.) The volume of the falling space was calculated under the condition that the particle would sink straightly down. In fact the relative way of a particle through the medium can never be a straight one, but in consequence of the turbulence and the phenomenon of "Browns motion" the real way and therefore the falling space at the same time would be multiplied especially at higher temperatures.
- 4.) In the Berlin experiments screened particles were gasified and an increase of the time required for small particles was not observed, quite the other way, small particles showed a slightly better behaviour. The consequence that an increase of temperature of the steam would cause an increase of time required was not observed too. An increase of temperature showed a clear reduction of the time required in any case.
- 5.) The results of this theory would be in contradiction with the experience of all chemists using fine grinding and high temperatures to accelerate the reactions.

III. The Berlin experiments.

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The following experiments were made in Berlin in 1940/41 with a small experimental plant diagrammatically shown in the attached figur.

Fuel: Powdered bituminous coal from middle Germany (Geiseltal near Merseburg) with about 8% ash contents was used. The water was evaporated before the experiments.

Description of the apparatus: A ~~little~~ small gas fired boiler produced superheated steam at 5 kg/cm² which was reduced to atmospheric pressure. The steam entered an electric preheater (4). The last stage of this superheater was made of platinum wire wound through the holes of a ceramic filter and allowed a steam temperatur up to 1200°C. ~~han the steam entered the reactor consist~~ing of a vertical steel tube which was heated by low voltage current. The tube material was a steel composition with Si, Cr and Al (Sicromal) which can resist high temperature and sulphur corrosions. The tube was 6 m long an when heated to 1000°C it

3" ind.

grew longer by 10 cm. At such a high temperature the steel tube would sink down under its own weight and therefore it was to be kept in tension by two counterweights.

The steam entered the reactor tube through a filter and immediately above the filter the fuel entered the tube. The feeding apparatus (5) consisted of a small coal bin, a feeding screw and a tube below the feeding screw in which an axis with spikes was rotated by a motor in order to keep the dust in suspension and to maintain a continuous flow of the coal dust this way. The further transport of the coal dust into the reactor was carried out by steam of about 150°C.

After the reactor was passed, the steam and the reaction products had two ways controlled by the tube switch (9). Firstly the measuring equipment consisting of a dust discharger with filter (10) and a condenser for the steam; secondly a by-pass with a rough discharger only.

The reactor tube was carefully insulated by fire-clay, asbestos and slag wool. The whole insulated tube was covered by a sheet iron tube of 500 mm ϕ .

Operation of the plant: On the evening the low-tension transformer (7) was switched on to heat the reactor during the night. The temperature was registered by a writing apparatus controlling the temperatures of 6 spots of the tube at the same time and writing it up in different colours. In the morning when the temperature scheduled was nearly reached the boiler was heated and the coal bin was filled. When the boiler gave steam the tube switch (9) was put to connect the discharge pipe (8) with the by-pass. (Different from the position as shown in the figure) Then the whole equipment was regulated to normal pressure and temperature conditions. Then the test was started by switching on the feeding apparatus and putting the tube switch into a position to connect the discharge pipe with the dust-discharger as shown in the figure. The time of operation was about 30 minutes. When the coal bin was cleared all electric energy was switched off and the dust discharged was removed and cooled immediately being very reactive. Gas samples were taken before the gas had passed the filter and sometimes after the gas meter as a control only.

The proper working of the plant was controlled by calculating the material balances. (Carbon in fuel = carbon in gas + carbon in dust recovered) The losses were found to be below 10%, generally about 5% and most likely due to the smallest particles which are difficult to be discharged completely.

The experiments:

1.) Powdered bituminous coal of a composition as it is used for combustion in boiler-houses was fed. (Maximum below ϕ 0,1 mm ϕ) Operation temperatures 800, 900 and 1000°C. The results were compared with the results of carbonising the same coal at the same temperatures. The quantity of the steam was always very high and about the double quantity ~~needed~~ required for complete gasification of the fuel. This was done to become free from the influence of the varying composition of the medium along the reactor.

Results: At 800°C ^{only} the volatile materia was gasified and ~~max~~ nearly the same results were obtained as when the coal was heated in ~~an~~ a china pot, at 800°C. - At 900 and 1000°C the attack of the steam on the fixed carbon could be clearly observed, but the figures according to the diffusion and heat transfer border

(Fig. 3)

was found to be too optimistic. Complete gasification could not be reached and the discharged dust ~~was~~ was found to contain about 40% of its original carbon after having been suspended for about 2 seconds in pure steam of 1000°C. The whole idea begins to become interesting at temperatures only which are considerably above 1000°C.

