Thermal Characterization of Extracts Obtained from Different Parts of *Nigella damascena (Ranunculaceae)* by TG and DSC

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The increased search for herbal products has generated an increasing interest in improving the quality control of extracts by pharmaceutical industry since these are raw materials of great importance by their quality and versatility. Thermal analysis such as thermogravimetry (TG) is a technique of high sensitivity, reproductibility and rapid response to variations in mass, obtaining results related to the composition and thermal stability of the sample, being important to the characterization of raw plants. Thermogravimetric techniques and differential scaning calorimetry (DSC) used for the study of pre-formulation of drug-excipient compatibility have been gaining importance. These techniques are being used for the verification of possible interactions between drugs and excipients. Aiming at studying the behavior of a plant extract, using these thermoanalytical techniques, the plant species Nigella damascena semen and Nigella damascena herba was used. This plant has healing and anti-inflammatory properties. The methodology for obtaining the extract followed the European Pharmacopoeia methodology. The TG and DSC curves were obtained under nitrogen and air atmosphere (20 mL/min) at a heating rate of 10°C/min. The TG and DSC tests were analyzed within a temperature range from 25 to 1000°C.

Keywords: herbal products, thermogravimetry, differential scaning calorimetry, Nigella damascena

The nigellas, popularly named black cumin or love-in-the-mist are represented by a genus of about 20 species of annuals from Northern Afrika Countries, Mediterranean countries and western Asia. The genus *Nigella*, *Ranunculaceae* family, includes some important species with aromatic and medicinal properties [1-3].

Nigella damascena is an annual garden flowering plant, belonging to the buttercup *Ranunculaceae* family. *Nigella damascena* is grown as an ornamental plant. Its seeds are used in african and Asian traditional medicine and, due to their sweet scent of strawberry, to prepare food. These plants are in the spontaneous flora under the popular name Fennel flower or black cumin. Because of her beauty, another popular name of *Nigella damascena* is love-in-the-mist. This fact in the signature theory of Paracelsus speek about a possible warning on toxic effects on species. Considering the chemical composition, there are several groups of compounds: active compounds - timokinona, nigellona, constant oils, compounds with nutritive role - proteins, carbohydrate, fat, other compounds - fat acid, amino-acids, vitamins, minor elements, alkaloids. This plant has healing and anti-inflammatory properties [4-7].

Thermogravimetric (TG) techniques and differential scanning calorimetry (DSC) are used during the pre-formulationstudies to help in the choice of the adjuvants used in a pharmaceutical formulation in order to check possible incompatibilities between drugs and adjuvants [8–12]. Such techniques have attracted great interest from researcher sand technologists because of their use for characterization of drugs from natural or synthetic origin as well as in foods, polymers, pharmaceuticals, and cosmetics [13–16]. And also the use of these techniques is important for a better characterization of components come from natural plant extracts. TG is likely to replace classical techniques (such as gravimetric weightloss) used in quality control of natural products, due to its rapid performance and reliability of the results and due tothe accuracy with which it determines the moisture contentof the material, useful in biological control for properstorage of natural products such as plant material [17].

Experimental part

Materials and methods

From the aerial parts and the seeds of *Nigella damascena* tinctures of 1:10 in 70 deg. ethyl alcohol have been prepared. The vegetal material from *Nigella damascena* was harvested from Arad County, Romania, july 2015- non poluated area. A sample of vegetal product was keept in the herbarium of the Pharmacognosy Departement, Faculty of Pharmacy of Arad. The plant material was air dried at room temperature in the shade, separated and ground to a fine ($\leq$ 300 μm) powder and then extracted. One gram of each sample was weighed andmacerated with 5 mL of 70% ethanol, for 10 days.

Simultaneous TG, DTG and DSC curves were obtainedin a dynamic air or nitrogen atmosphere (20 mL·min^{-1}) under nonisothermal conditions at a heating rate β=10 °C·min^{-1} using thermal analysis system, model Netzch-STA 449 TG/DTA. The TG and DSC tests were analyzed within a temperature range from 25 to 1000°C.

Results and discussions

Thermal behaviour of *Nigellae damascenae* herba in air and nitrogen atmosphere is similarly, for the most part. Of course, there is a slight difference in the temperatures characterizing the processes that occur, including a difference in the nature of a secondary process. This is normal and expected because of the different nature of the atmosphere.

Thus, in air, decomposition takes place in the temperature range 25.0 - 120.7 °C with $T_{peak\ DTG}$ = 87.0 °C and mass loss of 97.22%. The decomposition process is accompanied by a strong endothermic effect and $T_{peak\ DSC}$ = 92.0 °C. Next, take place (on TG curve) a slightly decreased of mass so that the residual mass at 575.0 °C is zero. This process corresponds to burning carbon and is

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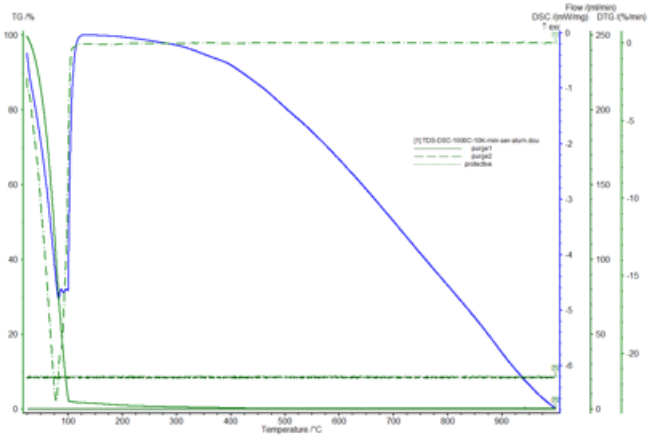

