UNIVERSITY „POLITEHNICA“ OF BUCHAREST

FACULTY OF BIOTECHNICAL SYSTEMS ENGINEERING

BIOTECHNICAL SYSTEMS ENGINEERING DOCTORAL SCHOOL

Doctoral Thesis Abstract

RESEARCHES ON OPTIMIZING THE PROCESS OF BIOMASS DENSIFICATION

(Keywords: biomass, pelleting, consumed energy, optimization)

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Rezumat

În ultimele decenii, înlocuirea combustibililor fosili cu cei din surse regenerabile a devenit o preocupare majoră la nivel mondial, cu rol în reducerea emisiilor de gaze cu efect de seră şi a poluării. Biomasa reprezintă resursa regenerabilă cu cea mai mare răspândire şi are avantajul că poate fi utilizată cu uşurinţă de către un sprectru larg al populaţiei. Peletizarea reprezintă unul din cele mai bune moduri de utilizare a biomasei drept combustibil, în urma procesului rezultând biocombustibili cu caracteristici similare lemnului.

Obiectivul general al cercetărilor teoretice şi experimentale îl constituie determinarea experimentală în laborator a influenţei parametrilor de intrare şi control asupra celor de ieşire (calitativi) ai peletelor, cu ajutorul unui stand automatizat, respectiv validarea experimentală, în condiţii de exploatare, a unor modele matematice şi optimizarea procesului de peletizare. A fost propus un model matematic ce exprimă densitatea finală a peletelor, ca fiind determinată de către presiunea finală aplicată în timpul proceslui, umiditatea iniţială a materialului şi densitatea iniţială a materialului şi un model matematic, obţinut prin analiză dimensională utilizând teorema Π, care exprimă densitatea peletelor care este influenţată de presiune, căldură, densitatea iniţială a materialului, viteza de peletizare şi volumul iniţial al materialului. S-au realizat cercetări experimentale utilizând un stand experimental pentru o singură peletă cu două matriţe de peletizare (cu orificiul de 8 mm şi de 10 mm), pentru 3 umidităţi ale materiei prime (10%; 13%; 16%), 3 viteze de peletizare (1,3 mm/s; 2,1 mm/s; 2,8 m/s), 3 temperaturi ale matriţei (70 °C; 80 °C; 90 °C) şi 3 forţe diferite de compactare (10 kN; 20 kN; 30 kN). Parametrii de ieşire urmăriţi, cei care exprimă calitatea peletelor şi eficienţa procesului de peletizare, au fost: energia consumată pentru obţinerea peletelor lungimea peletelor, volumul peletelor, densitatea peletelor, umiditatea peletelor, durabilitatea peletelor în timp. Metodologia de realizare a cercetărilor experimentale prin intermediul standului presupune parcurgerea următoarelor etape: se cântăreşte rumeguşul; se porneşte încălzirea matriţei şi se aşteaptă atingerea temperaturii dorite; se introduce rumeguşul în matriţă; se poziţionează pistonul în interiorul orificiului de presare şi se fixează în capul de presare al maşinii de forţă; se introduce viteza şi forţa de peletizare în programul software; se iniţiază procesul de peletizare a biomasei concomitent cu pornirea măsurării energiei consummate; procesul este oprit automat în momentul atingerii forţei maxime de peletizare setate; moment în care se opreşte şi măsurarea energiei consummate. A fost realizată validarea celor două modele propuse atât pe datele experimentale obţinute pe stand, cât şi pe un set de date obţinute pe o maşină de peletizat cu matriţă inelară. De asemenea, a fost realizată modelarea statistică şi optimizarea procesului de peletizare a biomasei.

Abstract

Over the last few decades, the replacement of fossil fuels with those from renewable energy sources has become a major concern worldwide, with a role in reducing greenhouse gas emissions and pollution. Biomass is the renewable resource with the largest spread on the surface of the earth and has the advantage that it can be easily used by a broad spectrum of the population. Pelleting is one of the best ways to use biomass as a fuel, the process resulting in solid biofuels with characteristics similar to wood destined for burning.

The general objective of the theoretical and experimental researches is the experimental determination in the laboratory of the influence of the entry and command parameters on the output (qualitative) of the pellets, with the help of an automated stand, respectively the experimental validation, under operating conditions, of mathematical models and the optimization of the pelleting process.

A mathematical model expressing the final density of the pellets was proposed, as determined by the maximum force applied during the process, the initial moisture of the material and the initial density of the material, and a mathematical model obtained through dimensional analysis using the Π theorem, which expresses the density of the pellets that is influenced by pressure, heat, the initial density of the material, the speed of pelleting and the initial volume of the material. Experimental researches were conducted using a single pellet stand, on two pelleting dies (with 8 and 10 mm orifice diameter), for 3 raw material moistures (10%; 13%; 16%), 3 pelleting speeds (1.3 mm/s; 2.1 mm/s; 2.8 m/s), 3 die temperatures (70 °C; 80 °C; 90 °C) and 3 different compression forces (10 kN; 20 kN; 30 kN). The output parameters followed, those that express the quality of the pellets and the efficiency of the pelleting process, were: the energy consumed to obtain pellets, the length, volume, density and moisture of the pellets and their durability over time. The methodology for conducting the experimental researches on the stand involves the following stages: weighing the sawdust; starting the heating of the die and waiting for the desired temperature to be reached; inserting the sawdust into the die; positioning the piston inside the pressing orifice and fixing it in the head of the force machine; the pelleting speed and the force are introduced in the software program; the process is initiated concomitantly starting the measurement of the energy consumed; the process is automatically stopped when the maximum pelleting force is reached, also stoping the measurement of the consumed energy. The validation of the two models proposed was performed both on the experimental data obtained on the stand, as well as on a set of data obtained on a ring die pelleting machine. Also, the statistical modeling and optimization of the biomass pelleting process was performed.

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CONTENTS

FOREWARD

Over the last few decades, the replacement of fossil fuels with those from renewable energy sources has become a major concern worldwide, with a role in reducing greenhouse gas emissions, and pollution in general. Biomass is the renewable resource with the largest spread on the surface of the earth and has the advantage that it can be easily used by a broad spectrum of the population. Pelleting is one of the best ways to use biomass as a fuel, the process resulting in solid biofuels with characteristics similar to wood destined for burning.

The doctoral thesis "*Researches on optimizing the process of biomass densification*" presents a synthesis of experimental researches performed by the author in the field of compacting woody biomass represented by fir tree sawdust by pelleting, using a special device designed and built by the author for the purpose of conducting experiments, respectively modelling. the phenomenon of pelleting in order to produce pellets with good qualitative attributes.

The paper is structured in 7 chapters, developed in 182 pages, contains 181 figures and graphs, 120 mathematical relations, 51 tables as well as a bibliography made up of 158 references. The paper also includes a list of notations (2 pages), and at the end, 4 annexes (30 pages), where the data obtained in the experimental research, data analysis and pelleting programs in the pelleting machine software are presented in detail).

The general objective of the theoretical and experimental researches is the experimental determination in the laboratory of the influence of the entry and command parameters on the output (qualitative) of the pellets, with the help of an automated stand, respectively the experimental validation, under operating conditions, of mathematical models and the optimization of the pelleting process.

In **Chapter 1**, entitled "*Introduction. The importance and the objectives of the doctoral thesis*", the general aspects regarding the operation of compaction of biomass by palletization are briefly presented, respectively the influence factors of the process. Also, both the importance and the specific objectives by which it is possible to achieve the main objective are presented, both theoretically and experimentally.

In **Chapter 2**, "*Study of the properties and characteristics of the biomass used in the pelleting process*", introductory notions are presented regarding: renewable resources and the importance of their use, the types of biomass and their directions of use, physical and chemical properties, as well as the biomass properties that influences its densification process by the pelleting process. At the end of the chapter, the conclusions regarding the importance and the properties of the biomass are presented.

In **Chapter 3**, "*Current stage of researches on biomass densification by pelleting*", the theoretical considerations on the constructive solutions of biomass pelleting equipment, the analysis of the working process of the pelleting machines with flat or ring die are presented, respectively, the theoretical and experimental researches conducted worldwide on the pelletization of biomass and the mathematical models that describe the process. A series of new constructive solutions identified in international patents are also presented. The chapter ends with the presentation of the conclusions of the solutions used to obtain solid biofuels by pelleting.

In **Chapter 4**, "*Theoretical researches on the mathematical modelling of the biomass pelleting process*" are presented a series of theoretical notions on the modelling of the compaction process, detailing the best-known models for this type of process. A mathematical model expressing the final density of the pellets was proposed, as determined by the maximum force applied during the process, the initial moisture of the material and the initial density of the material, and a mathematical model obtained through dimensional analysis using the Π theorem, which expresses the density of the pellets that is influenced by pressure, heat, the initial density of the material, the speed of pelleting and the initial volume of the material, which will be subsequently validated by experimental determinations.

Chapter 5, "*Experimental research on the process of pelletizing biomass on the single pellet stand* " includes own experimental researches, conducted at the The National Institute of Research – Development for Machines and Installations Designed to Agriculture and Food Industry – INMA Bucharest. The objectives and the methodology of the experimental determinations of pelleting biomass in the laboratory, the experimental stand designed and built especially for the realization of the tests and the equipment used for the accomplishment of the experimental researches, as well as the determination of the physical characteristics of the material from biomass are presented. The stages of experimental researches on the stand built for pelleting the biomass represented by fir sawdust are described. For the experimental validation of the mathematical model obtained in the previous chapter, 243 test samples (pellets) were made for each die used (with the pelleting orifice of 8 and 10 mm), in which the following parameters were varied: the moisture of the material, die temperature, compressive force and pelleting speed. The following output data of the process of pelleting the fir sawdust were recorded: energy consumed, length of pellets (later calculating their density) and moisture of pellets. The samples obtained were monitored for 90 days, during which 14 measurements were made of the pellets, in order to determine the evolution in time of the length and implicitly of the density. The chapter ends with presenting the conclusions regarding the factors of major influence on the quality of the pellets obtained in the experimental researches.

In **Chapter 6**, „*Researches on the optimization of the biomass pelleting process based on experimental results*", the main objective was to validate the mathematical models proposed in chapter 4, based on the experiments conducted using both the single pellet stand and a ring die pelleting machine. The systemic structure by means of which the description of the process of pelleting the sawdust in the cylindrical die was made, is a modern and suitable description of the technological processes, providing tools for prediction and optimization, useful in the rational exploitation of technologies. At the end of the chapter, conclusions about model validation and process optimization are presented.

Chapter 7, „*General conclusions. Contributions. Recommendations and perspectives*", presents the general conclusions drawn from the theoretical and experimental research regarding the densification of biomass by pelleting. Also, personal contributions are presented regarding the studied process and the experiments conducted within the doctoral thesis, as well as new research directions and recommendations, which may represent topics for theoretical and experimental studies that can be approached by other researchers.

The author considers that the present doctoral thesis represents a modest contribution to the clarification of important aspects related to the optimization of the process of biomass densification by pelleting, which can be deepened in further research.

SYMBOLS AND NOTATIONS

SRE	renewable energy sources
EE	energetic efficiency
ET	thermal energy
$\rho\ [kg/m^3]$	density
$\rho_a\ [kg/m^3]$	apparent density
$\rho_v\ [kg/m^3]$	bulk density
$m\ [kg]$	mass
$V\ [m^3]$	volume
$V_a\ [m^3]$	apparent volume
$V_v\ [m^3]$	Bulk volume
$\alpha_M\ [^\circ]$	natural slope angle
$Q_i\ [MJ]$	lower calorific value
$LC\ [\%]$	lignin content related to the dry and ash-free state
$C_f\ [\%]$	fixed carbon
$F_{CF}\ [\%]$	fixed carbon fraction
$Fv\ [\%]$	fraction of volatile substances
$F_i\ [\%]$	fraction of inert
$\lambda\ [W\ m^{-1}\ K^{-1}]$	thermal conductivity
$D_p\ [m]$	pellet diameter
$L_p\ [m]$	pellet length
$C\ [\%]$	ash content of pellets
DM	mechanical durability of pellets
DV	bulk density pellets
$r\ [m]$	radius of the pelleting roller
$R\ [m]$	radius of the pelleting die
$E_c\ [Wh]$	energy consumed
$I_{AVE}\ [A]$	average current
$U\ [V]$	electrical voltage
$MSR\ [-]$	methodology of response surfaces
$RNA\ [-]$	artificial neural network
$p\ [MPa]$	pelleting pressure
$D\ [kg/m^3]$	the relative density of the tablet
$\rho_f\ [kg/m^3]$	final density
$V_0\ [m^3]$	volume at pressure 0
$V\ [m^3]$	volume at pressure p
$V_s\ [m^3]$	volume of solid material free of voids
$V_R\ [m^3]$	volume ratio
$V_1\ [m^3]$	volume at pressure 1
$c\ [-]$	the degree of volume reduction
$\mu\ [-]$	coeficientul de frecare
$L_c\ [m]$	coefficient of friction
$p_m\ [MPa]$	average pressure
$\rho_r\ [kg/m^3]$	the relative density of compacted sawdust
$V_\infty\ [m^3]$	net volume of powders
$p_0\ [MPa]$	pressure before compression
$\theta\ [^\circ C]$	die temperature during the process
$U_i\ [\%]$	raw material moisture content
$\rho_0\ [kg/m^3]$	raw material density
$\varnothing_m\ [m]$	die orifices diameter

F_{max} [kN]	maximum applied force (compression force)
V [m/s]	pelleting speed
L [m]	pellet length
ρ_p[kg/m^3]	pellet density
U_p [%]	pellet moisture
V_p [m^3]	pellet volume
Q [J]	quantity of heat
CU [%]	moisture content of the biomass mixture
V_d [%]	content of volatile matter
M_{ad} [%]	humidity, as a percentage of the total mass of the material, determined for the sample to be analysed
A_d [%]	ash content
g [mm]	sawdust granulation
dF [kN]	the elemental frictional forces acting on the material
L_c [mm]	the perimeter of the cross-section of the pressing cylinder
β [-]	lateral expansion coefficient
A_c	the surface of the pressing cylinder
P_0 [MPa]	atmospheric pressure
e_g [-]	global error
e_{max} [-]	maximum error
c [J/g °C]	mass thermal capacity
m [g]	mass of the system (mass of the biomass sample)
ΔT [°C]	temperature variation due to heat exchange
R^2 [-]	coefficient of determination
q [kWh]	specific electricity consumption
P_c [kW]	consumed power
Q_p [kg/h]	quantity of processed product
η_{me} [-]	efficiency of the electric drive motor

CHAPTER 1
INTRODCUTION. THE ROLE AND IMPORTANCE OF BIOMASS DENSIFICATION. OBJECTIVES OF THE DOCTORAL THESIS

1.1.Introduction

Currently, researches in the field of obtaining solid biofuels is a major concern in the field of mechanical engineering, whose main aim is to optimize the densification processes, improve the quality of biomass products, minimize costs, increase the yields of compression equipment, as well as contribute to the protection of the environment.

