

Supplementary Material

for the

Letter to the Editor

on

On the Pelletier and Caventou (1817, 1818) papers on chlorophyll and beyond

by

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The first paper by Pelletier and Caventou (1817) is the longer of the two, and a translation into English is provided here. The words within [square brackets] are from the original text, for which notes by the authors are appended following the translation. *Italicized* words in parentheses have been added by the authors in an effort to improve clarity. A few instances of punctuation in the original text (use of colons and semi-colons) have been altered for clarity in the translation.

On the green matter of leaves

by Mr. Pelletier and Mr. Caventou

The substance responsible for the green color of the leaves of trees and herbaceous plants is not well known: no expert work [“travail *ex-professo*”]¹ has been undertaken on this subject; and this green matter, also referred to as *resin* or *green starch* in some plant analyses, has not yet been classified among the substances [“matériaux immédiats”]² of the plants, and is not the subject of any chapter in the most modern chemistry treatises. However, considering the role it plays in plant function [“l’économie végétale”]³, its abundance in nature, its interesting properties already recognized by several chemists who have performed plant analyses, particularly by Mr. Vauquelin, should have (*already*) led to a more specific research on this substance. We do not intend to fill the lacuna we have observed; but by reviewing the already known facts, and reporting several of our own observations, we bring together some material for the history of this remarkable substance.

Initially, we have attempted to obtain the green material in its pure form. To achieve this, we have treated with distilled alcohol, at room temperature, the well-pressed and well-washed pulp

[“marc”]⁴ of several herbaceous plants. The filtered alcoholic liquid was of a beautiful green color; and, after a controlled evaporation, it provided a dark green substance of a resinous aspect. This material, when powdered and treated with hot water, had acquired a high degree of purity, yielding a little bit of brown colored extract. A small amount of the green matter also dissolved in boiling water, but it separated partly from it upon cooling. The green matter, thus obtained, showed all its already known properties; it completely dissolved in alcohol, ether, and oils, while chlorine immediately destroyed its green color.

When exposed to air for several weeks, the green matter did not undergo any alteration at all: it produced alcohol-based dyes that were as colorful as when freshly prepared. When heated, the sample softened, but it did not melt; and if the temperature was increased, the sample decomposed and produced water, oil, a little of acetic acid, and ethylene, but no trace of ammonia was found.

When exposed to a candle flame, a dried fragment of the green matter ignited; it then continued to burn by itself with a less elongated flame than that of resins. The sample yielded a charred residue, which retained the shape of the original fragment, and was partially incinerated in the atmospheric air.

The action of acids on the green matter is quite remarkable. Cold concentrated sulfuric acid dissolves it without altering it; and this acid, when mixed in equal parts with an alcoholic solution of the green matter, does not change it at all. When the green matter is dissolved in sulfuric acid, it becomes cloudy; and when water is added to it, it loses a fraction of the coloring matter, although a notable amount remains in the solution, which can be extracted by saturating the acid with an alkali or an alkaline carbonate.

The ability of the green matter to be dissolved in sulfuric acid, without being altered, would seem to it being related to that of indigo. However, the experiments we have conducted to convert these two substances into each other were unfruitful; we do not believe that it is necessary to report them.

Hydrochloric acid significantly alters the green matter, giving it a yellowish tinge that is not lost anymore.

Nitric acid acts energetically on this substance and in a very particular manner. It first destroys its green color, replacing it with a greyish yellow hue; there is release of nitrous acid, and the matter disappears almost entirely, by dissolving in the acid, especially when heated. As a last

result, we obtained a matter having a dirty white color, which has no taste, nor any smell; it is soluble in concentrated nitric acid, but insoluble in alkalis and in water—it did not yield any trace of oxalic acid, nor mucic acid. We intend to return to these unique results.⁵

Chlorine destroys most rapidly the green color of the substance of this matter: it separates it from its solvent in the form of a fluffy, yellow material, which no longer has any relation to the substance from which it originated. This fact had been (*already*) observed by M. Proust (*Journal de Physique*)⁶.

Iodine acts in an analogous manner to that of chlorine; but its action is extremely slow and insensitive in its first moments.

The action of the alkalis on the green matter is partly known already: it is said that alkaline solutions dissolve it without altering it, and that they even seem to revivify its color. If one saturates the alkali with a weak acid, the green matter is partly precipitated, without any alteration.

Neutral salts have no action when cold on the green matter, but the muriate of tin⁷ nevertheless makes a light precipitate; however, if after adding an earthy or metallic salt in an alcoholic solution of a green matter diluted with water, an alkaline carbonate is poured into it, there is an abundant precipitate of the base, which, in most cases, entrains the green matter into a composite [*l'état de combinaison*]⁸. In this way, with this substance extracted from different plants and salts of lime, alumina, magnesia, lead and tin, we have prepared green lacquers of various shades, depending upon the plant and the salt used.

We also succeeded, and with much less cost, to prepare these lacquers⁹ by adding to the juice of the plants, obtained simply by pressing them out extensively, an earthy salt that is decomposed by an alkali or an alkaline carbonate [*sous-carbonate alcalin*]¹⁰. We have prepared by this procedure more than twenty different colored lacquers, depending on the species of the plant. We notice here that the same plant, under the same circumstances, always provides the same shade or color of the lacquer.

Most of these lacquers, prepared several weeks ago, have not suffered any change due to light; however, the green matter extracted from resinous trees, such as pine or fir, gave lacquers whose color changed. Was this phenomenon due, as we assume, to a little bit of resin that may have remained mixed with the green matter, and that is very difficult to be removed completely?

