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PREPARATION OF SOME PLANT CRUDE EXTRACTS AS SPRAYABLE AND DUST POWDER FORMULATIONS

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ABSTRACT

Crude extracts of different five plant varieties, *i.e. Artemisia alba* (wormwood), *Citrullus colocynthis* (Bitterapple), *Datura annoxia* (downy thornapple), *Urginia maritima* (Squill) and *Aloe vera* (aloe) were extracted and prepared as suitable formulations. The first four plant varieties were extracted using ethanol 95 %, n-hexane and chloroform, while the fifth one was obtained by cutting its leaves and collecting the bleeding juice. Some physico-chemical properties of these crude extracts, such as solubility and free acidity or alkalinity, were determined and consequently they were formulated as soluble liquids, and flowable suspensions or dust powders.

Results obtained indicated that the ethanolic extracts of *Artemisia herba alba, Citrullus colocynthis* pulp, *Datura annoxia* and *Urginia maritima* as well as that of *Aloe vera* showed good solubility in water. Therefore they were prepared as soluble liquid formulations. The rest crude extracts which did not possess such property were prepared as flowable suspension. Moreover, all the crude extracts were prepared as dust powders. Soluble liquid formulations showed good miscibility in water, flowable formulations exhibited good suspensibility and dust powder formulations were fine to degree less than 40 µ. All formulations showed suitable free acidity or alkalinity and passed successfully through heat storage test. Soluble liquid and flowable formulations may be used for controlling pests on vegetative growth plants, while dust powders may be utilized against stored grain pests.

INTRODUCTION

Since the last three decades, several researchers all over the world conducted researches on plant extracts as suitable alternatives of traditional pesticides against pests of economic importance. Such extracts contained several biologically active materials such as : alkaloids, saccharides (waches produce hydrocyamic acid), saponine and toxic acids such as tannic and cyanamic acids (El-Khadem and Abdel-Rahman, 1962 & 1965; Kloss and Schindler, 1966; Jermy *et al.*, 1981; Hussein and Yankov, 1981; El-Nomrossy, 1982; Saleh, 1984; Abid *et al.*, 1997; Chithra and Chandrataran, 1998; Pascual and Fermandez, 1999; Ramirez *et al.*, 1999; Kwon and Lee, 2001).

Most of researches, however, dealth with extraction, concentration, fractionation and identification of the included active materials. Besides, simple laboratory tests were conducted to evaluate the pesticidal effects of these extracts or their plant ingredients with no true attempts for preparing true alternatives suitable formulations for field application (Merz, 1987; Chouhan and Qadri, 1989; El-Naggar *et al.*, 1989; Niber *et al.*, 1992; Weaver *et al.*, 1995; Hatano *et al.*, 1999 and Wallaart *et al.*, 1999a & b).

MATERIALS AND METHODS

Materials :

1- Tested plants :

Five experimental plants were selected for this study, Wormwood (*Artemisia herba alba*), Bitterapple (*Citrullus colocynthis*), Downy thornapple (*Datura annoxia*), Squill (*Urginia maritima*), and aloe (*Aloe vera*) (Table 1). Several literatures found that these plants contain some biologically active materials (Table 2), which may utilize in the medicine purposes and possess some insecticidal activity.

Table (1): General information about tested plants.

No.	English name	Family name	Scientific name
1	Wormwood	Compositae	*Artemisia herba alba* (L.)
2	Bitterapple	Cucurbitaceae	*Citrullus colocynthis* (L.)
3	Downy thornapple	Solanaceae	*Datura annoxia*
4	Aloe	Liliaceae	*Aloe vera*
5	Squill	Liliaceae	*Urginia maritima*

Table (2): The biologically active materials presented in the experimental plants.

No.	Plant name	Biologically active components	Plant parts
1	*A. herba alba*	Alkaloids, phenol, glycosides, volatile oils, tannins, triterpenes, sterols, coumarins, flavonoids	Leaves
2	*C. colocynthis*	Pectin, saponins, alkaloids, resine, colocynthitin	Seeds & Pulp
3	*D. annoxia*	Atropine	Leaves & Roots
4	*U. maritima*	Steroidal glycosides, seillarin "a", seillarin "b"	Pulp
5	*A. vera*	Aloin (glycosides), anthraquinone	Leaves

2- Surfactants :

Two local surfactant agents were supplied by the Egyptian Company for Starch and Yeast Products. They are :

a- Polyethyleneglycol 600 monolaurate (600 m.l.)

b- Polyethyleneglycol 600 dilaurate (600 d.l.)

