**New aspect of gallic esters: marked effect on termite feeding**

Shunichi Kubota†, Tatsuya Mori*, Noritada Matsuo and Yoshinori Shono††

Agricultural Chemicals Research Laboratory, Sumitomo Chemical Co., Ltd., 2–1 Takatsukasa 4, Takarazuka, Hyogo 665–8555, Japan

Environmental Health Division, Sumitomo Chemical Co., Ltd., 5–33, Kitahama 4, Chuo-ku, Osaka 541–8550, Japan

†† Vector Control Division, Sumitomo Chemical Co., Ltd., 27–1, Shinkawa 2, Chuo-ku, Tokyo 104–8260, Japan

(Received June 28, 2010; Accepted October 1, 2010)

Some gallic esters have interesting effects on the feeding activity of Formosan subterranean termites (*Coptotermes formosanus* Shiraki). The feeding response of termites varied markedly according to the numbers of carbon in alkyl chain.

In particular, 1-decyl gallate exhibited the highest feeding deterrence, whereas 1-octadecyl gallate exhibited the highest feeding-stimulating activity. © Pesticide Science Society of Japan

**Keywords:**  gallic ester, termite, *Coptotermes formosanus*, feeding.

---

**Introduction**

Subterranean termites are serious pests to wooden materials in structures and it costs two billion dollars annually to prevent and control termites in the United States.3) Chemical control has been widely accepted as a long-lasting and cost effective measure, and various approaches, such as treatment with liquid termiticides and baiting are applied according to the purpose (prevention or remedy) and situation (inside or outside of a structure). One of those applications is to treat wooden materials with chemicals which repel termites or have adverse effects on them.

Some plant species are naturally resistant to termite feeding, such as Japanese cypress.2) Such tolerance is often attributed to the toxicity of secondary metabolites. Some natural products are known to have toxic effects on termites and often repel them.3–5) We found that some gallic esters deterred the feeding of termites. Gallic acid was originally found in plant gall and some gallic esters have been commercialized as food additives.6) Gallic esters are known to have various bioactivities, including anti-oxidative, anti-browning, anti-bacterial, anti-fungal activities.7–13)

We herein report that their effects on termite feeding are altered by carbon chain lengths.

**Materials and Methods**

### 1. Preparation of compounds

The melting points (mp) were measured on Yanagimoto micro melting point apparatus and are uncorrected. IR spectra were obtained with a HORIBA FT-720 spectrometer.1) 1H-NMR spectra were obtained with a JEOL EX-300 spectrometer (300 MHz) or JEOL AL400 spectrometer (400 MHz). Chemical shifts were recorded in δ (ppm) and the coupling constant J in Hz.

The preparation of (4) is typical of the procedure used.14,15) A mixture of 3,4,5-trihydroxy benzoic acid monohydrate (1.0 g, 5.3 mmol), 1-decyalcohol (0.84 g, 5.3 mmol) and p-toluenesulfonic acid monohydrate (0.1 g, 0.58 mmol) was heated at 150°C for 2 hr with continuous removal of water. The cooled reaction mixture was poured into sodium hydrogen carbonate solution and extracted with ethyl acetate. The organic layer was washed with saturated sodium chloride solution and then dried over anhydrous sodium sulfate. Evaporation of the solvent gave a residue, which was chromatographed on silica gel (Chloroform/Methanol: 100/1) to give 1-decyl gallate (4) as colorless crystal. Yield: 1.4 g, (82%). mp: 96–97°C. 1H-NMR δH (DMSO-d6): 0.85 (3H, t, J = 6.3 Hz), 1.25 (14H, br), 1.63 (2H, m), 4.14 (2H, t, J = 6.5 Hz), 6.93 (2H, s). IR νmax cm⁻¹: 3448, 3234, 2915, 2829, 1652, 1610, 1540, 1446, 1396, 1315, 1232, 1018.

1-Propyl gallate (1). mp: 147–148°C. 1H-NMR δH (DMSO-d6): 0.95 (3H, t, J = 7.3 Hz), 1.67 (2H, m), 4.11 (2H, q, J = 6.6 Hz), 6.94 (2H, s). IR νmax cm⁻¹: 3500, 3453, 2967, 1687, 1614, 1538, 1465, 1405, 1301, 1240, 1193, 1033.

1-Hexyl gallate (2). mp: 92–93°C. 1H-NMR δH (DMSO-d6): 0.86 (3H, t, J = 6.2 Hz), 1.26 (6H, br), 1.63 (2H, m), 4.14 (2H, t, J = 6.5 Hz), 6.93 (2H, s). IR νmax cm⁻¹: 3459, 3338, 2829, 2586, 1683, 1610, 1535, 1446, 1396, 1311, 1238, 1099, 1024.