The dust recovered was screened and the samples of different grains were examined separately in order to find out the different behaviour of the particles of various size. No considerable difference can be found and the increase of the ash contents is nearly equally distributed over all sizes.

2.) The powdered bituminous coal was screened and the separated sizes of particles were gasified separately. A set of screens was used containing wire screens with holes of 0,03 0,06 0,075 0,09 0,12 and 0,15 mm square. The best results were obtained with the smallest particles at 1000°C ~~where~~ the dust recovered contained about 20% of the original carbon contents. The analysis of the gas samples showed no difference between the various sizes of the grain and the composition of gas was determined by the temperature of the process only. This would be to ~~prove~~ suggest the constancy of the surface temperature as it is described before. After all the "falling space" theory does not hold good because according to that theory a better gasification of the bigger particles was to be expected. (Even if there is admitted that the results cannot be expected to be as clear as the hyperbolic law of the "falling space theory" demands for all results are a little covered by the influence of the volatile ~~and~~ constituents there must have been at least a slight improvement when particles of say 0,12-0,15 were gasified instead of particles between 0 and 0,03 mm, but ~~quite~~ quite the other way: Small ~~and~~ particles showed always, a better result than the bigger ones.)

Such experiments would give an interesting sight into the mechanism of endothermal reactions, if they are operated at proper conditions i.e. mainly between 1000 and 1500°C. There is a simple way to make this but it is of theoretical interest only: As in Fig. 9 is shown natural gas or the like is burned by pure oxygen in a combustion chamber and the flame is cooled down by steam. This way a gasifying medium is generated consisting mainly of steam and ~~in~~ small quantities of CO and CO₂. Into this medium the powdered fuel is fed and the adjoining tubular reactor consists of tubes made from a fireproof material. An external heating must be applied if small diameters are used and the best way is to put the reactor into a furnace which is filled with crushed fire-clay. The external heating can be run by using the same natural gas and oxygen as it is ~~used~~ burned in the combustion chamber and the porousness of the material where the reactor is made from will cause no troubles for there is nearly the same composition of the gases inside and outside the reactor.

Fig. 1.