Encouraging researches in the field of biomass pelleting aims at determining and using optimal solutions in designing, building and using pelleting machines, with final result in obtaining final products – pellets with better quality, complying with the standards in the field.

It is important to correlate the researches in the field with the current tendencies to give up fossil fuels and use renewable energy, with the best combustion attributes and with the highest durability.

Theoretical and experimental researches in the field of biomass pelleting aim to increase the quality of the finished products, correlated with the decrease of the costs and the impact on the environment.

In order to obtain solid biofuels with adequate density and durability, there must be a very good correlation between the physical-chemical properties of the biomass used as raw material and the characteristics of the pelleting equipment (type of die, size of orifices, type of drive on the material, speed of material pelleting, etc.).

1.2. Biomass resources

Biomass represents a promising renewable energy source for our country, both in terms of abundance and in terms of multiple possibilities of use.

An important part of the biomass used to obtain bioenergy originates from plant material and animal products. A first classification of biomass can be made according to the origin of the biomass according to the sector, such as: agriculture, forestry, industry and the urban sector. Another type of classification can be made according to its nature: energy crops, agricultural or forestry residues and different types of waste, [9, 23].

From the point of view of its use in energetic purposes, biomass is divided into the following types:

- Energy crops:
    - Annual herbaceous crops (cereals, sunflower, etc.);
    - Perennial herbaceous crops (reed, cane, giant miscanthus, etc.);
    - Lignocellulosic crops (willow, poplar, miscanthus, etc);
- Agricultural residues (straws, husk, stalks, etc);
- Forestry residues (sawdust from tree cuttings, branches, leaves, etc.);
- Zootechnical crops (manure).

Biomass can be used for obtaining:

- Food;
- Animal feed;
- Raw materials and auxiliary materials for various industries;
- **Energy.**

1.3. Use of biomass for energy purposes

Various types of wood (sawdust, branches from grooming trees, vine ropes, walnut and peanuts shells, etc.) or agricultural (cereal straws, corn stalks and cobs, vegetable stalks, etc.) residues are suitable to be used for obtaining energy.

By using these types of waste, it is avoided to turn them into unused waste and ensures the full use of important resources, which can ultimately reduce or, in some cases where there are sufficient biomass resources, replace the demand for fossil fuels such as coal, oil or gas.

Thorough planning is needed at national and local level, as well as the installation of biofuel production infrastructures for efficient use of biomass, [13, 68, 92, 114].

The solid fuel in the form of pellets and briquettes is produced from agricultural and forestry residues (cereal straws, corns, corn stalks and corn cobs, soybean stalks and pods, rapeseed stalks, vine ropes, residues from pruning, sawdust, tree bark, etc.), both types representing an efficient alternative to classic fuels.

The major difference from conventional fuels is the small size and regular shape of pellets and briquettes, thus allowing them to be used as fuel for automated installations destined for obtaining thermal energy.

1.4. Biomass properties for densification

Biomass properties strongly affect the quality of the raw material for densification and ultimately its use in combustion applications, [12, 18]. These properties include density, particle size, flow, moisture, calorific power, ash content and colour, which are very important features for designing biomass handling, transportation, storage and conversion systems.

Table 1.1. Characteristics of biomass and impact on the processing-use chain, [83]

Properties	Application in the processing chain
Density	Logistics for the transport and storage of biomass in different forms: wood chips, briquettes, pellets, etc.
Particle size	Design parameters for efficient conversion
Natural slope angle	Design parameters for handling and storage
Moisture	Design parameters for drying and thermal conversion processes
Calorific power	Efficiency of energy recovery
Ash content	Estimation of the potential risk of slag formation or deposits during combustion / gasification
Color	Quality control and rapid estimation of fuel properties

1.5. Densification of biomass by pelletization

Biomass densification consists of two stages:

- Pressure compaction of the material for volume reduction and particle agglomeration;
- Activation of the lignin existing in the wood material by this pressure resulting in "gluing" the material without needing an additional gluing agent.

The process of obtaining pellets requires that the biomass be treated at high pressures and forced to pass through the orifices, generally cylindrical, of a special die. In the case of favourable conditions, the biomass "fuses", resulting in a solid product. The process of obtaining pellets is called "extrusion". Some types of biomass (generally woody) naturally lead to qualitatively good pellets, but for other types of biomass (most types of agricultural biomass) it may be necessary to add some "binders" to maintain the pellets in a bound form after cooling.

1.6. Objectives of the doctoral thesis

General objectives

The general objective of the thesis is the theoretical and experimental study of the influence of the input and control parameters on the output (qualitative) parameters of the biomass pellets through an automated stand, the experimental validation, under operating conditions, of mathematical models for the optimization of the pelleting process.

Specific objectives

Throughout the thesis are presented some personal results obtained through experimental researches on stand in the laboratory and in operating conditions on a pelletizing machine, the fulfilment of the main objective of the doctoral thesis requiring the achievement of the following specific objectives:

- Study of the properties and characteristics of the biomass used in the pelleting process;
- Analysis of scientific papers in the field, both theoretical and experimental;
- Prospective study of equipment and installations for the compaction of biomass by pelleting;
- Study of the constructive solutions of pelleting equipment from patents;
- Identification of established mathematical models to estimate the density or volume of the pellets based on the input and control parameters process;
- Establishing the factors that influence the pelleting process and the density of the pellets, depending on the characteristics of the raw material (biomass);
- Development of own mathematical models for estimating the density of wood pellets;
- Design, realization and calibration of the experimental stand for laboratory researches;
- Conducting experimental pelleting researches using the experimental stand;
- Determining the density of the pellets obtained for two dimensions of the press orifices through the experiment stand;
- Monitoring the evolution of pellet length over time;
- Conducting experimental determinations under operating conditions, on a ring die pelleting machine;
- Validation of the mathematical models proposed based on the results obtained through the experimental researches performed on the stand for a single pellet and on the pelleting machine with a ring die;
- Elaboration of a set of recommendations for an optimized pelleting process;
- Broadly disseminating the results of theoretical and experimental researches on the pelleting process by publishing scientific papers in profile journals and in the volumes of specialized conferences.

CHAPTER 2
STUDY OF THE PROPERTIES AND CHARACTERISTICS OF THE BIOMASE USED IN THE PELLETING PROCESS

2.1. Biomass resources

According to the definition provided by Directive 2009/28 / EC, biomass is "the biodegradable fraction of products, waste and residues of biological origin in agriculture (including plant and animal substances), forestry and related industries, including fisheries and aquaculture, as well as the biodegradable fraction of industrial and municipal waste". This means that under proper industrial processing, freshly harvested biomass can be converted into products similar to natural gas or liquid or solid fuels. It represents the most abundant renewable resource on the planet. By applying various transformation processes, such as combustion, gasification or pyrolysis, biomass can be transformed into "bio-fuels" for transport, "bio-heat" or "bio-electricity". Biomass is listed among the first forms of energy used by man, since the discovery of fire, [20,51].

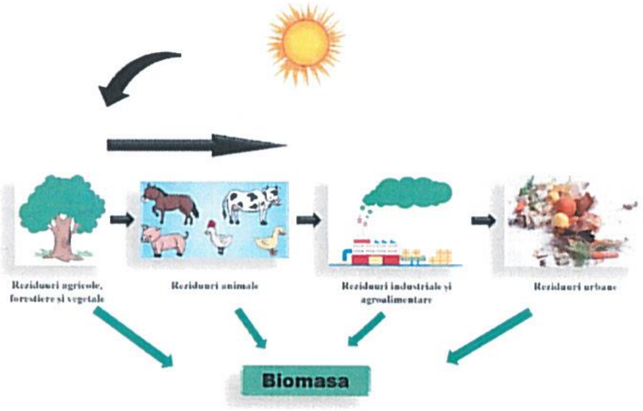


Fig. 2.1. Sources of biomass production, [77]

Biomass is composed of a series of building blocks made up of carbohydrates obtained from carbon dioxide and ground water. In this way, through photosynthesis, solar energy is stored in the chemical extract of biomass. By burning the biomass, a reaction occurs between the carbon in the plants and the oxygen that sustains the combustion, forming carbon dioxide and water. This process is cyclical, with carbon dioxide being able to participate in a new biomass formation process.

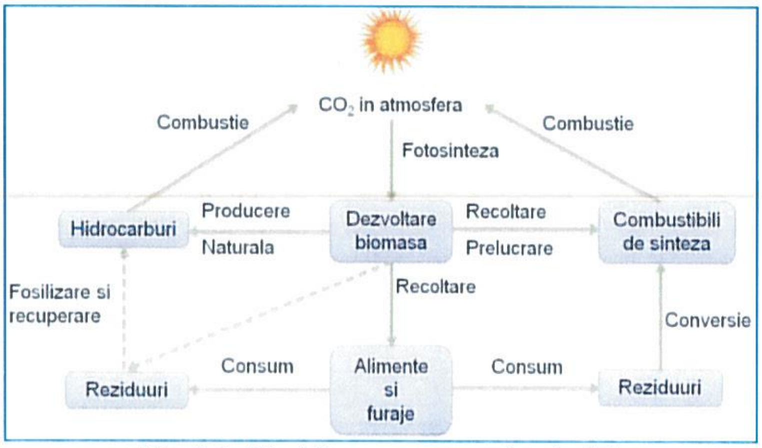


Fig. 2.2. The biomass energy circuit, [7]

Biomass contains 7-34% lignin and 66-93% carbohydrates represented by cellulose and hemicellulose, [120].

The energy stored in the biomass can be released through multiple methods, all essentially representing the chemical combustion process.

2.2. Main directions for energy use of biomass in Romania

The main types of biomass conversion processes are:

- **physical** (pelleting, briquetting, etc.);
- **biological / biochemical** (aerobic, anaerobic or alcoholic fermentation);
- **thermal** (combustion, pyrolysis, gasification, hydrogenation);
- **chemical** (biodiesel), [54].

Energy (bioenergy) from biomass is obtained through:

- direct combustion, generating heat;
- pyrolysis burning, generating syngas;
- fermentation, resulting in biogas (CH_4) or bioethanol (CH_3-CH_2-OH);
- chemical transformation with generation of esters, for example, methyl esters (biodiesel) and glycerol; In the next step, the purified biodiesel can be burned in diesel engines; also, vegetable oil can be burned in diesel engines as such or in mixture with diesel in different proportions, but with lower biodiesel qualities.
- enzymatic degradation (ethanol or biodiesel), [53].

2.3. Biomass properties related to the compaction - pelletization process

The inherent properties of biomass are those that determine both the choice of conversion mode and any problems that may arise from processing. Equally, the choice of biomass source is influenced by the form in which the energy is required and this interconnection between the two aspects is what brings the flexibility in the use of biomass.

2.3.1. Physical properties of biomass

A) Dimensions and particle size distribution of the mixture

In general, the density and durability of the pellets is inversely proportional to the size of the particles, because the small particles have a larger surface area during densification;

The absence of coarse particles in the mass of material will significantly affect the efficiency of the production of pelleting machines. Too high a percentage of very small particles can block pelleting machines, affecting production capacity. The coarser ground material tends to result in less durable pellets, as they can create natural cracks in the pellets, which are then susceptible to breakage.

B) The moisture content of the biomass mixture

- ➢ facilitates starch gelatinization, protein denaturation and fiber solubilization processes during compression;
- ➢ greatly influences the moisture content of the products resulting from the densification;
- ➢ acts as a binder during the densification process and increases binding by van der Waals forces, thus increasing the contact area of the particle.

C) Density

Density is an important characteristic of biomass, which directly influences the raw material to be processed. The uneven dimensions and shapes of biomass in its raw state, including the leaves and the stalks, lead to high costs of transport, storage and feeding of the material in each unit of the conversion operation.

D) The natural slope angle

The natural slope angle represents an indicator that characterizes the flow of the material, which is a function of the particle shape, frictional forces and cohesion. This is defined as the angle that the free surface of a mass of granular material makes when dropped onto a surface, with the horizontal plane.

E) Lower calorific value (combustion heat)

It represents the number of heat units released by the complete combustion of a mass unit; this differs from species to species being higher to those with higher lignin content (resinous) and lower to deciduous and agricultural biomass.

2.3.2. Chemical properties of biomass

A) Ash content

It is the product remaining after combustion and is composed of non-combustible inorganic substances (mineral salts). These remain as residues in the form of dust stored in the places where the fuels burned; ash content is a very important characteristic of quality, because ash causes problems in biomass processing and combustion.

B) The content of volatile matter

Volatiles are the components, apart from moisture, which are released at high temperatures in the absence of air.

C) Fixed carbon and inert content

It is the carbon found in the remaining material after the volatile materials are released; the fixed carbon content in agricultural biomass is generally in the range: 15-25% and in woody biomass between 14-23%.

D) Heavy metal content

Particular importance in the energy recovery of biomass fuels - waste type - is represented by the content in heavy metals; heavy metals have a negative impact on the environment, aiming that their percentage is as low as possible.

E) Starch and protein content

The starch softens and increases its volume in the presence of heat and moisture through a process known as gelatinization; proteins, like starch, soften under the action of heat and pressure and increase the strength of pellets.

F) Fiber content

There are water soluble fibers and water insoluble fibers in biomass. In general, soluble fibers increase the viscosity of the material and have a positive effect on the structure of the pellets, while the insoluble fibers may tangle with each other.

G) Lipid content

It helps during the pelleting process because it acts as a lubricant, but too much increase in lipid content results in a decrease in the mechanical properties of the pellets.

H) Cellulose

It is a polysaccharide that represents an important structural component of the primary cell wall of green plants.

