CHAPTER II
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2.1 Literature Review on Insect Repellents and Their controlled Release Formulations

2.1.1. Chemically derived Insect Repellents

In general, the chemical repellents have a broader spectrum of efficacy and a greater duration of action than botanical repellents. Bueschr et al [1] reported that 50% DEET provide only about 4 hrs protection against Aedes aegypti mosquitoes, but increasing the DEET concentration to 100% provided only 1 additional hour of protection. In another study, 12.5% DEET provided over 6 hours of protection against Aedes albopictus; doubling the DEET concentration to 25% increased the protection time only to about 8 hours [2].

Literature reported the application of permithrin directly to clothing or other fabrics such as tent wall [3] or mosquito nets [4]. The combination of permithrin treated clothing and skin application of DEET based repellent produced a formidable barrier against mosquito bites had also been reported in the literature [5, 6]. Dimethyl phthalate used as a repellent against the ticks responsible for Lyme disease was reported [7].

In field studies of tick repellents, indalone was an effective repellent and appeared to be safe for use either on clothing or on direct application to the skin.
Field test of DEPA with *Culex quinquefasciatus*, *Simulium himalayense* and the leech *Haemadipsa zeylanica* showed 1.5, 2 and 1.5 hours of complete protection, respectively. [8]. DEPA had been formulated in a commercial preparation by the Defence Research and Development Establishment (DRDE).

In one laboratory comparative study of the efficacy of insect repellents against mosquito bites, Avon Corporation's IR3535 (3-[N-butyl-N-acetyl]-aminopropionic acid)-based 7.5% repellent provided an average complete protection time of only about 23 minutes (range, 10-60 min) [9]. In another laboratory and field study, Thavara and his coworkers [10] conducted evaluations of IR 3535 in Thailand. In the laboratory, both IR3535 and deet showed equal repellency (P>0.05) for 9.8 and 9.7 h against *Aedes aegypti*, for 13.7 and 12.7 h against *Culex quinquefasciatus*, and for 14.8 and 14.5 h against *Cx. tritaeniorhynchus*, respectively.

Yap et.al [11] reported the field efficacy of Bayrepel® i.e., 1-piperidine carboxylic acid, 2(2-hydroxyethyl)-1-methylpropylester against vector mosquitoes.

Research on A13-35765 showed that it had similar efficacy to DEET against *Anopheles albimanus*, *An freeborni*, *An gambiae*, *An. stephensi* and *Phlebotomus papatasi* [12], *Prosimulium* and *P. fuscum* [13], *Anopheles stephensi* and *Culex quinquefasciatus* [14] as well as *Culex p nipiens* both in the laboratory and the field [12].

The effectiveness of a repellent was reported to be decreased by the abrasion from the clothing, evaporation and absorption by the skin surface, wash-off from the sweat or rains, higher temperature, windy environment etc [15-17].
2.1.2. Plant Derived Insect Repellents

Citronella oil (C. nardus) consists mainly of citronellal (3,7-dimethyl-oct-6-enal) and geraniol (3,7-dimethyl-octa-2,6(2,7)-dien-1-ol)\(^1\). Both the components were found to be effective alone \([19,20]\). Curtis et al \([18]\) calculated the ED\(_{50}\) of citronella in laboratory experiments to be 11.8 \(\text{nl/cm}^2\) for *Anopheles stephensi*, 20.2 \(\text{nl/cm}^2\) for *Anopheles albimanus*, and 42.1 \(\text{nl/cm}^2\) for *An. Gambiae*, which are similar to the effective doses of freshly applied DEET.

