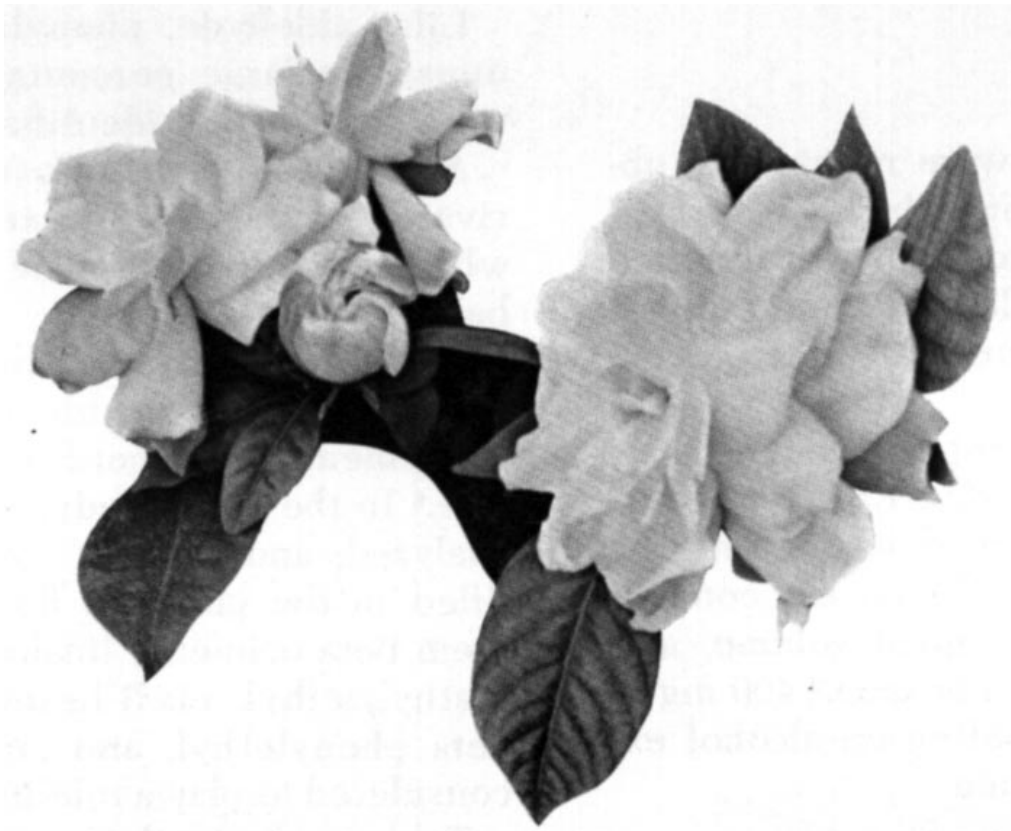

Gardenia in Perfumery

By Danute Pajaujis Anonis, Consulting Chemist Perfumer, Forest Hills, NY

Named after Dr. Alexander Garden, an 18th century Scottish naturalist, the gardenia is a latecomer to perfumery, following jasmin, rose, and violet.[1] Gardenia flower oil was used in fragrances in the early part of the 20th century. Today gardenia flower oil is scarcely produced, and synthetic compounds are used more often.

Botanical Origin

Many species of gardenias grow in various parts of the world, particularly in the Far East. It was reported that gardenias grow well in the Chungking region in China, where tuberose and osmanthus are also cultivated.[2] In Japan, *Gardenia jasminoides* is widely distributed. *Gardenia grandiflora Lour* and *Gardenia florida L.* (fam. Rubiaceae) originated in equatorial and South Africa.

Mode of Production, Yield, and Type of Oil

In the beginning of the century, gardenia was cultivated in Reunion Island. As in the case of other delicate flowers, steam distillation of gardenia flowers does not give any results. In the past enfleurage was used, and on extraction with alcohol numbered infusions were obtained,[3] giving on evaporation of alcohol concentrated extracts of pomade lavages. A more modern method of extraction was the use of volatile solvents, resulting in concrete oils and, on treatment with alcohol, absolutes.

Using petroleum ether as solvent, 3000 to

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4000 kilograms of flowers were required to obtain one kilogram of concrete which yielded 0.5 kg of the absolute. Naves and Mazuyer reported that 2 to 2.6 metric tons of flowers were required to obtain above quoted amounts of concrete and absolute oils.[4]

In more recent experimental research work, 2.5 kg of *Gardenia jasminoides, Ellis var. Hara* flower petals were immersed in acetone for three days, the extract was filtered and concentrated to one-tenth of its original volume, and then further extracted with n-hexane.[5] 400 mg of concrete was obtained, yielding on alcohol extraction 250 mg of the absolute.

