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Particle Films: A New Technology for Agriculture

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I. INTRODUCTION

Scientific evidence that chemically active pesticides are residually present on food, in water supplies, in the soil, and that these chemicals may interfere with animal growth and development, together with the public demand for reduced-risk pesticides, resulted in a Congressional mandate for USDA-ARS to develop reduced risk alternatives to chemical pesticides in 1985 as part of the Low Input Sustainable Agriculture (LISA) program (Jawson and Bull 2002). In the 1980s and 1990s it was clear that new paradigms were needed to control plant pests in an economically sustainable and environmentally safe manner. Particle film technology is a combined synthesis of knowledge on mineral technology, insect behavior, and light physics as they apply to pest control and plant physiology.

Feldspar and quartz are naturally occurring inorganic substances that are referred to as primary minerals. Upon weathering, primary minerals such as feldspar give rise to secondary minerals such as aluminosilicate clays. Current particle film technology is based on kaolin, a white, non-porous, non-swelling, low-abrasive, fine-grained, plate-shaped, aluminosilicate mineral [$Al_4Si_4O_{10}(OH)_8$] that easily disperses in water and is chemically inert over a wide pH range. Water-processed kaolin is >99% pure and has a brightness of >85%. Mined, crude kaolin has traces of Fe_2O_3 and TiO_2 that are removed during processing to increase brightness. In addition, crystalline silica, SiO_2, a respirable human carcinogen, must be removed to insure human safety (Harben 1995). Technical advances in kaolin processing within the past decades have made it possible to produce kaolin particles with specific sizes, shapes, and light reflective properties. Kaolin particles can be engineered with specific properties in paper, paint, cosmetic, and plastic applications. Potential uses of kaolin, however, have been largely ignored by the agricultural industry except for use as carriers for wettable powder formulations of pesticides. Recent advances in kaolin processing, formulating, and plant surface deposition properties have opened new opportunities for its use in agriculture.

An effective particle film on plant tissues has certain characteristics: (1) chemically inert mineral particle, (2) particle diameter < 2 µm, (3) formulated to spread and create a uniform film, (4) porous film that does not interfere with gas exchange from the leaf, (5) transmits photosynthetically active radiation (PAR) but excludes ultraviolet (UV) and infrared (IR) radiation to some degree, (6) alters insect/pathogen behavior on the plant, and (7) can be removed from harvested commodities. Many of these characteristics are similar to natural plant defenses consisting of increasing cuticle thickness and pubescence to reduce water and heat stress (Levitt 1980) and to interfere with disease and insect damage (Barthlott and Neinhuis 1997; Neinhuis and Barthlott 1997). An effective particle film can be applied to a plant surface in such a way that a nearly uniform layer is deposited over the entire plant without blocking stomates (Fig. 1.1A, B, C and Plate I, Top). At the present time, a commercial particle film material, Surround® crop protectant, is being used in about 90% of the Pacific Northwest pear market for the early season control of pear psylla and approximately 20% of the Washington State apple market to reduce sunburn damage. The pears and apples are sold in the fresh food market after being washed in a standard grading line. An effective fruit washing line will utilize a dump tank, often with surfactants added, a minimum of a 10 m bed of brushes, and overhead high-pressure sprayers. Waxing the fruit obscures trace amounts of kaolin residue that did not wash off (pers. observ.). Residue removal from the stem and calyx end of fruit is not easy because it is in a difficult area to clean, but brush and sprayer criteria as described above are effective (Werblow 1999; Heacox 2001).

The purpose of this paper is to bring together the historical and current literature related to the use of particle films in agriculture and to discuss their present and future use in crop protection and production.

II. PARTICLE FILM TECHNOLOGY FOR ARTHROPOD PEST CONTROL

A. Historical Review of Mineral Use in Agriculture for Pest Control

Soil dusts have long been used as insect repellents by primitive people, mammals, and birds that took “dust baths” regularly to ward off biting insects (Ebling 1971). However, recent efforts to control insects mainly

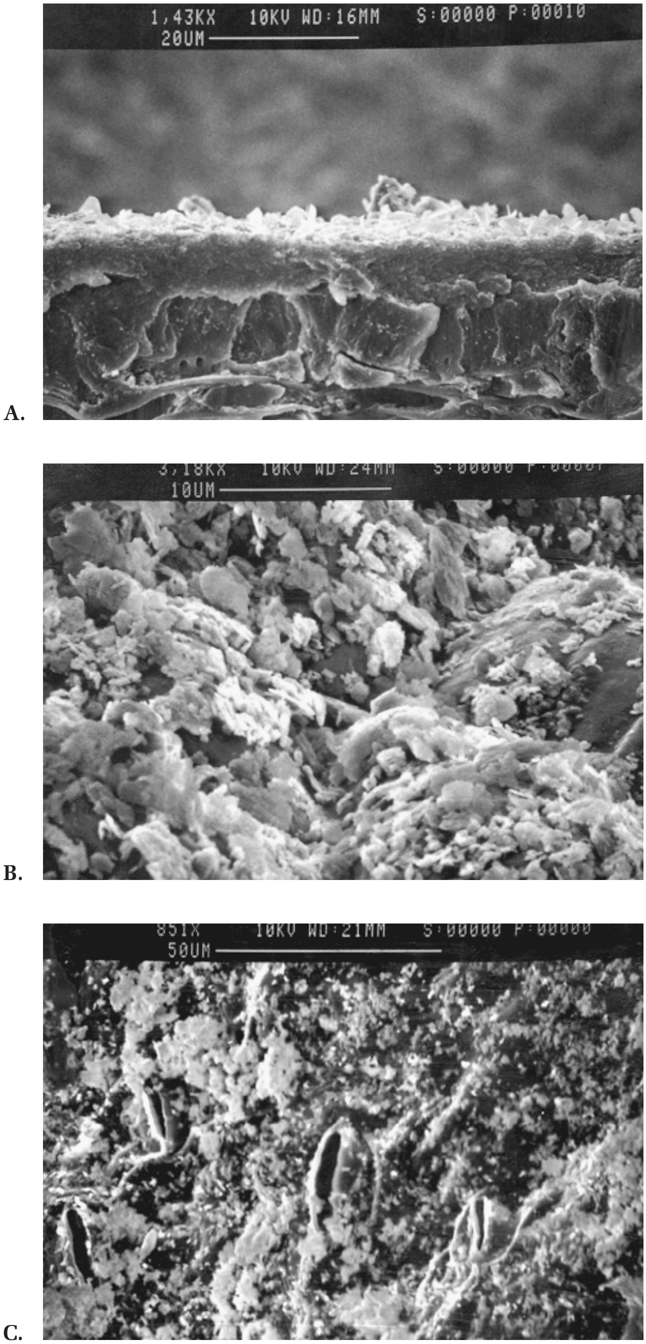

Fig. 1.1.
A. Scanning Electron Micrograph (SEM) of Surround® on a leaf cross-section of apple.
B. SEM of a particle film, Surround, on the upper surface of an apple leaf.
C. SEM of a particle film, Surround, on the lower surface of an apple leaf.

focused on toxic minerals or chemical compounds rather than inert mineral particles. In antiquity, elemental sulfur or sulfur compounds mixed with bitumen were heated to produce fumes that repelled insects from vines and trees (Smith and Secoy 1975, 1976). Diatomaceous earth (diatomite), which originates from fossilized sedimentary deposits of phytoplankton (diatoms), was applied to plants and structures for pest control in China as early as 2000 B.C.E. (Allen 1972). Toxic preparations of arsenic and arsenic salts were used around 900 C.E. in China and incorporated into ant baits in Europe in 1699 (Casida and Quistad 1998). Powdered limestone (calcium carbonate) was added to grain to deter insects in the 1st century. One of the primary insecticides and fungicides of early agriculture, dating to the Hellenistic Era, was the mixture of hydrated lime [$Ca(OH)_2$] with sulfur (S) (Secoy and Smith 1983). Chemically reactive hydrated lime and sulfur were applied independently or together in mixtures with a range of other materials such as tobacco, wood ash, linseed oil, soap, and cow dung. These concoctions were applied as paints or washes to fruit trees and grape vines to protect them from insect and disease damage. From the late 1500s to the 1800s, slaked lime (calcium hydroxide) and burned lime (calcium oxide) were used against household, stored grain, and crop insect pests. Sulfur mixed with limestone was also burned for use as a fumigant for trees in the late 1500s, while lime-sulfur preparations became popular in the latter part of the 18th century. In the 1800s a lime-sulfur combination was developed and replaced the application of the individual minerals. Lime-sulfur, slaked lime, and sulfur were the primary materials used as pesticides in the 1800s because these materials were readily available and easily prepared.

The discovery of the insecticidal properties of the pigment Paris green in 1897 marked the beginning of the modern use of insecticides (Little 1972). The bright green powder, prepared by combining copper acetate and arsenic trioxide to form copper acetoarsenite, was extremely poisonous and had to be made and used with caution. The minerals schultenite (lead arsenate) was first prepared as an insecticide and used against the gypsy moth in 1892 and was a widely used general insecticide for crops up to 1940, when it was replaced with the synthetic insecticide, diclorodiphenyltrichloroethane (DDT) (Peryea 1998).

Inorganic chemists were unknowingly synthesizing chemical compounds such as hexachlorocyclohexane during the early 1800s that were later found to be insecticidal in 1942 (Cassida and Quistad 1998). The discovery of this and other insecticidal compounds such as tetrahethylthiuram disulfide (Guy 1936) and DDT in 1939 (Cassida and Quistad 1998) spurred a major exploration into inert mineral carriers. Lead arsenate, sulfur, nicotine, and hydrated lime, alone or in mixtures, were still the predominant insecticidal materials used in agriculture in the early 1900s. During the first quarter of the 20th century few other insecticidal materials were used and pesticide delivery was also in its infancy. Pesticidal materials were applied as spray solutions using steam- or gas-driven spray gun systems that became available around 1900 (Fronk 1971). The labor involved in spraying orchards and other crops by handgun and using large volumes of water required for acceptable coverage motivated researchers to investigate particle dusts as insecticidal carriers in the early 1900s (Table 1.1).

Dust applications gained favor over liquid sprays in the 1920s because of the speed of dusting operations, economy in labor, good plant coverage, and comparable insect control with liquid sprays (Giddings 1921; Headly 1921; Parrot 1921). Other research that increased interest in using dusts to deliver insecticides proposed that chemically active particles of sodium fluoride and borax (Shafer 1915) and toxin impregnated minerals (Marcovitch 1925; Mote et al. 1926) reacted with the insect cuticle and caused a "self-cleaning" response due to the irritation, and, in the process, insects ingested particles and died. Particle ingestion led to a more rapid killing action by insecticide-laced dusts than by the insecticide (lead arsenate) alone (Mote et al. 1926).