Several articles have multiscreened a number of essential oils for mosquito repellency with mixed outcomes. Twenty essential oils were screened by Cora et al. \([21]\) against the mosquito (*A. aegypti*), fruit fly (*Drosophila domestica*), and the house fly (*Musca domestica*), where the oils of *Juniperus communis*, *Valeriana officinalis*, *Thymus vulgaris*, *Solidago graminifolia*, *sylvestris*, *Coriandrum sativum*, *Larix decidua*, *Pseudotsuga menziesii*, *Tanacetum vulgare* and *Abies alba* were all found to be highly repellent to mosquitoes. Tawatsin et al. \([22]\) investigated the repellency effects of essential oils of turmeric (*Curcuma longa*), keffir lime (*Citrus hystrix*), citronella grass (*Cymbopogon winterianus*) and hairy basil (*Ocimum americanum*), against the mosquitoes *A. aegypti*, *Anopheles dirus* and *C. quinquefasciatus* both in caged and open room conditions. Tumeric, citronella and hairy basil oils especially with the addition of 5% vanillin, repelled the three species under cage conditions for up to eight hours, comparing well against the DEET standard, which provided protection for at least eight hours against *A. aegypti* and *C. quinquefasciatus*. Amer and Mehlorn et al \([23]\) studied the repellency of 41 essential oils and their mixture against *Aedes aegypti*, *Anopheles*
*stephensi* and *Cx. quinquefasciatus* using the skin of human volunteers to find the protection time and repellency. Trongtokit et al [24] studied the mosquito repellent activity of 38 essential oils from plants at three concentrations (10%, 50%, 100%) against the mosquito *Aedes aegypti* under laboratory conditions using human subjects. When the tested oils were applied at a 10% or 50% concentration, none of them prevented mosquito bites for as long as 2 h, but the undiluted oils of *Cymbopogon nardus* (citronella), *Pogostemon cablin* (patchuli), *Syzygium aromaticum* (clove) and *Zanthoxylum limonella* were the most effective and provided 2 h of complete repellency.

Tiwari [25] tested lemongrass oil against the housefly *Musca nebulo* and female mosquitoes of *Culex fatigans* and *Ae. aegypti* and reported that lemongrass oil was less efficient than dimethyl phthalate and protection lasted only 40 min as opposed to 300 min for dimethyl phthalate. Ansari and Razdan [26] studied the effects of *Cymbopogon martini* var. *sofia*, *C. citratus*, *C. nardus* and *Cinnamomum camphora* against local Indian mosquito species (*Anopheles cujicifacies* and *Cx. quinquefasciatus*) by exposing the feet, forearms and faces of local volunteers from dawn till dusk, dosed with 1 ml of each of the oils. Repellency of each of the *Cymbopogon* oils lasted 11 h against *A. cujicifacies* and 6–7 h against *C. quinquefasciatus*, (but less for the *Cinnamomum* oil). The oils were found to be comparable in efficacy to dimethyl phthalate and dibutyl phthalate. Oyedele et al [27] studied the ointment and cream formulations of lemongrass oil in different classes of base and the oil in liquid paraffin solution for mosquito repellency. The 1% v/v solution and 15% v/w cream and ointment preparations of the oil exhibited 50% repellency lasting 2–3 h. This activity was comparable to that of a commercial mosquito repellent.
Li et al [28] tested extract of eucalyptus oil on *Aedes aegypti* mosquitoes and found that the protection only lasted for 1h. However, p-manthane-3, 8-diol (PMD) discovered in the waste distillate of the extract of the lemon eucalyptus plant was determined to be the active ingredient for the repellent activity of mosquitoes. Laboratory studies by Trigg and Hill showed that 30% PMD was almost as effective as DEET, the most widely available synthetic repellent [29,30].

Choi et al[31] investigated the essential oil of *Lavandula officinalis* as well as other oils such as *Eucalyptus globules, Rosemarinus officinalis* and *T.vulgaris* for their individual repellent activities against *Culex pipiens* and found that all the named oils actively repelled adult mosquitoes on hairless mice, with thyme oil showing particularly good activity in the confines of the protocol, giving a protection rate of 91% at 0.05% concentration in topical treatment, significantly extending the duration of protection.