3- Diluents and carriers :

Aswanly clay and talc powder were supplied by El-Nasr Co. for Phosphate, Cairo, Egypt.

4- Chemicals :

Acetone 99 %, ethanol 95 %, chloroform 99.4 %, n-hexane, xylene, hydrochloric acid, sodium hydroxide and methyl red were supplied by El-Nasr Co. of Pharmaceutical Chemicals, Cairo, Egypt.

Methods :

1- Extraction of plant parts :

The desired plant parts from *A. herba alba, C. colocynthis, D. annoxia* and *U. maritima* were extracted using ethanol 95 %, while *A. vera* was obtained by cutting their leaves and collecting the bleeding juice.

The selected plant parts of each plant were ground in high speed grinder (FARIBON 300). Plant powders (250 g of each) were placed into bottle (2 L) and soaked in the solvent (1000 ml) for 48 hours. The contents of each bottle were thoroughly mixed using high speed mixer and the mixture was filtered through filter paper Whatman No. 1 and anhydrous sodium sulfate (Na_2SO_4) in Buchner funnel. The filtrate was evaporated by rotary evaporator under vacuum till dryness. The crude extract (residues) were transferred quantitatively into a small vial (15 ml) using 10 ml acetone and dried by air steam at room temperature and kept in freezer.

2- Physico-chemical properties of crude extracts :

a- Free acidity or alkalinity : Determined according to method of WHO specifications (1993).

b- Solubility : Determined by measuring the volume of distilled water, acetone and xylene in which 0.5 g of crude extracts may completely soluble or miscible, then percentage of solubility was calculated, according to method of WHO specifications (1993)..

3- Formulation of crude extracts :

Type of formulation is mainly depends on the solubility rate of crude extracts in the examined solvents. Crude extracts that showed high solubility in water were prepared as soluble liquid formulations, while those exhibited high solubility in xylene were prepared as flowable formulations. On the other hand, crude extracts which neither soluble in water or xylene were prepared as dust powder formulations.

a- Soluble liquid formulation :

Formulation was prepared by mixing 10 gm of the wetting agent "polyethyleneglycol-600 monolaurate" with 90 gm of crude extracts. Free acidity or alkalinity, cold and heat storage tests of these preparations were determined according to WHO (1993).

b- Flowable formulation :

30 % of plant extracts as flowable formulations were prepared according to the procedure outlined by Heath *et al.* (1983).

Physico-chemical characteristics of the prepared flowable formula-tions such as free acidity or alkalinity, heat stability test, freeze thaw stability test as well as their suspensibility rates were determined according to WHO specifications (1993). Suspensibility rate was calculated according to the following equation :

$$\% \text{ Suspensibility} = \frac{b - a}{b} \times 11.1$$

where :

a = weight of formulation in the retained one-tenth of suspension (g).
b = weight of formulation in the initial sample (g).

Freeze-thaw stability was determined by cooling the samples at 0.0°C for 8 hrs to check out the spontaneous flowability of product. Suspensibility test was carried out after thawing.

c- Dust powder formulations :

Crude extracts of tested plants were prepared as dust formulation containing 3 % crude by mixing solution of crude extracts in a volatile solvent (acetone) with suitable diluent (talc) 97 %. After precise mixing and complete evaporation of acetone, prepared dusts were sieved using 40 µ sieve to obtain a dust of particle size less than 40 µ and their pH values were determined according to Watkins and Norton (1955).

RESULTS AND DISCUSSION

1- Extraction of plant parts using different solvents :

The four plant varieties, *i.e.*, *Citrullus colocynthis* (L.), *Artemisia herba alba* (L.), *Datura annoxia* and *Urginia maritima* were extracted by three solvents; ethanol 95 %, n-hexane and chloroform, while the crude extract of *A. vera* was obtained by cutting the plnt leaves and collecting the bleeding juice. The percentages of crude extracts obtained from examined plants were calculated and listed in Table (3). Results obtained indicated that *U. maritima*/eth. extract gave the highest yield of extraction (38.9 %), followed by *C. colocynthis* seed/eth. (30.05 %), *C. colocynthis* seed/chloro. (25.74 %), *C. colocynthis*/seed/hex. (18.78 %), *C. colocynthis* pulp/eth. (17.0 %), *A. herba alba*/eth. (8.94 %), *Datura*/eth. (7.15 %), *A. herba alba*/hex. (4.66 %, and *A. herba alba*/chloro. (3.23 %) extracts. On the other hand, *C. colocynthis* pulp/hex. and pulp chloro. gave the lowest extarction percentage yields 2 and 3 %, respectively, which unsuitable values to be formulated from economic considerations.