Table 1.1. Examples of minerals used either as insecticide dust carriers or insecticides.

Class of mineral	Subclass	Group	Hardness	Reference
Elemental		Sulfur	2.0	Watkins and Norton 1947
Oxide	Silicon	Quartz	7.0	Alexander et al. 1944b
Carbonate	Calcium	Calcite	3.0	Alexander et al. 1944b
Sulfate	Calcium	Gypsum	2.0	Alexander et al. 1944b
Silicate	Mica	Muscovite, biotite	2.5	Alexander et al. 1944b
	Clays	Talc	1.0	Alexander et al. 1944b
		Pyrophyllite	1.0–1.5	Watkins and Norton 1947
		Montmorillonite	1.2	Watkins and Norton 1947
		Kaolinite	1.5–2.0	Watkins and Norton 1947
		Attapulgite	1.5	Watkins and Norton 1947
		Palygorskite	1.5	Watkins and Norton 1947
Phosphate	Calcium	Apatite	5.0	Watkins and Norton 1947
Organic mineral	Silicone oxide	Diatomite, diatomaceous earth	7.0	Watkins and Norton 1947

Research in the 1930s established that certain "inert dusts" alone had toxic activity against insects when ingested during the process of self-cleaning (Boyce 1932; Richardson and Glover 1932). Suffocation by inhalation was not an important factor, and it was found that the inert dust itself had a desiccating action (Hockenyos 1933). This highly significant observation would later become regarded as one of the major mechanisms of how dusts kill insects. Research on inert mineral dusts (e.g., lime, kaolin) continued to demonstrate that dust had contact toxicity to insects (Maxwell 1937). A number of "so-called inert materials" caused high mortalities of stored grain weevils by desiccation (Chiu 1939a,b). Chiu (1939a,b) summarized the modes-of-action of inert materials as: (1) ingestion of the dust into the digestive system (Boyce 1932; Richardson and Glover 1932), (2) desiccation (Zacker and Kunike 1931; Hockenyos 1933), (3) chemical reaction with the body wall of the insect (Shafer 1915; Makie 1930), and (4) direct mechanical action (Germar 1936). Another important discovery related to mechanisms was that as particle size decreased from 37.0 to 2.9 μm in diameter, insect mortality increased (Chiu 1939a,b). Research in the 1930s brought about the realization that fine mineral dusts were misclassified by insect physiologists and that inert dusts had many unexpected properties in relation to insects (Briscoe 1943). Briscoe (1943) established that mortalities by dust ingestion and suffocation were negligible in grain weevils and that dusts increased water transmission through the insect's cuticle causing desiccation. Alexander et al. (1944a,b) established that the desiccating action of dusts was due to their absorbance of or penetration into the insect epicuticle and that this action was independent of their chemical reactive properties. Insect mortalities increased as particle size decreased and as intrinsic hardness of the materials increased. The mechanisms of how particles caused desiccation of insects was finally attributed to either their adsorption of the epicuticular waxes of the cuticle or abrasion of the cuticle (Kalmus 1944; Wigglesworth 1944). However, if absorption was a factor, many researchers believed it must be augmented by cuticular abrasion in order to cause desiccation in most insects (Beament 1945; Wigglesworth 1944; Hurst 1948).

While many researchers had focused efforts on determining the mechanisms of how "inert" dusts killed pest insects (Beament 1945; Kalmus 1944; Wigglesworth 1944; Hunt 1947; Hurst 1948), others had noticed that inert dusts affected insects in different ways and could actually cause increases in pest infestations (Callenbach 1940; Flanders 1941; Halloway et al. 1942; Halloway and Young 1943). Crops coated with dusts from dirt roads or intentional dust applications exhibited increased levels of codling moth, *Cydia pomonella* (L.) (Callenbach

1940), Citrus red mite, *Panonychus citri* (McGreggor) (Halloway et al. 1942), and purple scale, *Lepidosaphes beckii* (Newman) (Halloway and Young 1943). Flanders (1941) proposed that the pest increases were a result of dusts interfering with the efficacy of natural enemies. The efficacy of natural enemies was influenced by dusts via at least four mechanisms: (1) dusts impeded movement of legs and mouthparts (Germar 1936), (2) dusts invoked the "self-cleaning" response (Marcovitch 1925; Mote et al. 1926), (3) the mineral film presented a physical barrier to natural enemy attack (Driggers 1928), and (4) dusts caused direct mortality of natural enemies (Zacker and Kunike 1931).

Insecticidal dusts were the primary means of delivering insecticides in the 1940s and interest in the toxicity of mineral dust diluents established the need to better classify these diluents. Watkins and Norton (1947) found diluents and carriers fell into two basic categories, botanical flours (e.g., walnut shell flour) and minerals (e.g., attapulgite). A cornerstone study by David and Gardiner (1950) on the physical properties of dust carriers for insecticides summarized that particle size, shape, specific gravity, bulk density, surface area, hardness, and moisture relations were all factors that affected the toxicity of dusts alone or in combination with DDT. These results were confirmed by Alexander et al. (1944a), who established that abrasive dusts with sharp angular structure caused insects to die from desiccation most rapidly and that low mortalities were associated with high humidities. Watkins and Norton (1947) also found that abrasive dusts like alumina-aluminum oxide (Al_2O_3) or silica oxide (SiO_2) were the best carriers for DDT. Surprisingly, soft nonabrasive minerals like talc and slate dust, alone or in combination with DDT, attached to insects as well as Al_2O_3, but these minerals were not as lethal to insects as DDT or Al_2O_3. After World War II, the development of synthetic pesticides superceded the use of minerals in the control of plant pests. Despite the common usage of synthetic pesticides, diatomaceous earth (Celite®), wettable sulfur, and hydrated lime are still used as insecticides in some crops.

The ability of finely divided particles to adsorb and remove the cuticular waxes of insects was proven by Ebling and Wagner (1959), who developed several techniques to quantify this phenomenon. They found that nonabrasive sorptive dusts like montmorillonite and attapulgite removed the thin lipid layer covering the epicutical of dry wood termites, *Incistermes minor* (Hagan). Sorptive-dust treated termites died from desiccation more rapidly than through contact with insecticides like parathion. Certain silica aerogels (synthetic oxides of silicon), especially those impregnated with fluoride, were more lethal than mineral dusts at high humidities (Ebling and Wagner 1959). Further, they believed silica gels had less health issues for humans than crystalline silica because crystallized silicates in natural mineral dusts could cause the lung disease silicosis. Ebling (1961) later established that particle pore size of ≥ 20 Å strongly correlated with insect mortalities, regardless of the particle's size, or abrasiveness. Pore sizes of 20 Å or larger were required in order to adsorb the larger wax molecules (ca. C_{30} chain length) that are characteristic of most insect waxes. Synthetic silica gels were far better than sorptive minerals like attapulgite. Ebling (1971) later modified his statement on 20 Å pore size in mineral particles as being most critical for sorptive action and included particle surface area (particle size) as also being equally important. He also found that stored grain pests such as the rice weevil, *Sitiophilus oryzae* (L.), household pests such as the western drywood termite, or American cockroach, *Periplanta americana* (L.), and ectoparasites affecting livestock such as the northern fowl mite, *Ornithonyssus sylvarium* (Can. and Fan.), were ideally suited for control by sorptive dusts. In particular, silica aerogel dusts were effective against this wide range of pests. Although not mineral-based, Ghate and Marshall (1962) suppressed eggs and mobile forms of European red mite and two-spotted spider mites with a combination of buttermilk and wheat flour.

Interest in the control of insects with inert dusts transitioned from minerals to synthetic compounds like silica aerogels and fumed silicas by 1970. Although dusts for insect control may have had the greatest potential for the pest control needs of the grain industry, inexpensive fumigants became widely used instead. Much of the research on mineral particles after 1970 was limited to pesticide formulations where mineral particles were used as carriers for synthetic insecticides (Kirkpatrick and Gillenwater 1981; Margulies et al. 1992) or microbial agents (Studdert et al. 1990; Tapp and Stotzky 1995) and in the use of minerals as whitewash sprays for preventing plant virus diseases that were vectored by aphids (Moore et al. 1965; Johnson et al. 1967; Adlerz and Everett 1968; Bar-Joseph and Frenkel 1983) and thrips (Smith et al. 1972).

Moericke (1952) was first to demonstrate that aphid alight on plants in response to color (phototaxis). This discovery opened up a new field of entomological study, and provided a means of monitoring aphid movement and protecting plants from aphid transmitted diseases. Aphids respond strongly to yellow and alight on this color; they respond less so to green and orange, and few respond to white, red, blue, black or violet (Moericke 1955). Thrips, another important plant disease vectoring insect, did not respond to the same colors as aphids, except for blue, which was attractive (Wilde 1962). Within this time period, horticulturalists investigated aluminum foil mulches and also found vegetable yields markedly increased, possibly due to water conservation (Pearson et al. 1959). Further study into aphid response to color and light revealed that light reflected by foil and other surfaces repelled aphids (Kring 1962). This discovery led to a proposal by Kring (1964) that reflective mulches could prevent aphid infestation and the diseases that they vector. Aphids (Moore et al. 1965; Johnson et al. 1967; Adlerz and Everett 1968) and thrips (Smith et al. 1972) were repelled and the diseases they vectored were reduced by aluminum foil, white polyethylene, and other light-reflecting mulches. However, not all aphids respond to colors similarly. White mulches increased aphid levels (Brown et al. 1989) and thrips in tomato (Csizinszky et al. 1999). The drawbacks of using mulches included the high cost for material and labor and disposal problems (Greer and Dole 2003). Solutions to this problem include degradable mulches that include sprayable forms. It was not until the 1980s that a kaolin-based sprayable mulch was demonstrated to be effective against the spirea aphid, *Aphis spiraecola* Patch, in citrus (Bar-Joseph and Frenkel 1983). Spraying whitewashes for insect control, however, did not become popular and was of little scientific interest until the recent development of particle film technology. Particle film technology is partially based on the concept that reflective mulches and whitewashes repel certain arthropod pests and prevent pest vectored plant diseases.