Several species from genus *Ocimum* had been popularly used to repel or kill insects. *Ocimum americanum* (synonym: *Ocimum canum*) and *Ocimum basilicum*, for instance, have been widely employed in Africa [32]. *Ocimum* spp. leaves were reported to be rubbed on the skin as a method of repelling mosquitoes [33]. Seyoum et al.[34] reported that thermal expulsion and or direct burning of *Ocimum americanum, O. kilimandscharicum* and *Ocimum suave* were effective in repelling *Anopheles gambiae* in experimental huts within a screen-walled greenhouse. Seyoum et al [35] demonstrated that potted *Ocimum americanum* repelled *Anopheles gambiae* in experimental huts under semi-field conditions. Furthermore, *Ocimum americanum* volatile oil was shown to repel *Aedes aegypti, Anopheles dirus* and *Culex quiquefasciatus*, under cage conditions up to
Interestingly, when mixed with 5% vanillin, the protection times increased greatly for each mosquito species since it reduces the evaporation rates of repellents [36].

Jeyabal et al [37] considered methanol extracts of Pelargonium citrosa leaf, testing for biological, larvicidal, pupicidal, adulticidal, antiovipositional activity, repellency and biting deterrency against *An. stephensi*. At 4%, the extracts evoked strong repellent action.

Ezeonu et al. [38] used statistical studies of randomized complete block design with four replicates to show that the volatile peel extracts of *Citrus sinensis* (sweet orange) and *C. aurantifolia* (lime) possessed insecticidal activities against mosquitoes, cockroaches and houseflies.

A complete protection for 12h from the bites of all the anopheline mosquitoes species was reported by using 2% neem oil in coconut oil on the exposed part of the body [39]. However, Rajnikant and Bhatt [40] reported only 89 and 98% protection against *An. fluviatilis* and *An. culicifacies* respectively and only 68% protection against all anopheline species by using 2% neem oil. The protection from Culex and Aedes mosquitoes ranged between 76-86%. But Moore et al [41] did not find any significant protection from *An Darlingi* by using 2% neem oil, while a eucalyptus based repellent provided 96% protection for 12h. Prakash et al. [42] recorded 66.7% protection after 9 h using 2% neem oil diluted in mustard oil. Vanishing cream with 5% neem oil also provided 67 to 100% protection against malaria mosquitoes in different terrains in India [43-45]. Application of the neem cream for protection against mosquitoes was more acceptable because of its easy application, pleasant odor and more effective repellency up to 4 h after the application.
Several plant essential oils which had been demonstrated to exhibit good repellent activities against mosquitoes were Conyza newii, Plectranthus marrubioides, Tetradenia riparia, Tarchonanthus camphoratus, Lippia javanica and Lippia ukambensis [46], Artemisia vulgaris [47], Lantana camara [48,34,49], Zanthoxylum limonella [50], Vitex rotundifolia [51-52], Zanthoxylum piperitum [53], Ocimum selloi oil[54], Eucalyptus camaldulensis, Mentha piperita, Ocimum basilicum, Laurus nobilis [55], Curcuma spp.[56], Cedrus atlantica or Juniperus spp. (cedar, juniper) [57], Pinus sylvestris [57], Syzygium aromaticum (cloves)[57], Tanacetum vulgare (thujone)[58] etc.

2.1.3. Microcapsules / microspheres for Controlled Release formulations

Polymeric Materials

The use of controlled release technology with topical repellents provided extended protection against mosquitoes as reported by Gupta and Rutledge [17]. Formulations based on creams, polymer mixtures or microcapsules could prolong the effectiveness of repellents. Mixing of DEET with polymers increased its water-resistance fourfold and the duration of its effect against Aedes aegypti by 2.5 [59]. The duration of the effect against Aedes aegypti and Anopheles albimanus with microcapsule formulations was also up to 24 h longer than that of simple lotions [60]. The efficacy of a 35% DEET cream formulation lasted longer than that of a 75% lotion against Culex sitiens and Aedes vigilax [61] against Anopheles flavirostris[62]. A polymer cream formulation (33% Deet) and a microparticle formulation (42% DEET), tested against Aedes aegypti, Aedes taeniorhynchus, Anopheles albimanus and Anopheles stephensi,
were superior to the 75% lotion. The microparticle formulation had a longer lasting effect against *Aedes aegypti* and *Aedes taeniorhynchus* than polymer and microcapsule formulations [17].