Composition

In the first decade of the century, the following components of natural gardenia flower oil obtained by maceration in paraffin were known:[6]

Benzyl acetate
Linalool
Linalyl acetate
Methyl anthranilate
Styrallyl acetate (methyl phenyl carbinyl acetate)
Terpineol

Benzyl acetate was found to be present in the largest amount, but it was styrallyl acetate which was most suggestive of gardenia odor.

In 1924, additional components were known:[7]

Acetic acid
Benzoic acid
Lactone ($C_{10}H_{16}O_2$)
Tiglic acid

More recent research was done in Japan on *Gardenia jasminoides Ellis var. Hara* using a combined GC/MS, gas chromatography, IR and NMR. In one study, 130 components were identified in gardenia absolute.[8] No nitrogen-containing components were included because the basic fraction was too small for analysis of such compounds.

The following compounds were found to be the basic constituents for the gardenia odor: benzyl, cinnamyl, cis-3-hexenyl, and 2-phenylethyl tiglates, cis-3-hexenol and its esters with C_{1-6} aliphatic and aromatic acids, linalool oxides, rose oxides, and four isomers of lilac alcohol (found in nature only in lilac flowers).

Lilac aldehyde, phenylacetaldehyde, and an unusually large percentage of jasmin lactone were among other identified components.

Among the more unusual cis-3-hexenol derivatives, was cis-3-hexenyl cis-3-hexenoate which was reported to be of a powerful green-bamboo odor.

In the second study, alpha farnesene and cis-jasmin lactone were identified, and sixty other components were confirmed in gardenia absolute.[9] In the same study, the headspace gas was analyzed, and seventy components were identified in the gardenia flower volatiles. Among them beta ocimene, linalool, cis-jasmin lactone, methyl, ethyl, cis-3-hexenyl, n-hexyl, benzyl, beta phenylethyl, and cinnamyl tiglates were considered to play a role in the gardenia odor.

Table I shows the main components of gardenia absolute and the headspace gas identified in the first and second studies.

Table I. Main Components of Gardenia Flower Oil

	Study 1 (Reference 8)	Study 2 (Reference 9)	
Identified Components	Gardenia Absolute	Gardenia Absolute II	Headspace Gas
	%	%	%
alpha-Farnesene	18.0	12.1	2.2
Linalool	12.0	7.5	24.6
cis-3-Hexenyl tiglate	10.0	3.1	3.1
cis-Jasmin lactone	8.0	17.0	tr
cis-3-Hexenyl benzoate	5.0	4.6	-
beta Ocimene	1.4(trans)	0.2	45.8
cis-3-Hexenol	1.0	0.2	0.1
Methyl benzoate	1.0	0.8	6.4
Hexyl tiglate	1.0	0.5	1.7

The comparison of components in both studies in Table I shows that alpha farnesene, linalool, cis-3-hexenyl tiglate, and cis-jasmin lactone are the main ingredients of gardenia absolute, but their proportion is different.

Beta ocimene and linalool are shown to be the main components of the headspace gas.

In reference to Table I, an observation may be made regarding benzoates. In the headspace gas

a significant amount of methyl benzoate is present, while in the absolute comparable amounts of cis-hexenyl benzoate are found. It may be pointed out that styrallyl acetate (methyl phenyl carbinyl acetate), which was found in the early part of the century, mentioned before, in gardenia absolute obtained by maceration, was not identified in the headspace gas or gardenia absolute in either of the above studies.

Synthetic Compounds

In the past, styrallyl acetate combined with hydroxy-citronellal and small amounts of alcohol C-9, or phenyl ethyl heptylate was considered as a base for gardenia.[10] In some cases, styrallyl acetate was combined with dimethyl benzyl carbinyl acetate.

Styrallyl acetate remains an important ingredient of synthetic gardenia compounds. Basically, styrallyl acetate combined with jasmin, rose, lily of the valley, lilac, and a small percentage of orange flower or their components will produce gardenia odor characteristics. Sometimes woody notes and methyl ionone are added. Aryl carbinols or salicylates are used to round out the harshness of styrallyl acetate. Linalyl benzoate and phenylethyl anthranilate are sometimes included in the formula, and aldehyde C-14 and C-18 are added as modifiers. Aldehydes C-8 to C-12 are used for the top notes, as are citrus oils, bergamot, orange, lemon and mandarin. Among conventional fixatives are heliotropin, coumarin, musk ketone, cinnamic alcohol, labdanum, myrrh and tolu resinoids. The new synthetic ambergris compounds are valuable modern fixatives which enhance the floral-green notes.

Some gardenia compounds of the past were built on a tuberose note; they did not contain any styrallyl acetate. Aldehyde C-18 (nonalactone) was used in such types which were suitable as cream fragrances.