B. Development of Particle Film Technology for Pest Control

Particle film technology for arthropod pest control represents a combined knowledge of the benefits of reflected light, mineral barriers, and toxic properties of minerals. Key to this technology was the recognition that mineral particles can have significant effects on insect behavior that were not previously recognized (Glenn et al. 1999; Puterka 2000a). Although previous researchers (Moericke 1952, 1955; Kring 1962, 1964) established that aphids were repelled from highly reflective surfaces, Puterka et al. (2000a) demonstrated that mineral particle films on plants repelled insects that were not known to be repelled by reflective light. Insects were agitated by particle film treated plants through contact with the film where particles attached to insects as well as having other effects on insect biology and behavior (Glenn et al. 1999; Puterka et al. 2000a,b, Puterka and Glenn, in press). Just as important were the effects of particle films on plant photosynthesis where, as described in this chapter, it was crucial that these mineral particle films did not have adverse effects on the plant. Particle film research began in 1994 originally in an attempt to control fruit diseases with hydrophobic kaolin films. In field trials, it was quickly realized that hydrophobic films reduced insect damage, marking the beginning of the entomological research on particle film technology.

Particle film technology was originally based on a kaolin [$Al_4SI_4O_{10}(OH)_8$] made hydrophobic by a silicone coating that was originally developed for disease control in tree fruits. Hydrophobic kaolin (M96-018, Engelhard Corp.) was initially applied as a dust using various hand-operated dusters or modified sand-blasters for large-scale studies because the hydrophobic material could not be mixed and delivered in water. Plants coated with hydrophobic particle films exhibited repellence, ovipositional deterrence, and reduced survival of insects and mites on apple and pear (Glenn et al. 1999). However, the drift associated with dusting operations, plus lack of adhesion to the plant, made M96-018 dust applications impractical. Within a year, a methanol (MEOH)–water system was developed where M96-018 could be pre-slurried with 99% MEOH (11.3 kg M96-018 + 15.1 L MEOH premixed then added to 363.4 L water) and delivered as a spray to trees (Puterka et al. 2000). Yet, this formulation was difficult to pre-slurry, too expensive for practical use, and handling and transportation of 99% MEOH was restrictive because MEOH was listed as a hazardous material by the U.S. Department of Transportation. The need for an easier formulation brought the development of a two-package hydrophilic kaolin formulation, M97-009, that required a non-ionic spreader sticker, M03 (Engelhard Corp., Iselin, New Jersey). M97-009 contains the same kaolin material of M96-018 but without the silicone coating; both have particle sizes of about 1.0 µm in diameter. Laboratory (Puterka et al. 2000a) and field studies (Puterka et al. 2000b) determined that formulations based on M97-009 plus M03 spreader sticker were just as effective as M96-018 hydrophobic kaolin dusts or aqueous sprays in controlling insects and diseases. Advantages to using hydrophilic kaolin formulations were: (1) ease of mixing, (2) economical features, (3) compatibility with other materials for tank-mixes, and (4) formulation flexibility to alter spreading and rainfastness. M97-009 + M03 became commercially available in 1999 under the name Surround® crop protectant (Engelhard Corp., Iselin, New Jersey). Although this formulation worked well against pear psylla in pear, shipping and handling a two-package system (particles plus spreader sticker) had logistical problems that pushed research efforts to develop a single-package system. In 2001, Surround® was replaced by Surround® WP crop protectant, a single-package system that uses the same kaolin-base particle as M96-018 and M97-009, but has the sticking and spreading agents incorporated. Surround® WP is now the primary commercial formulation used for insect protection as well as for sunburn and heat stress control. Another single package particle film formulation that became commercially available in 2002 was Surround® CF, which is similar to Surround® WP but has a different spreader-sticker system to speed tank-mixing under cold weather conditions (4 to 10°C). Surround® WP is listed for use in organic food production by the Organic Materials Review Institute (OMRI). Surround® CF is listed for use in organic production by the Washington Department of Agriculture.

C. Efficacy of Particle Films to Control Arthropod Pests

Particle films are effective against many key orders of arthropod pests affecting crops, including homopterans, coleopterans, lepidopterans, dipterans, and rust mites, as well as the family Eriophyidae (Table 1.2). Most research trials using particle films were conducted with applications of 3–6% solids in water and were applied to trees or other crops until the leaves became thoroughly wetted. The exception is M96-018, which was usually applied at 3% solids because particle to particle repulsion of the silicone-coated particles produced very thick fluffy films in comparison to hydrophilic particle formulations. Applications are typically made to "near-drip" and are considered to be almost a "dilute application" where 3700 L/ha is applied to mature fruit trees 8 m in height. The popularity of dwarfing rootstocks results in smaller trees where particle film applications are often applied at 935 L/ha. Studies that compared 3 and 6% solids application rates showed no significant rate differences in the lab or field, indicating that rates of 3% solids for hydrophilic particle films were adequate for insect control in season-long programs where numerous (7–13) applications are made. However, we have observed that sprays of 6% solids produce films on leaves that are more rainfast and weather far better than two 3% solids sprays on apple and pear trees in the eastern United States where frequent rains are encountered in the spring.

Laboratory bioassays on the effectiveness of kaolin particle films against pests often correspond closely to results obtained in the field (Glenn et al. 1999; Puterka et al. 2000a,b; Knight et al. 2000; Unruh et al. 2000). Exceptions to this correlation are results using the silverleaf whitefly, *Bemesia argentifolii* Bellows and Perring, and two-spotted spider mite, *Tetranychus urticae* Koch. Liang and Liu (2002) report that Surround® WP sprays of 6% solids repelled adults by 50% in melons compared to untreated controls, yet Poprawski and Puterka (2002a,b) observed no control of this pest in the field. Particle film materials

Table 1.2. Efficacy (%) of particle film formulations against key insect pests of various crops.

Formulation^z	Crop	Pests	% Efficacy	Mechanisms	Comments (rate)	Reference
M96-018 dust	Apple	*Aphis spiraecola* Potch	50%	Mortality	Lab (dust@100 ug/cm^2)	Glenn et al. 1999
		Tetranychus urticae Koch	50%	Mortality	Lab (dust@100 ug/cm^2)	
		Empoasca fabae (Harris)	50%	<Damage	Field (dust@100 ug/cm^2)	
	Pear	*Cacopsylla pyricola* (L.)	>75%	Repellence, < oviposition	Field (dust@100 ug/cm^2)	
M96-018 dust and MEOH, Surround+M03	Pear	*Cacopsylla pyricola* Foerster	>90%	Repellence, < infestations	Field (dust@100 ug/cm^2 liquid @ 3% solids)	Puterka et al. 2000b
		Epitrimerus pyri (Nalepa)	60%	< Damage	Field (same as above)	
		Conotrachelus nenuphar (Herbst)	100%	< Oviposition suppression	Field (same as above)	
		Cydia pomonella (L.)	50–80%	< feeding damage	Field (same as above) M96-018 best	
M96-018+MEOH	Apple	*Choristoneura rosaceana* (Harris)	75%	Mortality, reduced mating success, repellence	Lab and Field (3% solids)	Knight et al. 2000
M96-018+MEOH, Surround+M03	Pear	*Cydia pomenella* (L.)	90–99%	< feeding damage, repellence, < oviposition	Field (1.5, 3 and 6% solids) No rate effect	Unruh et al. 2000
	Apple	*Cydia pomenella* (L.)	53–87%	< Damage, < oviposition, and repellence	Lab and Field (1.5 and 3% solids) No rate effect	Lapointe 2000
Surround+M03	Citrus	*Diaprepes abbreviatus* (L.)	68–84%	< feeding damage	Lab (3% solids)	Liang and
Surround® WP	Melon	*Bemesia argentifolii* Bellows and Perring	50%	Repellence, < oviposition	Lab (6% solids)	Liu 2002
Surround® WP	Cotton	*Anthonomus grandis*	75%	Repellence, < oviposition	Lab and Field (6% solids)	Showler 2002a
Surround+M03	Pecan	*Aphis spiraecola* Potch	70–90%	Repellence	Lab (6% solids)	Cottrell et al. 2002

(*continued*)

Table 1.2. *(continued)*

Formulation[z]	Crop	Pests	% Efficacy	Mechanisms	Comments (rate)	Reference
M96-018+MEOH, Surround®+M03	Collards	*Bemesia argentifolii* Bellows and Perring	No control	—	Field (3% solids)	Poprawski and Puterka 2002a
	Pepper	*Bemesia argentifolii* Bellows and Perring	No control	—	Field (3% solids)	Poprawski and Puterka 2002b
Surround® WP	Citrus	*Diaphorina citri* Kuwayama	75%	Repellence	Field (3 and 6% solids) No rate effect	McKenzie et al. 2002
M96-018 and Surround® dust	Stored grain	*Atribolium confusum* (du Val), *T. castaneum* (Herbst)	0–55% depending on RH	Mortality, dessication	Lab bioassays (0.5 mg/cm^2)	Arthur and Puterka 2002
Surround® WP	Grape	*Homalodisca coagulata* (Say)	>95%	Repellence, host camouflaging, < oviposition	Lab and Field (4–6% solids)	Puterka et al. 2003a
Surround® WP	Olive	*Bactrocera oleae*	>90%	< oviposition	Field (6% solids)	Saour, in press
M96-018 dust and MEOH, Surround+M03	Pear	*Cacopsylla pyricola* (L.)	>90%	Repellence, oviposition deterrence, fall-off, < nymphal survival, host camouflaging	Lab (3 and 6% solids) No rate effect, formulation	Puterka and Glenn, in press

[z] Formulation and rate: M96-018 dust (hydrophobic film)—100 g/tree; M96-018/MEOH (hydrophobic film)—3% solids, 4% MEOH, 100 gpa; Surround/M03 (hydrophilic film)—also called M97-009/M03—3% solids, 1 pt. M03 spreader/100 gal water, 100 gpa; Surround® WP—3% solids, 100 gpa.

coated peppers and collards well but the lack of coverage on the undersides of the leaves was likely the reason for its failure in whitefly control. Other insects that were controlled at least 50% in laboratory bioassays but were not controlled in the field were two-spotted spider mite and aphids. Again, when leaves are completely coated on both surfaces with particle films, the two-spotted spider mites are controlled under laboratory conditions (G. J. Puterka, unpubl. data), however, thorough coverage, particularly on the adaxil sides of leaves, is difficult to achieve and maintain adequately under field conditions. In contrast, we have observed that aphids escape the effects of films by moving progressively onto untreated newly emerging terminal leaf growth. San Jose scale [*Quadraspidiotur perniciosus* (Comstock)] was not controlled in apple with particle film treatments. This pest is generally controlled by natural predators and parasites in orchards, which indicated that the particle film reduced the efficacy of these beneficial organisms. Yet, from the trials we have conducted or observed, particle films have the potential to suppress to some degree nearly any arthropod pest species if adequate coverage can be maintained on the target plant parts.

