

GENEVER (GIN): A SPIRIT DRINK FULL OF HISTORY, SCIENCE AND TECHNOLOGY

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Introduction

On February 20, 1979 the Court of Justice of the European Communities passed a decree that would be registered by history under the name "Cassis de Dijon". This obliged the former Federal Republic of Germany to indulge the sale in Germany of the French blackcurrant liqueur with an alcohol amount of 20% vol although German law prescribed an alcohol amount of at least 25% vol. From that point onwards all products produced by an European Member State conform its legislation, could be sold in other European Member States. In fear of a declining quality of the traditional products a law concerning the European distilled drinks was drawn up under the pressure of Scottish whisky and French brandy distillers. This led to the promulgation of the Council Regulation (EEC) n° 1576/89 of 29 May 1989, laying down general rules on the definition, description and presentation of spirit drinks.

Describing genever unambiguously appeared to be a difficult task. Genever (shortened by the English to gin) is the national distilled drink of Belgium and the Netherlands and it is also being produced in the North of France (former French Flanders). The Dutch and the Belgians both have a distinctly different view on gin. For example the alcohol amount of the Dutch and the Belgian gin is respectively 35 and 30% vol. Even within Belgium there is a considerable difference in both aroma and taste between a gin from East Flanders and one from Hasselt.

While drawing up the European regulation several questions were asked. Why is gin the national drink of Belgium, the Netherlands and French Flanders, in fact the Low Countries by the sea? Do these gins have a common origin and how was the original composition of gin?

When looking for an answer to these questions we were surprised by the amount of prominent scientists who, through times, have been involved in studying the alcoholic fermentation and the distillation of alcohol. Their research contributed to the development of general and applied chemistry, biochemistry and microbiology. Without exaggeration, alcohol distilleries can be considered the cradle of biotechnology.

The genesis of genever¹

Until recently the discovery of gin was attributed to the Dutch professor from Leyden, Franciscus de le Boë, Sylvius (1614-1672).² This was understandably but wrongly attributed. Understandably, because Sylvius, founder of the iatrochemistry, was particularly trained in the art of distillery and preparation of drugs in which he made abundant use of juniper berries. Wrongly, because the Dutch *States' ordinance on brandy* already levied taxes in 1606 (which was 8 years before Sylvius was born) on all distilled wine, anise, gin or fennel water and alike which were sold as alcoholic drinks. The mentioning of "distilled anise, gin or fennel water sold as alcoholic drinks", has a particular relevance. In the *States' ordinance on brandy* of October 1, 1583 there is still no mention of a tax on the sales of anise, gin nor fennel water. Government still looked upon these drinks as medicinal drinks. The fact that a tax on these medicinal drinks is introduced in 1606 indicates that these drinks are no longer seen as a medicine, but rather as a largely consumed stimulant.³

Among these three medicinal waters, gin was the most purchased one, mainly because of the ancient belief in the therapeutic effect of the juniper berry. This belief is shown in several medieval manuscripts praising the medicinal properties of the juniper berry hence turning it into a wonder drug.^{4,5} The oldest tracts on juniper berries date from the fourteenth century, most of the tracts date from the fifteenth century. Especially in the German area the amount was high: until now 28 different German tracts on juniper berries are known. Some were translated into Danish, Swedish, Norwegian and Icelandic. The origin of this interest was undeniably to be situated in the Netherlands. Already in the thirteenth century Jacob van Vitri († 1244), from the French Flemish town of Atrecht, wrote an entire chapter about the juniper in his *Historia Orientalis*. Jacob van Vitri was inspired by Pliny the Elder, Avicenna and the Circa Instans. The medicinal properties of juniper berries and juniper oil were also emphasized in *Liber* de natura rerum by Thomas van Bellingen alias Thomas van Cantimpré (1201-ca.1270). Thomas, born in Sint-Pieters-Leeuw near Brussels, became a regular canon in the order of Saint Augustin in the abbey of Cantimpré near the French Flemish Kamerrijk, hence his name. Later on he became a Dominican in Louvain and during several years he was also a pupil of the famous Albertus Magnus (1206-1280) in Cologne. His *Liber de natura rerum* is a compilation of works of ancient masters like Aristotle, Hippocrates, Galen, Avicenna and works like the Circa Instans.

The oldest reference known in Dutch literature to the use of juniper berries in drinks can be found in Der Naturen Bloeme by Jacob van Maerlant (1235-1300).^{6,7} Jacob wrote this rhyming nature encyclopaedia between 1266 and 1269 in Damme, an outport of the city of Bruges. It is not an original work, but a compilation of earlier books, which he thoroughly compared with each other. His greatest source of inspiration was Liber de natura rerum, which had been written a few years earlier by Thomas van Cantimpré. Der Naturen Bloeme contains thirteen chapters which the author calls books. In book VIII on trees originating from seeds van Maerlant elaborately describes the properties of the juniper bush (fig. 1). His readers must have known this juniper bush (Juniperus) well because he regularly refers to it in order to describe certain properties of spices. He says for instance, that the cinnamon tree has the same purple leaves as the juniper bush, that the fruit of the sweet chestnut is of nearly the same size as the juniper berry, that the clove tree is as big as the juniper tree and that the wood and the leaves of the pepper tree resemble those of the juniper tree. One gets the same impression when reading the fifteenth century travel story of the pilgrim traveller Knight Joos van Ghistele from Ghent.⁸ In his story he always refers to, as a comparison, the juniper bush when describing the innumerable spices and trees he had seen in the Near East, as if it was the standard measure (and he did not know any other plant in his country).

The medicinal properties of the juniper berry are reflected in the following verses of Jacob van Maerlant:

Jeghen buuc evel van leden So salmen jenewere sieden

unipers dies neem ic goen Dint mi velen die ienen bo En die es dus glenatuert Dat con cel mers viaco ducot Datme met linen alden uet A 110 als playmet Willic ienew unlieven h eer en drogte ua manieven Aer tom es ua vechter nature O utbind en útern humuere ogten byt cuel ua leven o Calme ienettere lieven In reinwate en tat ontfaen Diemer lanceuel es buaen s text renetire in winc Ets goet iegten line pine H an Wen hutemaet mi mete D lie us good mæglenthed Teerlte mor gledroghet welen n de lone thout uan delen Dat letme enen pot glehed

Fig. 1. Properties of the juniper busch (Juniperus), 13th century. In: Jacob van Maerlant, Der Naturen Bloeme, Library of the University of Leiden, Ms. BPL 14A., fol. 115^v

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In reynwatre ende dat ontfaen. Die met lancevel es bevaen Siede jenewer in wine, Ets goet jegen sine pine.

In these verses rainwater used to boil juniper berries is recommended to cure abdominal pains. Wine in which juniper berries had been boiled was considered a good remedy for abdominal cramps. This last drink can be seen as a digestive and as the immediate predecessor of the present-day gin. Medicinal wines were also propagated by Jacob's contemporary Arnaldus de Villanova (ca.1255-1311). In his *Liber de vinis*, a book translated several times into Middle Dutch, he describes 28 different medicinal wines.⁹ A wine recipe using boiled juniper berries, cannot be found in his work.

According to the fifteenth century Middle Dutch manuscript 697 that is being kept at the academic library of the University of Ghent, juniper berries were also being used during the brewing process of beer.¹⁰ The juniper berries were sun-dried and burnt to powder. That juniper powder was used in order to make bad beer better and to make it resemble Harlem beer. Juniper wood and juniper berries are, still today, being used as a preserver in certain Norwegian beers.

Besides juniper berries also the oil extracted through dry, downward distillation of juniper wood, was being used. This oil was considered to be a work of art and it was used as seasoning or as an ointment. Jacob van Maerlant called this oil a *medicine rike* (a rich medicine) and advised its use to combat fever, epilepsy, arthritis and abdominal cramps.⁶ Jacob believed the juniper oil to be good for digestion and effective in suppressing melancholy. The preparation and the use of this oil is also described in *Herbarijs* and in *Den Herbarius in Dyetsche*, the first Dutch book printed on medicinal herbs which has been published in 1484 in Louvain by Jan Veldener.^{11,12} That book advises bathing in rainwater in which juniper berries are boiled to treat skin disease and stomachache. The same advice is given in the fourteenth century Middle Dutch translation of the Circa Instans.¹³

In several fifteenth century tracts on the plague the smoke of burning juniper berries en burning juniper wood is being used to clear the "evil air" of the rooms in which plague victims had been staying.^{14,15} In his *Cruydeboeck* (Antwerp, 1554) Rembert Dodoens will still advise the use of smoke of burning juniper wood and burning juniper berries to fight the plague and vermin.

The juniper berry tract Sequentur propietates et virtutes granorum juniperi et olei granorum is innovative.⁴ The tract, a part of manuscript n° 618 of the Wellcome Historical Medical Library in London, was written in 1496. It is partly written in Latin and partly written in Middle Dutch. Some recipes in the manuscript come from the practice of Jan Spierinck, the personal physician of the Duke of Brabant. Jan Spierinck was also attached to the University of Louvain of which he was the dean in 1457, 1462 and 1479. Jan Spierinck was a strong advocate of the use of indigenous drugs which he preferred to exotic ones. The juniper berry tract calls upon doctor Hubertus, hence it was called the juniper berry tract of doctor Hubertus. Supposedly, doctor Hubertus was an ambulant physician who, like he himself wrote, resided in an inn in 's-Hertogenbosch. The tract excels in simplicity and its sense for details. Hubertus attaches great importance to the type of juniper berry: the berries from the Orient are better than those from the Occident. Furthermore the berries need to be ripe and black. Therefore they should be plucked between two name days of the Holy Mother: August 15 and September 8, during the constellation of Virgo. Hubertus abandons the traditional way of administering the juniper berry. Instead of boiling it in wine, he leaves the berries to macerate during one night in aqua vitae or in clareyt, a strong wine mixed with honey and herbs. Afterwards the juniper berries are sun-dried. The aqua vitae (aqua ardens) and the sun enable the juniper berry, thanks to its hot and dry nature, to reach the fourth or highest degree. Such berries were very good to dissolve and digest cold and moist humours. Eating five to six of such berries in the morning before breakfast and at night before going to bed, could prevent and/or fight the most important diseases (twenty are being named). In other words, it was a true panacea. The same ailments can also be cured using oil distilled from juniper berries: this also is an innovation of doctor Hubertus. The oil is acquired by squashing the juniper berries, sprinkling them with

aqua vitae and then distilling this. In 1552 Philippus Hermanni describes a recipe of juniper berry water in *Een Constelijck Distileerboec* that strongly resembles a recipe of doctor Hubertus.¹⁶ According to Hermanni the juniper berry water can be consumed for digestive disorders, colds, the plague and bites of venomous animals. It can also be used to wash out bites.