Slow release microcapsule based formulations containing DEET considerably reduce dermal absorption [63]. In an effort to develop a new topical formulation, a liposphere lotion formulation (20%) and an alcohol solution (20%) with two insect repellents DEET and DEPA were evaluated for the extent of protection on rabbits against *aedes aegypti* [64]. The lotion formulation of DEPA and DEET were found to enhance the repellency by 1.5 and 1.25 times respectively compared to the alcohol solution of the repellents. Gupta et al. [65] reported that the protection time of DEPA against mosquitoes may be increased by developing its slow release formulations using polysiloxanes as matrix systems.

The 3M Company (St. Paul, Minnesota) developed a slow-release, polymer-based product containing 35% DEET. When tested under laboratory and several different environmental and climatic field conditions, the 35% DEET polymer formulation by the 3M Corporation was as effective as 75% DEET in repelling mosquitoes [66-68]. One study showed that Minnetonka Brands' 6.5% liposphere microdispersion of DEET was effective for up to 2.5 h and that their 10% product was effective for about 1 h longer [68].

Urea-formaldehyde microcapsules containing lemon oil were prepared by in situ interfacial polymerization. The particle size and their distribution under different experimental conditions were measured and reported [69]. Thimma and Tammishetti [70] studied the complex coacervation of gelatin with carboxymethyl guargum and applied it
for microencapsulation of clove oil and sulphonmethoxazole. Microcapsules containing fragrant oil were synthesized by in situ polymerization method and the microencapsulation efficiency and other physical properties were analyzed by Lee and his coworkers [71]. Heng et al [72] studied the encapsulation of wheatgerm oil and evening primrose oil using sodium alginate by emulsification method. They also investigated the physical appearance of microspheres, amount of oil to be encapsulated, flow property, size distribution and mean size of microsphere produced. Bachtsi et al [73] synthesized and studied the release behaviour of oil from oil containing poly(vinyl alcohol) microcapsules prepared by simple coacervation technique. Rosenblat et al [74] studied the effect of electrolytes, stirring rate and surfactant concentration on coacervation and microencapsulation process of gelatin. Effect of wall thickness of microcapsule on the release characteristics was studied and reported by Madan [75]. Sun et al [76] prepared a series of gelatin microspheres by emulsification–coacervation method and studied the influence of preparation parameters like concentration of gelatin, emulsifier, emulsifying time, stirring speed etc. on particle size, surface morphology and dispersion of gelatin microspheres. Shu and Zhu [77] studied the interaction of chitosan with three kinds of anion (tripolyphosphate, citrate and sulphate) by turbidimetric titration and reported that the electrostatic interaction took place in a certain region of solutions. Yan et al [78] prepared crosslinked chitosan / poly (vinyl alcohol) blend with high mechanical strength. Chitosan microspheres were prepared using sodium sulphate as precipitant. The microspheres were loaded with drugs and the loading property was investigated by spectrophotometry. The loaded microsphere were characterized by SEM and DSC [79].
Mani and Jun [80] developed a method, employing salt and wetting agents, to improve the loading efficiency of a water-soluble drug. Microencapsulation of hexadecane in a vegetable protein by salting out method and the effect of different process parameters on microcapsules characteristics were investigated by Mauguet et al [81]. Kulkarni et al [82] reported the use of urea formaldehyde resin for the controlled release of diclofenac sodium. Iwanga [83] and coworkers studied the release rate of insulin from gelatin microspheres with crosslinking densities. The release rate of insulin showed initially a burst effect followed by a slow release phase regardless of the crosslinking density.