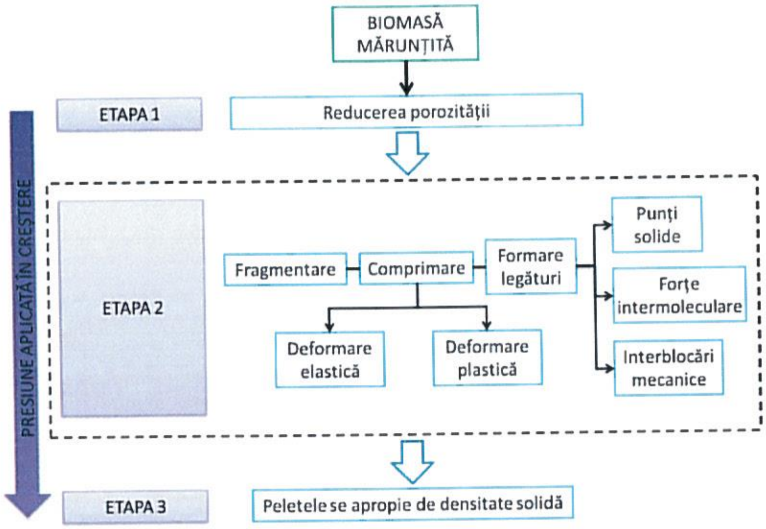
I) Hemicellulose

Woody tissue combines the surface of cellulose microfibrils, and linked together, constitute hard fibers of interconnected cell networks.

J) Lignin

It is a component part of wood, the third after cellulose and hemicellulose. Lignin is particularly important in the formation of cell walls, especially in wood and bark, giving them rigidity and helping them not rot easily.

The process of pellet formation consists in subjecting the biomass to high pressures, during which the particles are forced to agglomerate. The compression process is usually obtained through three distinct stages.



CHAPTER 3
CURRENT STAGE OF RESEARCHES ON BIOMASS DENSIFICATION BY PELLETING

3.1. General considerations on biomass densification

Pellets represent the solid biofuel normally produced from wood and agricultural residues. They look like cylindrical granules of standard sizes, with diameters generally between 5 and 8 mm (sometimes even up to 25 mm) and with variable length, from 5 to about 40 mm and irregular (broken) ends, [103]. They have high mechanical strength and good combustion characteristics. The process of obtaining pellets is similar to that of making briquettes, except that the biomass passes through much smaller orifices and the finished product has much smaller dimensions. The pellets are used most often when the process of feeding the combustion plant is required to be automated.

A high-quality pellet is dry, hard and durable, with small quantities of ash remaining after combustion.

Fig. 3.1. Pellet samples

The actual pelleting operation represents a small part of the technological flow for obtaining pellets.

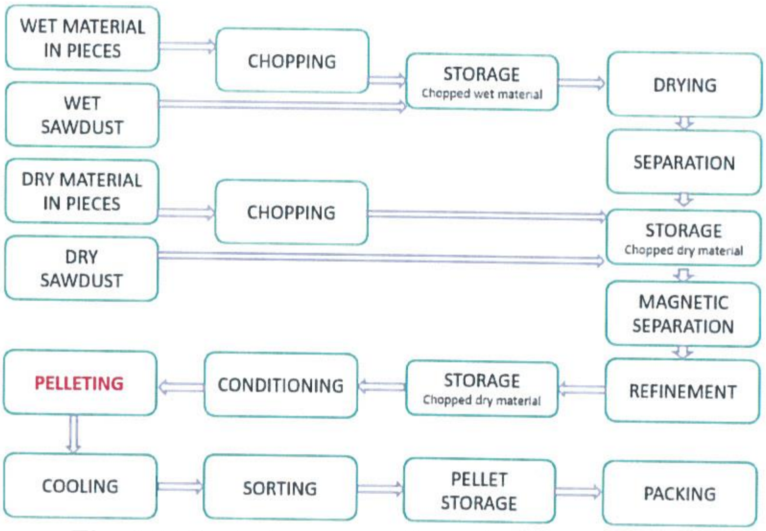


Fig. 3.2. The technological flow of pellets, [69, 86]

Pelleting machines have as main body the die, characterized by the ratio between length and diameter (L / D). The length refers to the depth of the orifice and the diameter refers to the diameter of the channels (orifices) of the die. In general, the durability of the pellets increases with the increase of the L / D ratio due to the increase of the frictional forces resulting from the increased friction between the material and the die.

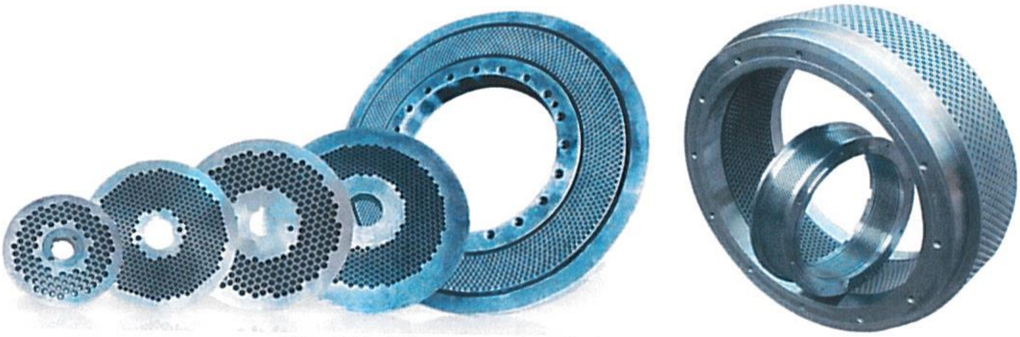
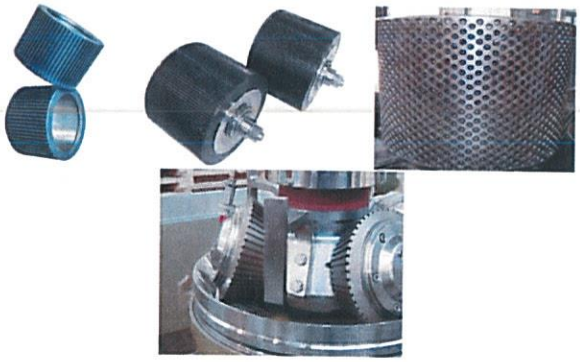


Fig. 3.3. Die examples. left – flat; right- ring

The press rollers may be cylindrical or cone-shaped. The surfaces of the rollers are corrugated or can be provided with different forms of indentations. During their movement, the rollers press the material into the orifices of the die. Each orifice in the die is only active for as long as it is near a press roll.



Fix. 3.4. Examples of pressing rollers

3.2. Analysis of constructive solutions regarding biomass pelletizing equipment

3.2.1. Pelleting equipment with flat die

In general, there are two types of flat-die equipment on the market, one with a rotary die and fixed rollers and one with a rotary rollers and a fixed die. The first type has a stationary roller and a rotary die and the second type has a stationary die, while the roller assembly rotates.

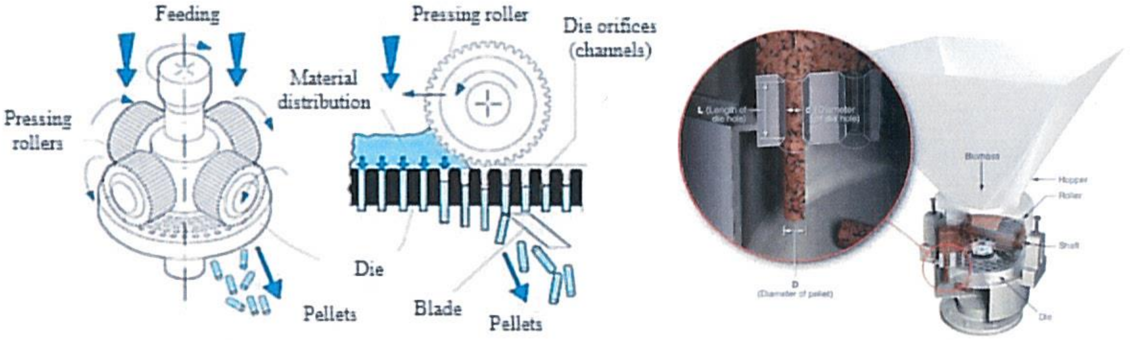


Fig. 3.5. Diagram and operating principle of the flat die pelleting machine, [39]

Flat die pelleting equipment is used to process materials with high adhesion forces, producing pellets intended for both biofuel use and animal feed. They are used in small and medium-sized units. They are characterized by a safe operation, have high mobility, produce a low noise, have a low energy consumption. They also have a low productivity, compared to pelleting equipment with ring die.

3.2.2. Pelleting equipment with ring die

Pelleting machines with ring die were designed based on the design of the ring die. The basic principle of this equipment is the operation by which the material is distributed on the inner surface of a perforated ring die, the material reaching near each pressing roll, being forced to pass through the orifices of the die, thus forming pellets.

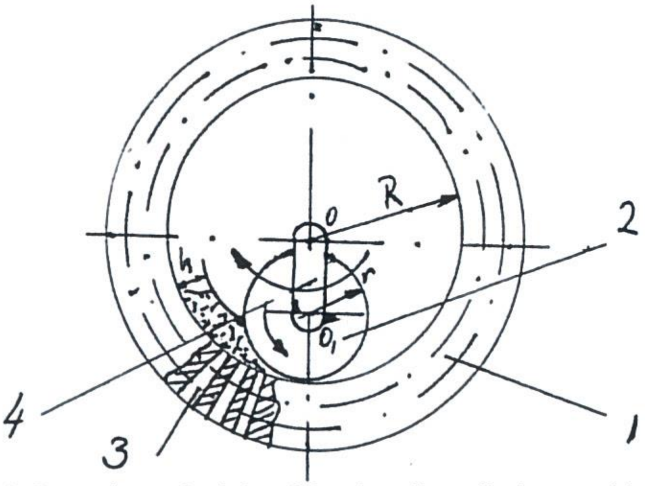


Fig. 3.6. Operating principle of the ring die pelleting machine, [14]
1. ring die; 2. Pressing rollet; 3. Pelleting orifices; 4. roller spindle

Pellet formation actually takes place at the "contact line" between the rolls and the die. All other activities related to this action, such as conditioning the material (drying, shredding), cooling the pellets, etc., practically support that moment in the system. In order to understand the process and to improve the overall quality of the pellets, there must be an in-depth understanding of what is happening at the point of attachment of the material by the rollers.

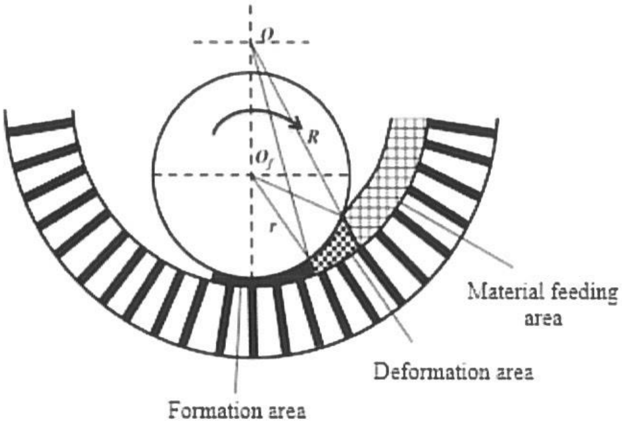


Fig. 3.7. Diagram of the pelletizing mechanism in ring die pelleting machines, [82]

Material feeding area: in this area, the material falls on the inside surface of the die. The material is not significantly affected by the mechanics affecting the materials. The density of the material in this area is 0.4-0.7 g / cm^3.

Deformation area: under the action of the rotation of the pressing rollers, the biomass gradually enters the area where the material is gradually compressed under the action of the extrusion force, forming the relative displacement within the mass of material. As the force exerted by the rollers increases, the distance between particles decreases rapidly. Plastic and elastic deformations take place simultaneously. In this area, the biomass density is 0.9-1.0 g / cm^3.

Formation area: in this area, the distance between the particles of material is further reduced, given the increase of the extrusion force, the contact area between the particles increasing rapidly. The density of the material in this area can reach 1.2-1.4 g / cm^3, [82, 129].

3.3. Stadiul actual al cercetărilor teoretice privind procesul de peletizare

The study of a system is important to understand the relationships between its components or to predict how the system will work in the event of changing or adding parameters. Sometimes it is possible to experiment with the system itself, but not in all cases. The study of the systems is done in many cases by modelling the system.

- Within paper [99], a predictive model for energy consumption is determined using the response surfaces methodology.

The authors have developed a predictive model for energy consumption in UV-assisted pelleting (UV-A). The experimental data obtained were used to optimize the process parameters through the response surface methodology (MSR), which aimed to determine the optimal working parameters of the system.

The software used recorded the average current (I_{AVE}). The voltage (U) was 120 V. The energy consumed can be calculated using the following equation:

$$E_c=U\cdot I_{AVE}\cdot\frac{120}{3600}, \text{(Wh)}$$

The ultrasonic power, the size of the die orifices and the mass of the pellet significantly affected the energy consumption, which increased when these three parameters increased. The effects of pellet mass on energy consumption were more significant at the higher level of ultrasound power and at the highest level of the size of the die orifices.

3.4. The current state of experimental research regarding the pelleting process

Frodesona S. et al. present a series of experimental investigations of pelleting seven types of biomass, in order to observe the effect of moisture on the quality of the pellets obtained.

In the research, 6 levels of moisture were tested: 0%, 5%, 7.5%, 10%, 12.5% and 15%. The pelleting was carried out using a device for a single pellet with pressing orifice size of 8 mm.

For the pellets obtained, measurements were made to determine the density and hardness. Following the study, it was found that for woody biomass, the lowest density was recorded for beech samples, and the lowest hardness was obtained for spruce samples. For all samples, it was observed that both density and hardness had the lowest values for moisture with the lowest and highest values (0, 2, 4% and 15%).

3.5. Constructive solutions of pelleting equipment from patents

The double die pelleting machine [147], is an invention from 2013, in the USA, which relates to the field of pelleting machines and in particular to double-die machines, having two die that rotate in the opposite directions that are connected by teeth that interlock, alternately compressing the material in oppositely arranged pelleting chambers while the die are in counter-rotation motion, pulling the material from the contact line down between the dies.