The late fifteenth century manuscript Sloane 345, fols. 51^r-51^v that is being kept at the British Museum in London is also of considerable importance.⁴ It contains two brandy recipes which are not mentioned with the medicinal waters, but which are included with the kitchen recipes. The title Gebrande wyn te maken (making brandy wine) is accompanied by the note de aqua vina in the margin and next to the title of the second recipe Een ander manyr om brande wyn tmaken (an other way to make brandy wine) it reads de aqua vitae. The fact that the brandy recipes could be found among the kitchen recipes and also the fact that the terminology aqua vitae is replaced by brandy, inclines us to believe that by the end of the fifteenth century the drug aqua vitae has become a stimulant called brandy. The levy of the tax on brandy first issued in the Netherlands in Amsterdam in 1497 confirms that suspicion.¹⁸ In the recipe Gebrande wyn te maken, brandy is distilled out of a mixture of wine and beer. Nevertheless the author adds that in order to be medicinal, the brandy can only and solely be distilled from wine. The medicinal properties of a brandy distilled from wine can be enhanced by adding several ingredients. These herbs are properly proportioned wrapped in a little piece of cloth, immersed in the brandy and jointly distilled. As ingredients are mentioned sage leaves, nutmeg, clove and gorsbeyn of dameren. Several authors think these terms refer to ashes (dameren) of frog bones (gorsbeyn).¹⁷ Gors is probably a corruption of the spelling of the words ghurst, gurst or goist which in several Middle Dutch texts are synonymous with juniper and beyn can be read as beyen or berries. Gorsbevn of dameren would in fact be juniper powder which, in that time, was often used to stabilise beer and it was also used in medicinal recipes. Flavouring brandy with herbs, berries and seeds was already introduced at the end of the fifteenth century.

In the sixteenth century the consumption of brandy rises rapidly. Many cities introduce taxes on brandy, which is increasingly being distilled from beer and mead. This resulted from the disappearance of the vineyards in our regions following the bad harvest between 1511 and 1524 and the 100

so-called cold wave that could be felt around 1540 and extensively from 1590 onwards.¹⁹ Hence wine became more scarce and expensive. However, the upper class continued to drink brandy from wine while the commoner resorted to drinking cheaper brandy distilled from beer and mead. Already in 1552 a physician from Antwerp. Philippus Hermanii protested against the growing consumption of brandy distilled from beer in Een Consteliick Distileerboec and in 1588 the pastor Casper Jansz. Coolhaes (1536-1615) did so in Van seeckere seer costelijcke wateren.^{16,20} According to these authors brandy from grains has less flavour, wine destillate is the only healthy one, one can only speak of brandy if it is distilled from wine, and grains can only be used for the purpose of making bread. Notwithstanding these warnings the production and consumption of brandy made from beer and mead rise enormously in the second half of the sixteenth century. The distillers no longer process the beer, but they immediately use the grain, mainly rye and wheat. These grains are left to germinate first, allowing the starch to transform easily into fermentable sugar during mashing. After the sugar's fermentation into alcohol distillation happens twice to three times hence obtaining corn brandy or malt spirit.

In 1588 the Northern Low Countries (Republic of the Seven United Netherlands) get severed from the Southern Low Countries. In the Southern Low Countries the archdukes Albert and Isabella issue an edict in 1601, because of the excessive brandy consumption, prohibiting the production, sales and consumption of brandy from grains, apples and putrid pears (fig. 2). This prohibition will remain operative during the entire 17th century. In the Northern Low Countries distillation can freely continue; this results in a sensational development of the malt spirit production in the ports (especially Schiedam) with sufficient grain supply. The upper class appreciates less the malt spirit's organoleptic characteristics than brandy distilled from wine. This urges the producers to aromatise the malt spirit, especially with the juniper berry flavour. This is also the origin of a speciality, viz. distillers produce malt spirit while liquor distillers are involved in aromatizing it. Hasselt, which is not a part of the Southern Low Countries but which belongs to the Principality of Liège, must not abide by Albert and Isabella's edict. Therefore it continues to distil brandy and "wachtelwater", its own kind of gin.²¹

ORDONNANTIE ende PLACEAETZ VANDE EERTZHERTOGHEN TEGEN d'abuyfen dier gefchieden in fommige plaetfen, ende Landen van hervvertfouer, deur dé ommatigen dräck ende flete vande Ghebrande-vvynen, anderfins geheeté Lauende-vvateren, ende ander gelycke diftillation.

Fig. 2.

Siené

sentes.

Edict of the archdukes Albert and Isabella prohibiting the production and sales of brandy from grains (March 20, 1601). Library of the University of Gent.

With the introduction of the government of the Austrian Habsburger family the Southern Low Countries are allowed to recommence their distilling activities.²² Moreover, distilling is even stimulated to re-energise the dying agricultural sector. When producing alcohol out of grains a valuable additional product is obtained, namely draff. This non-volatile residue remains in the still after distillation and consists of chaff, remainders of flower and yeast. This draff, rich in proteins and cellulose, is valuable food for cattle and pigs. The manure obtained was used to fertilise the fields so increasing the corn yield. This early ecological cycle was implemented in many farms (especially in East Flanders). Distilling activities were allowed under Austrian Habsburger rule if one disposed of a patent paying a yearly contribution regardless of the production volume. This led to expeditious,

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multiple and neglectful distilling: the tax had already been paid anyway! The farmers used thick mashes which were very sensitive to burning and there was no sufficient separation of the foreshots and the feints from the middle run. Furthermore the farms had a larger interest in the draff than the alcohol which seldom was flavoured with juniper berry although this brandy was already called gin. In 1752 the government established its own public distillery in Waasten (Warneton), a small village near the linguistic border between Hainaut and West Flanders. Dutch distillers who produce gin the Dutch way (viz. aromatised with juniper berries) were being lured and they were asked to pass on their expertise to the agricultural distillers. It did not go beyond the well-meant attempts. Up until this day some distillers in East and West Flanders are still producing gin which does not contain any juniper berries.²³

(Grain) distilleries: the cradle of biotechnology^{1,24, 25}

The oldest reference to distilling known in Dutch literature can be found in the manuscript *Der Naturen Bloeme* written in Damme between 1266 and 1269 by Jacob van Maerlant.⁶ In his voluminous oeuvre he only wrote once about distillation and miraculously enough – or is it not a coincidence – it concerned the dry distillation of juniper wood out of which juniper oil is extracted.

Jacob van Maerlant described the distillation process in the following verses:

Van desen houte maectmen mede Olie van groter moeghenthede Teersten moet ghedroghet wesen In die sonne thout van desen Dan setmen enen pot gheheel In daerde diep ghenoech een deel Ende enen andren op sinen mond Ende die ydel oec ter stont Die hevet in den bodem een gat Vol van den houte lechmen dat Ende stoppet boven so tien tiden datter ghene lucht uut mach liden Dan maect man an den pot een vier Also staerc ende also fier Dat dat hout binnen versmacht Also coemt met groter cracht Uut dien houte een lettelkijn Olien diere en fijn In den nedersten pot tier stede

These verses describe a 'pot-on-pot' process that was already described by Dioscorides in the first century and was later called *distillatio per descensum* or downward distillation. About the *distillatio ascensum* or the upward distillation Jacob van Maerlant does not say a word. Nevertheless he must have known this classical way of distilling because he describes several applications of *rosen watre*, a distillate of freshly plucked rose petals. Van Maerlant for instance writes about nard boiled with sugar in rose-water and says it is good against *hertvanc en syncopsis* (anxiety and fainting) while mint with rose-water prevents *spuwen en syncopsis* (vomiting and fainting). These little remedies make us spontaneously think of the drop of *eau de Carmes* on a lump of sugar, which is still being used against oppression.

The West copied the use of rose-water from the Arabs to whom the invention of the distillery art is attributed. The cradle of this art stood in Alexandria where Greek and Egyptian scholars established the Alexandrian higher institution. Among their assistants were Maria the Jewess (2nd Century), Zosimos (ca.350-ca.420) and Synesios (ca.365-ca.415). The invention of the distilling instrument and the water-bath (bain-marie) was attributed to Maria the Jewess. She probably knew the work of Aristotle (384 BC-322 BC) and Dioscorides (1st Century AC) who described for the first time a primitive form of distilling. For example: in his Meteorologica Aristotle tells about the sailors who boiled see water, caught the vapour in a sponge and obtained potable water after squeezing it. (In his book Dageraad ofte Nieuwe Opkomst der Geneeskonst Van Helmont (1578-1644) talks about sponge water meaning in fact distilled water)²⁶. Dioscorides, the personal physician of Emperor Nero, describes in his Materia medica how the solid, vermilion cinnabar (mercury sulphide) is transformed into a silver-white liquid (mercury). In order to obtain this effect he heats the cinnabar in a ceramic jar to which a lid with an internal channel is attached. The mercury vapours condense on the lid and flow into the channel. After having cooled off the mercury is removed out of the channel and it is used as a medicine.

The distillation instrument of Maria the Jewess was made out of copper, glazed ceramics or glass and consisted of three elements: the cucurbit (boiling kettle), the alembic (still-head) and a phial (receiving flask). The alembic, equipped with an internal gutter or rim with an outlet-tube (spout), was attached to the boiling kettle. The distillate ran through the spout into the phial. This phial was an elegantly shaped bottle with a long neck and a small belly. From the 10th century and later the "alembic" does not refer any longer to the still-head, but to the entire distillation instrument.

The alembic of Synesios is to this day the oldest known depiction of an alembic (fig. 3).

It is described and pictured in a 15th century manuscript (2327 fol. 33°) that is being kept at the *Bibliothèque Nationale* in Paris.²⁷ It was certainly possible to obtain rose-water using that distillation instrument. The main substance of rose-water, the geraniol, boils at 230 °C and is sufficiently cooled in this air-cooled alembic, condensed and then collected. For ethanol production (boiling point 78 °C) this alembic was not adequate since the ethanol vapours were insufficiently cooled and hence left the alembic largely as vapour. This deficient cooling process was the reason why the Arabs were unable to prove the existence of alcohol. Thus the Arabs did not use the word *al kuhl* to refer to ethanol, but they used it to refer to the fine, black antimony sulphide powder that the Eastern women used to colour their eye lids. It is Paracelsus (1493-1541) who was the first to use the word alcohol to describe wine spirit or ethanol.