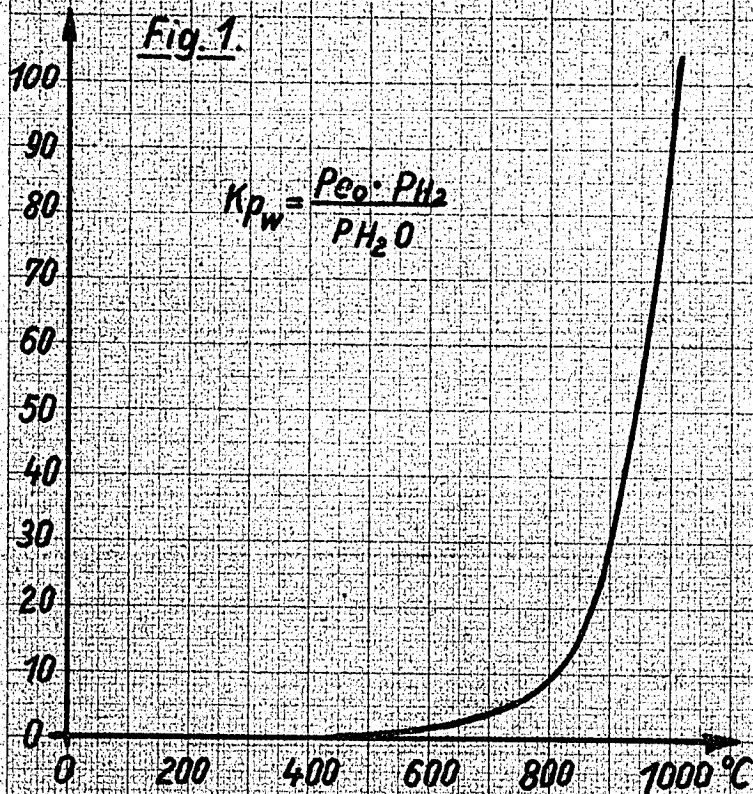
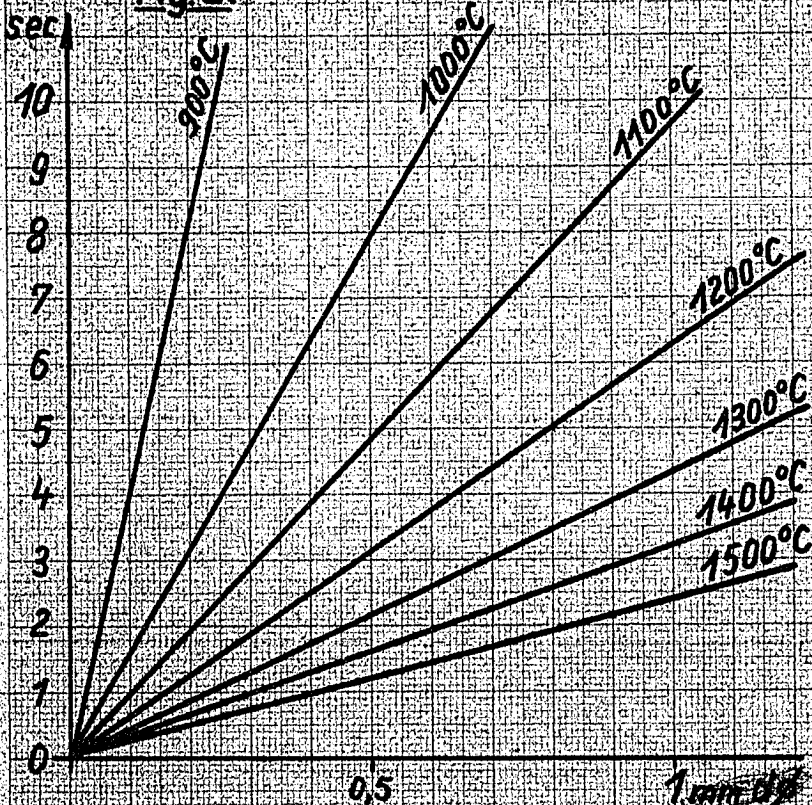


Fig 2.



sec

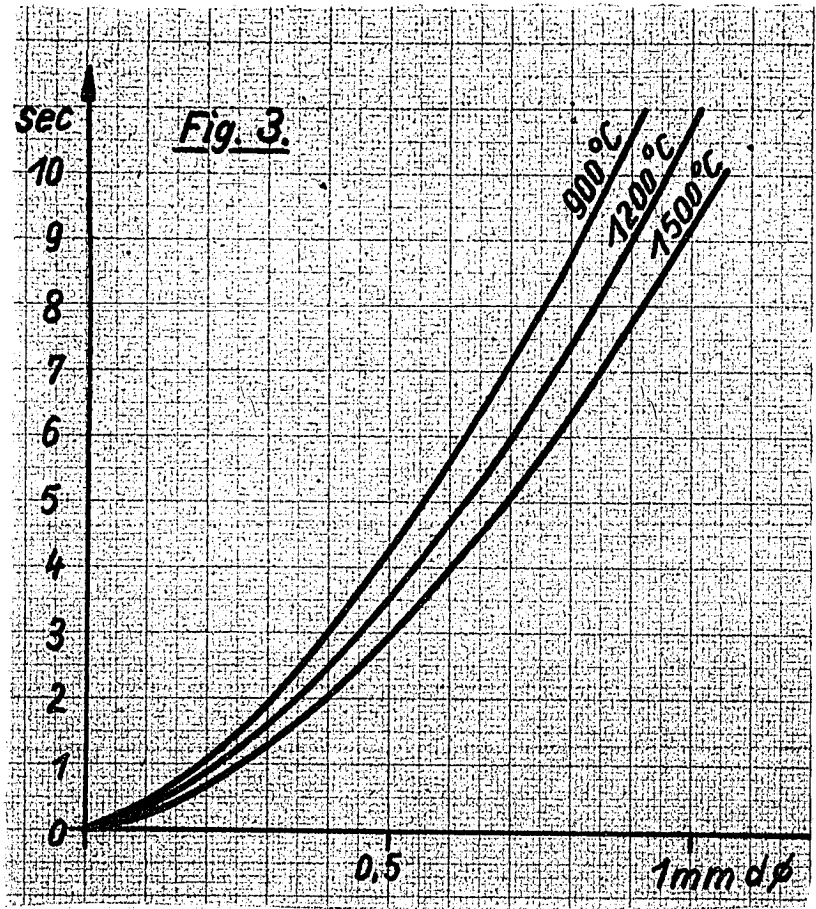
Fig. 3.

