

THE PRESENT CONDITION OF THE COAL-TAR INDUSTRY.¹

As the end of the century approaches it is interesting and instructive to look back and note the wonderful growth of the chemical industries and the great changes they have undergone, especially in the last few years, in which many of them have been revolutionized.

It may be of interest to some to consider the growth and present condition of one of the greatest of these, that of coal tar and its products.

This industry which originated in the endeavor to utilize a troublesome by-product, has grown to be the one which without doubt employs more scientific men in its study and development than any other, and a description of all its products and their manufacture would be a review of the greater part of the organic chemistry of to-day.

About 1822 coal tar was first distilled in England, but it was not until 1856, the date of the discovery of the first aniline colors, that the industry became an important one; and it is in the last twenty years only that it has attained its present gigantic proportions.

At first the entire supply of tar was derived from the distillation of coal in the manufacture of illuminating gas, but under the constantly increasing demand, new sources have been developed, and these latter may in time become our main supply.

The coal tar produced in modern gas plants is quite different from that of some years ago. Cast-iron retorts were then used and the temperature of carbonization was comparatively low. The tar in consequence was generally quite fluid and contained considerable toluene, xylene, and phenols, some naphthalene and free carbon. With the introduction of fire-clay retorts, a marked change was noted and to-day the extreme high heat produced by the use of regenerators yields a tar which, considered from the standpoint of the distiller, is of much poorer quality.

The aim of the gas manufacturer is to get the largest amount of gas of good quality that can be obtained from the kind of coal carbonized. The high temperatures employed to do this cause the volatile products as they are evolved during the distillation to be decomposed in contact with the intensely heated sides of the retort with separation of free carbon, and, as exhausters are universally used, this is drawn out with the gases and deposited with the tar.

¹Chairman's address at the annual meeting of the Philadelphia Section, May 17, 1900.

The higher phenols and hydrocarbons are split up with formation of large quantities of naphthalene; therefore, the tar is heavier in gravity and thicker in consistency on account of the presence of more naphthalene and carbon and less of the liquid portions.

As the use of coal-tar products became more general, the demand for tar increased and in consequence the price advanced. The supply being to a certain extent fixed, manufacturers were stimulated to look for other sources of supply. Several plants were built in Europe and one in America having the production of tar and ammonia as their chief aim, but these were soon found to be unprofitable. Attention, however, had been called at an early date to the waste of these by-products in the manufacture of coke, and inventors without number entered the field with ovens designed to produce a good coke and at the same time save the tar and ammonia. A number of these, varying in minor details of construction, have been found to work well in practice,—besides, many beehive ovens have been provided with condensing plants for collecting the by-products. The tar obtained from these latter ovens and also from the gases of blast-furnaces is, however, quite different in composition from that obtained from gas works or improved coke ovens.

The temperature at which the coal is carbonized in the former being much lower, the tar contains practically no aromatic bodies, and when distilled yields oils which are free from naphthalene, benzene, and phenol, but instead contain compounds of the paraffin series, large amounts of higher complex phenols and paraffin; such tars are, therefore, of no value to the manufacturer of refined coal-tar products.

In the United States, owing to the extended use of oil and carburetted water-gas, the output of coal tar has been very much less in proportion to the population than in Europe, and it has been heretofore necessary to make up the deficiency by importing creosote oil and pitch. In the last few years, however, there has been a considerable addition to the domestic supply. A number of beehive ovens have been fitted with condensing plants and large batteries of improved coke ovens have been erected, and from these a fine grade of coke and considerable tar is obtained. The American coals appear to be much richer than those carbonized abroad.

In these ovens none of the charge is burnt to supply the heat necessary for the carbonization, as is the case with the older forms of ovens, and therefore, the yield of coke is very much larger, and may even amount to more than the analysis of the coal would seem to warrant, this being due to the deposition of carbon on the surface of the coke by the decomposition of a part of the tar as it is distilled.

The fact of a larger yield and purer quality of coke must, it seems, finally force other manufacturers to adopt improved ovens, and if even a part of the 20,000,000 tons of coke yearly produced in this country was made in such ovens with recovery of tar, we would pass from the condition of a shortage of supply to a veritable flood of tar, for even a small portion of which there would not appear to be a market.

One of the forms of ovens successfully introduced is the Semet-Solvay, of which about 300 are in operation in this country. Those interested in the subject will find a full description of them in an article by J. D. Pennock, which appeared in this Journal¹ in 1899.