The knowledge of the Alexandrian alchemists was refined and completed by Arab savants like Djâbir (ca.722-ca.803), Rhazes (865-932), Abû Al-Qâsim or Albucasis (936-1013) and Avicenna (980-1037). Djâbir (Ge-

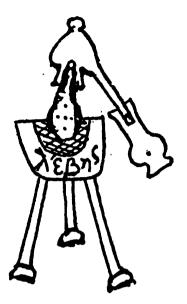


Fig. 3. Alembic of Synesios, 4th century. Bibliothèque Nationale de Paris, Ms. 2327, fol. 33^v.

ber in Latin) used the alembic to study *ghosts*, which are volatile mineral substances like mercury, sulphur and ammonium chloride and mineral acids like hydrochloric acid, sulphuric acid and nitric acid. Rhazes, Albucasis and Avicenna however, were more interested in distilling medicinal waters.

Through crusades and Moorish settlements in Sicily and Spain the Western world came into contact with the Arab culture and science. In the twelfth and thirteenth centuries the translation schools of Toledo and Salerno translated many alchemist writings from Arabic to Latin. Doing this they left many Arabic words un-translated, words that later on were also assimilated in Dutch. Some examples: alembic (al anibcq), derived from the Greek word ambix (vase) and the Arabic article al; alcohol (al kuhl); elixir (al iksir); alcali (al qali); alchemist (al khemia).

Presumably there was already knowledge of some form of wine distillate, called *aqua*, in the 8th century. The manuscript *Mappae clavicula* would indicate such notion, although in the form of a cryptogram.

From the 12th century onwards the distillation of wine is gaining interest. A small tract on *De aqua ardente* is known of Magister Salernus who pertained to the famous medical school of Salerno. It was probably written between 1140 and 1160. Salernus says that an inflamable liquid can be obtained out of wine, in the same way rose-water is made. Before distilling he adds salt and tartar to the wine. We know now that these salts do not only bind water but that they also help to expel alcohol which increases the alcohol concentration of the distillate. To raise the flammability of the distillate sulphur was added, a recommendation from the *Liber ignium ad carburendos hostes* that was presumably written by Marcus Graecus in the 11th century.

One century later, around 1280, Thaddeo Alderotti (1223-1303) wrote one of the most influential tracts on wine distillery: *De virtutibus aquae vitae et eius operationibus*. This Florentine physician (hence his name Thaddeus Florentinus) was a professor at the university of Bologna. He was the first to equip the copper still-head with a spiral spout which was continuously cooled with running water. This cooling made it possible to strongly increase the alcohol concentration of the distillate. He did not only improve the distillation equipment but also the distillation methods: when half of the wine had already been distilled, the procedure was stopped. Then the distillate was distilled for the second time processing 7/10 of the liquid. Thaddeus Florentinus believed that the distillate was not only very inflammable, but that it also had several powers: it was used as a medicine, as *aqua vitae*, and in the *alkimia* (chemistry) it was used as a solvent, a means of fixing and extracting.

The knowledge of Thaddeus Florentinus was disseminated all over Europe via universities and monastic orders. Arnaldus de Villanova (ca.1255-1311) and Johannes de Rupescissa (ca.1300-1365) played a prominent role in this dispersion. Both of them belonged to the fraticelli, a franciscan sect which advocated complete poverty and used the alchemy to search for the mystic gold and the production of medicines. The Spaniard Arnaldus de Villanova wrote the *Liber de vinis* of which several versions in Middle Dutch exist, the oldest dating from the 14th century.⁹ The book is not only about medicinal wines, but also about *water of life* in which he leaves multiple spices to macerate. However de Villanova does not describe the distillation process. The Frenchman Johannes de Rupescissa does. In his *De consideratione quintae essentiae omnium rerum* he writes about the preparation of a non-perishable *quinta essentia*, the Elixir of Life that can assure eternal life. It is multiple distilled *aqua ardens* that without interruption is being heated, vaporised in the circulatorium and then ascends, cools and condenses. Its force can even be increased by dipping two golden coins in the *aqua ardens* while being heated. Johannes de Rupescissa, whose work was also translated into Middle Dutch, does not only relate about wine distillation but also about distillation of minerals, metals and mineral acids.²⁸ In this case it is not about the transmutation of base metals into gold with the help of the Philosopher's Stone, but it is about the application of metals and mineral acids in medicine.

It is far from a coincidence that many Latin written chemical tracts were translated into Middle Dutch. In the Low Countries existed a strong alchemy tradition. Jacob van Maerlant knew alchemy but he was no fan. In his book XII of his *Der Naturen Bloeme* he resists the transmutation: he states that it is only to the Lord to give eternal life.⁶ He is also suspicious of the many alchemic writings without an author. It is possible he feared Church. For the Church argued that eternal life could only be provided for by the sacraments, not by the Elixir of Life. The Church mainly aimed at the franciscan monks who counted several alchemists among their members. In 1323 the Church forbade them to practise the art of alchemy on penalty of excommunication.²⁹

The mingling of alchemy and religion is very obvious in Constantinus' *Bouc der heimelicheden van mire vrouwen alkemen* and in Gratheus' title-less manuscript.²⁹ Both tracts have been written in Middle Dutch and are being kept in the *Österreichische Nationalbibliothek* in Vienna. Although in the tracts the date 1224 appears, a graphological study indicates that it was probably written in the 14th century. They are of the utmost importance because up to this day they are the oldest Western illustrated alchemy tracts known. Constantinus' tract deals with the transformation of mercury. To the man living in the Middle Ages the transformation of solid, red mercury sulphide into fluid, silver-white mercury was a truly miraculous event. A solid *stone* was transformed into a volatile *spirit* after being heated in a distillation flask and then after letting it cool off it turned into a *liquid*! The resurrection of Christ and the transformation of bread and wine into the body and blood of Christ could be explained in an analogue way. Gratheus' tract contains peculiar representations with among other things Christ's face surrounded by a circle of different chemical vases (fig. 4). So far, they are the oldest, although very primitive representations of distilling flasks that can be found in alchemic literature.

Besides the speculative alchemists in search of gold, there were also very practical alchemists in the Low Countries who used the latest insights in chemistry and chemical techniques in the most diverging production procedures following the traditional methods. A lot of attention was spent on gaining and purifying base materials like salammoniac, alum, ammoniac-borax, sodium carbonate, salt and potash. These base materials soon became commercial goods and they were transported from one country to another. At the beginning of the fourteenth century the production of three 'more recent' chemical products was started: saltpetre, alcohol and mineral acids.³⁰ The production of alcohol and mineral acids had only become possible thanks to the improved distillation technique. The three new products themselves created new technologies and trades, for instance saltpetre and alcohol were applied during the production of gunpowder. The mineral acids like nitric acid and sulphuric acid made the division of silver and gold possible, which gave origin to the fine-metallurgy. Alcohol was not only used as a means of extracting or as a solvent of medicinal substances, but also as a universal solvent for things like oils, lacquers and some paints, smell and taste additives.

The production and the use of explosive, burning or inflammable chemicals led to the promulgation of a series of new rules.³¹ For instance regulation n° 36 of the *Keure van Deelmans* (Bruges, 1305) prohibited to burn *wijnas* (grape-vine charcoal) within the city to prevent fire risk. The greyish white grape-vine charcoal obtained through evaporating and calcining grape marc in open stone jars, contains a high concentration (40-60%) of potash, besides the carbon. This preparation method resulted in a new Dutch name for *wijnas*: "potas" of which the Latin derivative potassium. This grape-vine charcoal was probably used to produce gunpowder, a mixture of saltpetre, sulphur and carbon. In the accounts of the city of



Fig. 4. Christ's face surrounded by a circle of different chemical vases, 14th century. Tract of Gratheus, Österreichische Nationalbibliothek, COD. 2372, fol. 61^r.

Bruges of 1345 (fol. 141, n° 10 and 11) we can read that the city purchased saltpetre, sulphur, powder of black amber and turpentine but also an alembic from master Niclaeis Bollaerd. Presumably the alembic was used to distil turpentine, hence obtaining camphor. The powder from the black amber was used to increase the inflammation ability of the gunpowder while the camphor held all powder together and reduced the gunpowder-smoke. These additives were mixed with the gunpowder in an alcoholic solution. In the city accounts of Bruges of 1476 (fol. 12[°]) an indication can be found that wine was being distilled to make gunpowder: *levende watre omme poedre te makene* (water of life to make gunpowder). Who was responsible for distilling the water of life is not known. What we do know is that distilling was already a profession there in 1447 because a certain *Baptiste de Gambaro, stokere* (distiller) was summoned in the courts of Bruges.

The water of life also called *aqua ardens or aqua ignea*, could be poured from the town walls onto the enemies, hence burning them. The preparation of *aqua ignea* is already described in a 14th century chemical tract, *Boec van .XIJ. goeden wateren* that is being kept in the *Koninklijke Bibliotheek* in Brussels (manuscript 4260-4263, fol. 32^r). Aqua ignea is obtained through distilling in an alembic of white wine to which almizadir (ammonium chloride), sulfuris (sulphur), tartari (wine stone) and salt were added.³²

Also dating from the 14th century is the first Middle Dutch tract Aqua vite, dats water des levens of levende water (manuscript 15624-15641, fols. 6^v-8^r, Koninkliike Bibliotheek in Brussels).³³ Until now it is the oldest Dutch publication known about alcohol (fig. 5). The author of this alcohol tract is unknown, but it is a fact that in 1351, two years after the pest epidemic broke out in the Low Countries, Johannes de Aeltre (Jan van Aalter) copied the document. A dialectic study reveals that the author must be of West Flemish origin. The production of aqua vitae goes as follows: a jar of 9 stopen (1 stoop in Bruges amounts to about 2.23 litres) is filled with wine and equipped with an alembic. The jar and the alembic are glued together with flour and egg white. The wine is boiled and the inflammable part of the distillate is caught in a recipient made of glass. The inflammation ability is regularly checked by immersing a piece of cloth in the distillate from time to time: as long as the cloth remains burning, the distillate is collected. After the first distillation the jar is cleaned and refilled with the last distillate. The distillation process is repeated up to 4 and 5 times. Aqua vitae could be used to treat the most divergent diseases. The medicinal characteristics of it could be reinforced by adding different spices, berries and seeds. Aqua vitae was also used to concentrate wine and preserve foods from putrefaction. Furthermore Van Aalter also writes about the aqua vitae characteristic to float on oil. To the man of the Middle Ages who had always seen that oil was able to float on water, the phenomenon of water of life floating on oil was equally miraculous. The most important characteristics are mentioned here below:

Het doet oec den mensche droefheit vergeten Ende maecten van hertten vro ende oec stout ende coene. (It makes people forget about sadness and it makes the hearts happy and brave.)