10
9
8
7
6
5
4
3
2
1
0

900°C
1200°C
1500°C

0.5

1mm dφ



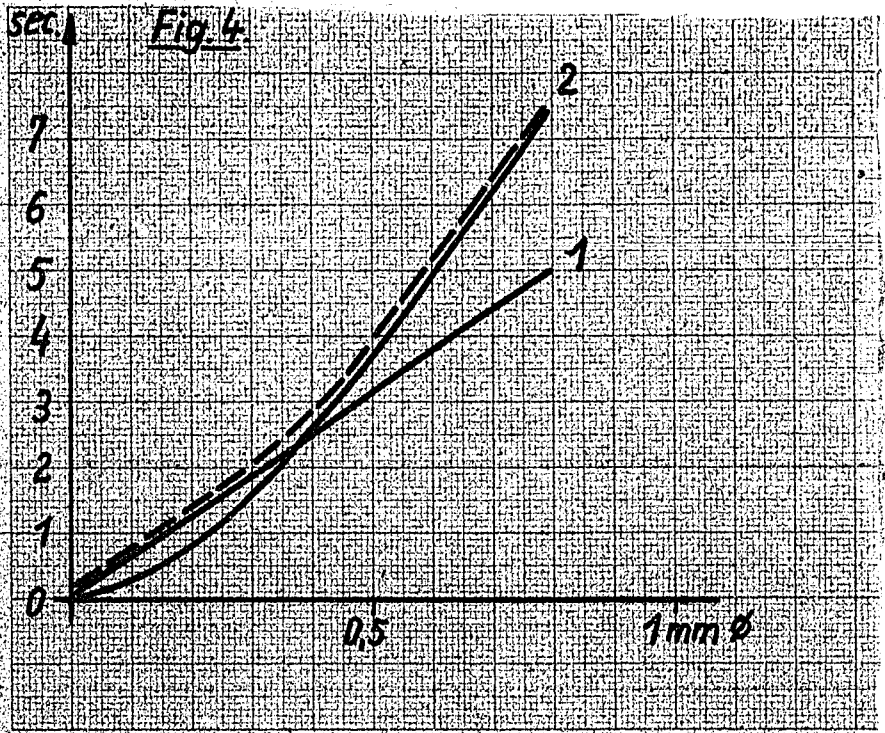


Fig. 5

$$t_H = 1200^\circ\text{C}$$