Table (3): Percentages of crude extracts with different solvents.

No.	Plant	Solvent	% Yield
1	*Artemisia herba alba*	Eth.	8.99
2		Hex.	4.66
3		Chloro.	3.23
4	Seed	Eth.	30.05
5	*Citrullus colocynthis*	Hex.	18.78
6		Chloro.	25.74
7	Pulp	Eth.	17.0
8	*Citrullus colocynthis*	Hex.	2.0
9		Chloro.	3.0
10	*Datura annoxia*	Eth.	7.15
11	*Urginia maritima*	Eth.	38.9

2- Physico-chemical properties of crude extracts :

2.1- Solubility :

It was determined by measuring the volume of distilled water, acetone and xylene required to complete solubility or miscibility of 0.5 gram of crude extracts. Table (4) shows that ethanolic extract of *U. maritima* showed the highest solubility in water followed by ethanolic extract of the pulp of *C. colocynthis*, while the rest extracts showed low or not solubility water. The hexane extract of seeds of *C. colocynthis* exhibited the highest solubility in xylene comparing with the other crude extracts which showed low or not solubility in xylene. Hexane and ethanol extracts of the seeds and pulp of *C. colocynthis*, respectively, declared the highest solubility in acetone (50 %), followed by ethanolic extract of *U. maritima* (20 %), while the other crude extracts showed low or no solubility in acetone. Crude extracts of high solubility in water should be prepared as soluble liquid formulations (S.L.), while those did not soluble in water should be prepared as flowable formulation (F.L.). On the other hand, crude extracts exhibited high solubility in acetone (the high volatile solvent) may be prepared as dust powder formulations.

2.2- Free acidity and alkalinity :

Results shown in Table (4) indicated that *C. colocynthis* seed/ hexane extract was neutral, while the rest crude extracts were acidic.

C. colocynthis pulp/ethanol extract was the highest acidic one followed by those of *Aloe vera*/ethanol, *D. annoxia*/ethanol,and *A. herba alba*/hexane, while that of *C. colocynthis* seed/hehsane was the lowest acidic one.

Results obtained indicated that only neutral or slightly acidic carriers, solvents, emulsifiers but not any only alkaline additives, should be used for preparing suitable formulations from these crude extracts.

Table (4): Some physico-chemical properties of different plant crude extracts.

Type of crude extracts	% Solubility (wt./v.) in			Free acidity as % H_2SO_4	Free Alkalinity as % NaOH
	Water	Xylene	Acetone		
C. colocynthis pulp/eth.	25	-	50	1.070	-
A. vera/ethanol	10	-	10	0.490	-
D. annoxia/eth.	10	-	10	0.390	-
A. herba alba/hex.	-	3.0	1.9	0.370	-
C. colocynthis seed/eth.	-	1.3	-	0.294	-
A. herba alba/chloro.	-	2.0	10.0	0.280	-
U. maritima	100	-	20	0.210	-
A. herba alba/eth.	10	1.6	2.0	0.196	-
C. colocynthis seed/chloro.	-	2.5	10	0.050	-
C. colocynthis seed/hex.	-	33.3	50	0.040	-

3- Preparation of crude extracts as suitable formulations :

3.1- Soluble liquid formulation :

Since *A. vera*, *U. maritima*/ethanol, *D. annoxia*/ethanol, *C. colocynthis pulp*/ethanol and *A. herba alba*/ethanol extracts showed suitable solubility in water, therefore, they were prepared as soluble liquid formulations, by mixing 10 % slightly acidic wetting agent (polyethyleneglycol 600 monolaurate) with 90 % of these extracts. Results in Table (5) indicated that the local prepared soluble liquid formulations passed successfully the miscibility test according to WHO specifications (1993). When these formulations were added to hard water at 3 % concentration, they showed good miscibility and they did not show any separation or precipitation.

Table (5) shows also that all the soluble liquid formulations which were derived from the prepared crude extracts exhibited slightly free acidity less than 0.3 % comply to WHO recommendations (1993). Also, these formulations passed successfully cold and heat storage tests.