These euphoric properties turn the medicine *aqua vitae* into the stimulant *brandy* over a period of one century.

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Aqua vite, dats water des levens..., 1351. Koninklijke Bibliotheek Albert I, Ms. 15624-15641, fol. 6^v.

In the second half of the 14th century many Middle Dutch *aqua* vitae tracts resembling the one of Jan van Aalter, are issued.^{17,28} The late 15th century recipe *Gebrande wyn te maken* (making brandy wine) (manuscript Sloane 345, fols. 51^r-51^v, British Museum in London) is really innovative.¹⁷ The basic ingredient is wine, diluted wit Hamburg beer, at the time considered to be the best beer available. The dilution can not happen at random, but most be dosed to a thickness equal to that of buttermilk. We

know now that this dilution is important to be able to remove the alcohol from the liquid in an effective way. To prevent burning the mixture was stirred until just before the boiling point; then the still-head is attached to the jar with a mixture of egg white and chalk or buckwheat-floor. Afterwards wet towels are put on the still-head and the cooling-coil is immersed in a tank of water. This improved the rectification and the alcoholic vapours are better cooled and condensed, hence increasing the alcohol concentration. Checking the alcohol amount still happens as it did in the times of Jan van Aalter: when a piece of cloth immersed in the distillate catches fire, it is considered to be alright.

It is not strange that many artists were interested in alchemy. For alchemists disposed of the technical procedures that were useful to goldsmiths, potters, glassblowers and painters. For instance, we know that the painters Jan van Eyck (ca.1390-1441) and Leonardo da Vinci (1452-1519) were experienced and skilled alchemists. Famous writers like Giorgio Vasari and Carel van Mander think that the revolutionary oil painting procedure invented by Jan van Eyck, existed thanks to his knowledge of alchemy.^{34,35}

There is even a sketch known of a distillation instrument by Leonardo da Vinci and also in his literary work we can read: since varnish is made from the juniper wood's resin, the varnish can be dissolved in the distilled product after distillation of the resin.²⁹

The distillation of turpentine from the larch or other conifers' resin is described in the 15th century Middle Dutch manuscript Sloane 345, fols. 40^{v} - 43^{r} .¹⁷It also mentions how to prepare *quicsilver* from cinnabar, acetic acid from wine vinegar and *aqua fortis* from alum, saltpetre and green vitriol. For the condensation of the mercury fumes ice cubes were already being used. These distillates were being used by the goldsmiths: mercury was used to dissolve gold and with *aqua fortis* (strong water or nitric acid) it was possible to separate gold and silver. *Aqua fortis* was used during etching. For this purpose the artist covered the copper plate with an acidrepressing layer of wax, resin or tar. The drawing was carved into the etching layer and then the copper plate was dipped into a bath of *aqua fortis*, which eroded the etching lines.

Painting recipes can be found in the late 15th century manuscript 517 of the Wellcome Historical Medical Library in London. For instance, fol. 64^v

reveals the recipe of a "water" with which you can dye a cloth in greenish blue or yellow. To obtain this effect a crushed mixture of saltpetre, blue vitriol, alum and Spanish green were distilled. The first water distilled coloured the cloth greenish blue, the next yellow. This recipe illustrates how far the art of distillation had evolved at the end of the 15th century.³⁶

Around the same period during which the recipe Gebrande wyn te maken was written, the first printed booklet on medicinal waters is published in Augsburg in 1476: "Hienach volget eyn nüczliche materi von manigerley aussgeprannten wassern wie man die nüczen und prauchen sol zu gesuntheyt der menschen". The booklet (15 pages folio) was written by the physician Michael Puff von Schrick (ca.1400-1473), professor at the university of Vienna. The booklet deals with the preparation and the medicinal characteristics of medicinal waters. Furthermore it contains a chapter on geprannten Wein (distilled wine) highlighting its medicinal properties. In fact, the booklet does not disclose any new knowledge and the author was mainly inspired by Alderotti's work and Middle Dutch texts.

From now on books on distilled waters are being continuously published. For instance, in 1500 the Liber de arte distillandi de simplicibus oder das Buch der rechten Kunst zu Distillieren die eintzigen Dinge, a book by the physician and pharmacist from Strasbourg, Hieronymus Brunschwijgk (ca.1450-1512), is issued. The book consists of three parts and was partly written in Latin and partly written in German. The first part describes distillation instruments and furnaces and contains 79 illustrations. That is why the book has enormous relevance to the history and the distilling techniques. The second part is a herbal and it presents a adjusted distillation method for each plant. The third part is about the medicinal properties of the distillates. In 1512 Hieronymus Brunschwijgk published a more elaborate work: Liber de arte distillandi de compositis. The addition mainly covered the preparation of distillates out of herbs, berries and seeds. Both books would continue to be thé works on medicinal water preparation. Several reprints and translations followed. For example in Brussels in 1517 a translation of the booklet is published by Thomas van der Noot with the title Die distellacien ende virtuyten der wateren. The Dutch translation contained only 104 pages instead of the original 230 pages. Perhaps the restriction happened due to financial reasons, because books were very

expensive at that time. It is largely the technical part that had been shortened: did the translator think or know that the distillation techniques were sufficiently known in the Low Countries?

In Antwerp in 1520 Willem Vorsterman publishes *Dit is die rechte conste om alderhande wateren te distilleren ende oock van die virtuten van alle ghedistileerde wateren seer goet ende profitelyck.* It is a pharmaceutical manual (48 pages) that can be considered as an appendix to the book of Hieronymus Brunschwijgk. In the introduction the writer describes the alembic and the furnace. The alembic is made of lead (something that will be prohibited later) or glazed pottery and it consists of a *scotele*, a dish in which the crushed herbs are placed and a *cleyn torreken*, a small still-head with a spout that can be put on the dish. The alembic itself is put on a *cleyn ooveken*, a small furnace under which a good fire without any smoke is made. The preparation of *aqua vitae* out of wine is not discussed. However the author does praise the many properties of this water of life and he calls it *ghemeynlyc vrouwe van alle medecinen* (common wife of all medicines). He does add the phrase *alsment drinct bi maten* (if it is consumed with measure).

In Antwerp in 1552 Jan Roelands publishes *Een Constelijck Distileerboec* written by the physician and Antwerp resident Philippus Hermanni. The book was such a best-seller that it was reprinted several times in the same city: in 1558 by Simon De Cock and in 1570 by Guilliaem van Parijs. After the disintegration of the Low Countries in 1585 the book was published in Amsterdam in 1612 and 1622 by Broer Jansz. The reason for this was the prohibition rule of the archdukes Albert and Isabella following excessive use of alcohol which had made distilling of corn brandy in the Southern Low Countries illegal and which had caused the distillers to flee northwards. The main focus in *Een Constelijck Distileerboec* is on the preparation of botanical, animal and mineral medicinal waters. Philippus writes in chapter XXI about the way to prepare juniper water or aqua juniperi.



Fig. 6. Alembic for the production of brandy, 1552. In: Philippus Hermanii, Een constelijck Distileerboec, Antwerpen, 1552.

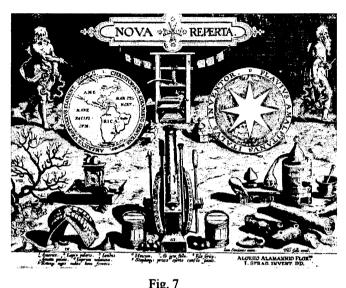
For this purpose juniper berries are crushed, sprinkled with wine and then distilled. The book also contains a small, decorated tract of the master himself about how to distil brandy (fig. 6). This tract cannot be matched and will be considered the manual for distillers during many years and it will contribute to the explosive growth of the malt spirit industry in the Northern Low Countries. The tract starts with the description of a furnace. Its construction's concept is such that with a minimum of coal or peat a maximum heating efficiency is obtained. The copper distillation kettle is hanging detached in the furnace allowing the fire's warmth to heat up the kettle's sides sticking out of the furnace. A small grid makes a small hearth that can be fanned with adjustable air holes. To alleviate the work an automatic supply of coal and peat is provided. The size of the copper distillation kettle depends on the amount of fermented mash one wants to distil. According to the representation in the book, the still-head fitting on the kettle can have the form of a cone (the so-called Rosenhut) or a watercooled onion shaped still-head (the so-called Moor's head). Philippus Hermanni gives much attention to the cooling of the vapours. He writes

about *spirits* or forces of wine, which are driven out by the heat. It is the first time in Dutch literature that alcoholic vapours are called *spirits*. To obtain a maximal cooling of the alcohol vapour the copper pipe attached with a mixture of egg white and flour to the spout of the still-head, has to run through a barrel filled with water. The size of the distillation kettle determines the size of the cooling barrel. When a kettle of two *amen* or more is used (1 aam is about 135 litres), the cooling barrel has to be eight or nine *amen*. For a small kettle of ten or twelve *potten* (1 pot is about 1.15 litres) a cooling barrel of a *quart* (1 quart is about 57 litres) suffices.

Although Philippus Hermanni advises the use of oprechten welrieckenden wijn (wine with a good smell), it is admissible to use wine that lanck oft onclaer gheworden is (turbid wine) because of economical reasons as long as it has not turned sour. Because the sweeter the wine, the stronger and better the brandy will be. Philippus warns for the many impostors who use other substances than wine like grape marc, beer yeast and diergelijcken onreynicheyt with which he is probably referring to turbid beer and alcoholic drinks made from fruit.

Besides the tools and base materials Philippus also describes the distillation procedures. For instance, only two thirds of the kettle is filled with wine. Next the kettle, still-head and pipe are attached to each other. The distilling itself has to happen slowly because the slower the procedure, the better the brandy will turn out. The concentration of the distillate is checked with a taste test. Philippus makes a distinction between the best brandy and brandy. The best brandy is obtained by collecting only the strong fraction when distilling the wine. The residue of different distillations is gathered and distilled again hence obtaining brandy.