D. Action of Particle Films on Arthropod Biology and Behavior

Arthropods use the senses of touch, taste, sight, and smell in the processes of locating and accepting plants as a host for feeding and reproduction (Miller and Strickler 1984). During the process of locating and accepting hosts, the four senses interact in such a manner that insects sense positive and negative cues, the sum of which provokes a positive or negative behavior in insects. For example, when the accumulation of positive cues outweighs negative cues, an acceptance behavior (e.g., feeding, oviposition) will occur. Plant tissues coated with particle films are obviously altered visually and tactilely to insects. Particle films also could alter the taste or smell of the host plant (Puterka and Glenn, in press). Choice and no-choice laboratory bioassays with various insects revealed that the primary mechanism of action was repellence of adults from treated foliage that results in reduced feeding and oviposition (Table 1.2). Repellency is only used tentatively as a mechanism since it has not yet been demonstrated whether insects orient away from particle films before film contract (repellence) versus after film contact, which is more appropriately termed a deterrent (Puterka and Glenn, in press). These mechanisms will be dependent on the insect species. Other mechanisms include: (1) reduced survival of adults or immature insects (larvae) when born into the particle film coated leaf environment (Knight et al. 2000; Unruh et al. 2000; Cottrell et al. 2002; Puterka and Glenn, in press), (2) reduced mating success of adult lepidoptera exposed to particle films (Knight et al. 2000; Puterka and Glenn, in press), (3) impeded movement/host finding ability within plant canopies (Unruh et al. 2000), (4) camouflage of the host by turning the plant foliage white with the particle film (Puterka et al. 2003a; Puterka and Glenn, in press), and (5) impeding the insect's ability to grasp the plant (Table 1.2). In impeding an insect's ability to grasp the plant, insects simply "fall-off" the host plant (Puterka and Glenn, in press). Most of the effects particle films have on insects result from particle attachment to the insect's various body parts (Plate I, bottom).

The lethal effects of particle attachment to insects have been well documented (Alexander et al. 1944a,b; David and Gardiner 1950; Ebling 1971). Yet, one should not underestimate the effects particle films have on altering the insect's visual and tactile perception of the host as key aspects in host finding and acceptance (Miller and Strickler 1984). Although repellence of aphids (Kennedy et al. 1961; Kring 1962, 1965; Nawrocka et al. 1975) and thrips (Wilde 1962; Ota et al. 1968; Smith et al. 1972) by reflective mulches has been demonstrated, the effect of reflected light on other arthropod species has not been well studied. Many other arthropod species besides aphids are attracted to specific colors, such as yellow for glassy-winged sharpshooter [*Homalodisca coagulata* (Say)] (Puterka et al. 2003a), pear psylla (*Cacopsylla pyricola* Foerster) (Puterka and Glenn, in press), and red for apple maggot [*Ragioletis pomonella* (Walsh)] (Prokopy and Hauschild 1979). Many arthropods have been shown to be attracted to specific colors that are believed to represent a "super-normal" colored host, where, for example, yellow represents super-normal foliage mimics (Prokopy and Owens 1978). Masking host plant color with reflective white particle films could conceivably have major effects on arthropod pest behavior.

E. Examples of Successful Particle Film Use to Control Arthropod Pests

Particle film technology became commercially available to growers in 2000. Surround® WP is registered for control of a broad range of arthropod pests on nearly all major groups of agricultural crops and has been successfully used against many more pests than summarized in Table 1.2. Particle film technology has had a major impact on two arthropod pests in particular, pear psylla (*C. pyricola*) in pear, and the glassy-winged sharpshooter (GWSS) (*H. coagulata*). These two successes will be reviewed in more detail.

The pear psylla is a key pest of pear whose feeding causes leaf necrosis, defoliation, and reduced yields (Hibino et al. 1971). This pest rapidly develops resistance to insecticides (Follett et al. 1985; Pree et al. 1990). Much of the original entomological research on particle films used this organism as a model pest species (Glenn et al. 1999; Puterka et al. 2000a,b; Puterka and Glenn, in press). Processed kaolin repelled adult pear psylla and reduced oviposition greater than minimally processed, air floated kaolin (Puterka et al. 2000a). Both hydrophobic M96-018 and hydrophilic M97-009 + M03 (Surround®) particle films were based on the same purified and processed kaolin, and both have demonstrated comparable efficacy against pear psylla. This efficacy operated through at least six mechanisms: repellence, ovipositional deterrence, reduced feeding efficacy, impeded grasping of the host (fall-off), host camouflaging, and direct mortality (Puterka et al. 2000a,b; Puterka and Glenn, in press). Repellence is the most obvious effect that particle films have on psylla adults and several factors are thought to influence repellence. Hydrophobic particle films cause greater particle attachment to pear psylla than hydrophilic particles, thus, hydrophobic particles have greater effects on pear psylla biology and behavior (Puterka and Glenn, in press). Despite such differences in particle attachment between formulations, those formulations that show lower particle attachment compared to M96-018 remain repellent to pear psylla adults. Repeated summer applications of Surround® can produce a white staining effect on tree bark that remains through the winter and effectively prevents oviposition on dormant twigs the following spring (March) (Puterka et al. 2000). This observation of carryover effect suggested that particle attachment may not be necessary to prevent oviposition of winter-form adults, and alterations in bark color or surface structure could deter oviposition. Psylla adults show no preference for color during March and become attracted to yellow only after pear begins to break dormancy and produce foliage (Puterka and Glenn, in press), which argues against white staining of the bark as a possibility in deterring oviposition. Thus, the alteration of the twig surfaces by the incorporation of kaolin particles may have been a key factor in reducing pear psylla oviposition. Horton (1990) noted that psylla adults prefer to oviposit in the grooves, lenticels or other areas of relief in the leaves or bark. Thus, it is possible that these areas of relief in the bark could be altered by the particle film treatments (Puterka and Glenn, in press). Once green foliage became available, the carryover effect on overwintering psylla adults was lost and eggs were deposited on untreated foliage (Puterka et al. 2000).

Initially, control of pear psylla with particle films was conducted on a season-long basis where up to 13 applications were used (Puterka et al. 2000b). However, commercial usage in conventional pear orchards in northern Washington State soon focused on early-season control where two to three applications of particle films were applied at 6% solids to dormant trees prior to bloom (Plate II, top left). Timing applications prior to bloom often resulted in greatly reducing pear psylla oviposition to the extent that applications after petal-fall were rarely needed. Usage of Surround® WP on U.S. pear crop area grew from 2% in 1999 to 14% in 2001, and its usage in 2002 and 2003 increased to nearly 50% of U.S. pear growers. The remarkable efficacy of particle film technology against pear psylla inspired the Washington State Research and Extension Service to organize an area-wide approach for psylla control called the Peshastin Creek Pear Growers Area-Wide Organic Project that was instituted in 2002. In this program, psylla is predominately controlled by Surround® WP, while other insects not controlled by Surround® WP, such as mites, are controlled using spray applications of light summer oils. This program has effectively reduced insecticide usage in pear by directly replacing conventional chemical insecticides.

The second successful example of particle film use is against the glassy-winged sharpshooter (GWSS) (Plate II, top right). The GWSS is a serious pest of grape that was recently introduced before 1990 in southern California via eggs on nursery stock (Sorensen and Gill 1996). By 1999, the GWSS had spread throughout coastal southern California and northward into the southern San Joaquin Valley where it utilized citrus as a primary host. GWSS is considered a minor pest in citrus and is generally not controlled. The GWSS has become a significant problem to California agriculture because it feeds readily on grape vines and, in doing so, transmits *Xylella astidiosa*, the causal agent of Pierce's disease (PD). PD causes leaf scorching, vine dieback, and eventually kills the vine within a few years (Phillips 1999). There is currently no cure for PD (Krewer et al. 2002).

This sharpshooter species was considered a significant threat to California's $40 billion grape industry because there were no known low-toxicity control measures available for preventing GWSS from feeding on grape vines and vectoring PD. Contact insecticides only offer short-term protection against infestations but the continual influx of immigrating sharpshooter adults from nearby citrus soon re-infests grape vines. Systemic treatment of grape vines with imidacloprid, Admire 2E (Bayer Co., Kansas City, Missouri), was found to slow the rate of disease incidence but only extended vineyard life by one year under high GWSS infestations (Krewer et al. 2002). GWSS is a particular problem in California where citrus borders grape vineyards and citrus trees are the primary reproductive host. GWSS reproduces in citrus orchards during the summer months and over-winters in citrus. When air temperatures begin to rise in the spring, GWSS migrates into grape vineyards where it feeds and reproduces. GWSS spreads PD during the feeding process. Research in Kern County, California established that when grape vines were treated with Surround® WP in a 247.5 m barrier where grape vines bordered citrus (Fig. 1.2, top), migration of GWSS was suppressed and oviposition was prevented (Puterka et al. 2003a). Furthermore, the Surround® WP barrier on grape vines had a sufficient depth to prevent GWSS from flying over the barrier and invading vineyards. In that study, three bi-weekly applications of Surround® WP outperformed six weekly applications of contact insecticides in reducing GWSS infestations, and Surround® WP nearly eliminated oviposition (Fig 1.2, bottom). The modes of action of particle films on GWSS include repellence, ovipositional deterrence, and host camouflaging (Puterka et al. 2003a). GWSS were found to be attracted to yellow, and to a lesser degree orange, while white was non-attractive during the grape growing season, making host camouflaging a possibility. A large-scale pilot study called the General Beale GWSS Management Program was initiated in 2001 in Kern County; it utilized Surround® WP as part of the IPM strategy. Surround® WP was used in this program as a 247.5 m barrier in grape vines that bordered citrus where treatments began in March prior to GWSS migration into vineyards. The strategy was to keep GWSS contained in citrus until temperatures increased to about 18°C, the minimum temperature needed for satisfactory levels of control with pyrethroid insecticides in citrus. This program decreased the GWSS number from up to a thousand per trap to undetectable levels in vineyards and citrus groves within a year. The success of the program resulted in its expansion to include most of Kern County the following year. Research is ongoing to determine whether reduced adult GWSS activity in Surround® treated plots resulted in PD reductions in vines.

III. PHYSIOLOGICAL AND HORTICULTURAL USES OF PARTICLE FILMS

A. Effects on Net Gas Exchange and Productivity

Practitioners learned that the application of mineral particles could greatly reduce disease and insect damage but this benefit was overshadowed by negative effects of light reduction and reduced photosynthesis.