We have attempted to apply these ground lacquers on paper with glue; we have had wallpapers whose color has not yet faded. In preparing these lacquers, we have noticed that those

made with the same vegetable and various bases, were all the more green, the more alkaline the base was; thus, the lacquers made with lime are generally more beautiful than those obtained by magnesia and alumina. We have no doubt that it may be possible, in the domain of arts, to take great advantage of these lacquers, and substitute them in certain cases for *Scheele green*¹¹, an expensive and above all dangerous preparation.

Among the lacquers prepared this way, we have remarked those provided by the common grass of the meadows, which, as is known, is almost entirely composed of various grasses; those provided by hemlock and other (*members of the*) Umbelliferae (*family*); the elderberry also gives a very beautiful one (*lacquer*); the vine gives a very remarkable canary yellow lacquer; (*and*) the alfalfa produces a very light green (*lacquer*).

We intend, in a more favorable season for vegetation, to prepare a greater number of such samples, and to continue with a little more success our research on ways to combine the green matter with fabrics of plant and animal origin; (*this will be*) an advantage of the most importance for dyeing, which we do not presume impossible to achieve.

We have also studied the possible action on the green matter of plant substances that can be considered as chemical agents. We are assured that, among the plant acids, only acetic acid dissolves it in a remarkable manner; water cannot precipitate it from its solution; it is soluble in sulfuric and acetic ethers; non-volatile oils also dissolve it; the action of volatile oils is less marked; (*and*) we finally know that it dissolves in fats.

From the facts contained in this Note, it follows that the green matter of plants, improperly called *starch* or *resin*, is a particular substance that must be classified among the highly hydrogenated vegetable substances, which should be separated from the resins; it is closer to several coloring agents, such as those of alkanet (*roots*), turmeric, red sandalwood; and it deserves, by its properties and the role it plays in plant function [*“l'économie végétale”*]³, to be considered as a constituent substance [*“principe immédiat”*]¹² of plants: it would be then necessary to be designated by a specific name.

We do not have any right to name a substance known for a long time, and to the history of which we have added only a few facts; however, we will propose, without attaching any importance to it, the name of chlorophyll [*“chlorophile”*]¹³, from chloros, color, and φυλλον (*phyllon*), leaf: this name would indicate the role it plays in nature.

As for the advantages that could be obtained from lacquers, of which we have prepared several samples, only time and usage will tell.

Comments and explanations pertaining to the superscripts ¹⁻¹² in the above translation. The following are provided to enhance clarity, particularly concerning several terms from 19th century French chemistry. The References cited are listed in the body of the paper or at the end of this section.

¹ “**travail *ex-professo***” – translated here as “expert work” although it is not clear if the Latin “*ex-professo*” was meant to convey additional or alternate meaning.

² “**matériaux immédiats**” – translated here simply as substance, but in so doing obscuring a deep intellectual struggle of the early 19th century. As recounted by Tomic (2010, 2012), Fourcroy proposed this term in 1800 to classify chemical substances by merging the long thread of natural history with emerging concepts in chemistry, but in doing so any such substance would confusingly have both a taxonomic classification and a chemical description. In the early 1820s, Chevreul swept away “matériaux immédiats” and defined the term “organic chemical species” as “a collection of entities identical in their nature, proportion and arrangement of their elements,” which in fact was equivalent to the term “principe immédiat” and in its atomistic sense provided a clean break from a natural history classification.

³ “**l'économie végétale**” – a commonly used term in the scientific literature of the late 18th century (Sigaud de la Fond 1781; Sigaud de la Fond, 1785; Mustel 1781) to refer to ‘the principles of vegetation, growth and development of plants, the organization of plants as much as vital activities: sprouting, root suction, and circulation of sap’ (Oghina-Pavie 2011, referring to du Monceau 1758) and hence is translated here as “plant function.” The term “végétale” was synonymous with “plant” at the time (Mustel 1781). An equivalent modern translation could be “plant physiology” but given that the word “physiologie” was known but not employed by Pelletier and Caventou, and 18th century reports spoke of “l'économie animale & végétale” (Mustel 1781), the translation “plant function” is used here. It warrants comment that the term “l'économie végétale” finds modern usage in the field of archeobotany, but to refer to all aspects of plant utilization (collection, cultivation, consumption, procurement) in pre-modern societies (e.g., Tengberg and Lombard 2001).

⁴ “**marc**” – the scientific term appears to be “pomace,” but because this is likely unknown outside of a specialized audience, the similar more common term “pulp” is used here and in the translation of the 1818 paper.

⁵ It is unknown to us if Pelletier and Caventou did so.

⁶ We are unsure of this reference.

⁷ Muriate of tin is used as a mordant in the generation of scarlet dyes (Berard 1810).

⁸ **“l'état de combinaison”** – translated here simply as a composite. A likely interpretation is that the chlorophyll from solution inevitably becomes entrapped with the voluminous precipitate to afford a green solid material, forming an inorganic–organic composite. The same term appears in the 1818 paper.

⁹ A lacquer is a liquid made of shellac dissolved in alcohol or of synthetic substances that dries to form a hard protective coating for e.g., wood or metal.

¹⁰ **“sous-carbonate alcalin”** – an alkaline salt of carbonate (CO_3^{2-}), perhaps Na_2CO_3 .

¹¹ *Scheele green* – a mixture of cupric hydrogen arsenites (e.g., CuHAsO_3) that served as a yellow-green pigment in the 19th century but fell out of favor due to toxicity concerns.

¹² **“principe immédiat”** – as described in the main text, a substance pre-existing in a natural source that is obtained by direct isolation without alteration and for which the intrinsic features are inseparable from its physiological function (Tomic 2010, 2012).

¹³ **“chlorophile”** – the term used in the 1818 paper was “chlorophyle” whereas the French spelling in modern usage is “chlorophylle.”

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