From numerous publications appears that the art of distillery reached a high during the second half of the 16th century. In this respect the Dutch took the lead. For example we know of several books on the preparation of medicinal waters and in the majority of the books on agriculture and apiculture we can find a chapter on the preparation of brandy from wine, mead or cider.^{37,38}



Nova reperta (new inventions), end 16th century. Engraving by Jan van der Staet (Stradamus), Amsterdam

In the engraving *Nova reperta* (New inventions), made at the end of the 16th century by the Fleming Jan van der Straet (Stradanus) the nine most important discoveries of the late Middle Ages and the Renaissance period are illustrated: one is the alembic (fig. 7). When one studies the paintings of that time, one concludes that mainly painters (Pieter Brueghel de Oude, Jan Steen, David Teniers de Oude, Jan van der Straet, Jeroen Bosch), engravers (Pieter van der Borcht, Maarten de Vos) and writers (Erasmus) from the Low Countries tackle the subject of alchemy and alchemy chambers.²⁹

When, in 1585, the union of the Low Countries disintegrated many distillers swarmed off to all parts of the world. They can be traced in several cities of the Northern Low Countries where they participated in the expansion of the malt spirit industry. This consisted of many so-called four-casks distilleries. Once in twenty-four hours 4 casks with grain mashes were made, 4 were fermenting and 4 others were being distilled. These casks had an average size of 13 barrels (1 barrel being 232 litres).

The distillation took place in two wash stills of about 6.5 barrels with cooling tanks and the rectification in an alembic of about 4 barrels. When the distillery expanded there was simply added a second (or a half) four-casks distillery. In such a distillery work 3 to 4 labourers.³

We also encounter distillers from the Southern Low Countries in Berlin, Cologne and Nürnberg where they established grain distilleries. much to the dismay of the local distillers.^{25,39} In France we can find them on the island Île de Ré and in La Rochelle where they produce brandevin (just as brandy derived from the Dutch word brandewijn).⁴⁰ In the town of Cognac they establish the first distillery of cognac.⁴¹ Still reminding this is the Quai des Flamands by the Charente. In 1604 the consuls of Bergerac met to discuss the plan de faire venir certains hommes flamands pour faire une grande quantité d'eau-de-vie et à ces fins dresser leurs fourneaux.⁴² In London resides a strong colony of protestant refugees from the Netherlands (in 1570 there number exceeded 6000!) with among them distillers who produced brandy (also derived from the Dutch word brandewijn) and gin (geneva or Dutch courage).⁴³ The book The Whole Art of Distillation was written in 1692 by W.Y-worth, a physician born in Schiedam and citizen of Rotterdam, and illustrated by the Flemish engraver M, vander Gucht. The Dutch established rum distilleries in Brazil and Barbados and in 1644 they established the first North American grain distillery in Staten Island.^{44,45}

The world trade in brandy was completely monopolised in the first half of the 17th century by the Dutch and the sailors of the United East and West Indian Company were the biggest consumers...⁴⁶ The brandy was transported in barrels the volume of which was globally expressed in *vier-tels* and the alcohol strength in *Hollandse proef (Dutch test)*. Until the 17th century there were several ways to determine the alcohol amount. A brandy had only been rightly distilled if

- it completely burned without leaving any humidity behind,
- a piece of cloth immersed in it did not burn up,
- an oil drop sank in it,
- a drop of it put on a hand evaporated entirely,
- a bucket filled with it completely burned up,
- camphor melted in it.

In the 17th century one was familiar with the gunpowder test: a spoonful of gunpowder was showered with brandy and held in a burning candle. If he

brandy contained too much water, the gunpowder did not catch fire. The most used checking method however, was the *Hollandse proef (Dutch test)*. This implied filling more than the half of a small bottle with brandy and shaking it vigorously. This shaking formed many little bubbles of air. If the brandy was too strong, the bubbles would rise very quickly; if, on the contrary, it was too weak the bubbles disappeared slowly and the brandy became white. If the brandy was *op proef* (on proof, about 50% vol alcohol) a *kleyn schuymjen* (some foam) appeared in the middle and the bubbles did not disappear quickly.⁴⁷ This Dutch checking method was accepted all over Europe; it would only disappear in the 19th century when the hydrometer was introduced.

How corn brandy was produced in the 17th century, can be read in Den Volmaekten Brandewijnstooker en Distilateur. The book was written by J.K.B.P. (Jacob Bols?) and published in Maastricht in 1794. A second revised edition titled Een uytvoerig en omstandig bericht van de nieuw ontdekte distilleerkonst was published in Amsterdam in 1736. On the matter of the preparation of corn brandy the author refers to the recipe Koorn-Brandewyn uvt Koornvruchten te maeken by Johann Rudolf Glauber (1604-1668). This German physicist and chemist settled in Holland in 1648 and died 20 years later in Amsterdam. He was possessed by the art of distillery and exercised this art in a very practical manner. His contemporaries called him the Paracelsus of distillery art. In 1648 he published his Furni novi philosophici oder Beschreibung der neu erfundenen Destillierkunst in Amsterdam and in Leyden: this work was reprinted several times. Glauber developed an iron alembic for the industrial production of nitric acid. This led to the establishment of distilleries that were able to produce over 10,000 kg of nitric acid (aqua fortis) a year and by adding hydrogen chloride were able to prepare aqua regia (fig. 8).

The nitric was very popular among European engravers at the time and it was known by the name of *Hollands zuur* (Dutch acid). Glauber is also considered to be the inventor of the vapour distillation: he boils water in a retort and allows the vapour to effervesce through a barrel filled with wine. The steam carries the volatile alcohol and the vapours leaving the barrel are cooled in a spiral tube, immersed in a cooling barrel. In a different setting up he lets the vapours run through a battery of cooling barrels thus obtain



Fig. 8 De Stoockerey van Gerrit Kraandyk en Cie, manufacture of aqua fortis and aqua regis, 1732. Engraving by Abraham Zeeman, Amsterdam.

ing different alcohol fractions (fig. 9). This invention is nevertheless soon to be forgotten and will come into vogue again during the first industrial revolution. Until then the distillers continue to cherish their alembics.

The first industrial revolution does not leave the distillers unaffected. Stimulated by B. Thompson Rumford, in England around 1802 steamers are used to heat the grain mashes. In this respect a distinction is made between direct and indirect heating. When heating indirectly the mashtuns are equipped with a steam-jacket or with an internal, spiral steam tube. When heating directly the mash is heated by direct steam injection, also called living steam. In large distilleries steam engines were used to drive the malt mills, steam pumps, mixers and lifting machines; all these tools made manual labour easier and speeded up the production.

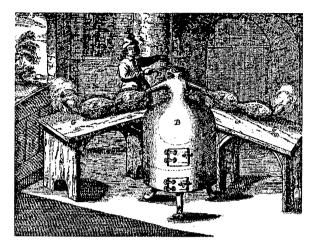


Fig. 9 Alembic with dephlegmators, 1666. In: J.R. Glauber, Von den Dreyen Anfangen der Metallen, Prague, 1666.

Of even more importance than the heating of the mash and the driving of the machines, was the introduction of the continuous working steam column. In this area the French inventors were very important. In a period of 30 to 40 years the transition of the classic alembic to the continuous working steam column had been achieved.^{24,48,49,50}

The first substantial step in this respect was the invention in 1780 of *le chauffe-vin* by the brothers Argand. Between the alembic and the water containing cooling barrel, they placed a barrel with wine not yet distilled. They made the cooling spiral run through the wine barrel first and only then through the cooling barrel. The vapours condensed in the cooling spiral running through the wine barrel hence preheating the wine. The use of this heat exchanger led to a considerable fuel saving and a reduction of the distillation time. Thus the invention of the Argand brothers was called *cuve de vitesse*. The heat exchanger could also be used to distil a thick grain mash; then the heat exchanger was provided with a mixer that prevented the sedimentation of flour particles, chaff and yeast cells.

The next significant step was made in 1801 by Edouard Adam who took over the ideas of Glauber. Adam boiled wine in an alembic and then let the vapour effervesce through three egg-like containers filled with wine (fig. 10). Next the damp was condensed in a closed cooling spiral that was placed in two drums arranged on top of each other. The upper vessel was filled with wine and thus preheated. The lower vessel contained cooling water. The damp carries the volatile wine alcohol along in the egg-like containers hence enriching the damp with alcohol. The great advantage of Adam's distillation device was that it was possible to produce a high alcohol concentration using little fuel.

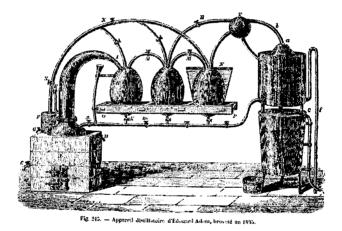


Fig. 10 A simple Adam still, 1805. In: L. Figuier, Les merveilles de l'industrie, Paris.

In 1805 Isaac Bérard took a patent on a *appareil distillatoire propre* à retirer du vin dans une seule opération de l'eau-de-vie épreuve d'Hollande, de l'esprit trois-cinq, trois-six, à la volonté du fabricant. Bérard placed a condenser between the alembic and the cooling barrel (fig. 11). The condenser was cooled in a water trough and consisted of two parallel, horizontal, cylindrical tubes which had been internally compartimented with perforated plates. When running through the condenser the least volatile part of the vapour was condensed. The non-condensed vapour contained more alcohol and was made liquid in the cooling barrel. The condensate, rich in water and poor in alcohol, ran back to the alembic. From the title of Bérard's patent it appears that at the beginning of the 18th century the strength of brandy was still indicated in the *Hollandse proef* like it had been the custom to do so since the 17th century. An *eau-de-vie of trois-cinq* means that three parts of this eau-de-vie diluted with two parts of water result in an alcoholic solution with a strength of the standard Dutch gin (*Hollandse proef*).

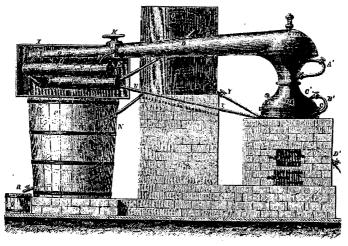


Fig. 251. - Deuxième appareil d'Isaac Bérard.

Fig. 11 The Bérard apparatus, 1805. In: L. Figuier, Les merveilles de l'industrie, Paris.

In 1813 Jean Baptiste Cellier-Blumenthal took a patent on a continuous working steam column. He had discovered this when investigating the production of granulated sugar out of sugar beets. It was the German chemist Andreas Margraff who demonstrated in 1747 that beets contained crystallizable sugar. Forty years afterwards Karl Archard succeeds in preparing beet sugar and with the support of king Frederic II of Prussia he opens the first European sugar factory. Germany is the first European sugar producer but it is swiftly surpassed by France. The reason for this was the English blockade of the continental ports. Consequently the cane sugar from the colonies was no longer obtainable. Napoleon promised a premium of 1 million French francs to the inventor of a good method to produce on an industrial scale white granulated sugar from beets. Like others, Cellier-Blumenthal also tried to refine beet sugar with alcohol, a method already described for the refinement of sugars from grapes and honey. However for this procedure large quantities of alcohol needed to be vaporised, which encouraged Cellier to develop a distillation instrument that could produce even faster and more economically than the devices of Adam and Bérard, He combined Adam's principles (enriching vapours relatively poor in alcohol through contact with an alcoholic liquid) and Bérard's (enriching vapour by controlled partial condensation and by removing the condensate). Cellier located a vertical column consisting of two parts on the alembic. The upper part, the rectifying column, was equipped with different perforated plates; the lower part, the distillation column, contained plates with bubble caps. Cellier let the wine stream downward out of the supply tank with height control, through a small and large condenser, into the rectifying column. The wine streaming downwards came into contact with the ascending vapours out of the steam-heated alembic filled with wine. The good contact between the downward-streaming wine and the ascending vapours produced a continuous condensation and evaporation. The vapours leaving the rectifying column were very rich in alcohol and were being cooled in the condensers while in the meantime the wine was being preheated. The non-volatile wine fraction, the vinasse ran down the column and left it at the bottom.