1-Octyl gallate (3). mp: 96–97°C. 1H-NMR δH (DMSO-d6): 0.83 (3H, t, J = 6.1 Hz), 1.24 (10H, br), 1.63 (2H, m), 4.13 (2H, t, J = 6.5 Hz), 6.94 (2H, s). IR νmax cm⁻¹: 3446, 3340, 2915, 2850, 1666, 1608, 1533, 1465, 1375, 1303, 1241, 1101, 1024.

1-Dodecyl gallate (5). mp: 95–96°C. 1H-NMR δH (DMSO-d6): 0.85 (3H, t, J = 6.2 Hz), 1.22 (18H, br), 1.64 (2H, m), 4.15 (2H, t, J = 6.5 Hz), 6.97 (2H, s). IR νmax cm⁻¹: 3466, 3346, 2913, 2846, 2818, 1666, 1608, 1531, 1465, 1409, 1378, 1299, 1255, 1195, 1029.

1-Tetradecyl gallate (6). mp: 95–96°C. 1H-NMR δH (DMSO-d6): 0.85 (3H, t, J = 6.7 Hz), 1.23 (26H, br), 1.62 (2H, m), 4.14 (2H, t, J = 6.5 Hz), 6.93 (2H, s). IR νmax cm⁻¹: 3446, 3342, 2915, 2848, 1683, 1608, 1533, 1465, 1409, 1378, 1303, 1257, 1195, 1029.

---

**Note**

* To whom correspondence should be addressed.

E-mail: mori7@sc.sumitomo-chem.co.jp

Published online November 10, 2010

© Pesticide Science Society of Japan
Bioassays

2.1. Insects

Undifferentiated larvae (workers) of C. formosanus were obtained from a laboratory colony maintained at the Agricultural Chemicals Research Laboratory of Sumitomo Chemical Co., Ltd. (Hyogo, Japan).

2.2. No-choice feeding test

Filter paper disks treated with 0.5% (wt/wt) of each compound were air-dried and weighed. Each disk was placed in a small plastic cup (ca. 14 ml) with small entry holes to allow termite access. Plastic cups were then placed in separate 200 ml plastic containers, each holding 100 termite workers. The bottom of this larger container was covered with 2–3 mm of plaster and several small holes were made in the base. Assembled units were placed on a damp cotton pad in an incubation chamber so that termites could take up water through the plaster. The units were maintained at 25±2°C in the dark for 1 week. After the 1-week no-choice test, each disk was cleaned, air-dried and then weighed to calculate the amount consumed by termites. Three replications were made for each compound and untreated control.

Results and Discussion

The feeding response of termites varied markedly according to the numbers of carbons in the alkyl chain (Fig. 2). Compound (3), (4) and (5) significantly deterred termite feeding. Anti-feeding efficacy was obtained but was weak in Compound (1), (2) and (6), which indicates that 8–12 carbons in the alkyl chain provide the strongest anti-feeding efficacy against termites. (6) An unpublished study showed that 0.05% (wt/wt) of Compound (4), which showed the strongest activity, reduced termite feeding by approximately 50%; therefore, the IC50 of Compound (4) should be close to 0.05%. The anti-bacterial efficacy of alkyl gallates against Bacillus subtilis was strongest with 8–11 carbons in the alkyl chain, and their effect was considered to come from inhibition of the respiratory system. (13) The anti-fungal efficacy of alkyl gallates against Saccharomyces cerevisiae was strongest with 8 and 9 carbons in the alkyl chain, and their effect was considered to come from their action as a surfactant. (11) Such bio-activities might have adverse effects on termites or deter termites as they recognize them as toxic materials. It remains unclear as to whether feeding inhibition is caused by the adverse effects of the chemicals or the intrinsic repellency of the chemicals. Topical application of a high dose of Compound (4) showed acute toxicity to termites; (16) however, natural products generally repel termites at much lower doses than those shown to be fatal to them. (3–5)

In contrast, Compound (9) showed a feeding-stimulating effect on the termites. The amounts fed by termites significantly increased in the filter paper disks treated with compound (9). Compounds with less than 15 carbons in the alkyl moiety were consumed less than the untreated control; therefore, the flexible chemical structure is extremely small for a feeding-stimulating effect. (17)

One of the possible explanations of the phenomenon is its similarity to chemicals such as known phagostimulants or trail pheromones, which may induce greater consumption of materials treated with them; (18–21) however, such known compounds, for instance hydroquinone and (Z,Z,E)-3,6,8-dodecatrien-1-ol, are not specifically similar to Compound (9). (18,21) This contradicts the much smaller structural flexibility of the phagostimulant described above; therefore, it is unlikely that the compound mimics such phagostimulants and pheromones. Systematic research...
on the structure-activity relationship remains to be conducted. The chemical ecology of termites remains unclear, but extensive research in this field is expected to bring about great progress in better understanding termite ecology and to facilitate efficient termite control.

References