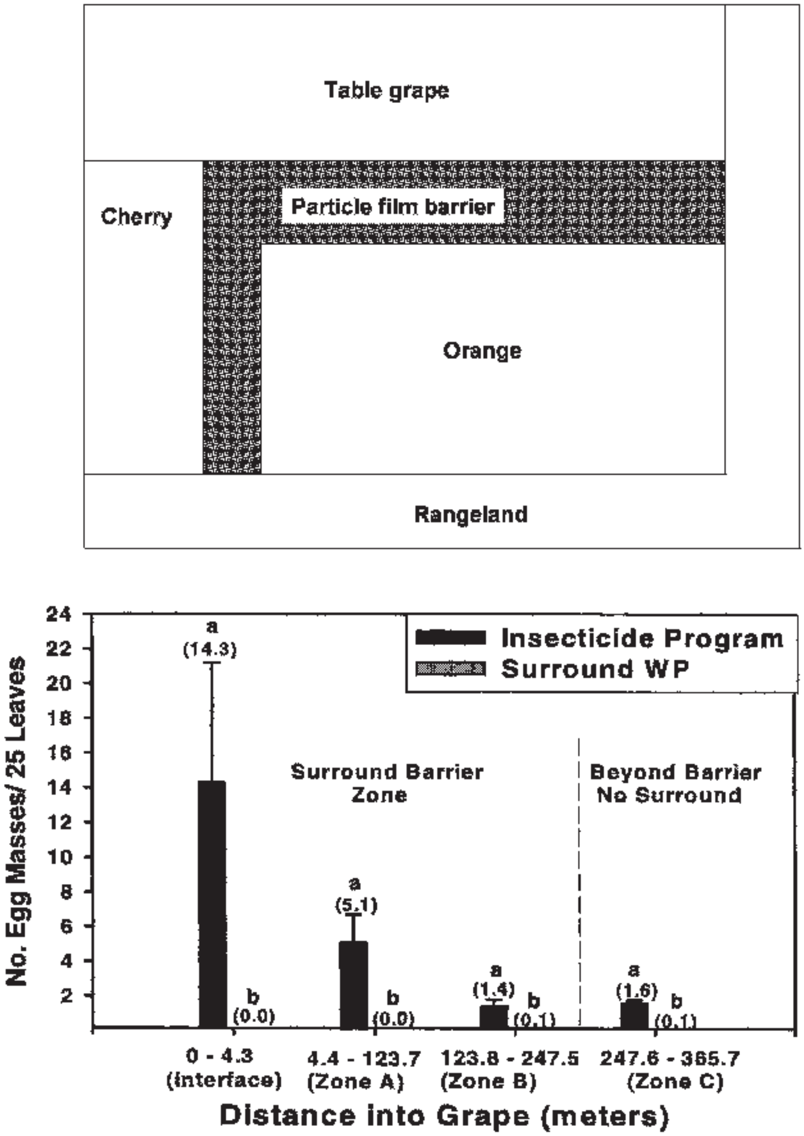

Fig. 1.2. (Top) Typical cropping system in Kern Co., California where citrus borders grape. A 247.5 m buffer zone of Surround® WP particle film as a barrier was applied in grape to prevent glassy-winged sharpshooter (GWSS) from migrating out of citrus orchards into grape vineyards during the spring. GWSS infestations were contained by the barrier until insecticides were applied in citrus to eliminate GWSS. (Bottom) Effect of biweekly Surround® WP and weekly contact insecticide applications on GWSS oviposition three weeks after the last insecticide application, Kern Co., CA. Contact insecticides were also applied beyond the Surround® WP barrier.

Lime sulfur applications reduced photosynthesis for several days following application (Hoffman 1934). Bordeaux mix, particularly the copper sulfate portion, physiologically reduced photosynthesis by chemical interference, and not by blocking light (Southwick and Childers 1941). Lime sulfur reduced photosynthesis more than a "dry mix (wettable sulfur)" which had little or no effect on leaf photosynthesis. Mills (1937) demonstrated improved vigor and long-term yield using wettable sulfur agents for disease control compared to lime sulfur. Heinicke (1937) confirmed the reduction of photosynthesis by lime-sulfur and advocated the use of milder agents such as wettable sulfur that would have "cumulative benefits resulting from the greater photosynthetic activity of the leaf surface." Heinicke (1937) noted "improvement in color and size of fruit where the leaf surface is not handicapped by the application of materials that tend to inhibit photosynthesis." Current agrichemicals such as surfactants and foliar urea (Orbovic et al. 2001), fungicides (Wood et al. 1984), and insecticides (Wood and Payne 1984) can also reduce photosynthesis on a short-term basis. Yet, the temporary reduction in photosynthesis apparently is acceptable because the value of the pest control outweighs the transient reduction in photosynthesis. Particle film technology builds on this idea of using mineral particles that are chemically inert in order to reduce any deleterious effects on leaf physiology and to safeguard human health.

The deposition of fine particles on plant surfaces from natural and human activities, such as mining and road traffic, generally decreased plant productivity due to light blockage that reduced photosynthesis and interference with stomatal activity that increased leaf temperature when sufficient residue develops (1 to 10 g/m2) (Thompson et al. 1984; Armbrust 1986; Farmer 1993; Hirano et al. 1995). Yet reflective antitranspirants, historically termed whitewashes, have been used in agriculture to reduce heat stress. Reflective antitranspirants, unlike polymer film antitranspirants that physically block the stomates, have antitranspirant properties because they can lower leaf temperature (Gale and Hagan 1966) by increasing reflection of infrared radiation (IR). Lowered leaf temperature reduces the vapor pressure gradient between the leaf and the bulk air which is the driving force behind transpiration (Pennman and Schofield 1951) and reducing the vapor pressure gradient reduces transpiration. Abou-Khaled et al. (1970) conducted the first systematic evaluation of reflective minerals as antitranspirants by applying a minimally processed kaolin mineral whitewash to bean, citrus, and rubber plants. They observed that most of the radiation reflected was in the visible region rather than the infrared (IR), transpiration was reduced 20–25%, and leaf temperature was reduced up to 5°C over a wide range of photosynthetically active radiation (PAR). In addition, photosynthesis (Pn) was reduced by the kaolin coating at low light intensities but Pn was equivalent or higher at high light intensities. Carbon dioxide assimilation/transpiration ratios or water use efficiency (WUE) increased with the kaolin treatment, indicating improved WUE under high light intensity. These reflective antitranspirants would be beneficial under conditions of high light intensity where the Pn rate was light saturated. Abou-Khaled et al. (1970) stimulated considerable research in the following three decades. In a series of five publications [Doraiswamy and Rosenberg (1974); Lemeur and Rosenberg (1974, 1975); Baradas et al. (1976a,b)] a group headed by Rosenberg examined the energy balance components of soybean coated with kaolin mixed with guar gum plus a surfactant and demonstrated that net radiation was reduced because reflection of short wave and long wave radiation was increased. The reduced net radiation could potentially reduce transpiration but there were conditions in which leaf temperature could increase or decrease with the application of kaolin depending on how much transpiration and the vapor pressure deficit (VPD) were affected. Basnizki and Evenari (1975) applied a commercial reflectant to globe artichoke and reduced leaf temperature, increased water use efficiency, and increased plant survival. Stanhill et al. (1976) increased sorghum yield 11% over a 3-year period with kaolin formulations similar to Doraiswamy and Rosenberg (1974), yet they measured a long-term reduction in CO_2 assimilation and early leaf senescence. Moreshet et al. (1979) used a gum binder with kaolin applied to cotton and measured an 11% lint yield increase in one year and no effect in a second year; however, total biomass was unaffected in either year. Their kaolin treatment of 25% (w/w) did reduce $^{14}CO_2$ uptake due to both a reduction in light absorption and partial blockage of stomata, yet these presumably negative effects did reduce water stress. Mungse and Bhapkar (1979) applied three reflectants (kaolin, calcium silicate, and a commercial whitewash) to both the plants and soil in dryland sunflower and found that all three reflectants increased grain and oil yield. Seasonal water use of the three reflectants was slightly higher than the untreated control, but yield increases were proportionally larger, resulting in improved water use efficiency with the use of the reflectants. Souondara Rajan et al. (1981) applied 3% and 6% kaolin to peanuts and increased yield with both concentrations, yet the 6% kaolin treatment had yield less than the 3% kaolin treatment (732 vs 1755 vs 1010 kg/ha for control, 3%, and 6% kaolin, respectively). These data suggest that 6% kaolin residues were excessive and were in some manner limiting photosynthesis. Rao (1985) applied 5% kaolin with a surfactant to non-irrigated tomato and increased yield compared to untreated controls. In subsequent work, Rao (1986) suggested that the yield increase and improved water status was due to decreased transpiration caused by reduced stomatal opening (4.1 vs 3.5 μm for control and kaolin treatments, respectively). In contrast to previous work, Nakano and Uehara (1996) found that kaolin applied to leaves and fruit increased cuticular transpiration and they suggested that the kaolin particles may combine with the waxy components of the cuticle to facilitate water movement through cuticular layers. Anandacoomaraswamy et al. (2000) applied kaolin to tea and slightly reduced transpiration from 10:00 to 15:00 hr; however, yield was unaffected.

It is critical that any product applied to a plant not interfere with the exchange of carbon dioxide through the stomates, otherwise primary productivity will be reduced. Antitranspirants increase stomatal closure to maintain high plant turgor by reducing transpiration, but obstructing stomates will also reduce photosynthesis when stomatal conductance is the limiting factor for carbon assimilation (Weller and Ferree 1978; Gu et al. 1996). Moreshet et al. (1979) applied hydrous kaolin to cotton and reduced $^{14}CO_2$ uptake within 2 days by more than 20% and they attributed the reduced carbon assimilation to reduced stomatal conductance since transpiration was reduced more than photosynthesis. However, not all formulations of kaolin applied to leaves will reduce stomatal conductance. Glenn et al. (2001c, 2003) demonstrated that stomatal conductance, transpiration, and photosynthesis are increased with the application of both hydrophobic and hydrophilic particle films based on heat activated and purified kaolin. While there are mineralogical differences in the kaolin used by Moreshet et al. (1979) and Glenn et al. (2001c, 2003), a key difference was the formulation. The formulation of Glenn et al. (2001, 2003) was friable (loosely bound), porous, and allowed the opening and closing of the stomates to dislodge particle fragments from the stomatal opening (Fig. 1.3). The formulation of Moreshet et al. (1979) utilized a gum agent as a binder that blocked stomatal openings.