To construct this steam column Cellier called upon the Parisian machinery builders Derosne and Cail. Cellier's steam column was soon copied and some like Baglioni from Bordeaux claimed the paternity of the invention of the steam column. This issue was taken to court and Baglioni lost. Irritated by all these trials Cellier left Paris in 1820 and settled in Koekelberg near Brussels.

Cellier also had success abroad. In 1817 the German professor Johann Pistorius took a patent on an instrument that allowed him to distil thick grain and potato mashes. In England in 1830 Aeneas Coffey patented a steam column that is very suitable to distil grain mashes. The German and English instruments were able to produce industrial alcohol in one step. (These steam columns, primarily Coffey's, will later also be used to distil petroleum). In France and in Belgium industrial alcohol was produced in two steps. First wine or an other alcoholic liquid was distilled to an alcohol percentage of 50-52% vol and consequently rectified to 80% vol, which was the industrial standard at the time.

In Belgium there was a lot of interest in the steam column.⁵¹ The fact that Cellier-Blumenthal had settled in Koekelberg in 1820 was certainly one of the reasons. Also his later friendship with king Leopold I had a stimulating effect. Leopold I, who owned a potato distillery in Niederfülbach, Saxony, was very interested in Cellier's work. In Belgium Cellier mainly worked with the coppersmiths Delattre, Dubois and Camal from Brussels. In 1828 Cellier performed tests in the distillery Dooms in Lessen together with coppersmith Dubois. He rebuilt his steam column, developed for wine distillation, so it would be suitable to distil thick grain mashes. For instance, he replaced the perforated plates in the rectifying column by plates with bubble caps in order to obstruct the column less. In 1829 he installed a steam column in the sugar refinery / distillery Van Volsem in Halle and in 1830 one in the distillery Claes in Lembeek. This steam column was entirely satisfactory and meant a real breakthrough in the world of distillery, where it was known by the name la colonne belge (fig. 12). This column that could mostly be found in industrial distilleries, was made out of red copper and contained depending on the distiller's wish 12, 14 or 20 dishes. In Belgium, Cellier also collaborated with Pierre Savalle with whom he had been friends since 1813. This French engineer spent a large part of his life in Belgium and owned three sugar refineries with distillery. The story goes that Cellier and Savalle escaped death by a hair's breadth when an explosion happened during a test distillation. This incident would have driven Savalle to look for a steam control, research he ended successfully in 1857. François Savalle, born in Louvain in 1838, assisted his father and inherited the sugar refineries with distilleries and the construction workshop Savalle et Cie when he died in 1864 in Lille. F. Savalle built numerous steam columns, which were not only used in industrial distilleries and agricultural distilleries, but also in the emerging petroleum industry.

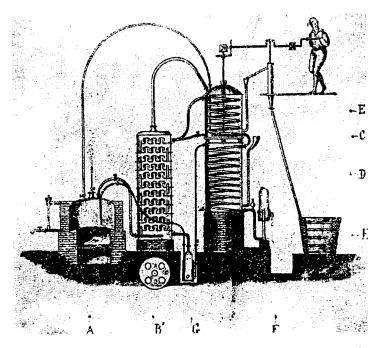


Fig 12 The Cellier column (La colonne belge), 1841. Brevet de perfectionnement n° 816, 30 août 1841, Brussels.

The most impressive industrial distillery was Louis Meeus' distillery *De Sleutel* in Wijnegem (fig. 13). It was established in 1869 but its roots were in the Antwerp distillery *Het Anker* which dated from 1753. During the world exibition in Antwerp in 1895 the distillery had a prestigious stand and important business figures were revealed at that time.⁵² The company had a build-on surface of 20,543 m² and disposed of a steam engine park that supplied 1,084 h.p. The grain was brought in by ship and stored in one of the 19 silos. These silos were four storeys high and could contain 50,000 hl of grain. The malt-house had a malting floor of 8,000 m² and the green malt was dried in an one-layer kiln equipped with mechanical turners. The company had 12 grain mills. The unmalted grain was released with steam under pressure in cylindroconical Henze-cones. The saccharification took place in lying macerators rotating on their axe. The fermentation process took 48 hours and took place in one of the 34 copper tubs of 120 hl,

equipped with an internal cooling spiral. The company had 11 different distillation columns to separate volatile from non-volatile components. The rectification was done with alembics (for gin) or rectification columns (for spirits). It was possible to produce 50,000 litres of alcohol of 50% vol on a daily basis. The year after its establishment the company already produced 7,154 hl of alcohol at 50% vol.

The production rose continuously and in 1884 it mounted to 101,359 hl. The company had its own steam vessel, l' *Etoile*, which transported gin from Wijnegem to Antwerp where the enterprise had large warehouses. Part of the gin production was exported to Coruna, Rio de Janeiro, Liberia, Sourabaya, Dunedin, Sydney, Hongkong and Melbourne. In 1884 42,665 tons of imported grain was consumed, 3,600 pieces of cattle were fatted, 1,300 of which in stables of their own, 574,000 hl of manure was sold and 25,998 francs of taxes was daily paid. The company employed 250 workers, had its own fire brigade and even a factory chapel.

Since the previous century the steam column has known no substantial changes. However it has become bigger, made out of rustproof steel, computer-controlled and more energy economical.⁵³ Today the largest distilleries are in Brazil where colossal amounts of water-free alcohol are being produced as fuel for vehicles as part of the *Programa Nacional do Alcool.*⁵⁴ In this respect distilleries were built which starting from the fermented cane sugar molasses can produce up to 240,000 litres of water-free alcohol per day!

Despite the introduction of heat pumps and the distillation under increased pressure distillation remains very energy-consuming (10 to 13 MJ per litre water-free alcohol) and (at the current rate) the use petrol of is cheaper. Therefore a less expensive separation technique is being searched, possibly it will be the membrane technology. For the production of pure industrial alcohol this might be possible; for the manufacture of the more complex strong drinks this is not for tomorrow, not even for the day after.



Louis Meeus' distillery *De Sleutel*, Wijnegem (Antwerp), beginning 20th century. Postcard, Nationaal Jenevermuseum, Hasselt.

Alcoholic fermentation: the cradle of biochemistry and microbiology^{55,56}

The phenomenon that grape juice or some other sweet liquid starts to bubble up spontaneously during a shorter or longer period of time to finally end up being a drink that has a fuddling effect, has intrigued many researchers throughout the centuries. Although the art of making wine, beer, mead and other alcoholic drinks has been existing for centuries, the insight in yeast and the fermentation process was not to be gained until the 19th century. The study of yeast and the fermentation process is one of the most interesting chapters in the history of science. Many famous scientists were interested in the phenomenon of alcoholic fermentation and their research is part of the origin of general and applied biochemistry and microbiology, a scientific discipline nowadays referred to as biotechnology.

Until the 15th century the alchemists believed that fermentation (*fermenta-tio*) was being caused by a ferment (*fermentum*). They compared the ferment with the Philosopher's Stone: just like the Philosopher's Stone transformed base metals into gold, such did the ferment to the dough, made it rise and transformed grape juice into wine. Although one was not familiarised with the true nature of the ferment, knowledge of the fermentation parameters was relatively well managed. In a 15th century beer recipe (Ms. 697, fols. 72-84, Library of the University of Ghent) the use of *heve* (yeast floating on the surface) and *onderghist* (flocculated yeast) is already being mentioned. These different types of yeast are first being tempered in *werse* (wort) and only then the inoculated yeast is casked and mixed with the chilled grain mash. The chilling continues until *bloet leau* (body temperature). The fermentation process (*liggen heffen*) takes three days.¹⁰

In the first Dutch bee book Van de bven, hare wonderlicke oorspronc, natuer, evgenschap, crachtige, ongehoorde ende seltsame wercken two recipes to make mead can be found.³⁸ The bee book, first published in 1597 by Jan Claesz. in Leyden, was written by Theodorus Clutius (1550-1598) in the form of a dialogue with the famous botanist Carolus Clusius (1526-1609) from the French Flemish town of Atrecht (present-day Arras). In 1593 the board of governors of the university of Leyden entrusted Clusius with the planning of the Universitevts Kruyt-Hof. Clusius was not very mobile so he had Theodorus Clutius (Dirk Outgaerz Cluyt), who was a pharmacist in Delft, assist him. Their mead recipes show that they had a profound knowledge of the fermentation parameters. For example 90 parts of white honey are boiled down and skimmed off with 10 parts of clean river water. This result in a triple effect: the mead must becomes sterile, the sugar concentration rises and the proteins solidify. The sterilisation of the must does not only prevent flavour deviations following infections, but together with the increased sugar concentration it leads to an increased alcohol formation. Skimming of the coagulated proteins prevents the yeast from getting foul and results in a clear mead. The choice of white honey is

remarkable: we know now that honey contains less proteins and hence produces less turbidity. Boiling down the mead must to the density desired is being checked with an egg: as soon as the egg starts to float the boiling process is stopped. After being chilled the mead must is put in a drum and it is mixed with brewer's yeast. The drum has to be full at all times, so it must be refilled in order to allow the yeast to remove all non-soluble substances, which improves the clarity and taste of the mead. After the primary fermentation process the drum is closed causing an anaerobic maturing, so excluding an acidification by acetic acid bacteria.

In the 16th and 17th centuries the iatrochemists advocated the use of *spiritus vini* for making medicinal tinctures and extracts. The most important representatives are Paracelsus (1493-1541), Joan Baptista van Helmont (1579-1644) and Franciscus de le Boë, Sylvius (1614-1672).

Paracelsus joins the alchemists and writes his *Paragranum* in 1530: "the alchemist proceeds like a baker making bread with flour and leaven". Paracelsus is the first to call *spiritus vini* alcohol, a term which will be largely introduced by Herman Boerhaave (1688-1738).