**Crosslinking Agents**

Aminabhavi et al [84] studied the effect of various crosslinking agents on the release behaviour of diclofenac sodium encapsulated chitosan microspheres. The effect of crosslinking agent on the release of lactic acid from gelatin microsphere was studied by Dinarvand et al [85]. Varieties of crosslinking agents like glutaraldehyde [86], formaldehyde [87], epoxy compounds [88] were reported to be employed for improving controlled release behaviour of controlled release polymer. Genipin, a natural crosslinker whose cytotoxicity, feasibilility and biocompatibility were studied and reported [89,90]. Sung et al [5] reported that genipin was 10,000 times less toxic than glutaraldehyde. Genipin crosslinked alginate-chitosan microcapsules for live cell encapsulation was reported by Chen et al [91]. In another study, Chen et al [92] investigated the fluorogenic characteristics of chitosan-genipin reaction for microencapsulation purposes.
Deacetylated Chitosan

Li et al [93] reviewed the various aspects of chitosan and reported that the degree of deacetylation (DDA) could be employed to differentiate between chitin and chitosan. Baxter et al [94] reported that an increase in either temperature or strength of sodium hydroxide solution could enhance the removal of acetyl groups from chitin, resulting in a range of chitosan molecules with different properties. Different methods were available in the literature for characterization of degree of deacetylation of chitosan. These were ninhydrin test [95], infrared spectroscopy [96], linear potentiometric titration [97,98], circular dichroism spectroscopy [99], NMR [100,101], UV spectroscopy [102], elemental analysis [103] etc. Spherical beads of chitosan, crosslinked with different concentration of glutaraldehyde were prepared and used for controlled release of drugs [104]. The effect of molecular weight and degree of deacetylation of chitosan on inherent viscosity, crystallinity and swelling were evaluated by Taghizadeh and Davari [105].

2.2. Literature Review on Controlled Release Agrochemicals

The different classes of polymers viz., elastomers, plastics and fibres were extensively used in agriculture for varied purposes. The major application fields included CR pesticides, herbicides and fertilizers, soil conditioning, plant protection, seed coating and gel planting [106].

Several pesticides like sevin, dimethoate, ethyl trithion, methyl trithion, diazinon, malathion, chloropyrifos and temephos could be incorporated in plasticized poly(vinyl chloride) to obtain CR products [107].
El-Refaie and coworkers [108] prepared controlled release formulations based on crosslinked polyacrylamide derivatives. The release data of the herbicide 2,4-D in vitro from formulations were described.

Micro-or macro encapsulation of active agents using polymers is one of the methods widely used for the preparation of CR products. Condensation polymerization reactions yielding polyamide, polyester, polyurea, polyurethane, polycarbonate and polysulphonamide could be well utilized to prepare CR formulations. Crosslinking of the polymer wall provided durable and storage stable capsules [109,110]. Several controlled release pheromone formulations were also synthesized by microencapsulation. The utilization of starch as a polymer matrix for CR agrochemical was reported [111].

Pfister, Bahadir and Korte [112] claimed another system based on calcium alginate with a series of herbicides. Starch was used as an encapsulating material for S-ethyldipropylthiocarbamate (EPTC), atrazine and trifluralin [113-116]. Teft and Friend [117] synthesized controlled-release polymeric microspheres of herbicides DiChamba(DA) based on ethylcellulose, polyarylsulfone or a combination of the two.

Kulkarni, Kumbar, Dave and Aminabhavi [118] reported the release kinetics and encapsulation efficiency of urea-formaldehyde (UF) crosslinked matrices of starch, guar gum (GG) and starch+guar gum (St+GG) for controlled release of solid (chloropyrifos) and liquid (neem seed oil) pesticides. In another report, Kulkarni and his group [119] claimed the synthesis of novel polymeric sodium alginate interpenetrating network (IPN) beads for the controlled release of chloropyrifos. They also synthesized IPN beads of poly(vinylalcohol)-g-poly(acrylamide) with sodium alginate for the controlled release of cypermethrin pesticide[120].
Marei et al. [121] compared carbofuran encapsulated controlled release formulation with the granular formulation in terms of mobility of carbofuran and reported that leaching potential of alginate formulation decreased more than nine times compared with granular formulation. Chitosan gel beads and film were assessed for their ability to control the release of herbicide atrazine and fertilizer urea [122]. Elabahni et al. [123] developed a technique for encapsulation of herbicide inside ethyl cellulose microsphere and evaluated the shape and size of microspheres by scanning electron microscopy. Polysaccharides like cellulose, chitin, amylose and amylopectin were found to be useful natural polymers for the CR formulations of 2,4-dichlorophenoxyacetic acid and metribuzin [106]. CR formulation of kraft lignin and propachlor had been successfully prepared by Wilkins and Blackmore [124]. It was reported that rice husk lignin could be combined with 2,4-dichlorophenoxyacetic acid [125]. The application of lignin in CR formulations was reviewed by Wilkins [126].