Photosynthetically active radiation from 400 to 700 nm (PAR) is captured in the chemical pathway of photosynthesis and it is critical that PAR reach the chloroplasts in the mesophyll instead of being reflected or absorbed by a particle film on the leaf surface. Early research with reflectants attempted to reduce net radiation on the plant canopy under conditions of high PAR. Doraiswamy and Rosenberg (1974) applied a kaolin mixture to soybean and reduced net radiation about 8%, primarily by increasing reflection of PAR with little reflection of longwave radiation (IR). In contrast, Abou-khaled et al. (1970) found both high reflectivity in the PAR and near IR wavelengths by kaolin on orange, lemon, and rubber trees. The physical and optical properties of kaolin can be altered by processing to achieve specific particle size distributions and heating (calcination) to alter light transmission properties.

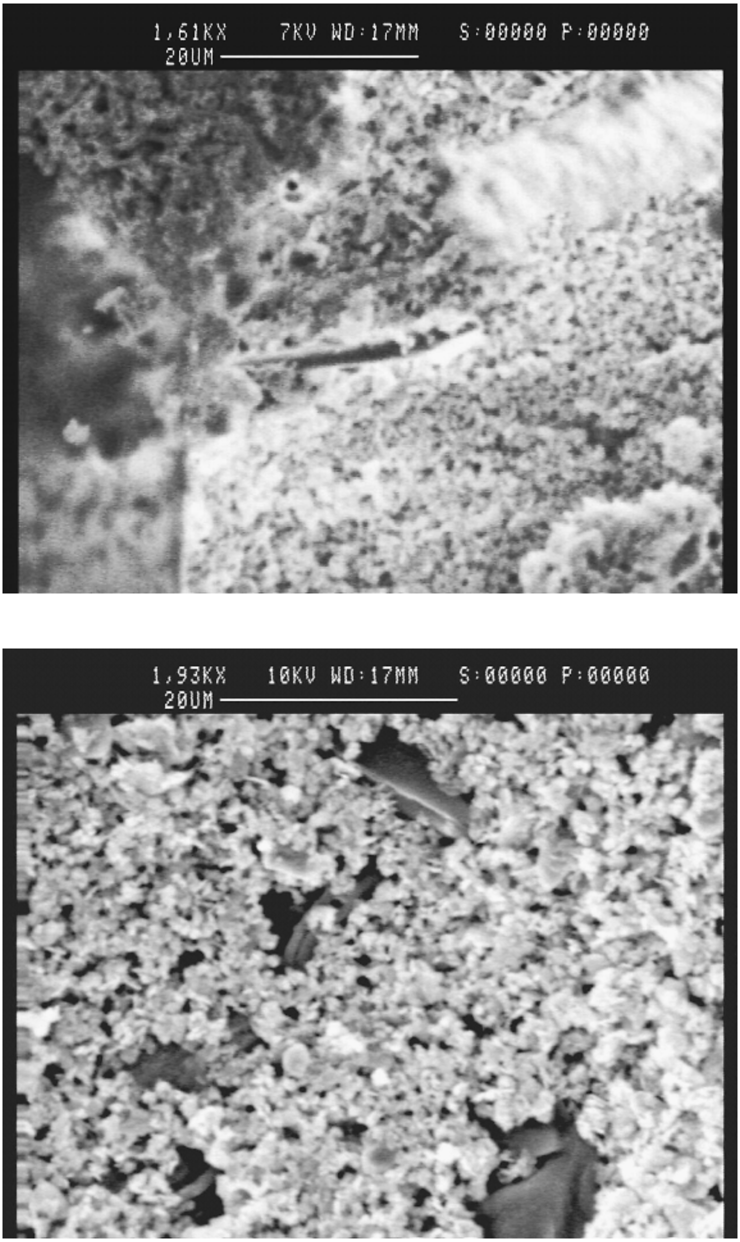

Fig. 1.3. SEM of stomata in apple. (Top) After initial application of Surround® WP Crop protectant. (Bottom) After 3 days. The particle bridges over the openings have been broken away by the opening and closing action of the guard cells.

The formulation of processed kaolin used by Glenn et al. (1999) and Jifon and Syvertsen (2003b) transmitted more PAR than the unprocessed kaolin of Abou-khaled et al. (1976) (Fig. 1.4). While the formulation used by Glenn et al. (1999) was hydrophobic and that used by Jifon and Syvertsen (2003a,b) was hydrophilic, both formulations are based on the same processed kaolin particles and have very similar optical properties. Both formulations deposit films similar in thickness and weatherability (Puterka et al. 2000).

Rosenberg (1974) stated that *"If reflectants can be developed that are more effective in the near IR, greater reduction in the energy load on the crop can result with less direct interference in photosynthesis. Although these advances await research and development, reflectant materials in use thus far already offer one very important advantage over most of the chemical antitranspirants. They are inert materials that pose no danger to the health of man or of domestic and wild animals."* The current state-of-the-art in particle film technology has achieved some of these predictions by reducing direct interference with photosynthesis through formulation and structural changes to kaolin.

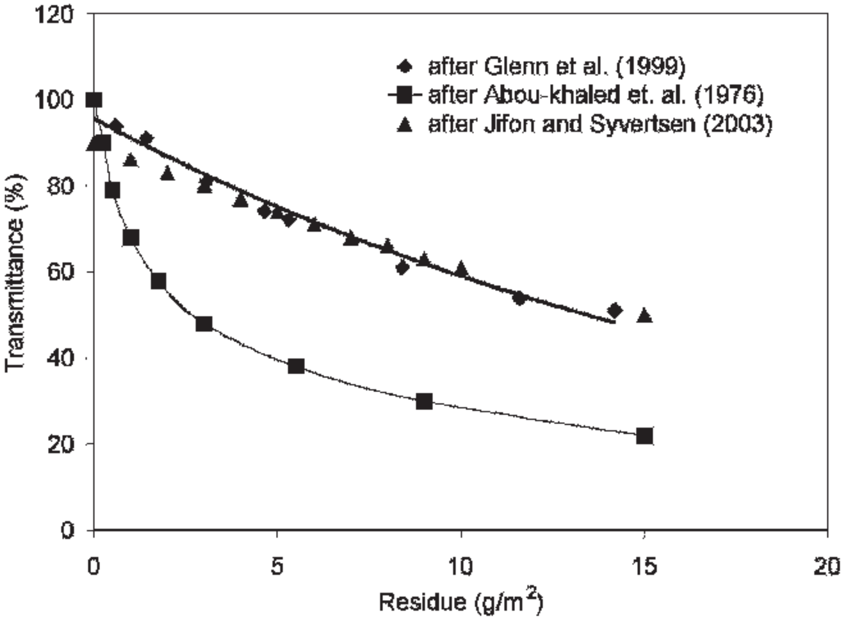

Fig. 1.4. Transmission of PAR through particle films of various kaolin sources.

Glenn et al. (1999) demonstrated that tree canopy temperature was reduced and peach yield and shoot growth were unaffected by dusting with hydrophobic particles (M96-018, Engelhard Corp., Iselin, New Jersey). Single-leaf studies did not indicate any reduction in photosynthesis with the application of M96-018 > 10 g/m^2 leaf area. Puterka et al. (2000b) compared hydrophobic and hydrophilic kaolin formulations in pear production and both formulations increased pear yield nearly 100% (Table 1.3).

Glenn et al. (2001c) used an aqueous formulation of hydrophobic kaolin (Glenn et al. 1999) to examine its effect on apple physiology in a number of locations. Single-leaf carbon assimilation was increased and canopy temperatures were reduced by particle sprays in seven of the eight trials. The trial that did not demonstrate an increase in single-leaf photosynthesis was conducted in Washington State when air temperature was less than 25°C, while all the other trials had air temperature greater than 30°C. Thus, it appears that when air temperatures are near the photosynthetic optimum (25°C), an increase in Pn should not be expected. Yet, in this trial there was an increase in yield when particle sprays were applied early in the growing season, when high air temperatures occurred. Yield and/or fruit weight were increased by the particle treatment in seven of the eight trials. There was no yield increase when fruit were severely hand-thinned to limit the size of the fruit sink despite an increase in single-leaf photosynthesis. Red fruit color was increased, but not consistently. Elkins et al. (2001) improved 'Red Sensation Barlett' pear color at harvest and after 1 and 3 months storage. The mechanisms of particle treatments affecting fruit color are not clear at this time and will require further study.

Surround® WP application to cotton reduced free amino acid content, specifically, alanine, arginine, isoleucine, phenylananine, and threonine, compared to untreated plants (Showler 2002b). The reduction of arginine in the absence of a change in proline suggested heightened

Table 1.3. Effect of hydrophobic and hydrophilic kaolin applications on 'Seckle' pear productivity and quality (after Puterka et al. 2000).

Treatment	Yield (kg/tree)	No. fruit/tree	Fruit size (g)	Red color (%)
Hydrophilic kaolin	54.8 a^z	1392 a	39.4 ab	56.5 a
Hydrophobic kaolin	54.0 a	1237 a	43.7 a	45.5 a
Conventional	28.3 b	793 b	35.7 b	27.5 b

^{z}Mean separation in columns by Duncan's Multiple Range Test ($P \leq 0.05$).

light reception or photosynthetic activity but did not indicate typical shade responses in the free amino acid profiles. In conjunction, Makus (2000) and Makus and Zibilske (2001) measured increased leaf transpiration, reduced canopy temperature, and increased biomass and lint yield in cotton with Surround® applications. Citrus leaves are light saturated at relatively low PAR levels and are vulnerable to overexcitation of the photochemical systems (Jifon and Syvertsen 2003b). High PAR levels can elevate leaf temperature and increase the VPD. Kaolin treatments increased citrus leaf reflectance, lowered leaf temperature and reduced the VPD (Jifon and Syvertsen 2003a) and similar responses were observed with shading (Jifon and Syvertsen 2003b). In single leaf studies, carbon assimilation, stomatal conductance, and water use efficiency were increased, particularly during midday hours, by 3 applications of Surround. They speculated that in warm climates with high PAR levels and high VPDs, where these conditions likely limit photosynthetic capacity, kaolin applications could improve carbon assimilation in young and small trees where most of the leaves are exposed to direct sunlight. In two of three years in California, citrus yield was increased by the application of 3 monthly applications of 3% Surround® beginning in April (Table 1.4, unpubl. data). Yield was increased due to an increase in fruit number from less fruit drop in 2001 and 2002 with no change in fruit size. Reducing heat stress with Surround® applications in 2001 and 2002 reduced fruit drop. Fruit drop, however, was not a limiting factor in 2003.