Joan Baptista van Helmont broadens the term fermentatio. According to this Flemish scholar from Vilvoorde fermentation (fermentatio), digestion (digestio) and putrefaction (putrefactio) are similar phenomena. In his Dageraad ofte Nieuwe Opkomst der Geneeskonst, published in 1660 by his son, van Helmont argues that every change needs a specific ferment (heve).²⁶ During these change warmth is released. It is not the heat that activates the ferment like Libavius (1540-1616) argues, but it is the drift des zaets (passion of seed). This seed or semen gives it specificity to each body. During the fermentation process van Helmont finds a siedende bobbelinghe (seething bubbles), a windt (wind) that he can see and hear. Above the bung of a drum filled with fermenting wine must he collocates the head of an alembic. Much to his surprise he notices that this "wind" cannot be condensed and therefore it cannot be a wine ghost. He compares this what he calls *bobbelinghe* with the effect of acids on rock, a process during which windt is also released. The same winds which he calls gas, also appear in the human abdomen and in Spa water and are being formed when burning wood (a century later the Scot Mac Bride will show this gas to be carbon dioxide).

The French Fleming Franciscus de le Boë, Sylvius, professor at the university of Leyden, does not quite agree with van Helmont. For instance, in his book *Opera medica* (Geneva, 1698) Sylvius makes a clear distinction between the phenomenon of releasing gas when an acid is affecting limestone and the phenomenon of fermentation. He calls the first phenomenon *effervescentia* or effervescence. It goes together with the origin of a new compound. This is not the case with *fermentatio* or the fermentation which resembles more a disintegration.⁵⁶

Antoni van Leeuwenhoek (1632-1723) was the first to see living yeast cells.⁵⁷ This cloth trader from the Dutch town of Delft was familiar with the use of lenses to check the quality of cloth. He developed a technique to polish lenses and with these he built little single microscopes. With some of them he accomplished to magnify 500 times. In 1680 he looks at yeast cells from wine and beer and he draws them as congealed bulbs. He does not consider them to be alive (but he does view bacteria as such) but as congealed bulbs originating from elements used in the preparation of wine and beer. The yeast cells isolated from beer seem bigger to him than those from wine or syrups he had been given by a pharmacist from Delft.

According to the English physician Thomas Willis (1621-1675), professor at Oxford university, a ferment is an element which transfers its inner movement onto other fermentable elements. The German physician and chemist Georg Ernest Stahl (1660-1734), professor at the University of Halle, shared this opinion. In his work *Zymotechnia fundamentalis* (1697) he states that the fermentation process is a kind of reaction during which the ferment collides with fermentable elements. This produces larger elements to be broken down into smaller, ever more stabile elements. This mechanical fermentation theory was advocated by many, partly because of the enormous prestige Stahl enjoyed. For he was the inventor of the phlogiston theory which stated that elements release a substance, phlogiston, when being burned (oxidation). The influential professor from Leyden, Hermann Boerhaave (1668-1738) also advocated this mechanical fermentation theory.

The French chemist Antoine Lavoisier (1743-1794) destroyed both Stahl's phlogiston theory and his fermentation theory.⁵⁸ On the basis of weight determinations he concluded that sugar is half transformed into alcohol and half into carbon dioxide during the fermentation process. He saw sugar as an oxide (*oxide végétal*). During the fermentation process of the sugar a part of the oxygen would compound with a part of the carbon forming carbon dioxide. With hydrogen and oxygen the rest of the carbon would produce alcohol. In 1815 French chemist Joseph Gay-Lussac (1770-1850) formulated these ideas in an equation:

$$C_{12}H_{24}O_{12} = 4 CO_2 + 4 C_2H_5OH$$

In 1828 his fellow countryman J.B. Dumas (1800-1884) argued that this was not a matter of sucrose (wrongly written like $C_{12}H_{24}O_{12}$ by Gay-Lussac) but one of glucose. He corrected this equation into:

$$C_6H_{12}O_6 = 2 CO_2 + 2 C_2H_5OH$$

The Austrian Christian Erxleven was the first to state that yeast is a living organism in 1818. More or less two decades later this statement is agreed with by the Frenchman Charles Cagniard Latour (1777-1859) and in 1837 the Germans Theodor Schwann (1810-1882) and Friedrich Kützing (1807-1893). Among these three Schwann excelled because his vitalistic fermentation theory relied on tests. Moreover he had quite a status because of the discovery of animal and plant cells about which he declared that the cell was the smallest unit of a living organism. Schwann demonstrated that boiled, sugar containing liquids started to ferment when they were being exposed to air. Furthermore he found that air looses its fermentation ability when it is being heated in a flame or washed in concentrated sulphuric acid. Following these tests he concluded that fermentation is not caused by oxygen but rather by a living organism that can be destroyed by heat or concentrated acids. He called this organism Zückerpilz or Saccharomyces. Kützing, who did his research on acetic acid bacteria, concluded with analogue findings.56

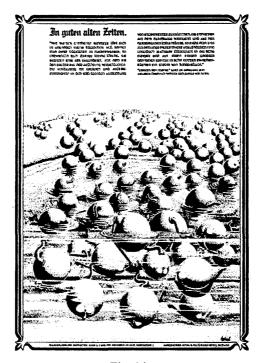


Fig. 14 Cartoon representing "living" yeast cells, F. Wöhler and J. von Liebig, 1839. In : Annalen der Chemie 29 (1839)

This vitalistic fermentation theory was strongly opposed by J.J. Berzelius (1779-1848), Friedrich Wöhler (1800-1882) and Justus von Liebig (1803-1873). These men were far from unimportant: in 1813 the Swedish professor Berzelius introduced the modern chemistry notation system. In 1828 the German professor Wöhler succeeded in synthesising urea and in doing so he laid the foundation of organic chemistry. His friend and fellow countryman von Liebig, professor at the university of Giessen is being regarded as one of the greatest chemists ever. In 1839 Wöhler and Liebig published a cartoon (fig. 14) representing yeast cells as small, globular creatures. They had a little trunk to suck up sugar, an anus to discharge alcohol and genitalia to secrete carbon dioxide...⁵⁹

Berzelius stated in 1839 that yeast was a lifeless, organic catalyst and Von Liebig shared this opinion. Both were familiar with the work of A.P. Dubrunfaut, a distiller from Lille, who wrote in 1824 how gluten, a nitrogen containing element in germinating grain, was able to break down quickly and entirely starch into fermentable sugars (in 1833 Payen and Persoz will demonstrate that gluten contains a diastase, able to liquefy and saccharify starch).⁶⁰ Dubrunfaut observed that the gluten worked best at a temperature of 62,5 °C and that the activity decreased when temperatures increased and finally it disappeared at 75 °C. Therefore he stimulated the use of a thermometer and he checked the efficiency of the saccharification with iodine: starch gives a blue colour with iodine. When the starch decomposition is finished the red-brown colour of iodine is recovered.

Berzelius' chemical fermentation theory also relied on the analogy between the fermentative conversion from alcohol into acetic acid and the catalytic oxidation of alcohol into acetic acid in the presence of colloidal platinum. According to Von Liebig the conversion of sugar into alcohol and carbon dioxide is catalysed by a nitrogen containing element. The speed at which it happens is influenced by temperature, degree of acidity and the presence or absence of inhibitory factors like heavy metals.

In 1857 the Frenchman Louis Pasteur (1822-1895) joins the vitalistic fermentation theory of Schwann.⁶¹ This happened after being asked by a distiller from Lille, Bigo, father of one of his students, to study a fermentation of molasses that had got out of hand. For several weeks Pasteur took different samples in Bigo's distillery and he analysed these in his laboratory (fig. 15).

He demonstrated that in fermentations that had gone wrong not only alcohol had been formed, but also lactic acid, something that did not happen with healthy fermentations. Under the microscope he also found that in bad fermentations besides spherical-shaped yeast, many smaller rod-shaped and other globular ferments could be seen. By adjusting the composition of sugar solutions Pasteur managed to direct the fermentations in the right

A 92 or: Sequermy a usine de 10 93. 90 . Two band ports an condage Tom to save supremulations . I us at rempli of qui and Day wour Trapidy . Ju pris ilaticios de lapes, culand Des sais à une pran Par se globules so format . Court sugar Sepetter ayour an mout settimulation & plur aprile , and rapide que tim by patto de la cure ans 7/8 de suprementation . Sendement Dais celly at la mombre de cense qui to invision of put the plus grand , If sign par Sugar global . Bitteran bould in caron , un per algisia no istal a synis planing, jours muchelaboration . Elland profestion of some , This bills about blander & C'interiour them to prate gogs rape . The Commence and the aports Conservages , Jepan a jus Dim mutore fine . Deaucoup de pilit gladuly à memoriet try rapity of parties a tom by watting presailiely you and beaun sugarie globules, specipie another in trace quart shale grassing Da globules Day was a saferment at ion a saveda , un primero in mente . Je you a airetrain So Dam, to olimy Jus antivand Distages & with an plat. Je parte monettintion but les so Drive Dag Colule, d Caferal tim very gratter and gross farmanests and in selvin - 1

Fig. 15 Notes of Louis Pasteur in the distillery Bigo, Esquermes (Lille), 1856, November 4. Collection Pasteur Vallery-Radot.

direction. For instance, he proved that yeast cells develop easier in acid sugar solutions, while lactic acid ferments develop easier in neutral sugar solutions. Besides that, Pasteur also found that apart from alcohol and carbon dioxide an equal amount of glycerol and succinic acid is formed and that yeast cells absorb ammonium salts from the fermentation liquid. ANALOGUE testing led to the proposition that every fermentation process is triggered by a specific living organism and that every micro-organism needs a specific nutrient for it. Pasteur also looked for the causes for infections occurring during the fermentation process. He proved that the air contains a lot of micro-organisms. To do this he sucked air through a sterile filter of cotton and then he added this contaminated filter to a sterile infusion. This resulted in the development of micro-organisms, something which did not happen when the aspirated air was heated first. Sterile infusions could be kept in open glass recipients indefinitely if only the long neck was sufficiently bent downwards (U-trap) so the air, but not the germs could end up in the infusion. With these tests Pasteur managed to enfeeble the theory of generatio spontanea that claimed germs could spontaneously be generated from non-life matter. During his research on butyric acid fermentation in 1876 Pasteur discovered the existence of micro-organisms which were only able to stay alive in absence of oxygen: la vie sans air. Pasteur thought that these anaerobic micro-organisms got their oxygen and energy from the degradation of organic matter. Some micro-organisms, the so-called facultative anaerobic, dispose of two energy supplying mechanisms. Pasteur demonstrated that yeast is inclined to form alcohol in an environment lacking oxygen, while in an environment filled with oxygen it is more likely to focus on multiplying (the Pasteur effect). These new insights in the fermentation process led to practical applications which Pasteur disclosed in his Etudes sur le vin (1866) and Etudes sur la bière (1876).