Zhu and Zhu [127] synthesized a new starch-g-poly(butylacrylate) for encapsulating carboxylic group containing herbicides. Polymerizable derivatives of pesticides containing acid groups could be prepared by a reaction with alcohols having a vinyl group [128,129]. Copolymers of vinyl 2,4-dichlorophenoxyacetate and trimethyl amine methacrylamide were reported to be used for CR application [125]. Increased release of herbicide was obtained as the hydrophilic co-monomer content increased.

Kenawy and his group [130] prepared controlled release systems based on polyureas and poly(Schiff's bases). The effects of structure and temperature of the aqueous environment on the hydrolysis rate of the obtained polymer had been reported. Cheillini and Akelah [131] synthesized polymeric herbicides containing 2,4-D and
MCPA by modification of oligoethylenoxideylated styrene/divinylbenzene(DVB) resins. The release features for these systems were greatly affected by the pH.

Akelah et al. [132] reported chemical modifications of a series of polyamides containing hydroxyl groups with 2,4-D in the presence of dicyclohexylcarbodiimide(DCC) as a condensating agent to yield a series of polymer. They reported that the rates of release of 2,4-D from the formulations were mainly dependent on hydrophilicity, the pH and the temperature of the release medium.

Pesticides containing acid groups were converted to more reactive acid chlorides, which could react with polymers containing pendant hydroxyl or amino groups. Acylation of synthetic and natural polymers were possible in this manner [133-136]. Pentachlorophenol intercalated on mineral clay was reported by Akelah and Rehab [137]. The release of pentachlorophenol from the formulations was studied in different media at 30°C and it was concluded that the release of pentachlorophenol from the formulations was dependent on the structure, swelling degree and the medium of release.

A series of preformed polymers modified with pesticides were reported [125]. Chitin [138-140] as a naturally occurring polymer was used as carrier for herbicide metribuzin and the system showed slow release when polymer was directly attached to metribuzin.

Fertilizers

Development of synthetic CR fertilizers had been conducted, mainly with nitrogen sources for many years [137]
Based on the synthetic method and nature of the products CR or slow release fertilizers are divided into the following categories:

a) Slowly Releasing Organic-N Compounds

Urea formaldehyde (UF) is the most popular organic-N compound used for the slow release of nitrogen, and the most widely used of all SRF/CRFs [140-141]. The release of N from these compounds thus depends strongly on soil properties such as biological activity, clay content, pH, and external conditions such as moisture content, wetting and drying, and temperature [140,142-144]. Additional nitrogen compounds based on the reaction of urea with other aldehydes were cited in the literature [140,143,145].

b) Coated Fertilizers

Resin, plastic, lac, silica, sulphur, natural rubber, polyolefin, starch and gypsum were reported to be used for preparing CR urea fertilizers [111,146-148]. The preparation of sulphur and gypsum coated urea (SCU) were reported by several workers [149-153]. The use of polymers to the sulfur-coated urea for improving attrition resistance of coated granules was reported [154].

The alkyd-type coated fertilizer was reported by Lambie [155] and Goertz [143]. Moore [156,157] developed a polyurethane-like coating on fertilizer. The release of nutrients from these products was mainly temperature dependent, while moisture content
in the soil, pH, wetting and drying, and soil microbial activity have little effect on the release [157-159].

Coating of granular fertilizers with thermoplastic materials such as polyethylene by dissolving the coating material in a chlorinated hydrocarbon and spraying it on the granules in a fluidized bed reactor was reported in the literature [160-163].

Polyolefin-coated urea fertilizer (POCU) was developed in Japan [148]. Several types of POCU with varying release rates were reported to be commercially available in Japan. Release was reported to be less sensitive to soil pH and soil moisture [148].