The Otto-Hofmann ovens are used very extensively abroad, and through the kindness of Dr. Schniewind, chemist of the company, I am informed that 680 ovens are in operation in the United States, and that 400 are now being erected in Canada. The most interesting of these plants is that at Everett, near Boston, Mass., where 400 ovens are in successful operation. These have been fully described and illustrated by Dr. Schniewind in articles which have appeared in the *Progressive Age*. The plant is so situated that coal from Nova Scotia can be discharged directly from the steamers, and labor is reduced to a minimum by the use of every modern appliance for handling the materials used and produced.

The gas as it distils being higher in candle-power in the first hours of carbonization, this portion is collected by itself and after removal of tar and ammonia, is supplied to the city of Boston and vicinity. The gas coming off in the latter part of the distillation is used, after the tar has been removed, to heat the ovens. The coke is equal in quality to the best metallurgical coke and is sold for this purpose or broken into various sizes for domestic use.

It would seem that this novel and interesting application of the coke oven is destined to come into general use for large cities.

The tar from these ovens resembles the average retort tar except that it does not contain as much low-boiling hydrocarbons or phenol.

A certain amount of tar is also obtained from the manufacture of oil- and water-gas but they are of much less value than pure coal tar. The water-gas tar is especially difficult to utilize as it contains from 50 to 75 per cent. of water in the state of an emulsion which will not separate on standing.

In the United States tar is usually distilled into light oil, heavy oil, and pitch. The light oil comprises the first portions of the distillate until the oil, as it runs off, sinks in water. The

¹ This Journal, 21, 678.

entire distillate when mixed has a gravity from 0.93 to 0.98. Its nature has varied with the change in the tar as noted above, formerly they were quite fluid, but now in most cases they are semisolid from naphthalene.

The pitch remaining in the still after the distillation is known as soft pitch, and is adapted for roofing or paving purposes.

Formerly it was customary to run to hard pitch which was sent abroad to be used in the manufacture of compressed fuel; the anthracene oil obtained by this distillation was filtered and the anthracene pressed in hydraulic presses, making 40 per cent. anthracene. This was shipped to England and used in the manufacture of alizarin. At that time anthracene was one of the most valuable of the crude tar products, but the price has now fallen so low that it does not pay to export it, and there is very little sale for hard pitch as the manufacture of briquettes for fuel is carried on in a limited way only. The manufacture of anthracene has, therefore, been abandoned for some years.

The demand for creosote oil has, on the contrary, been constantly increasing owing to its extensive use in creosoting lumber, a number of plants for this purpose being in operation here.

Oil for this purpose is required to conform to certain specifications; formerly it was deemed necessary to have a large amount of tar acids, but now it is recognized that the naphthalene is a much more important agent in the preservation of lumber; and in consequence, specifications require 40 to 50 per cent. to be present, and in cold weather the oils are often entirely solid from separated naphthalene. In this connection, it is well to call attention to the fact that many specifications call for 50 per cent. of naphthalene and yet require the oil to be limpid at 90° F., which is an impossibility when such a large amount of naphthalene is present.

When we come to consider the refined products of coal-tar we can, in the short time at our disposal, mention only the most important. In general it may be said that each year greater purity is demanded by consumers, and a large number of the commercial products are practically chemically pure.

This branch of the industry has not made as great strides in the United States as it should. A severe blow was dealt when nearly all the protective duties were removed a few years ago. Formerly there was on the most of them a duty of 20 per cent. which enabled the American manufacturer to compete notwithstanding the higher wages ruling here.

The use of special labor-saving machinery which has done so much to enable our manufacturers to enter the markets of the world is not an important factor in this industry, which, owing to its nature, does not permit the extensive use of such labor-

saving devices, and where machinery can be used, we find the German manufacturers, who are our chief competitors, have the most improved devices in this line.

If we compare the estimates of cost of manufacture by American and German experts, the item of labor is seen to be a very serious one. For ordinary labor we must pay from 14 to 15 cents per hour, German estimates are based on 6 to 7 cents. In the pay of skilled mechanics the difference is even more striking: carpenters, masons, machinists, boiler and tank makers, receive from 30 to 45 cents per hour, while the prices abroad are scarcely 50 per cent. of these. This has a twofold effect on the industry: first, in increasing the item of repairs and maintenance, which, owing to the nature of the business, is a very large one, necessitating the constant renewal of parts destroyed in the processes. Second, it handicaps the manufacturer by requiring a larger outlay of capital to erect the plant than is the case abroad. A conservative estimate would be that an average plant would cost twice as much here as in Germany.

Of all the refined products, benzene has probably been subjected to the greatest fluctuations. The demand for this article increased so rapidly that the price became very high, and efforts were made in all directions to perfect processes which would produce it independently of the gas works. It was at this time that the endeavor was made both in Europe and America to distil coal in retorts at the mines with the making of benzene as their prime object; but, as already stated, these were soon abandoned. A plant was also built in Ohio to manufacture benzene and its homologues from petroleum residues but without success.