The study of yeast and fermentation was accelerated in 1879 when the Danish researcher Emil Hansen completed a method to breed pure yeast cultures and in 1897 when the German chemist Eduard Buchner isolated the first yeast-free ferment, the zymase.⁶²

Emil Hansen was tied to the laboratory of the Carlsberg brewery in Copenhagen. In the same laboratory worked also Kjeldahl who was the first who knew how to dose a protein. Hansen developed a special slide with cavitations and using this he succeeded in isolating one single yeast cell out of a mixture of yeast cells under the microscope. Afterwards he cultivated the desired amount of these yeast cell. This technique had many advantages: from now on it was possible to work with any type of yeast like with a chemical reagent, hence enabling the registration of its specific chemical, cytological and morphological characteristics. For instance, specific colourings would indicate cell parts like the cell wall, the cell nucleus, the mitochondria, the vacuole, glycogen kernels and oil drops. It is observed that the cell's look (thickness cell wall, size of the vacuole, presence of glycogen kernels...) can be influenced by nutrition (both a lack and a surplus thereof), temperature and oxygen. This can explain the pleomorphism of some types of yeast. One demonstrates that the cell nucleus mainly consists of chromatine parts which causes the multiplication and caries the hereditary characteristics. It is observed that yeast does not only procreate asexually through budding, but also through sexual spores. Through fermentation and assimilation tests the system of the fermentation process is implemented. This scientific knowledge is being directly applied in breweries and later on also in yeast and spirit factories. Progressive brewers check their fermentation culture on cleanliness and viability. The determination of the fermentation rate and the fermentation degree is being introduced.

The isolation of the fermentation enzymes out of the living yeast cells was a mere coincidence. In 1897 E. Buchner (1860-1917) was studying the medicinal effect of yeast proteins. He washed the yeast with water, added sand and kieselguhr, crushed the cell membranes and finally he squeezed the moist mass under high pressure. The liquid obtained resulted to be very subject to putrefaction. Bearing in mind the fact that fruit could be conserved by adding sugar. Buchner added a certain amount of sugar to the juice. Much to his amazement this sugar began to ferment. Buchner concluded that the fermentation had been caused by fermentation enzymes which he called zymase. The fermentation ability of the juice varied strongly, which was brought about partly by the amount of enzymes in the veast used, partly by the lability of the free zymase. Zymase was not a secretory product of the yeast, but appeared within the yeast cell and its composition seemed to be strongly connected with the yeast cell's growth. Furthermore, the fermentation activity seemed to be connected with the yeast cell's metabolism and energy management. By discovering the zymase, Buchner reconciled Pasteur's vitalistic fermentation theory and von Liebig's chemical fermentation theory: fermentation demands a living organism (Pasteur) that operates through a nitrogen containing catalyst (von Liebig).

The zymase discovery stimulated chemical research into alcoholic fermentation. Relatively soon was found that zymase was not only able to ferment glucose, but also fructose, mannose, sucrose and maltose. The research mainly focused on the degradation of glucose, a process that was also studied in mammalian muscles. Parallel with the study of anaerobe degradation of glycogen to lactic acid (glycolysis) ran the study of the change of glucose in alcohol and carbon dioxide. The results obtained from the study of alcoholic fermentation contributed to the solution of the problems encountered when studying glycolysis and vice versa. The study of glycolysis and alcoholic fermentation was done between 1900 and 1950. Many known researchers were involved in this study: for instance A. Harden, W.J. Young, C. Neuberg, H. Euler, G. Embden, O. Meyerhof, O. Warburg, K. Lohman, C. and G. Cori and J. Parnas.^{62,63}

In 1905 the British researchers Harden and Young did a dialysis of the cell free fermentation juice on parchment. They concluded that the dialysed juice and the dialyse water were not able to trigger fermentation of the glucose separately, but they did so when jointly used. The nondialysable component (the protein or enzyme complex) was heat sensitive. the dialysable component (the co-enzyme) was not. (In 1933 H. Euler identified the co-enzyme as being nicotinamide adenine dinucleotide or NAD⁺). Harden and Young found that apart from the enzyme complex and the co-enzyme fermentation also needed phosphates. They demonstrated that CO_2 production from glucose starts very rapidly, but that it suddenly slows down unless an inorganic phosphate is added. They detected a disappearance of the added phosphate during the fermentation when they were unable to precipitate it with uranium acetate. They concluded that the inorganic phosphate is converted into organic phosphate and succeeded in isolating a hexose diphosphate. In 1913 Embden proved this to be the fructose-1.6-diphosphate. (Later on Robinson isolated the glucose-6-phosphate and Neuberg the fructose-6-phosphate). Harden and Young proposed the following correction of the Gay-Lussac and Dumas equation:

$$2 C_6 H_{12}O_6 + 2 H_3 PO_4 = 2 CO_2 + 2 C_2 H_5 OH + 2 H_2 O + C_6 H_{10}O_4 (PO_4 H_2)_2$$

When the alcoholic fermentation was coming to an end, the phosphate was released again under the influence of a phosphatase, which indicated that the phosphate was part of a continuous cycle.

Progress in organic chemistry made it possible to isolate and identify other fermentation products. Some researchers argued that the transformation of glucose in alcohol and carbon dioxide happened through lactic acid which would be changed into formic acid and acetaldehyde. Consequently the formic acid would disintegrate in carbon dioxide and hydrogen and this would reduce the acetaldehyde to alcohol:

 CH_3 -CHOH-COOH = CH_3 -CHO + H-COOH H-COOH = CO_2 + H_2 CH_3 -CHO + H_2 = CH_3 -CH₂OH

However, Harden and Young rejected this point of view because they were unable to show the presence of any lactic acid in alcoholic fermentations. In 1907 Wohl stated that glucose, by dehydration, was changed into a "hypothetical molecule" which then disintegrated in glyceraldehyde and methyl-glyoxal:

CH_2OH -CHOH

CH_2OH -CHOH-CHOH- CH_2 -CO- $CHO = CH_2OH$ -CHOH-CHO + CH_3 -CO-CHO

In 1907 P. Mayer contested this hypothesis because methylglyoxal could not be proved and was not fermentable. In 1910 E. Buchner and O. Meisenheimer proved that glyceraldehyde was fermentable and that is was easily transformable into its fermentable isomer ketose, the dihydroxyacetone. A. von Ledebew said in 1911 that the glyceraldehyde and the dihydroxyacetone before being fermented, were phosphorylated. Also in 1911 C. Neuberg was able to determine the presence of pyruvic acid (pyruvate). Under the influence of an enzyme he called carboxylase, the pyruvic acid is transformed into acetaldehyde and carbon dioxide. The acetaldehyde is then with hydrogen reduced to alcohol using a reductase:

$$CH_3$$
-CO-COOH = CH_3 -CHO + CO_2

$$CH_3$$
- $CHO + H_2 = CH_3$ - CH_2OH

In 1911 C. Neuberg formulated a scheme of the glucose disintegration putting forward methylglyoxal and pyruvic acid as the central components of the fermentation (fig. 16). In 1913 G. Embden argued that the fructose-1,6-diphosphate was changed into two triose phoshates, viz. the dihydroxyacetone phosphate and the D-glyceraldehyde-3-phosphate. In 1927 O. Meyerhof demonstrated that glucose was transformed in the presence of adenosine triphosphate (ATP) and a hexokinase into glucose-6-phosphate. Early 1930 several researchers found that adding fluoride to the fermentation fluid led to an accumulation of 2-phosphoglycerate and 3phosphoglycerate. Furthermore, the concentration of fructose-1,6diphosphate was raised because of an addition of iodoacetate. In 1932 L. Fischer succeeded in synthesising the D-glyceraldehyde-3-phosphate and proving its fermentability. This research data allowed G. Embden and O. Meyerhof to compose their Embden-Meyerhof cycle in 1933. Although the individual steps were known, the research was continued. In 1935 H. Euler and O. Warburg demonstrated that nicotinamide adenine dinuleotide (NAD⁺) played a role in the transformation of D-glyceraldehyde-3phosphate into 1,3-diphospho-D-glycerate. The same O. Warburg proved in 1937 that ATP was formed when transforming 1,3-diphospho-Dglycerate into 3-phospho-D-glycerate and phospho-enol-pyruvate into pyruvate. This meant that the energy released when changing glucose into alcohol and carbon dioxide was partly released in warmth and was partly stored in the form of ATP. Also in 1937 K. Lohmann and P. Schuster declared that the heat stable thiamine played a role in the decarboxylation of pyruvate to acetaldehyde and carbon dioxide. Negelein and Bromel isolated the labile 1,3-diphospho-D-glycerate in 1939. This allowed the statement that by 1940 the Embden-Meyerhof cycle was entirely known.

The formation of the higher alcohols (fusel alcohols) was studied in 1905 by F. Ehrlich. Yeast forms these higher alcohols out of amino acids in accordance with:

 $R-CH(NH_2)-COOH + H_2O => R-CH_2OH + CO_2 + NH_3$

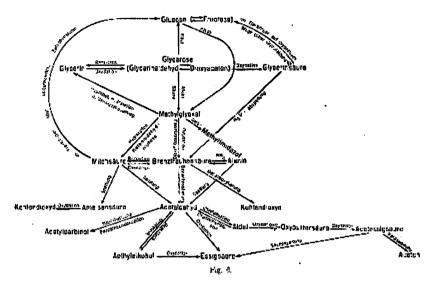


Fig. 16 The C. Neuberg scheme of the glucose fermentation, 1911. In: H. Euler & P. Lindner, *Chemie der Hefe und der alkoholischen Gärung*, Leipzig, 1915.

So leucine results in iso amyl alcohol, isoleucine gives the optic active amyl alcohol and valine gives isobutanol. These kinds of alcohol can be oxidised to acids which will form fats when combined with glycerol. Thus Ehrlich demonstrated already in 1905 the connection between carbohydrate, protein and fat metabolism.⁶²

Up to this day yeast (or is it because of the alcohol?) is still a favourite subject of many researchers. So it is no mere coincidence that *Saccharomyces cerevisiae* was one of the first micro-organisms of which the entire genetic information was listed. The complete DNA sequence of this yeast was deciphered in an international project. On one of the sites of the Max-Planck-Gesellschaft many results are summarised .(<u>http://speedy.mips.biochem.mpg.de/mips/yeast/</u>).

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Notes

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