Glenn et al. (2003) measured whole-tree carbon assimilation, water use efficiency, yield, and quality of apple treated with processed kaolin and calcium carbonate particle films. Whole-tree carbon assimilation was increased by processed kaolin applications only under conditions of excessive air temperature. Carbon assimilation was increased by the processed kaolin treatment but water use efficiency was reduced likely due to increased stomatal conductance associated with reduced leaf

Table 1.4. Effect of kaolin (Surround® WP) application in citrus production (D. M. Glenn, unpubl. data).[z]

	Yield (metric tons/ha)		
Treatment	2001	2002	2003
Conventional production	36.1 b	17.8 b	54.1
Surround® treatment	39.3 a	27.3 a	52.5 ns

[z]N=4. Plots were arranged in a randomized block design. Each plot was approximately 1 ha. (P=0.05).

temperature that increased transpiration more than photosynthesis. Calcium carbonate produced none of these effects and reflected more PAR from the tree canopy than processed kaolin.

In summary, many key horticultural characteristics such as fruit size, fruit color, and yield have been improved by the application of reflective kaolin particle film materials. The proper environment, plant species, and time of application interactions need to be refined on a regional or seasonal basis in order to assure that the predicted horticultural response will occur and be of economic value.

B. Reduction of UV Damage

Ultraviolet radiation is categorized in 3 bandwidths: UVa (315 to 400 nm); UVb (280–315 nm); UVc (195–280 nm). Deleterious ultraviolet radiation (UV) effects on plants include formation of DNA dimers, and inhibition of photosystem II and Rubisco activity. At plant temperatures >35°C, both UV damage and photoinhibition of photosystem II can be additive. However, under high PAR, photoreactivation can repair much of the DNA damage and UV damage is less than under low PAR conditions (Tevini 1999). Kaolin is reflective of UV radiation (Plate II, center) but the formulation and particle size distribution significantly influence the degree of its UV reflection. The formulation of the highly processed Surround® WP has greater UV reflection than unprocessed kaolin or calcium carbonate (Fig. 1.5).

UV reflection was increased by increasing amounts of Surround® residues on the fruit and leaf surfaces (Fig. 1.6 and Plate II, center) (Glenn et al. 2002). In 50% of the recent studies, ambient solar UVb imposed significant constraints on biomass accumulation for terrestrial plants, yet these reductions in productivity typically occurred without a reduction in photosynthetic rates per unit leaf area (Day and Neale 2002). In addition, UVb effects can be chronically deleterious to perennial crops by reducing leaf area. Plants generally respond to UVb by increasing leaf thickness through thickening of the cuticle in addition to synthesizing UVb absorbing compounds (Tevini 1999). The application of a particle film artificially increases leaf thickness so the path length of radiation to target cells within the leaf (Fig. 1.1 A) is increased, as well as reducing the UV radiation load at the cuticle level of the leaf.

Sunburn or solar injury (SI) is defined as damage to fruit exposed to direct solar radiation (Jones and Aldwinckle 1990). The biological value of reflecting UV to reduce SI is not established, because the role of UV in SI is not clear. Lipton (1977) demonstrated that UVa directly induced SI in cantaloupes and Renquist et al. (1989) found that SI in raspberry

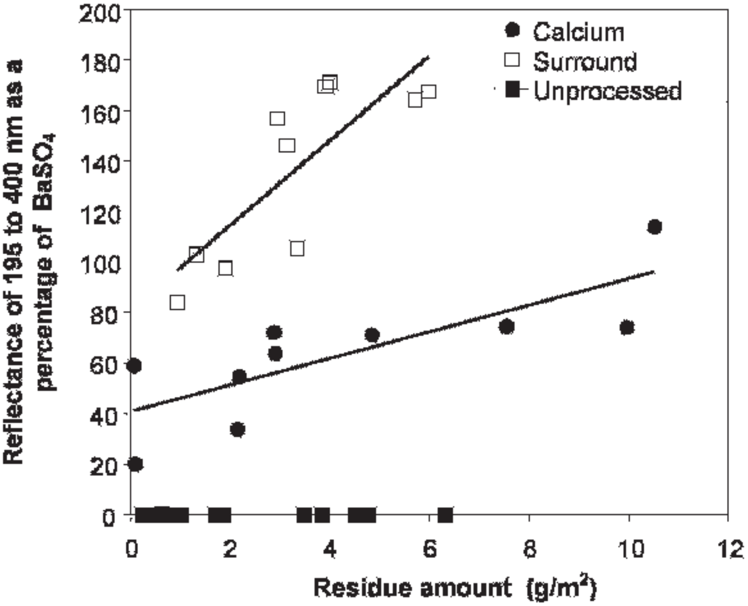

Fig. 1.5. Reflectance of ultraviolet radiation by Surround WP®, a highly processed kaolin, unprocessed kaolin and calcium carbonate.

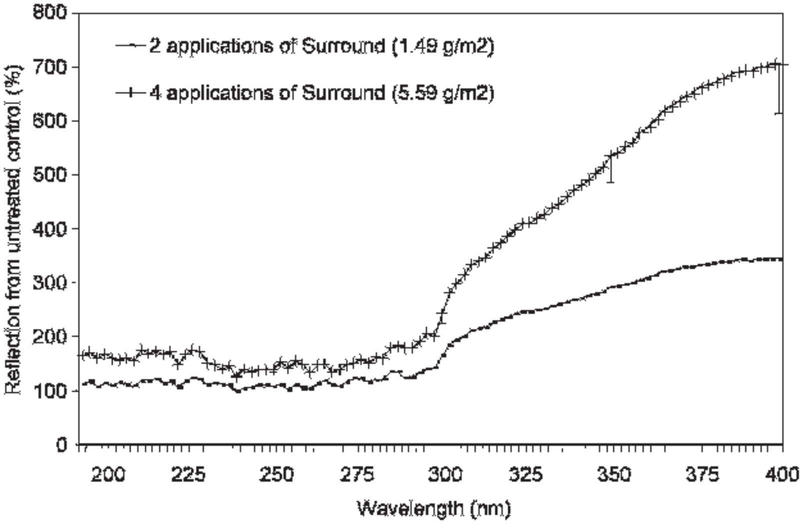

Fig. 1.6. Reflection of ultraviolet radiation from the surface of 'Fuji' apple.

was directly proportional to UVb dosage when exposed to air temperature of 42°C. In contrast, Lipton et al. (1987) found that UV radiation had minimal effect on SI in 'Honey Dew' melons, and Adegoroye and Jolliffe (1983) suggested that SI in tomatoes was due primarily to IR and that visible and UV radiation did not have an essential role. Both a critical temperature and solar radiation were necessary for SI in apple, but Schrader et al. (2001) did not distinguish the roles of UV and visible radiation. Wunsche et al. (2000) used mylar bags to exclude UVa and UVb from fruit surfaces and found no effect on SI development, although there was low SI severity in general. Yet, particle film application reduced SI in apple (Glenn et al. 2002) by mechanisms that include reflection of UV radiation. New uses of kaolin particle film under specific environmental conditions will likely be developed to exploit the mitigation of UV injury to plant tissue.

C. Reduction of Solar Injury

Whitewash and other materials formulated to have paint-like properties have been successfully used to reduce SI for decades. Serr and Foott (1963) applied a 6% mixture of $ZnSO_4$ and $Ca(OH)_2$ and a commercial whitewash of undisclosed composition to Persian walnut trees with no apparent injury to the trees. The temperature of the nut centers was reduced approximately 3°C by the treatments and sunburn damage of nuts, leaves, and twigs was reduced. Lipton and Matoba (1971) reduced sunburn of 'Crenshaw' melons with a 12% concentration of finely ground aluminum silicate by reducing surface temperature 8°C. This technology was incorporated by the California tomato industry in the early 1970s (Elam 1971). If there were reductions in fruit number or size, they were not measured and the benefit of increased yield of non-SI-damaged fruit outweighed any reduction in plant productivity.

The conditions that cause solar injury include high air temperature and solar radiation (UV 195–400 nm; PAR 400–700 nm; IR >700 nm). Serr and Foott (1963) felt that 38°C was a critical air temperature for walnut sunburn. Critical fruit surface temperatures include: 50°C for muskmelons (Lipton and O'Grady 1980); 42°C for raspberry fruit (Renquist et al. 1989); 40°C for tomatoes (Rabinowitch et al. 1974); 46–49°C for browning and 52°C for necrosis of apple skin (Schrader et al. 2001). Only Adegoroye and Jolliffe (1983) present data that solar radiation, either visible or UV, did not have a role in tomato sunscald. Schrader et al. (2001) present a strong argument that solar radiation is a key component of sunburn in apple and that sunburn can occur at air temperatures as low as 30°C under conditions of strong solar illumination. The VPD may also be a component of SI but it has not been documented. A secondary condition that exacerbates SI is the lack of foliar shade on a plant or the movement of fruit from shade to sunlight as the fruit increases in weight and changes position (Rabinowitch et al. 1986; Drake et al. 1991; Parchomchuk and Meheriuk 1996; Khemira et al. 1993).

Evaporative cooling is an effective means of reducing fruit temperature but there are concerns over expense, water quality, and the need to reduce agricultural water use (Parchomchuk and Meheriuk 1996; Unrath and Sneed 1974). The application of a Surround® particle film approached the effectiveness of evaporative cooling with intermittent water sprays in reducing fruit temperature (Table 1.5).

Reductions in fruit surface temperature can be correlated to the amount of Surround® residue on the fruit surface (Glenn et al. 2002). Midday fruit surface temperatures were reduced as much as 5–10°C by a Surround® WP particle film (Plate II, bottom). Solar injury was reduced almost 100% in some studies and had no effect in others, while the general trend was approximately a 50% reduction in SI fruit damage. The incidence of SI varied by location and cultivar. Schupp et al. (2002a,b) reduced sunburn in 'Fuji' apple in Idaho using Surround® but reduced fruit size and color at that location and also in 'Honeycrisp' apple at a New York location. They concluded that light in New York was more limiting to fruit growth and development than reduced temperature. Under New York conditions, the increased reflectance from the Surround® treatment may have reduced photosynthesis to the point that it

Table 1.5. Maximum daily fruit surface temperature (°C) of two apple cultivars treated with Surround® WP reflective particle film at Finley, Washington, 1999. (modified from Glenn et al. 2002)

	Scarlet Delicious			Fuji	
Treatment	21 Aug.[z]	22 Aug.	24 Aug.	25 Aug.	26 Aug.
Non-treated	40.9 a[y]	41.8 a	40.4 a	37.2 a	42.3 a
Surround®	36.9 b	37.5 b	38.1 b	35.8 b	38.8 c
Evaporatively cooled ± CI	32.8 ± 2.1[x]	36.9 ± 2.4	35.4 ± 1.2	36.3 ± 1.1	38.1 ± 1.2
Air temp. (C)	30.0	31.1	34.8	32.8	28.1

[z] Sampling date.
[y] Mean separation using Duncan's Multiple Range Test, $P < 0.05$.
[x] 95% confidence interval from a non-replicated, evaporatively cooled area adjacent to the study site.

diminished fruit growth and color development, especially when applied late season. In contrast, Garcia et al. (2003) increased fruit size and percentage red color in Vermont in a two-year study. R. Byers (pers. commun.) delayed fruit color development in 'Fuji' and 'Gala' but full color did develop, indicating that harvest dates may be changed by Surround usage. E. Fallahi (pers. commun.) has not found reductions in fruit size or color for a number of apple cultivars in subsequent years in Idaho but has observed reduced SI.