$$t_R = 600^\circ\text{C} - 800^\circ\text{C}$$

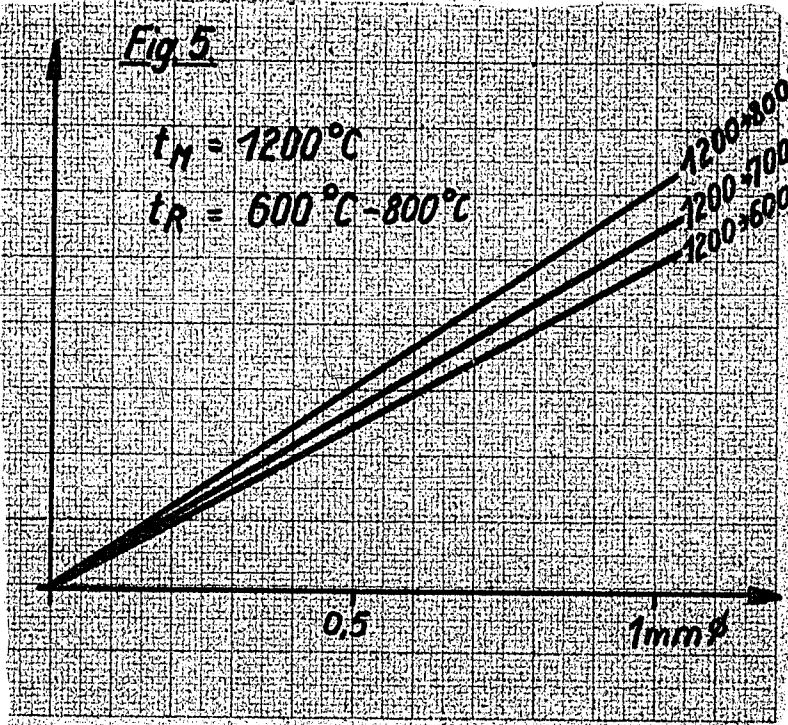
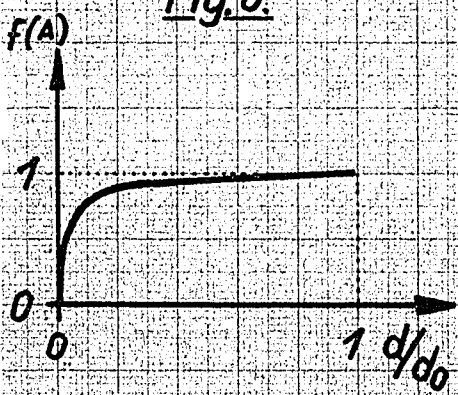
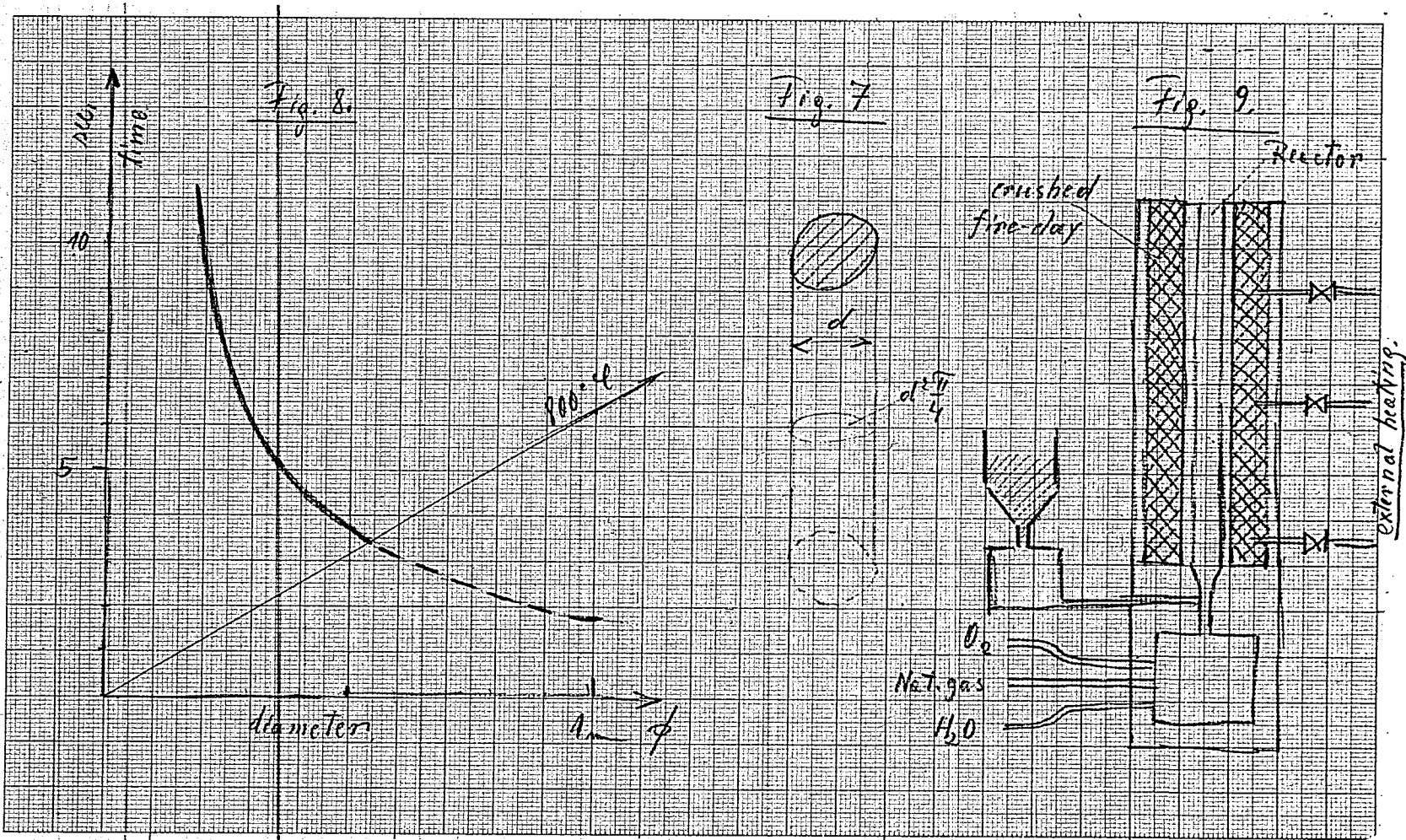