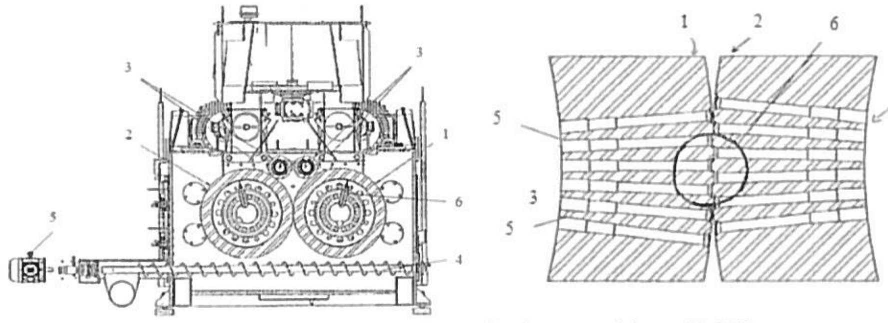


Fig. 3.8. Double die pelleting machine, [147]

Pellet forming system [150] is an invention from 2011 for the production of pellets from agricultural or woody biomass for use in combustion boilers or other similar equipment, the system comprising both pellet forming means and means for pre-processing of wood and / or other types of biomass to be transformed into pellets. The system is comprised of:

- a pellet formation system;
- a biomass processing module.

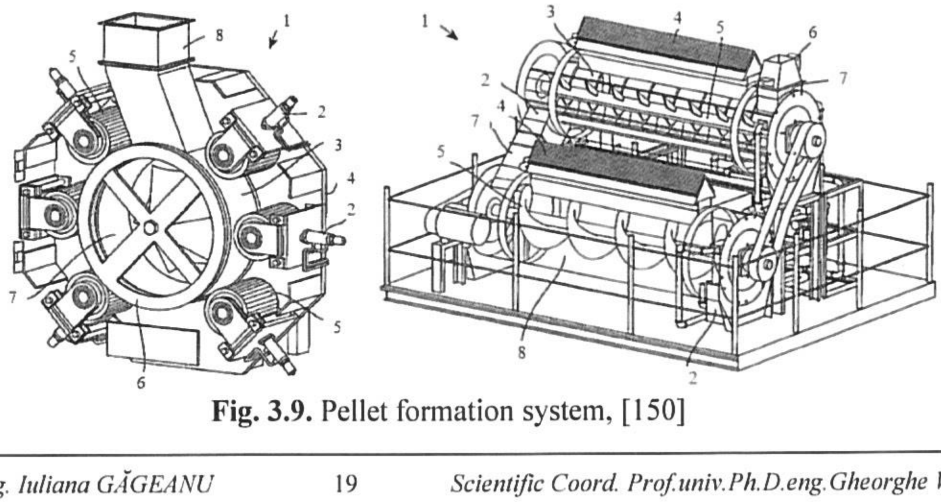


Fig. 3.9. Pellet formation system, [150]

CHAPTER 4
THEORETICAL RESEARCH ON THE MATHEMATICAL MODELLING OF THE BIOMASS PELLETING PROCESS

4.1. Overview

Compression models help to reveal the behaviour of biomass / particles during the compaction (pelleting) process and can help optimize the parameters needed to obtain good quality pellets. The pellets can be formed by the use of pressure agglomerations, in which the particles join together with or without the aid of binders.

Jones Model

Jones (1960), [55], expresses the pressure-density data of compacted powders in the form of equation (4.1):

$$\ln \rho = m \ln (p) + b \tag{4.1}$$

where: ρ – bulk density (volumetric mass) pf the mixture – kg/m^3;
p - applied compression pressure – MPa;
m şi b – model constants.

$$b = \ln\left(\frac{1}{1-\rho_0}\right) \tag{4.2}$$

where: ρ_0= the relative density of the powder mixture (kg/ m^3).

Heckel Model

The equation of the Heckel model, [43], is used to express density from the point of view of the fractions as a function of the applied pressure:

$$\ln\frac{1}{1-D} = mp + b \tag{4.3}$$

where: D – the relative density of the compressed material (the ratio of the density of the compressed product to that of the powder material).

Cooper-Eaton Model

Cooper and Eaton (1962), [17] have studied the behaviour during compaction of four types of ceramic powders. For each case, it was assumed that densification is achieved by two almost independent probabilistic processes, namely the filling of the orifices having the same size as the particles and the filling of the orfices with dimensions smaller than those of the particles. Based on these assumptions, the following equation (4.4) was given):

$$\frac{V_0 - V}{V_0 - V_s} = a_1 e^{-\frac{k_1}{p}} + a_2 e^{-\frac{k_2}{p}} \tag{4.4}$$

where: V_0 – volume at pressure 0, m^3;
V – volume at pressure p, m^3;
V_s - the volume of solid material free of voids, m^3;
a_1, a_2, k_1 şi k_2 - Cooper-Eaton model constants.

4.2. Contributions to the modelling of compression in the pelletization process

The first model (4.5) proposed starts from Jones's model, [55], based on the determination of pellet density as a function of the pressure applied to the biomass, but takes into account the correction factor that takes into consideration the moisture of the raw material.

$$\rho_p(\rho_0,p,U_i)=a\rho_0\left(\frac{p}{p_0}\right)^b U_i^c \tag{4.5}$$

where: - a, b and c are model parameters, dimensionless;
- p_0 atmospheric pressure;
- p is calculated using the maximum force in the compression process and the geometrical characteristics of the die:

$$p=\frac{4F_{max}}{\pi d^2} \tag{4.6}$$

The second model, which represents a more complete version of the process study, was determined using the dimensional analysis theory - Π method.

In the study of the process of compressing biomass materials, the theory of dimensional analysis was applied, for the mathematical modelling of this process. From the theory of dimensional analysis, Π theorem was applied, stated by Buckingham, [8].

From the theoretical investigations of the process of compression by pelleting biomass powder materials, a number of 8 main parameters that influence the pressing process were considered in the study, presented in table 4.1.

Table 4.1. Parameters of the pelleting process taken into account in the dimensional analysis

Parameter	Name	Notaion	Measurement unit	Dimensiune fizică
1	Quantity of heat	Q	kg m^2/s^2	ML^{2}T^{-2}
2	Compression pressure	p	N/m^2	ML^{-1}T^{-2}
3	Initial material moisture	U_p	%	-
4	Pellet density	ρ_p	kg/m^3	ML^{-3}
6	Volumetric mass of raw material	ρ_0	kg/m^3	ML^{-3}
7	Initial volume of the raw material	V_0	m^3	L^3
8	Pelleting speed	v	m/s	LT^{-1}

Thus, a **second model** was determined, representing a more complex version, taking into account both the moisture of the raw material and the temperature of the die during the pelleting process, is given in formula (4.7):

$$\rho_p(\rho_0,p,U_i)=\rho_0\left(\frac{Q}{\rho_0\cdot v^2\cdot V_0}\right)^a\left(\frac{p}{p_0}\right)^b U_i^c. \tag{4.7}$$

In model (4.7), the control parameter represented by the temperature of the die was introduced in a dimensionless factor considering the temperature as having an energetic nature (quantity of heat), in the second factor of the right member, which rises to the power of *a*.

CHAPTER 5
EXPERIMENTAL RESEARCHES ON THE PELLETING PROCESS ON THE SINGLE PELLET STAND

5.1. Objectives of experimental researches

The main objective of the experimental researches on the experimental stand was represented by the experimental determination of the output parameters depending on the input and control parameters. This was achieved by achieving the following specific objectives:

- preparation of raw materials for pelleting;
- designing, building and preparing the experimental test stand for conducting the researches within the present doctoral thesis;
- determination of the physico-chemical properties of the raw material;
- conducting the experimental researches using two pelleting dies (with the pelleting orifices of 8 and 10 mm), for three raw material moistures (10%; 13%; 16%), three pelleting speeds (1.3 mm/s; 2.1 mm/s; 2.8 mm/s), three die temperatures (70 °C; 80 °C; 90 °C) and three different compaction force (10 kN; 20 kN; 30 kN);
- determining the moisture and density of analysed samples;
- time tracking the length of pellets for 90 days, and calculation of their final density.

5.2. Preparation of the material for conducting the experimental determinations

In order to conduct the experimental researches on the pelletization of biomass, the material to be pelleted, consisting of fir sawdust was prepared. In order to transform the fir fragments into sawdust, machines for coarsely chopping and grinding are required. The sawdust was subsequently sieved at dimensions <2 mm and its particle size distribution was made.

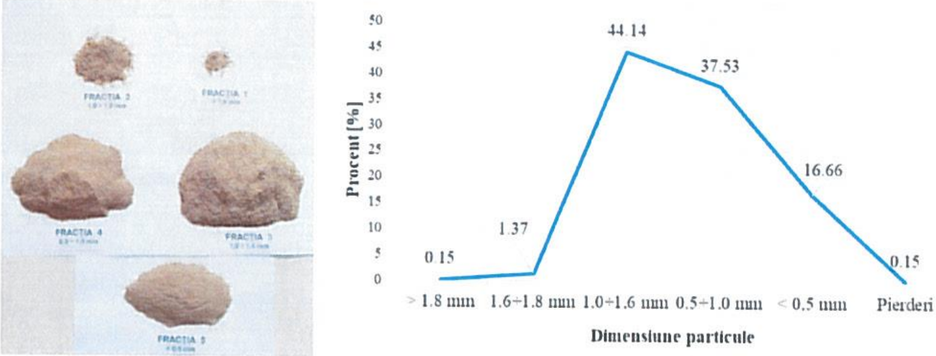


Fig. 5.1. Distribution of fractions for sawdust < 2 mm

5.3. Working methodology regarding the process of pelleting the fir tree sawdust biomass

5.3.1. Physico-chemical characterization of the biomass analyzed before and after the pelleting process

A) Determination of biomass moisture

Moisture was determined by drying using a UFE 500 oven, Model: 100 - 800, Memmert - Germany, in accordance with the standard SR EN ISO 18134-1: 2016.

For the rapid determination of moisture during the pelleting tests, in order to adjust the moisture to the desired values, a SHIMADZU MOC63u thermobalance was used.

Fig. 5.2. Moisture determination for the samples in the oven (left) and in the thermobalance (right)

B) Determination of the volumetric mass of the material

The bulk density of fir sawdust samples was determined by the cylinder method, according to standard SR EN ISO 17828: 2016.

Table 5.1. Determined values of the density of fir stree awdust

Moisture [%]	Bulk density [kg/m^3]
	Average for three samples
10	136.08
13	142.18
16	147.37

5.3.2. The experimental plan regarding the process of compaction of the biomass and the methodology used

During the biomass pelleting process, a series of input and control parameters of the compaction process, respectively output parameters of the quality of the finished product (for fir sawdust pellets) were followed.

Thus, for each pelleted sample, the following input data were established and determined:

- raw material granulation: < 2 mm;
- raw material moisture (**U_i**): 10%, 13% și 16%;
- raw material volumeytric mass (**ρ_0**): 136.08 kg/m^3; 142.18 kg/m^3; 147.37 kg/m^3;

The control parameters for the biomass compaction process were:

- die orifice diameter (**$\varnothing_m$**): 8 mm, 10 mm;
- maximum applied force (**F_{max}**): 10 kN, 20 kN și 30 kN.
- pelleting speed (**v**): 1,3 mm/s; 2,1 mm/s; 2,8 mm/s;
- die temperature throughought the process (**θ**): 70°C, 80°C, 90°C.

For each pelleting test (sample) were determinedthe following output (quality) data for the pellet obtained:

- consumed energy (**E_c**);
- pellet lenght (**L**);
- pellet density (**ρ_p**);
- pellet moisture (**U_p**);
- pellet volume (**V_p**).

5.4. Equipment used for experimental determinations

5.4.1. Equipment used to prepare the material used in the pelleting process

TCU hammer mill- INMA Bucharest

For the coarse chopping of biomass represented by fir tree wood fragments, a hammer mill was used.

TRV-0 vegetable waste grinder– INMA Bucharest

After being coarsely chopped, the material was grinded to sizes smaller than 2 mm using a vegetable grinded.

AS 200 basic sieving system– Retsch, Germany

This system is used for the separation by fractions and the determination of the granulometric fractions of the powder mixtures.

5.4.2. Equipment used to determine the physico-chemical properties of biomass

UFE 500 Model 100–800 laboratory over– Memmert, Germany, used to determine the moisture of materials.

MOC63u thermobalance – Shimadzu, Japan, allows the rapid determination of moisture.

AV 220 analytical balance - Shimadzu- Japan.

Calcination furnace - Naberterm Seria L, GmbH, Germany.

5.5. Equipment used in the pelleting process

The experimental stand for a single pellet, specially designed for conducting the experimental research within the thesis, is represented by a pelleting device (fig. 5.3.) connected to a force machine with a maximum capacity of 100 kN, (fig. 5.4.).

The force machine is in turn assisted by a computer, thus offering the possibility, through a specialized software program, to vary the pelleting speed and the compaction force.

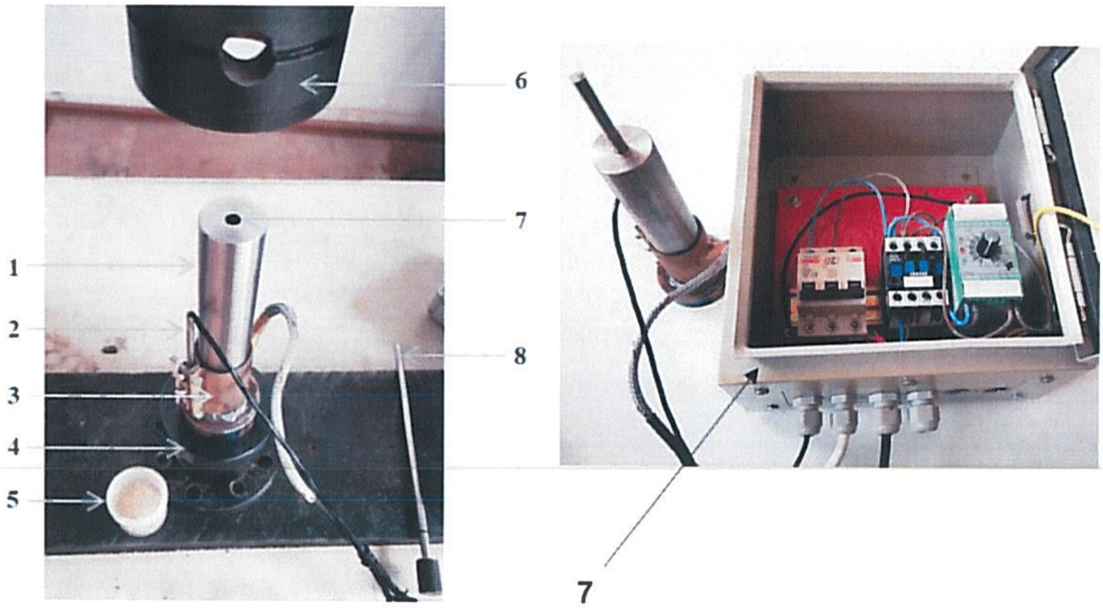


Fig. 5.3. Pelleting device used for conducting experiments

1.– changeable die (∅ 8, 10 mm for the pelleting orifice); 2. temperature sensor; 3. heating element; 4. blocking plate 5. pressing head; 6. piston; 7. automation and and temperature control box.