Korean Advanced Institute of Science and Technology (KAIST) had developed several batches of Silicate and Polymer Coated Urea (SPCU) and they had observed satisfactory results for their product on rice [164]. The dissolution rate was adjusted by varying the thickness of coating. The polymer latex used for coating was prepared with styrene, 2-ethylhexyl acrylate and acrylic acid.

c) Matrix-Based Slow Release Fertilizers

Efforts have been made to prepare SRFs or CRFs by mixing nutrients with materials that reduce their dissolution rate. Different materials were used for this purpose: rubber [165], gel-based materials [166], and thermoplastic polymers. Natural rubber based slow release fertilizer had been formulated by Hepburn et al, [172]. It was reported that the crosslinking influenced the rate of release of fertilizer from the matrix.

CR fertilizers were made by using water-absorbing polymers as a carrier for fertilizer solutions. Smith and Harrison [168] had carried out experiments on acrylate, vinyl alcohol and starch based polymers that can be expanded in fertilizer solution. Wu et
al. [169] prepared a double coated slow release NPK with a superabsorbent and water retaining crosslinked poly(acrylic acid)/diatomite-containing urea granules (the outer coating), chitosan (inner coating). The product had a water absorbency of 75 times its own weight if allowed to swell in tap water for 2 h.

2.3. Objectives and Plan of Work

Insect repellents are substances that protect animals, plants or products from insect attack by making food or living conditions unattractive. A large number of synthetic as well as herbal repellents are available. Both synthetic as well as herbal repellents have limited persistence on skin. The ideal repellent should have long-lasting effectiveness against a wide variety of arthropods. The use of controlled release technology with topical repellents has known to provide extended protection against mosquitoes. Controlled release formulation of an insect repellent can prolong the availability of the repellent on the treated surface by reducing the loss of repellent by absorption, evaporation and abrasion. Few reports are available regarding the development of controlled release formulation based on plant-based repellents. Plant-based repellents are ecofriendly, safe and lesser toxic compared to synthetic repellents.

The need for agrochemicals is absolute; thus, there exists an urgent necessity to improve the production and the use of active agrochemicals. This can be achieved by using and applying controlled release technology. Many of the controlled release formulations are highly efficient in sustaining the release of the biologically active components. It should be recognized that the polymeric material to be used in controlled release must degrade to some fashion before there can be any environmental impact in
the chemical, biochemical or biological sense. If polymers for use in controlled release were completely inert or their degradation rate was measured in geologic time, the cumulative aspect would be a matter of concern. However it is possible to use naturally occurring polymers or degradable synthetic polymers.

Keeping in view all the above backgrounds, the aim of the present work was set and the present work had been undertaken. In the present research work controlled release polymeric systems of an agrochemical, urea and an insect repellent based on plant essential oil, *Zanthoxylum Limonella* oil (ZLO) were developed and characterized. The polymeric materials used were naturally occurring polymers such as gelatin and chitosan. Microencapsulation technique was used for the synthesis of controlled release formulations of the agrochemical and insect repellent. Microencapsulation is a technique that reproducibly applies a uniformly thin polymeric coating around small solid particles, liquid droplets or solid dispersions. The polymeric wall is designed to permit controlled release of the encapsulated material under desired conditions. The release of the active agents can be controlled by crosslinking of the polymeric wall. Both synthetic and naturally occurring crosslinking agents were used in our present work. Synthetic and natural crosslinking agents used were glutaraldehyde and genipin, respectively.

The plan of work was divided into the followings:

- To develop and optimize the microencapsulation of active agents (mosquito repellent, urea) in crosslinked natural polymers for controlled release (CR) application.
• To study the effect of various parameters like concentration of active agents, polymer and crosslinking agent on percent encapsulation, active agent content and release rate.

• To characterize the encapsulated product by spectroscopy, differential scanning calorimetry (DSC), thermogravimetry and scanning electron microscopy.

• To study and compare the repellency of the developed CR formulations of mosquito repellent against standard repellent such as DEET.
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