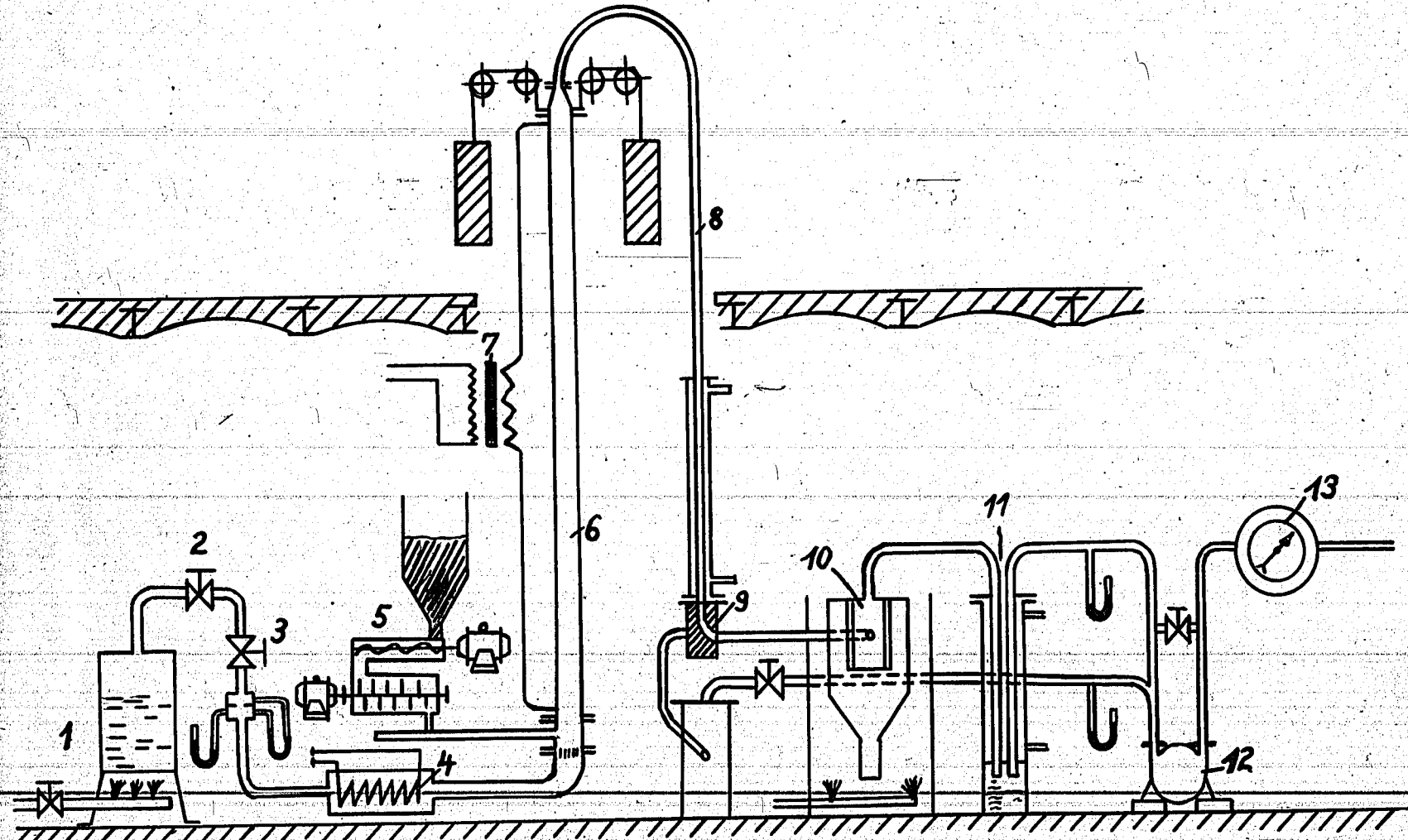


Fig. 6.





Reduce to 8 1/2"



Experimental Plant
for gasifying i. suspension (Nistler) *reproduced from photostat*