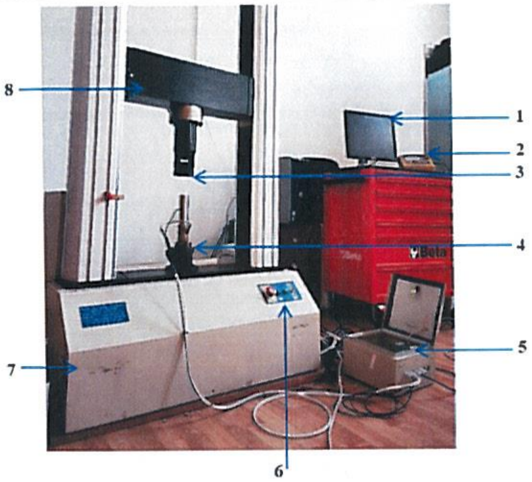


Fig. 5.4. Force machine connected to the pelleting idevice
1.– computer; 2. frequency and phase analyzer; 3. pressing head ; 4. pelleting device; 5. automation and and temperature control box 6. force machine control; 7. case; 8. weight.

Fig. 5.5. Example of force - time curve generated by software for a pelletization test

For the measurement of energy consumption for each sample, a phase and frequency analyzer was used.

The size of the pellets at the exit from the process was determined using electronic calipers.

The method of obtaining a pellet sample on the stand

The methodology of conducting the experimental researches on the stand involves the following stages:

- 2.5 grams of sawdust are weighed for the 10 mm die, respectively 1.65 for the 8 mm die;
- the heating of the die is started and is waited until the desired temperature is reached;
- the sawdust is inserted into the die using a metal funnel;
- the piston is positioned inside the pressing hole;
- the piston is fixed in the pressing head of the force machine;
- the pelleting speed and force are introduced in the program;
- the process of pelleting biomass is initiated at the same time as starting the measurement of the consumed energy;

the process is stopped automatically when the maximum pelleting force is reached, simultaneously stopping the measurement of consumed energy.

5.6. Experimental results of the process of pelletizing sawdust

5.6.1. Experimental results of the compaction process

After conducting the experiments, a set of 243 samples was obtained for each of the two dies used (with pelleting orifices of 8 and 10 mm). Examples of pellet samples are shown in figure 5.6.

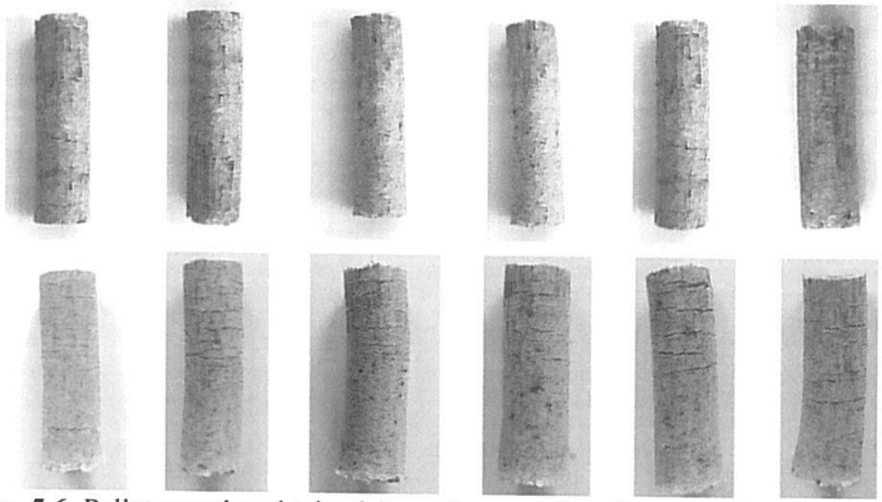


Fig. 5.6. Pellet samples obtained (top - 8 mm pellets; bottom - 10 mm pellets)

For the output parameters of the pelletization process, a histogram distribution was made for the 243 samples obtained for each die.

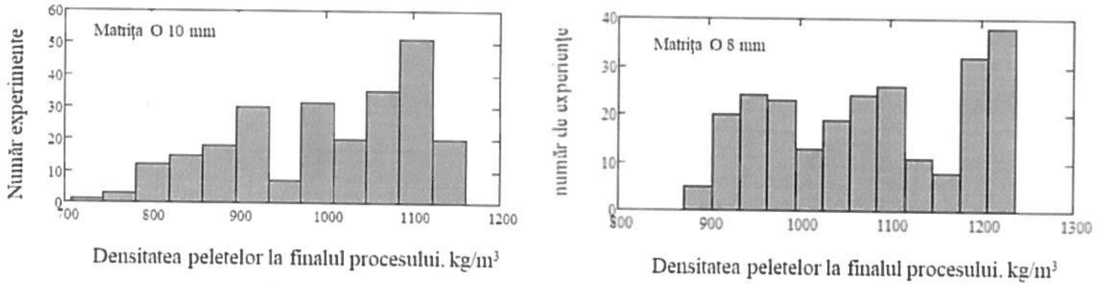


Fig. 5.7. Pellet density distribution for the two sets of samples obtained

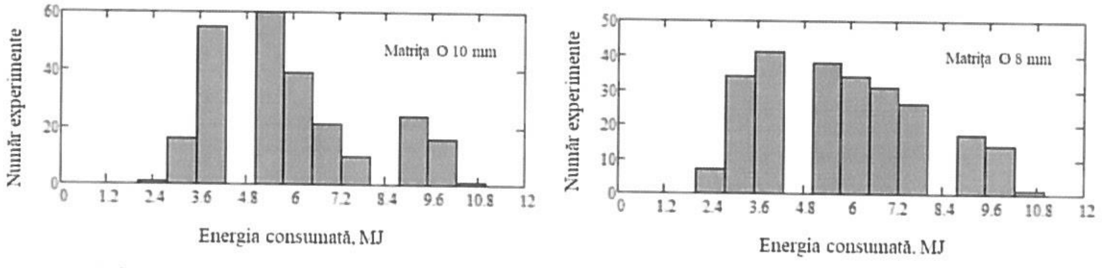


Fig. 5.8. Consumed energy distribution for the two sets of samples obtained

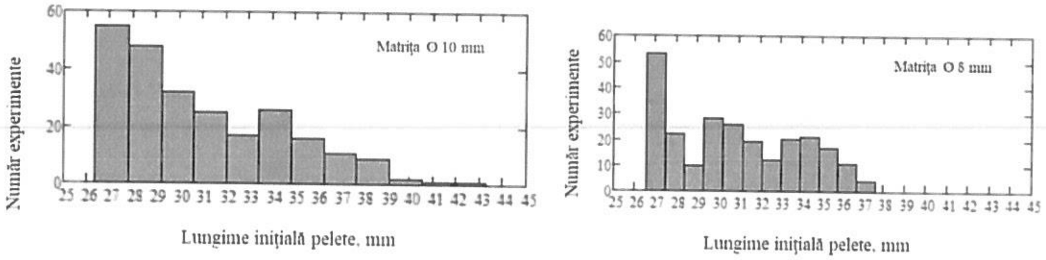


Fig. 5.9. Pellet length distribution for the two sets of samples obtained

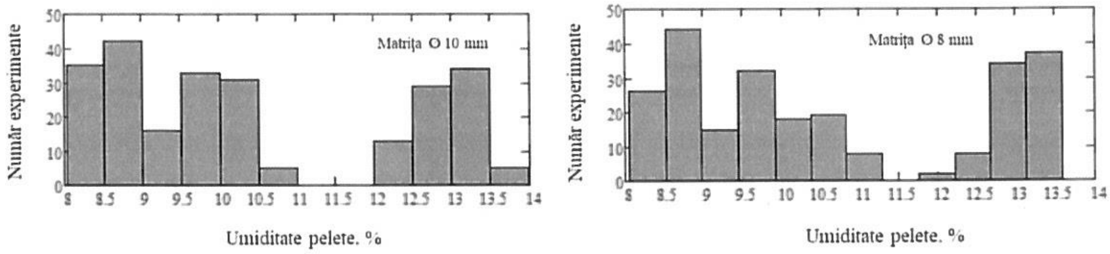


Fig. 5.10. Pellet moisture distribution for the two sets of samples obtained

5.6.2. Statistical analysis of experimental results

In order to obtain empirical relations between the parameters of the pelleting process, a statistical analysis of the experimental data was performed. Initially, the elementary statistical analysis is performed (for the values of the main estimators of the parameters involved in the process and the correlations between them).

The values of the correlation between the ten parameters involved in the process (parameters with acceptable variation, namely they take a satisfactory number of different values) are given in tables 5.2 and 5.3.

Table 5.2. Correlations between the variable parameters involved in the process for the 10 mm die

Correlations	ρ_0	ρ_p	F_{max}	U_i	θ	v	E_c	V_p	L	U_p
ρ_0		-0,78	0	0,999	0	0	-0,236	0,768	0,763	0,938
ρ_p	-0,78		0,333	-0,796	-0,007	-0,112	0,446	-0,994	-0,993	-0,878
F_{max}	0	0,333		0	0	0	0,644	-0,301	-0,305	-0,098
U_i	0,999	-0,796	0		0	0	-0,242	0,785	0,78	0,949
θ	0	-0,007	0	0		0	-0,027	0,028	0,023	-0,089
v	0	-0,112	0	0	0		-0,112	0,135	0,134	0,085
E_c	-0,236	0,446	0,644	-0,242	-0,027	-0,112		-0,43	-0,432	-0,314
V_p	0,768	-0,994	-0,301	0,785	0,028	0,135	-0,43		0,999	0,871
L	0,763	-0,993	-0,305	0,78	0,023	0,134	-0,432	0,999		0,867
U_p	0,938	-0,878	-0,098	0,949	-0,089	0,085	-0,314	0,871	0,867	

Table 5.3. Correlations between the variable parameters involved in the process for the 8 mm die

Correlations	ρ_0	ρ_p	F_{max}	U_i	θ	v	E_c	V_p	L	U_p
ρ_0		-0,75	0	0,999	0	0	-0,287	0,762	0,762	0,942
ρ_p	-0,75		0,295	0,762	-0,119	-0,024	0,395	-0,996	-0,996	-0,783
F_{max}	0	0,295		0	0	0	0,569	-0,281	-0,281	0,080
U_i	0,999	-0,762	0		0	0	-0,287	0,773	0,772	0,953
θ	0	-0,119	0	0		0	-0,116	0,121	0,121	-0,059
v	0	-0,024	0	0	0		-0,379	0,020	0,020	0,143
E_c	-0,287	0,395	0,569	-0,287	-0,116	-0,379		-0,390	-0,390	-0,389
V_p	0,763	-0,996	-0,281	0,773	0,121	0,020	-0,390		0,999	0,799
L	0,762	-0,996	-0,281	0,772	0,121	0,020	-0,390	0,999		0,799
U_p	0,942	-0,783	0,080	0,953	-0,059	0,143	-0,389	0,799	0,799	

Is noticeable the high value of the correlation between the volumetric mass of the raw material and its moisture, between the volumetric mass of the raw material and the initial moisture of the pellets, between the initial moisture of the raw material and the initial moisture of the pellets, between the volume and the length of the pellets (because they are geometrically linked), between the initial volume of the pellets and their length or moisture. High intensity inverse correlations are recorded between the volumetric mass of the raw material and the density of the pellets, the volume, the length and moisture of the pellets and the initial density of the pellets, the density and moisture of the pellets at the end of the process.

A comparative graphical representation of the results obtained after determining the output parameters of the pellets immediately after the die was made, for the two dies used (8 and 10 mm) depending on the input and control parameters.