Fig. 1.Thermoanalytical curves of the *Nigella damascena* semen (air atmosphere)

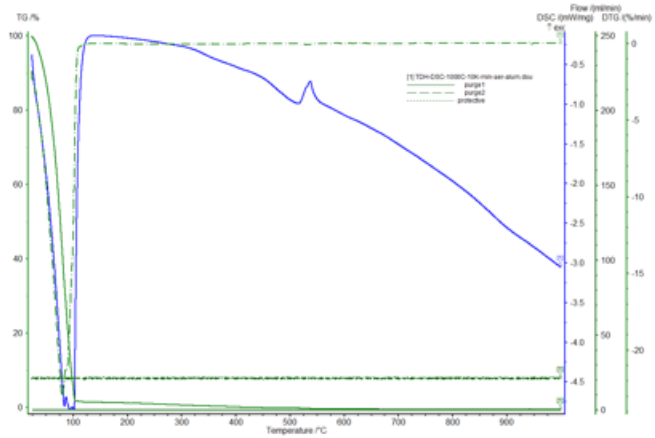

Fig. 2.Thermoanalytical curves of the *Nigella damascena* herba (air atmosphere)

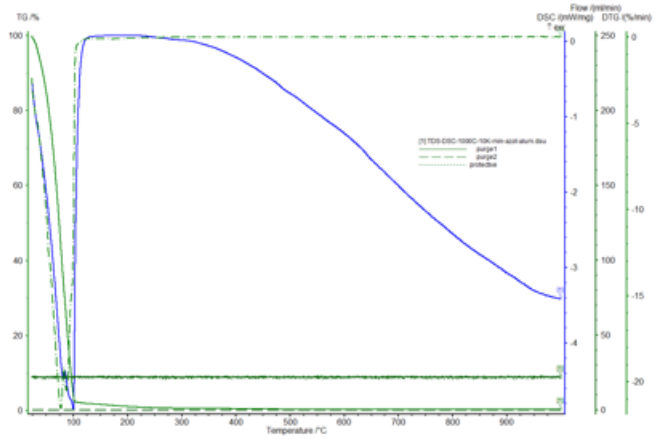

Fig. 3.Thermoanalytical curves of the *Nigella damascena* semen (nitrogen atmosphere)

accompanied by an exothermic effect with $T_{peak\ DSC}$ = 536.2 °C.

As regards the decomposition in nitrogen it occurs in the temperature range 25.0 – 119.6°C, with$T_{peak\ DTG}$ = 86.0 °C and a mass loss of 98.56%. The decomposition process is accompanied by a powerful endothermic effect, $T_{peak\ DSC}$ = 96.2°C. Further TG curve showed a slight decrease of mass so that the residual mass at 575.0 °C is zero. The process is accompanied by a slightly endothermic effect, with $T_{peak\ DSC}$ = 510.7 °C.

Thermal behaviour of *Nigella damascena* semen, in air and nitrogen, is very similar. The small differences between the characteristic temperatures of the decomposition process are insignificant.

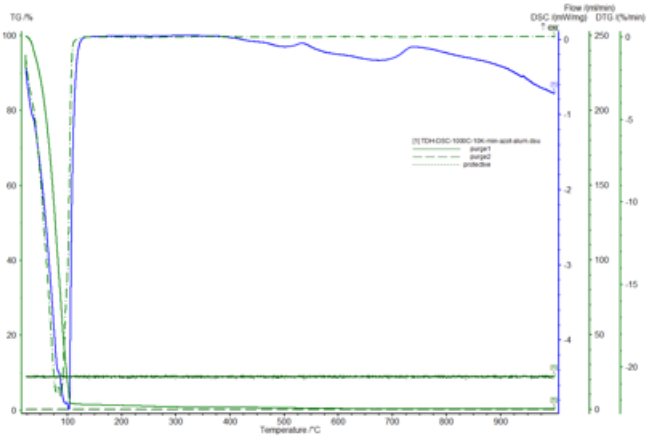

Fig. 4.Thermoanalytical curves of the *Nigella damascena* herba (nitrogen atmosphere)

There is a difference between the size of an endothermic peak appearing on the DSC curve. Thus, it is lower in decomposition in air, which would assume in the same time an easy exothermic-oxidative process.

Characteristics temperatures of the decomposition process are the following:

- in air: 25.0 – 112.5 °C with $T_{peak\ DTG}$ = 79.3°C and $T_{peak\ DSC}$ = 92.5 °C and mass loss (TG curve) is 98.11%; the nature of the decomposition was endothermic in nature;
- in nitrogen 25.0 to 115.7 °C with $T_{peak\ DTG}$ = 80.2°C and $T_{peak\ DSC}$ = 91.8 °C and mass loss (TG curve) is 97.85%; the nature of the decomposition was endothermic in nature.

In both cases the TG curve shows a loss of mass, so that the residual mass at 500.0°C is zero.

Conclusions

In this study, we set our goal in investigating the thermal behavior of *Nigellae damascenae semen* and *Nigellae damascenae herba* in both dynamic oxidative atmosphere (air) and inert one (nitrogen), in order to determine the stability.

Thermal decomposition occurs in a single step, within the temperature range 25 - 120 °C. Decomposition occurs with total loss of mass.

According thermal curves of *Nigellae damascenae herba* and *Nigellae damascenae semen* has a very similar thermal stability. Informations obtained by using thermal analysis allow the settlement of optimal conditions for storage of *Nigella damascenae herba* and *Nigella damascena semen*, respectively the preparation of products in the pharmaceutical industry.

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