Chlorophyll florescence of apple fruit can indicate heat stress and solar injury (Song et al. 2001; Wunsche et al. 2000). Flesh browning was negatively correlated with chlorophyll florescence in both studies. Shahak (unpubl. data, 2000) observed that photoinhibition in apple fruit surfaces was significantly reduced on 'Jonagold' apples when treated with Surround® WP (16 vs 30% inhibition for Surround® WP treated vs untreated control). This demonstrated the effectiveness of the particle film to reduce the heat and light load on the fruit surface that caused photoinhibition.

There are significant differences in heat flux between minimally processed kaolin that has had only coarse sand particles removed and highly processed kaolin used in Surround® WP that is purified and structurally altered by heat-treatment (calcination), thus the processing of kaolin is a key component in the reflection of heat (Fig. 1.7). The processing of kaolin increased both IR (Fig. 1.7) and UV (Fig. 1.5) reflection, which are key aspects of reducing solar injury in horticultural crops.

The demand for water in agriculture is in competition with urban, industrial, and recreational water demands. New tools are needed to reduce agriculture's consumption of water without jeopardizing yield stability or quality. We have demonstrated that kaolin particles can be engineered and formulated to reflect more heat and UV radiation than minimally processed kaolin so that SI can be effectively reduced. Particle film technology applied the knowledge of particle processing to the problem of SI and provided a tool to reduce or eliminate evaporative cooling of horticultural crops.

IV. DISEASE CONTROL WITH MINERAL AND PARTICLE FILM MATERIALS

Lime, sulfur, and lime-sulfur affect plant pathogens through chemical mechanisms (Secoy and Smith 1983). There are numerous citations of pH-altering minerals that are effective fungicides and include the common water-soluble minerals: hydrated lime, monopotassium phosphate,

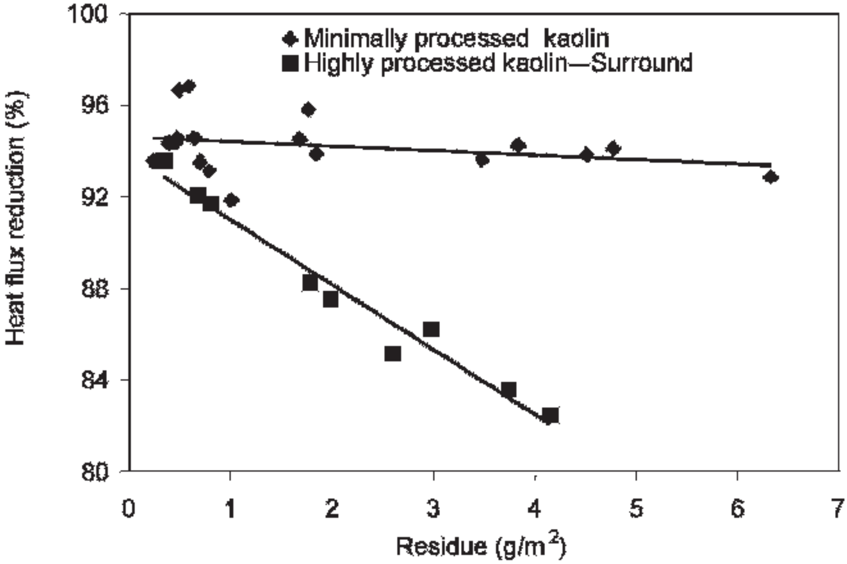

Fig. 1.7. Heat flux of highly processed kaolin, Surround® WP, and a minimally processed kaolin.

and various silicates, carbonates, and bicarbonates (Horst et al. 1992; Ziv and Zitter 1992; McAvoy and Bible 1996; Spotts et al. 1997; Reuveni et al. 1998a,b; Washington et al. 1998; Olivier et al. 1998; and citations included therein). Neinhuis and Barthlott (1997) examined over 200 plant species and found that a common plant adaptation that appears to suppress disease infection is production of water-repellent plant surfaces. Water-repellent surfaces facilitate the removal of particulate depositions (spores, conidia, hyphae) through the deposition and subsequent runoff of rain, fog, or dew. A cleansing action occurs when particulate depositions adhere to water droplets and are carried off the plant surface. Glenn et al. (1999) mimicked this mechanism by applying hydrophobic kaolin (M96-018) to plants in order to develop an artificially hydrophobic plant surface that would repel water. In single-leaf laboratory studies, fungal infection could be completely eliminated; however, on the whole plant and field plot scale, studies found complete coverage by the hydrophobic kaolin was impossible and so failed to control apple scab (*Venturia inaequalis*) (Puterka et al. 2000). Fabrea leaf spot (*Fabreae maculata* Atkinson) of pear was suppressed by both hydrophobic and hydrophilic kaolin particles, presumably through both a physical interference in the infection process and a lack of adherence of inoculum to the plant surface (Puterka et al. 2000). Powdery mildew in squash was suppressed by whitewash (Marco et al. 1994), in cucumber and grape by a chlorite-mica clay (Ehret et al. 2001), and in apple by processed kaolin (Glenn et al. 2001b). The bacterial disease, Fireblight (*Erwinia amylovora*), has been suppressed by both hydrophobic (Glenn et al. 1999) and hydrophilic kaolin particles (Glenn et al. 2001b) applied to flowers under conditions of artificial inoculation in greenhouse and field studies. Surround® also suppressed fireblight blossom infection under natural infection conditions. Surround® applications (3%) to 10-year-old 'Jonathan' apple trees 3 days prior to an infection event, the day of the infection, and 3 days following infection, reduced blossom blight from 28% in the untreated treatment to 5% in the Surround® treatment (N=21, $P \leq 0.05$). The mechanism of action is probably a physical interference of the initial infection of the hypanthium. Based on these results (Glenn et al. 1999, 2001b; Puterka et al. 2000), particle film technology has the potential to suppress some bacterial and fungal diseases; however, the environmental conditions and treatment timing have not been thoroughly documented.

V. FUTURE USES OF PARTICLE FILM TECHNOLOGY IN AGRICULTURE

A. Pest Control

The concept of manipulating inert mineral particles to alter plant surfaces for pest control and to improve the physiological properties of the crop has been expanded to address other problems in agriculture. Pesticide concentrations can be reduced by 50% when combined with particle films as a pesticide delivery system that provides the efficacy of a full rate of that pesticide (Puterka et al. 2003b). The delivery system utilizes the combined effects of improved plant coverage, attachment of pesticide coated particles to insects, combined action of quick knockdown with insecticides, and a durable physical barrier to insects.

B. Freeze Prevention

In another application, growth chamber and field studies have established that M96-018 hydrophobic particle films can prevent plant freezing by physically separating dew or frost from the plant surface and thus allowing the plant to supercool and not suffer freeze damage (Glenn et al. 2001). A hydrophobic particle film has effectively prevented ice nucleation and freeze damage (Wisniewski et al. 2002; Fuller et al. 2003) in whole tomato plants. The mechanism of action was the physical separation of water from the plant surface. When water freezes on the surface of a plant, it initiates ice nucleation within the plant by the physical growth of ice crystals into the internal portion of the plant. Growth of an ice crystal from outside the plant occurs through stomates, cracks in the cuticle, wounds, broken epidermal hairs, or other lesions. Blocking the activity of extrinsic nucleators and the subsequent growth of ice crystals into the plant allows the plant to supercool and provides some freeze protection. Further development will be required, but the potential to protect crops from spring frosts has tremendous potential in agriculture.

C. Improved Fruit Finish

Fruit finish has been improved by the application of both hydrophobic and hydrophilic kaolin. Glenn et al. (2001c) reduced russet in 'Golden Delicious' apple with a hydrophobic formulation and Fallahi (2003, unpubl. data) documented significant reduction of 'Fuji' russetting by Surround® in Idaho. In a 3-year study, russet in 'Comice' pear was reduced by both Mancozeb and Surround® WP applications with greater russet reduction when the two were combined. Applications were made at petal fall, 2 and 4 weeks after petal fall (unpublished data, David Sugar, Oregon State University, Medford, Oregon). The mechanism of action has not been identified at this time but suggests an interference with microbial activity on the fruit surface since apple russeting has been linked to epiphytic microbial populations (Matteson Heidenreich et al. 1997).

D. Conclusion

Particle film technology is based on the mineral kaolin, which has a long history of human safety from uses in pottery, paper, paints, and food processing and it is also used as a food additive. In principle, the inert particle film coating a plant creates a hostile environment for insects and a physical barrier to infestation, impeding insect movement, feeding, and egg-laying. The underlying mechanisms of this technology, which we have reviewed in this article, make it unlikely that insects will develop resistance. This particle film also acts as a physical barrier to prevent disease by separating the inoculum from the plant surface. The particle film allows the exchange of gases from the leaf during photosynthesis and transpiration, while its reflective properties reduce heat stress and increase photosynthesis, and fruit size and yield. Sunburn control of fruits currently relies on shade cloth materials or the extensive use of irrigation water for evaporative cooling of sensitive fruit. 'Surround® Crop protectant' is the first spray-on material to provide effective suppression of high heat damage and sunburn without the use of shade screens or evaporative cooling. In this way, particle film technology reduces the dependence of agriculture on expensive screens or irrigation water sources to mitigate heat stress.

Particle film technology has already displaced a significant percentage of the organophosphate and carbamate insecticides in pear and grape and has the potential to greatly reduce conventional insecticide usage in agriculture as mandated by the Food Quality Protection Act of 1996. In organic agriculture, particle film technology represents the first environmentally friendly, multi-functional material that provides effective insect control, mitigates stress, and produces high-quality organic fruits and vegetables. Its adoption by organic growers will further increase the growth of this expanding industry in the United States and globally. Commercialization of this concept has met with rapid acceptance in the agricultural industry. The broad effectiveness of particle film technology in controlling a large variety of insect pests will result in a global impact on agricultural production and reduced pesticide usage. As research and development on the various aspects of particle film technology continues, the mechanisms of how particle films affect pests and plants will become better understood. Based on the impact that this technology has had in only a few years, particle film technology could have a significant impact on crop production practices in the future, which could lead to reduced pesticide usage and improved yields.

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