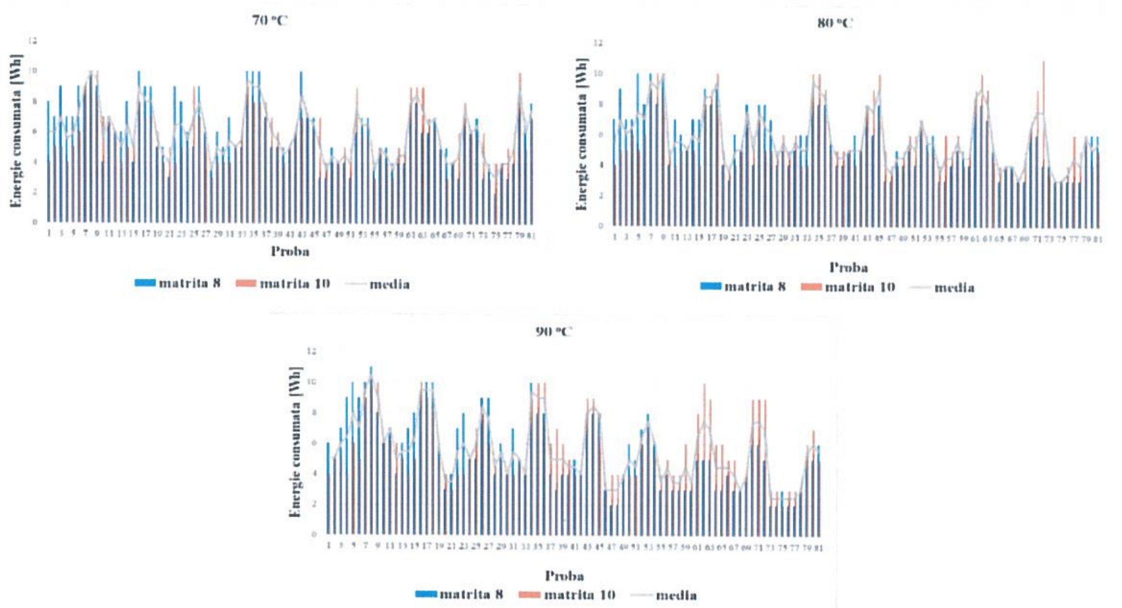


Fig. 5.11. Variation of the energy consumed depending on the temperature of the die, for the two dies

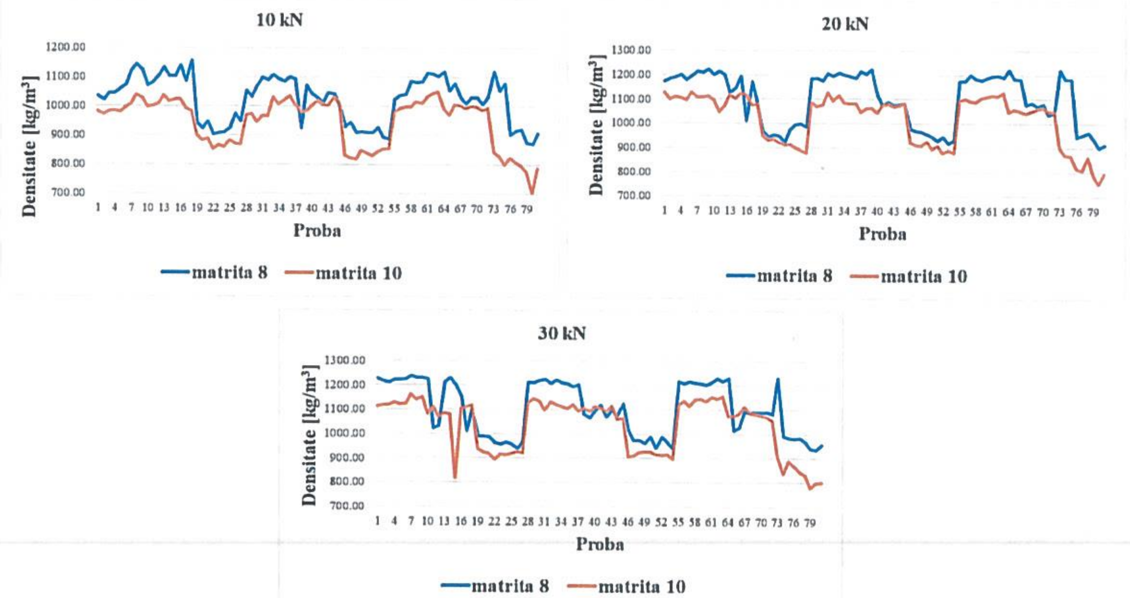


Fig. 5.12. Pellet density variation depending on the compression force for the two die

Figure 5.13 shows examples of compressive-displacement force curves for the three forces (10, 20, 30 kN) used during the experiments.

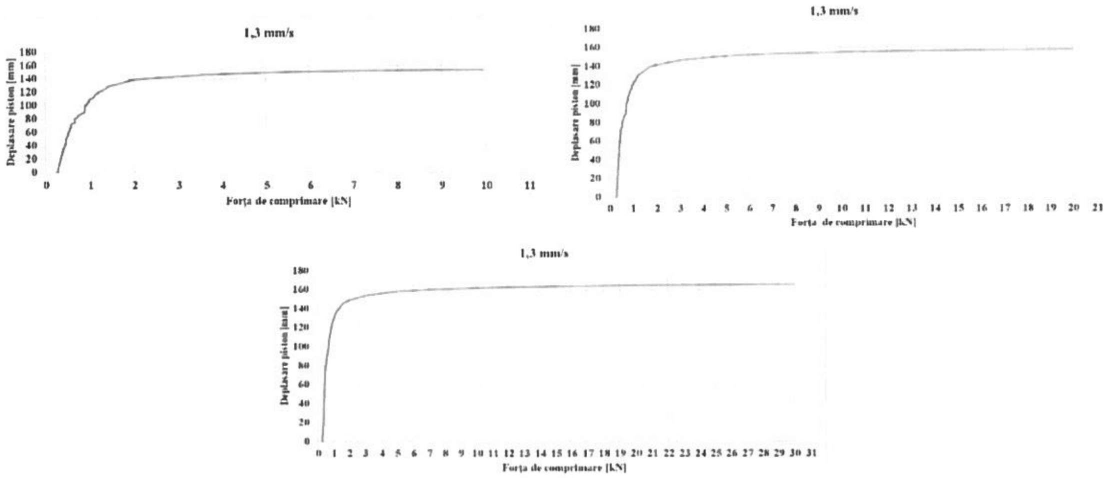


Fig. 5.13. Example of force-displacement curve

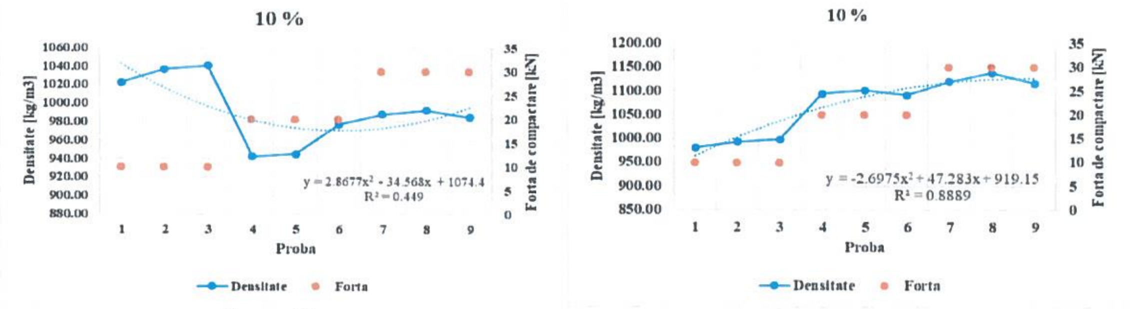
5.6.3. Analysis of the variations of the parameters measured with the parameters of the working regime

In Table 5.4., three experimental datasets are presented for pellet density in the case of constant temperature (70 °C), initial material moisture (10, 13 and 16%) and pellet speed (2.8 mm / s) while variating only the compaction force for both pelleting dies used.

Table 5.4. Pellet density while varying the compaction force and keeping the other parameters constant, for a temperature of 70 °C

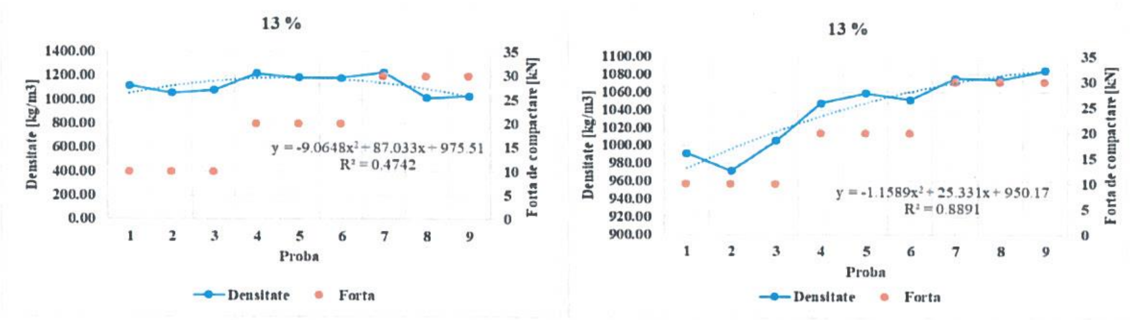
SAmple	Temperature [°C]	Initial moisture [%]	Pelleting speed [mm/s]	Compression force [kN]	Pellet density [kg/m³]	
					8 mm die	10 mm die
1	70	10	2,8	10	1022.61	980.61
2					1036.82	992.70
3					1041.09	997.88
4				20	942.47	1093.85
5					945.18	1101.33
6					977.32	1091.51
7				30	988.14	1119.46
8					992.61	1137.78
9					985.17	1116.20
10		13		10	1118.42	991.41
11					1055.83	971.58
12					1078.02	1005.42
13				20	1220.29	1047.77
14					1185.04	1059.02
15					1182.48	1051.73
16				30	1228.51	1075.39
17					1015.02	1074.26
18					1029.02	1084.54
19		16		10	921.22	844.46
20					935.71	827.11
21					937.45	801.75
22				20	942.47	901.97
23					945.18	869.67
24					977.32	865.73
25				30	988.14	908.67
26					992.61	840.29
27					985,17	888,61

In Figures 5.14.-5.16., the results obtained for the density with the variation of the compaction force and the constant maintenance of the other parameters are graphically compared, for the the two pelleting dies used.



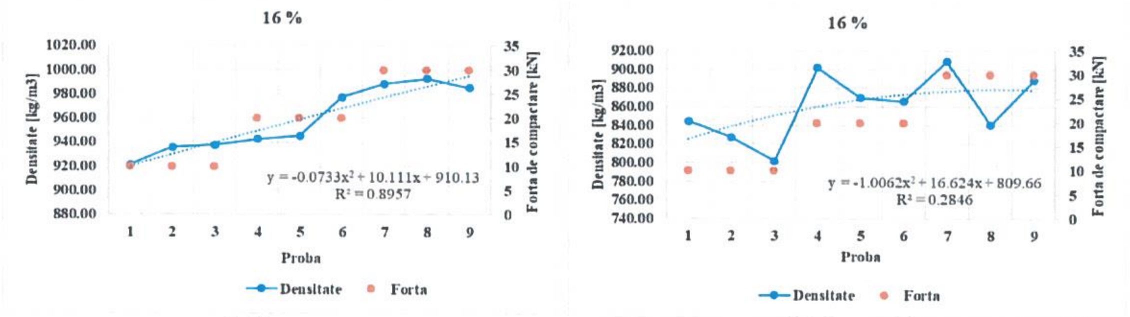
8 mm die **10 mm die**

Fig. 5.14. Pellet density for θ=70 °C, v = 2.8 mm/s and Ui = 10 %



8 mm die **10 die**

Fig. 5.15. Pellet density for θ=70 oC, v = 2.8 mm/s and Ui = 13 %



8 die **10 die**

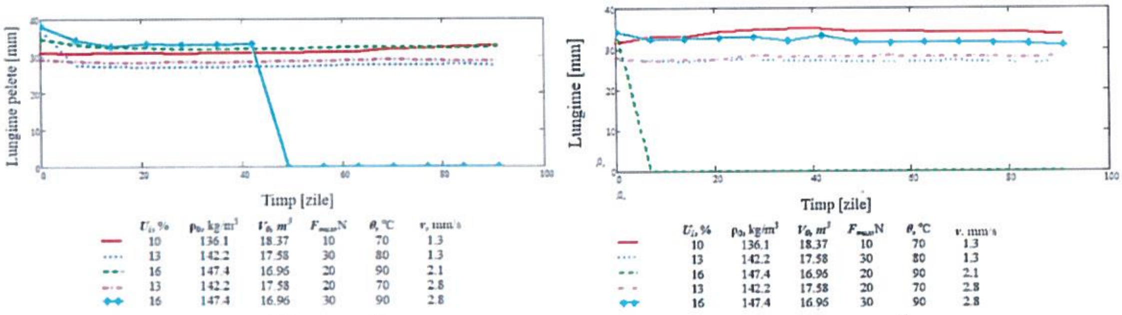
Fig. 5.16. Pellet density for θ=70 oC, v = 2.8 mm/s and o Ui = 16 %

From Figures 5.14-5.16, in the case of the 10 mm die, we can observe a strong correlation of the density obtained with the compaction force in the case of the initial material moisture of 10%, a good correlation for the initial moisture of 13% and a low correlation in the case of 16% initial moisture. For the 8 mm die, the results show the best correlation in the case of 16% moisture, and low correlations for the other two moistures.

5.7. Evaluation of the evolution of pellet length over time

For each sample, both length and diameter were measured, but considering that there were no variations in diameter, only length was taken into account. The monitoring was carried out over a period of 90 days, measuring every 7 days, resulting in a total of 14 measurements for each sample.

Fig. 5.17. Detail during pellet length measurements



10 mm die

8 mm die

Fig. 5.18. Examples of variations in time of the length of some pellets obtained with what 2 dies

The curves remain bordered in the range 25 - 40 mm. Only one exception from this behaviour is presented for each die, curves that according to the graph reach zero length, meaning that the pellets disintegrate during the tracking period. Therefore, some of the pellets lose their consistency and become unusable.

In Figure 5.19. are presented examples of pellets that were broken during the tracking period compared to their appearance on the day of obtaining.

Fig. 5.19. Examples of pellets that disintegrated (broke) during evolution tracking
sus – pellets on the day of obtaining; jos – disintegrated pellets

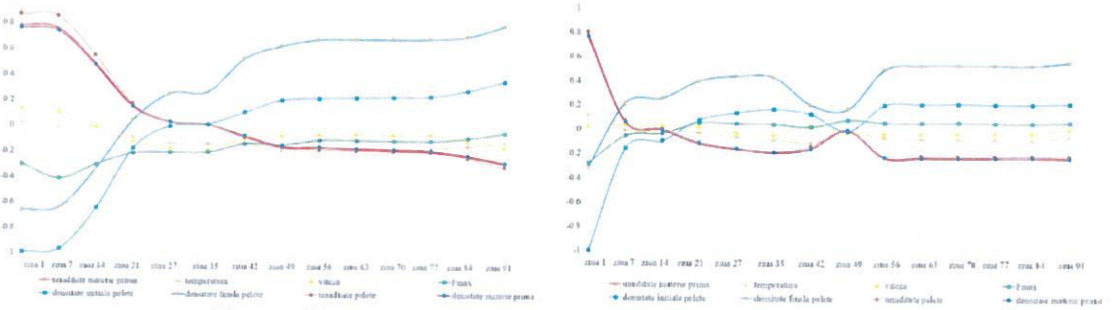
Table 5.5. Distribution of degraded pellets during the 91 days after formation for the 10 mm die

Initial moisture, %	**10**	**13**	**16**
Number of broken pellets	1	0	24
Raw material volumetric mass, kg/m³	**136.10**	**142.20**	**147.40**
Number of broken pellets	1	0	24
Raw material initial volume, cm³	**16.90**	**17.58**	**18.37**
Number of broken pellets	24	0	1
Maximum compression force, kN	**10**	**20**	**30**
Number of broken pellets	10	8	7
Temperature, °C	**70**	**80**	**90**
Number of broken pellets	5	9	11
Pelleting speed, mm/s	**1.3**	**2.1**	**2.8**
Number of broken pellets	6	3	16

Tabelul 5.6. Distribution of degraded pellets during the 91 days after formation for the 8 mm die

Initial moisture, %	**10**	**13**	**16**
Number of broken pellets	0	5	20
Raw material volumetric mass, kg/m^3	**136.1**	**142.2**	**147.4**
Number of broken pellets	0	5	20
Raw material initial volume, cm^3	**16.9**	**17.58**	**18.37**
Number of broken pellets	25	0	1
Maximum compression force, kN	**10**	**20**	**30**
Number of broken pellets	15	5	5
Temperature, °C	**70**	**80**	**90**
Number of broken pellets	7	9	9
Pelleting speed, mm/s	**1.3**	**2.1**	**2.8**
Number of broken pellets	7	11	7

In figure 5.20, the time correlation of the evolution of the length of the pellets with the input and control parameters of the pelleting process for the two dies used, is presented.