Attention had been called at an early day to the large amount of benzene present in the coal gas, amounting to about twenty times as much as is present in the tar from the same gas. It was manifestly impossible to remove it from the gas without destroying, to a large extent, its illuminating power, but this objection did not hold good with the gas obtained from the improved coke ovens, and appliances were added to these plants to remove this benzene. A large number of such plants are in operation abroad at this time; the gas from the ovens after removal of tar and ammonia is forced through specially designed towers, in which the gas is brought into intimate contact with a fraction of oil from coal tar. This oil absorbs the benzene, and when saturated is heated, and the crude benzene driven out. The product thus obtained is equal to about 90 per cent. benzene; that is to say, consists of about 70 per cent. benzene, 20 to 25 per cent. of toluene, and the balance of higher bodies. The introduction of these benzene extracting plants may be said to have been *too* successful, as the price has fallen to such an

extent that the extraction at present is hardly remunerative. We are, however, assured of a source of supply which can be readily increased as the demand warrants. The present supply is undoubtedly more than the demand; toluene, however, is not obtained in such large quantities by this method and as the demand is steadily increasing owing to many new uses, among them the manufacture of artificial indigo, the price has advanced so that it is now selling abroad at a higher price than benzene, while formerly the reverse was the case. At the present time there is no plant in operation in this country extracting the benzene from gas.

The low price of benzene permits it to be used in many industries as a solvent for the extraction of chemicals and drugs, and immense quantities are used abroad for enriching gas.

Naphthalene is one of the coal-tar products which is now manufactured on a very large scale in a great degree of purity. This article has advanced considerably in price owing both to the great demand, and to the fact that crude naphthalene is now readily sold without removal from the creosote oil for reasons stated above. Before the duties were removed, there were three factories manufacturing the article in this country; since it has been placed on the free list only one of these survives. The output is chiefly used as a substitute for camphor for destroying moths.

Crystal carboic acid or phenol is one of the few tar products which has not suffered the general decline.

In all the varied new sources of supply of other tar products, none has been found which promises increased output of phenol. The coke oven tar does not appear to yield much phenol, so that while the supply is constant or perhaps increasing slightly, it does not keep pace with the large demand, the chief of which is for the manufacture of picric and salicylic acids. A limited amount of phenol is made in this country, and considerable refined from imported materials, but the great bulk comes to us in a finished condition from Europe.

Nitrobenzene, nitrotoluene, and other nitro compounds are made in one factory, the output being mainly used by manufacturers of colors and explosives. A certain quantity of rectified nitrobenzene, known as oil of myrbane, is used by manufacturers of soaps and oils.

Aniline oil which is made in such large quantities abroad is not made at present in this country. Its manufacture was undertaken a number of years ago by makers of colors but abandoned as it was found the article could be imported at less cost. A factory, however, is now being equipped for its manufacture, and in a few weeks will be in operation.

Acetanilide is made by two or three factories and they supply nearly all the requirements of this market.

When the tariff on coal-tar products was revised and most articles placed on the free list, for some reason, salicylic and benzoic acids were singled out from all other compounds and favored with the specific duty of 10 cents per pound. In consequence, a number of works are now producing the former acid, using imported phenol.

The finer organic compounds are apparently not produced except vanillin, which is being made by at least two if not three firms.

Notwithstanding the great competition of Europe in aniline colors, the American manufacturers have in the last few years made decided strides, although as yet they appear to have confined themselves to the older standard colors. They are aided by a duty of 20 per cent. with practically all their raw materials free. They still labor, however, under the disadvantage of having to import a great deal of their supplies. There are at present five factories in operation.

It will be seen from this rapid review that there is still a wide field for the chemist and manufacturer in this country; and with a gradual increase in the output of crude materials, it is to be hoped and expected that in a few years we will produce a large amount of the enormous quantity of these products now imported.

England appears to have fallen out of the race in the competition for the American trade, especially in the finer coal-tar products, and Germany is now in the lead, even competing with England in the cruder preparations. To show the important position held by Germany, attention is called to the statistics compiled by the German government up to July 1898, on which date there were 25 factories devoted to anilin and anilin dyes, and 48 establishments which made dyestuffs, carbolic acid, and other coal-tar preparations. Among the exports for 1898 were:

- 9,321 metric tons, alizarin,
- 19,712 metric tons, coal-tar colors.
- 12,360 metric tons, aniline oil and salts.

In this latter class the exports of 1898 were nineteen times as much as in 1883.

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