Let us look at a few conventional illustrative formulas:

Gardenia G-2 (with a tuberose note)

350	Amyl cinnamic aldehyde
200	Hydroxycitronellal
200	Benzyl alcohol
160	Benzyl salicylate
160	Aldehyde C-18
120	Aldehyde C-14 pure
110	Linalool
100	Benzyl acetate
90	Methyl salicylate
70	Terpineol
60	Linalyl acetate
60	Petitgrain
60	Geraniol
50	Musk ketone
42	Tolu resinoid
30	Phenyl ethyl alcohol
22	Aurantiol
10	Isoeugenol
5	Ethyl vanillin 10%

Another gardenia type, containing styrallyl acetate:

Gardenia G-3

200	Benzyl acetate
80	Amyl cinnamic aldehyde
80	Phenyl ethyl alcohol
70	Ylang ylang
50	Linalyl acetate
50	Cyclamen aldehyde
50	Styrallyl acetate
40	Terpineol
40	Musk ketone
40	Orange sweet
40	Cinnamic alcohol
30	Ethyl vanillin 10%
30	Isoeugenol
20	Methyl anthranilate
20	Linalool
20	Anisic alcohol
20	Tolu resinoid
15	Aldehyde C-8 1%
10	Indol 10%

Besides older established aromatics, modern gardenia compounds may include more recent synthetics. Among the former, isoamyl benyzl ether, also called "Gardenia oxide," of a mild floral odor, blends with the harsher notes of aldehydes in a manner similar to the carbinols.[11] Phenyl ethyl cinnamate, anthranilate, and salicylate are useful components in gardenia. Linalyl isovalerate contributes a citrus-fruity top note. Methyl octine carbonate used previously in some gardenia formulas can now be replaced with the more modern nonadienol and its derivatives.

Hydroxycitronellal is now mostly replaced by more stable cyclamen aldehyde derivatives, or other hydroxycitronellal substitutes. Cinnamic alcohol and isoeugenol are used in limited amounts because of dermatological considerations.

Among newer aromatics, 2-ethyl hexanal cyclo

glycol acetate of a strong green-tart odor, linalool oxide, rose oxides, hexenol and its esters, especially tiglates, benzoates, and caproates, and cis-jasmin lactone may be mentioned. Methyl phenyl ethyl ether of a diffusive jasmin-rose note is effective in traces.

Application

In the beginning of the century, Chanel introduced a perfume named Gardenia Chanel, but it did not prove successful. Pungent florals came into vogue in the 30s, and several gardenia fragrances were created. Among them, Jungle Gardenia was a success, and it remains on the market. Gardenia was also used as a lotion fragrance in the past. In today's perfumer's workshop, a gardenia fragrance is offered among other single flower perfumes.

A study made by the author in an environmental exhibit in the Museum of Crafts in New York showed that a gardenia odor elicited a highly favorable response in both under 25 and 25-45 age female and male groups.[12] The 25-45 female age group showed a marked preference for gardenia over rose, lily of the valley, or orange blossom.

Although gardenia is considered a secondary floral in perfumery, it plays an important role in fragrances as a modifier. According to a noted French perfumer, the original Chanel No. 5 perfume contained a gardenia note.[13]

Coty's l'Aimant contained styrallyl acetate used in jasmin, cassie and tuberose infusions when the perfume was first created.

The addition of gardenia to chypre resulted in Crepe de Chine. A further development was Ma Griffe, an aldehydic chypre with a gardenia note.

Among other established and more recent perfumes containing gardenia or its components are: Aphrodisia, l'Air du Temps, Shocking, Vent Vert, Cabochard, Detchema, Charlie, Calèche, Aliage, Michelle, Adolfo, Tatiana, and Première.

In the 70s, the gardenia note came to prominence, and it was used to advantage in modern green and aldehydic fragrances.

Gardenia-carnation-lavender are considered osmically balanced floral components.[14]

Absolute gardenia was used in the past in luxury fragrances. It was also used as a modifier in rose and other floral bouquets.

Gardenia has been used in brilliantines, creams, lipsticks, powder, and soap. Today, gardenia compounds are also used in cosmetic, toiletry, and soap fragrances, but mostly as components of a perfume adapted to the line. In powder fragrances, phenyl ethyl cinnamate is used to advantage in gardenia, as it imparts sweetness and is a good fixative.

A modern source describes the use of homogenized gardenia oil in non liquid air-freshening compositions, using "ultra-amylopectin (I) or starch carboxy methyl ether in ethyl alcohol," the mixture being further homogenized with wax or waxlike materials or paraffin and soaked in polyurethane foam.[15]

Conclusion

Gardenia compounds are being modernized by the application of new aromatics, and by replacing dermatologically unacceptable components.

Fragrances have become more sophisticated, and today very few single floral gardenia perfumes are created.

Gardenia plays an important role as a fragrance modifier both in traditional and modern fragrances, and it will remain a valuable floral-green note in future fragrances.

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