10 mm die

8 mm die

Fig. 5.20. Correlation in time with input and control parameters of the pelleting process

In Figure 5.21., the evolution of the length of the pellets obtained with an initial moisture of 10% is presented for the three pelleting speeds used, compared to the samples obtained with the 10 mm die.

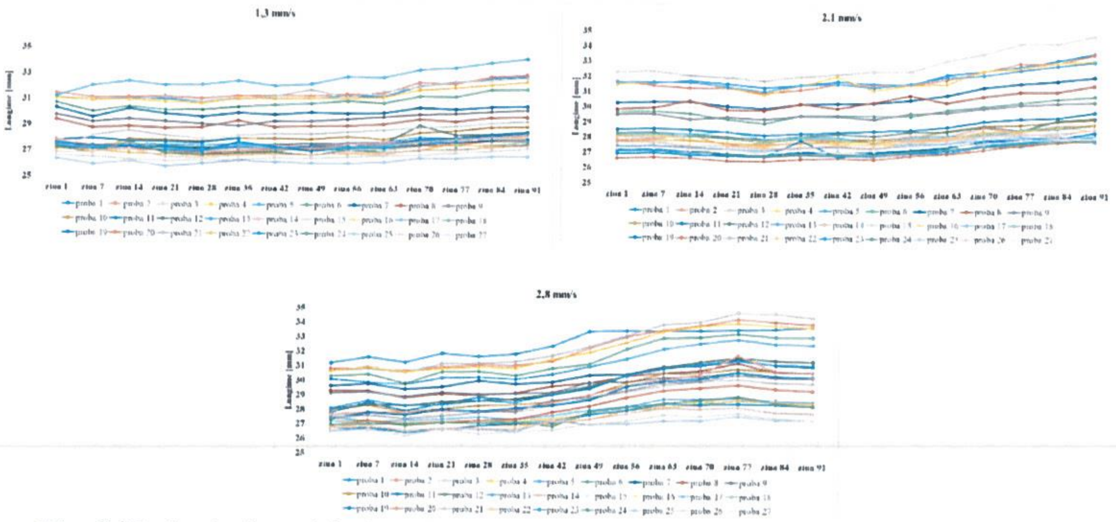


Fig. 5.21. Evolution of the length of pellets obtained with a 10% moisture, at the three pelleting speeds used, for the 10 mm die

In Figure 5.22., the evolution of the length of the pellets obtained with an initial moisture of 10% is presented for the three pelleting speeds used, compared to the samples obtained with the 8 mm die.

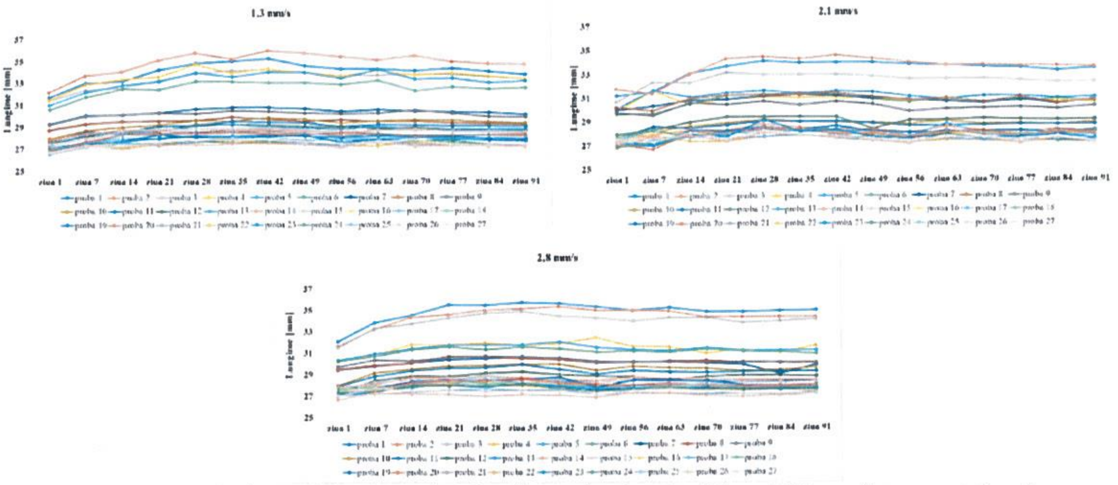


Fig. 5.22. Evolution of the length of pellets obtained with a 10% moisture, at the three pelleting speeds used, for the 8 mm die

5.8. Validation of a series of classical mathematical models for compacting powdery materials with experimental data obtained

A) Jones Model

The first investigated relation is Jones's formula, which, after calculating the model parameters, becomes:

$$\rho_p(\rho_0,P)=\rho_0 e^{1.321}\left(\frac{P}{P_0}\right)^{0.081} \tag{5.1}$$

A graphical comparison between the experimental and the calculated data using Jones model is shown in Figure 5.23.

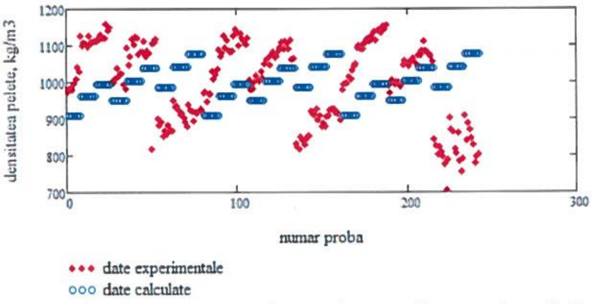


Fig. 5.23. Comparative graphical representation of experimental and the calculated data using the Jones model

B) Heckel Model

After calculating the model parameters, the Heckel model has the form:

$$\rho_p(\rho_0,P)=\frac{\rho_0}{1-e^{0.00000552\frac{P}{P_0}-0.17}} \tag{5.2}$$

A graphical comparison between the experimental and the calculated data using Heckel model is shown in Figure 5.23.

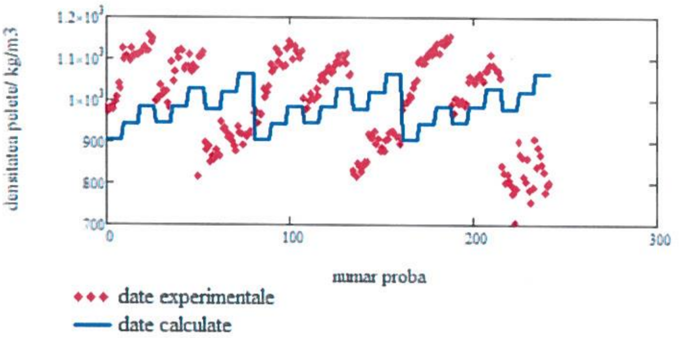


Fig. 5.24. Comparative representation of the experimental and the calculated data using the Heckel model

5.9. Conclusions

Following the analysis of the experimental results, it was found that:

- on average, the energy consumed for the production of a pellet was higher for the 10 mm die than for the 8 mm die;
- under the same conditions, 8 mm pellets of had a higher moisture than 10 mm pellets;
- pellets obtained with 16% material moisture had the longest length for both dies, with more than 5 mm compared to those obtained with 10% material moisture;
- pellets obtained using the 8 mm die had a density higher than those obtained using the 10 mm die, the average density of the 8 mm pellets being 1075.22 kg / m^3, and that of the 10 mm pellets was 999.66 kg / m^3;
- the greatest influence on the average density of the pellets was the moisture of the material, on average the density increasing from 964.30 kg / m^3 (8 mm die) and 870.10 kg / m^3 (10 mm die) in the case of 16% material moisture to 1099.54 kg / m^3 (8 mm die) and 1051, 16 kg / m^3 (10 mm die) at 13% material moisture, reaching 1161.83 kg / m^3 (8 mm die) and 1077.73 kg / m^3 (10 mm die).

Tracking the evolution of the physical characteristics of pellets over time is a mandatory test for estimating their quality.

An important conclusion is that the high moisture of the raw material leads to a high moisture of the pellets, which causes most of the pellets to disintegrate. In the same situation is the volumetric mass of the raw material. Its growth leads to pellets with higher density, but which can disintegrate more easily.

CHAPTER 6

RESEARCHES ON THE OPTIMIZATION OF THE BIOMASS PELLETING PROCESS BASED ON EXPERIMENTAL RESULTS

6.1. Validation of the models with the experimental data obtained using the single pellet stand and process optimization

From the experimental research of pelleting fir tree sawdust using the single pellet stand and analyzing and interpreting the data, it was possible to test the accuracy of the models proposed in chapter 4.

Model 1

By replacing the values of the model parameters in model 1, we obtained:

$$\rho_p(\rho_0, P, U_i) = 1,.698\rho_0 \left(\frac{P_i}{P_0}\right)^{0,081} U_i^{-0.61} \tag{6.1}$$

Figure 6.1 shows a graphical comparison between the experimental the calculated data.

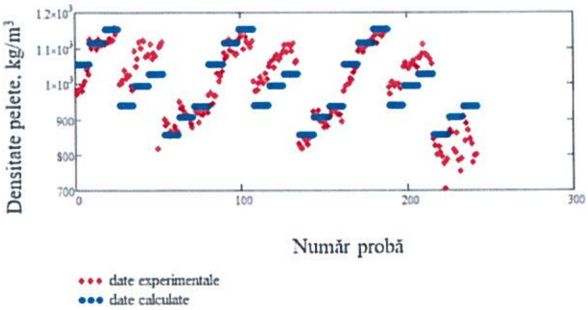


Fig. 6.1. Comparative representation of the experimental and the calculated data using **model 1**

The graphical representation of pellet density dependence on the moisture of the sawdust and the maximum applied pressure is given in Figure 6.2.

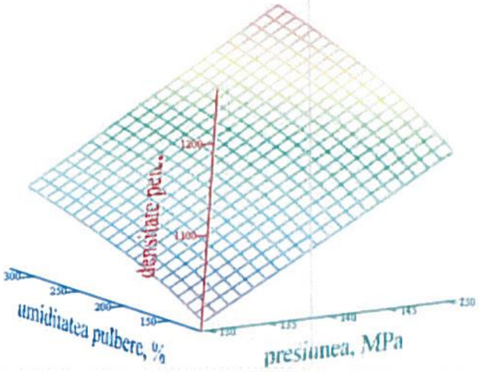


Fig. 6.2. Dependence of pellet density on material material moisture for fixed values of the volumetric mass of the raw material and the maximum pressure

In figure 6.3 it is represented, in the form of surface and isocline, the dependence of pellet density on two variables (according to formula (6.1)), the volumetric mass of the material and the maximum pressure realized in the compression plant, for the used moisture interval.

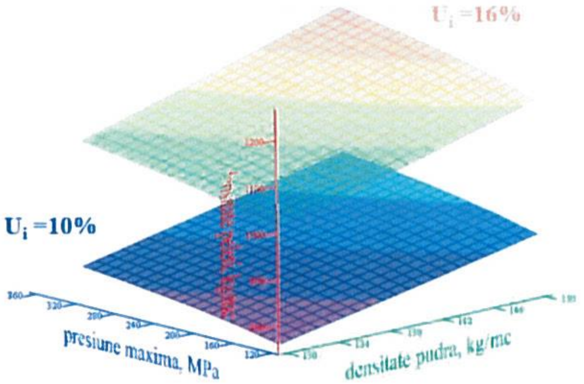


Fig. 6.3. Dependence of pellet density on the volume of the sawdust and the maximum pressure applied, for the limits of the moisture range considered for experiments

Model 2

By replacing the values of the model parameters in model 2, we obtained:

$$\rho_p(\rho_0, p, U_i) = \rho_0 \left(\frac{Q}{\rho_0 \cdot v^2 \cdot V_0}\right)^{0,097} \left(\frac{p_i}{p_0}\right)^{0,133} U_i^{-0,514}. \tag{6.2}$$

Figure 6.4. shows a graphical comparison between the experimental and the calculated data using model 2.

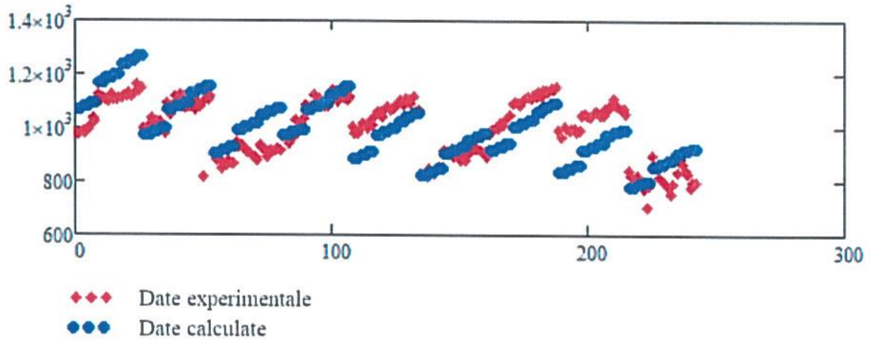


Fig. 6.4. Comparative graphical representation of the experimental and the calculated data using model 2

The graphical representation of the dependence of pellet density on the moisture of the sawdust and on the maximum applied pressure is given in figure 6.5.