3.2- Flowable formulation :

Data shown in Table (6) indicated that the best formula for preparation crude extracts as flowable formulation 30 % (wt./v.) which achieve suitable suspensibility (more than 70 % suspensibility) was consists of 30 % (wt./v.) crude extract, 35 % gelling agent (Aswanly clay), 14 % (wt./v.) wetting and suspending agent (polyethyleneglycol-600 dilaurate) and completing the total volume to 100 % by distilled water. This successful formula was comply with WHO specifications (1993) about flowable formulation.

Table (6): Important % constituents of locally prepared crude extracts as 30 % FL corresponding to its % suspensibility.

Crude extract	Important % constituents (wt/v)		% Suspension	
	Aswanly clay	Wetting agent	Soft water	Hard water
Seed *C. colocynthis*/chloro.	35 35	10 14	65 82	64 85
Seed *C. colocynthis*/hex.	35 35	10 14	41 77	40 75
Seed *C. colocynthis*/eth.	35 35	10 14	65 91	64 90
Artemisia/chloro.	35 35	10 14	58 81	52 80
Artemisia/hex.	35 35	10 14	45 81	44 80

Physico-chemical properties of the locally prepared flowable formulations :

The successful flowable formulations should fulfill the specifi-cations decided by WHO (1993). These specifications comprise good suspensibility before and after heat storage, free acidity or alkalinity should not exceeded

than 0.3 % before and after heat storage and passing through cold test (free and thawing). Results shown in Table (7) indicate that all locally prepared flowable formulations showed good suspension stability more than 70 % before or after heat storage, showed suitable free acidity (not exceeded than 0.3 %) before or after heat storage and passed successfully through cold test as recommended by WHO specifications (1993).

3.3- Dust powder formulations :

Crude extracts of tested plants were prepared as dust formulations containing 3 % crude by mixing solution of crude extracts in a volatile solvent (acetone) with suitable diluent (talc) 97 %. After mixing and complete evaporation of acetone, prepared dusts were packed and the following tests were determined : pH according to Watkins and Norton (1955), and particle size less than 40 μ using 40 microns sieve.

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تحضير بعض المستخلصات النباتية الخام على صورة مستحضرات قابلة للرش ومساحيق تعفير

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أجرى البحث بإستخلاص أجزاء مختلفة من خمس نباتات هى : الشيح (Wormwood)، الحنظل (Bitterapple)، الداتورة (Downy thornapple)، بصل العنصل (Squill)،الصبار (aloe) وتم تجهيزها على صورة مستحضرات مناسبة.

أستخلصت الأربعة نباتات الأولى بكحول الإيثانول ٩٥ % والهكسان العادى والكلوروفورم، فى حين تم استخلاص الصبار بقطع الأوراق وتجميع العصير النازف. تم دراسة الخواص الطبيعية-الكيماوية للمستخلص الخام مثل الذوبان والحموضة والقلوية، وتم تجهيز المستخلص على صورة مستحضر قابل للذوبان فى الماء أو صورة مستحضر قابل للتدفق أوصورة مساحيق تعفير.

دلت النتائج المتحصل عليها أن مستخلص كحول الإيثانول للشيح والحنظل والداتورة وبصل العنصل بالإضافة للصبار تعطى درجة جيدة للذوبان فى الماء ولذلك تم تجهيزهم على صورة مستحضر قابل للذوبان فى الماء. أما بقية المستخلصات الخام والتى لم تذب فى الماء تم تجهيزها علىصورة مستحضر قابل للتدفق، بالإضافة إلى ذلك فقد تم تجهيز كل المستخلصات على صورة مسحوق تعفير. أظهرت المستحضرات القابلة للذوبان إمتزاجا جيدا بالماء، أما المستحضرات القابلة للتدفق فقد أظهرت تعلقا جيدا عند تخفيفها بالماء، أما مساحيق التعفير فقد كانت ناعمة لدرجة أقل من ٤٠ ميكرون. علاوة على ذلك فقد أظهرت كل المستحضرات حموضة أو قلوية مناسبة وإجتازت إختبار التخزين الحار. المستحضرات القابلة للذوبان فى الماء والقابلة للتدفق يمكن استخدامها على النمو الخضرى للنباتات لمكافحة الافات عليها، بينما يمكن إستخدام مساحيق التعفير ضد افات الحبوب والمواد المخزونة.