

# UNITED STATES PATENT OFFICE.

JULIUS HERBABNY, OF OFFENBACH-ON-THE-MAIN, AND CHRISTOPH HARTMANN, OF FRANKFORT-ON-THE-MAIN, GERMANY.

## BLACK TETRAZO DYE.

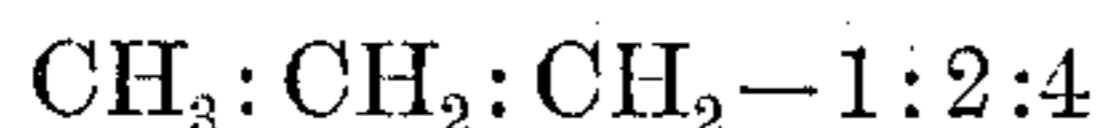
SPECIFICATION forming part of Letters Patent No. 639,041, dated December 12, 1899.

Application filed September 28, 1898. Serial No. 692,075. (Specimens.)

*To all whom it may concern:*

Be it known that we, JULIUS HERBABNY, doctor of philosophy, residing at Offenbach-on-the-Main, and CHRISTOPH HARTMANN, doctor of philosophy, residing at Frankfort-on-the-Main, Germany, have invented a new Black Tetrazo Dyestuff, of which the following is a specification.

The present invention relates to the manufacture of substantive poly-azo coloring-matters suitable for dyeing unmordanted cotton; and it consists in the reduction of the disazo dyestuff which results from the combination of a meta-diamin of the benzene series, such as meta-toluylenediamin



or meta phenylenediamin, respectively, with two molecules of para-nitro-diazobenzene, converting the thus-obtained tetra-amidodisazo body by sodium nitrite into a tetrazo compound, and, finally, combining the latter with two molecules of a suitable component.

The following example will illustrate the manner in which our invention may be carried out and brought into practical effect. The parts are, by weight:

Combine in a weakly soda-alkaline solution twelve and one-fifth (12.2) parts of toluylenediamin with twenty-seven and three-fifths (27.6) parts of paranitranilin diazolized in the usual way with thirteen and four-fifths (13.8) parts of sodium nitrite. The precipitated dyestuff may either be isolated or directly reduced by sodium sulfid. In the latter case proceed as follows: mix sixty-three (63) parts of the dinitrodisazo dyestuff with one hundred and eighty (180) parts of sodium sulfid dis-

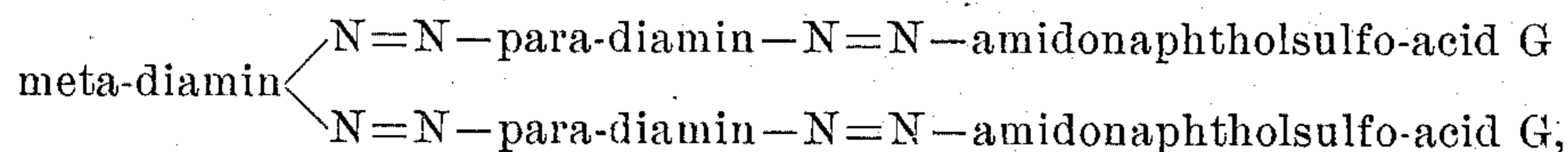
solved in eighty (80) parts of water and add, to quicken the reaction, two hundred and forty (240) parts of spirit and heat the whole to fifty to eighty degrees centigrade (50° to 80° C.) during four to six hours. The addition of spirit only serves to facilitate the reaction, and therefore is not absolutely necessary. After the reduction is finished distil off the spirit, dissolve the residue in diluted hydrochloric acid, filter, and precipitate with alkalis. Then tetrazolize while cooling in hydrochloric-acid solution eighteen (18) parts of the obtained tetraamidodisazo compound with six and nine-tenths (6.9) parts of sodium nitrite and pour the whole into a solution of twenty-three and nine-tenths (23.9) parts of amidonaphtholsulfo-acid G, keeping alkaline by means of soda throughout the operation, and precipitate with common salt.

The dyestuff thus obtained forms a black powder with a metallic luster insoluble in cold, but easily soluble in hot water with a violet shade, which turns reddish on adding soda, whereas on adding acids the dyestuff falls down as a violet precipitate. Its solution in concentrated sulfuric acid is pure blue. It dyes unmordanted cotton from acid or neutral baths deep black.

In the above example the meta-toluylenediamin may be replaced by the equivalent quantity of meta-phenylenediamin.

Now what we claim is—

As a new article of manufacture, the black coloring-matter which is obtained by combining the tetraamido disazo-dyestuff derived from a metadiamin of the benzene series and para-nitro diazo benzene chlorid with amidonaphtholsulfo-acid G, which has the general formula



is a black powder with a metallic luster, insoluble in cold but easily soluble in hot water with a violet shade which turns reddish on addition of soda, forms a violet precipitate by the addition of acids to its aqueous solution, produces a pure-blue solution in concentrated sulfuric acid, and which dyes unmordanted cotton from alkaline or neutral baths a deep black which is very fast to washing, substantially as described.

In testimony whereof we have signed our names to this specification in the presence of two subscribing witnesses.

JULIUS HERBABNY.  
CHRISTOPH HARTMANN.

Witnesses:

DEAN B. MASON,  
JEAN GRUND.