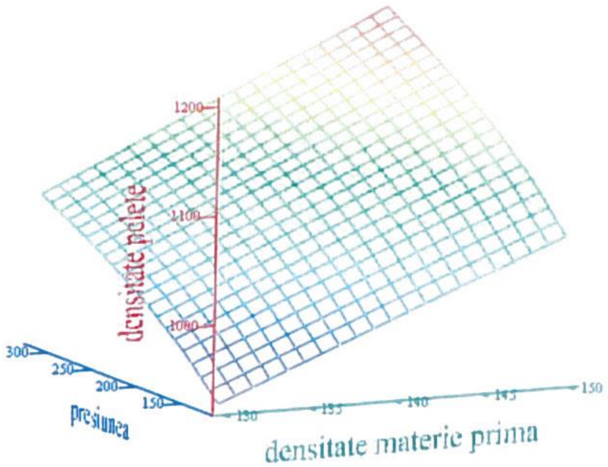


Fig. 6.5. Dependence of pellet density on the maximum pressure and the material volumetric mass

6.2. Validation of mathematical models using a ring die pelletizing machine

In order to validate the mathematical models proposed, experimental tests were performed for pelleting the sawdust using a ring die pellet press with two pressing rolls (fig. 6.6), in which the biomass was introduced at the top, the actual process of pelleting being achieved at the contact point between the pressing rolls and the mold.

The pelleting press with fixed ring and pressing rollers driven by a rotor shaft is an equipment intended for obtaining pellets that can be used in plants for the production of heat and domestic hot water, for small and medium-sized farms, as well as for households, aiming at ensuring energy independence.

The mass of material represented by chopped biomass is inserted into the bunker of the press feed conveyor, from here it is transported through the magnetic separator and the feed pipe into the pelleting press cyclone by the refiner. The cyclone evacuation for feeding the pelleting press is made by means of a shifter and a rotary valve.

Fig. 6.6. Ring die pelleting equipment used to conduct the experiments
1. mini bunker for material feed; 2. Pellet curring blade motor; pelleting die casing;
4. feeding bunker motor; 5. Command panel; 6. Press main motor

A number of 6 samples (2 samples for the three raw material moistures used: 10%, 13% and 16%) were obtained from fir tree sawdust with dimensions smaller than 2 mm. The sawdust had the same characteristics as the one used for conducting experimental research on the single pellet stand.

Using the pelleting machine, the pellets from fir sawdust were obtained using the following methodology:

1. The initial moisture of the raw material was determined using the thermal balance.
2. The required mass (10 kg for each sample) was measured for insertion into the pellet press using a balance.
3. The pellet blade casing and blade were demounted to allow the force measurement dose to be mounted on the ring die and a material feed funnel was fitted;
4. The tensometric dose was mounted on the ring die;
5. The dose was connected to a data acquisition system (MGCPlus - HBM, Germany), which in turn was connected to a computer to record the data;
6. The actual pelleting process was conducted using the pelleting press;
7. The size of 40 pellets in each pellet sample was measured.
8. The 40 pellets from each sample were weighed for further determination of density.

Fig. 6.7. Pellets obtained using the ring die pelleting machine

Following the experimental researches on the pelleting machine with fixed ring die and of the measurements and density calculations, the following results were obtained, presented in tables 6.1 - 6.3.

Table 6.1. The results obtained after conducting the experimental researches on the ring die pelleting machine for the moisture of **10%**, the temperature of **90 °C**, the volumetric mass of the material of **136.08 kg / m^3** and the pelleting pressure of **107 Mpa**

Proba	ρ_p	Proba	ρ_p	Proba	ρ_p	Proba	ρ_p
	kg/m^3		kg/m^3		kg/m^3		kg/m^3
1	1074,38	11	1145,12	21	1178,37	31	1152,13
2	1123,98	12	1121,47	22	1147,93	32	1198,78
3	1184,52	13	1152,32	23	1109,74	33	1138,29
4	1147,48	14	1147,81	24	1142,11	34	1183,09
5	1082,13	15	1141,16	25	1098,00	35	1149,33
6	1063,95	16	1083,19	26	1148,54	36	1161,95
7	1104,84	17	1142,30	27	1139,52	37	1150,21
8	1081,56	18	1108,22	28	1110,12	38	1123,18
9	1112,74	19	1074,85	29	1097,44	39	1141,04
10	1142,89	20	1156,08	30	1189,91	40	1139,90

Table 6.2. The results obtained after conducting the experimental researches on the ring die pelleting machine for the moisture of **13%**, the temperature of **90 °C**, the volumetric mass of the material of **142.18 kg / m^3** and the pelleting pressure of **107 Mpa**

Proba	ρ_p	Proba	ρ_p	Proba	ρ_p	Proba	ρ_p
	kg/m^3		kg/m^3		kg/m^3		kg/m^3
1	1054,36	11	1076,42	21	1050,85	31	1087,29
2	1081,22	12	1055,38	22	1071,35	32	1127,86
3	1131,65	13	1061,23	23	1058,52	33	1084,18
4	1115,34	14	1065,50	24	1088,11	34	1116,16
5	1026,61	15	1068,50	25	1022,91	35	1084,29
6	995,69	16	1035,08	26	1079,73	36	1086,56
7	1054,88	17	1073,84	27	1072,83	37	1057,42
8	1025,51	18	1042,62	28	1037,97	38	1046,16
9	1050,76	19	954,70	29	1035,55	39	1069,40
10	1085,66	20	1080,52	30	1127,15	40	1060,46

Table 6.3. The results obtained after conducting the experimental researches on the ring die pelleting machine for the moisture of **16%**, the temperature of **90 °C**, the volumetric mass of the material of **147.37 kg / m^3** and the pelleting pressure of **105 Mpa**

Proba	ρ_p	Proba	ρ_p	Proba	ρ_p	Proba	ρ_p
	kg/m^3		kg/m^3		kg/m^3		kg/m^3
1	877,31	11	883,94	21	888,93	31	904,33
2	878,85	12	914,85	22	907,83	32	903,63
3	901,85	13	878,48	23	898,29	33	893,84
4	901,20	14	875,92	24	900,47	34	889,25
5	902,85	15	884,28	25	910,78	35	897,43
6	910,72	16	901,87	26	886,71	36	901,72
7	904,28	17	907,20	27	917,24	37	884,62
8	909,75	18	889,84	28	879,39	38	871,59
9	891,28	19	885,40	29	888,86	39	893,74
10	907,28	20	864,96	30	894,62	40	905,70

Figure 6.8 shows a graphical comparison between the experimental and the calculated data using relation 6.1.

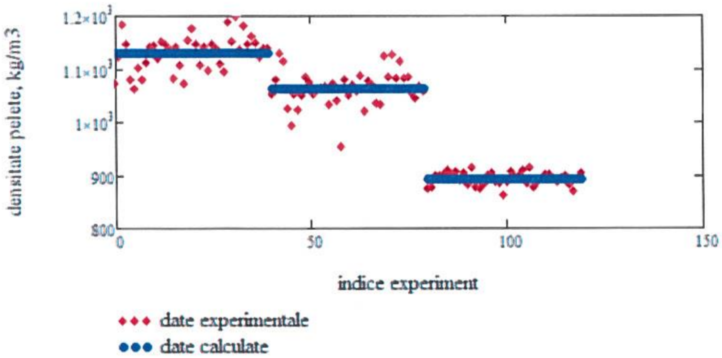


Fig. 6.8. Comparative graphical representation of the experimental data obtained with the pelleting machine and of the data calculated using the relation (6.1)

Figure 6.9 shows the dependence of pellet density on the input and control parameters, for relation 6.1.

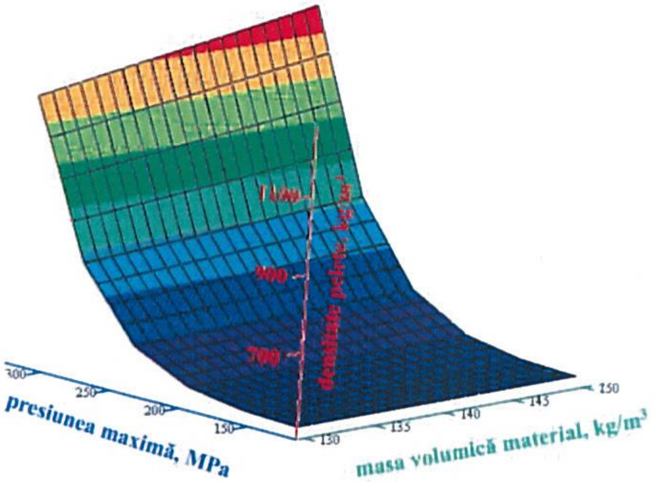


Fig. 6.9. Dependence of pellet density on the maximum pressure and the volumetric mass of the sawdust for the samples obtained using the ring die pelleting machine, for relation 6.1

Figure 6.10 shows a graphical comparison between the experimental and the calculated data using relation 6.2.

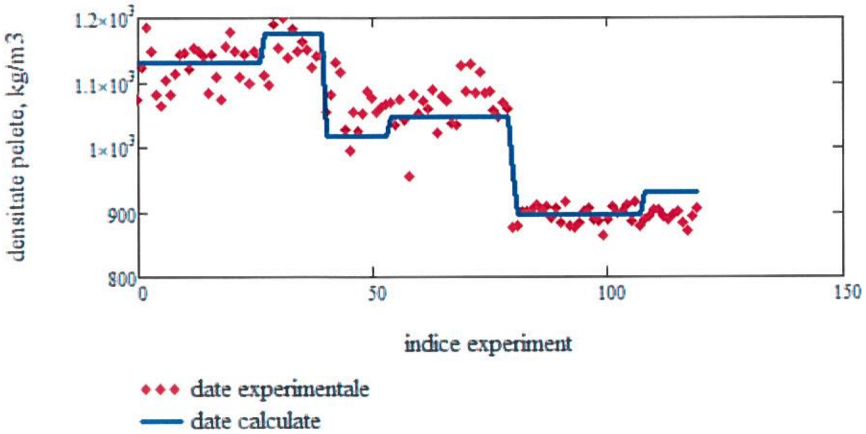


Fig. 6.10. Comparative graphical representation of the experimental data obtained with the pelleting machine and of the data calculated using the relation (6.2)

Figure 6.11 shows pellet density dependence of the pressure and the volumetric mass of the raw material, for the relation 6.2.

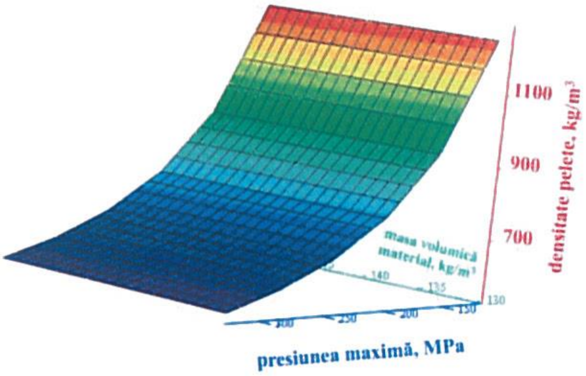


Fig. 6.11. Dependence of pellet density on the maximum pressure and the volumetric mass of the sawdust for the samples obtained using the ring die pelleting machine, for relation 6.2

6.3. Statistical modelling of biomass pelletization processes

The formulation of the problem in terms of systems theory, requires a sorting of the parameters that mathematically describe the system, such as the one in table 6.4.

Table 6.4. Parameters of the pelleting process and of the pellets obtained in the system type organization

Parameter type	Parameter name	Notation	Measurement unit	Physical unit
Input	Raw material granulation	g	m	L
	Raw material volumetric mass	ρ_0	kg/m^3	ML^{-3}
	Material moisture	U_i	%	-
	Raw material initial volume	V_0	m^3	L^3
Control	Die diameter	$\O_m$	m	L
	Maximum applied force	F_{max}	kN	MLT^{-2}
	Piston movement speed	v	m/s	LT^{-1}
	Die temperature	θ	°C	
	Pellet extraction force	F_e	kN	MLT^{-2}
Output	Consumed energy	E_c	Wh	ML^{2}T^{-2}
	Pellet length	L	m	L
	Pellet density	ρ_p	kg/m^3	ML^{-3}
	Pellet moisture	U_p	%	-
	Pellet volume	V_p	m^3	L^3

A selection of the input and control parameters based on their influence on the output parameters, can reduce the number of arguments of each output parameter function. As measure of influence can be taken, in the first approximation, the value of the correlation between the numerical series made by them within the experiments.

The estimations of the correlations of the output parameters related to the input and control parameters and the order considered in this process are given in table 6.5.

Table 6.5. Matrix of input / output correlations

	ρ_0	U_i	V_0	F_{max}	v	θ
E_c	-0.236000	-0.242000	0.232000	0.644000	-0.112000	-0.027000
L	0.763000	0.78000	-0.753000	-0.305000	0.134000	0.023000
ρ_p	-0.779655	-0.796201	0.770537	0.33332	-0.111758	-0.007409
U_p	0.937586	0.948841	-0.93105	-0.098442	0.085247	-0.08906
V_p	0.768073	0.785466	-0.75853	-0.301463	0.134581	0.027897

Considering the values of the correlations in table 6.4, the following dependencies were considered for the output parameters:

$$E_c=E_c(F_{max},F_e) \tag{6.3}$$

$$L=L(U_i,\rho_0,V_0,F_{max}) \tag{6.4}$$

$$\rho_p=\rho_p(U_i,\rho_0,V_0,F_{max}) \tag{6.5}$$

$$U_p=U_p(U_i,\rho_0,V_0) \tag{6.6}$$

$$V_p=V_p(U_i,\rho_0,V_0,F_{max}) \tag{6.7}$$

The regression calculation was made for all the models expressed in the form of polynomials of order 1 or 2, using for interpolation the data obtained from the experiments and the Mathcad program.

A) Polynomial regression for pellet density

Considering the structure of the pellets density function of the form (6.5), the expression of linear regression was obtained:

$$\rho_p(U_i,\ \rho_0,\ V_0,\ p)=-18784.920 - 357.766\ U_i+171.590\rho_0 -9.504V_0+3.41\cdot10^{-7}\ p \tag{6.8}$$

In support of the prediction made, the coefficient of determination $R^2 = 0.862$ is also advocated. In order to increase the estimation accuracy, linear regression can be calculated with respect to all six input and control variables varied within the experiment program, also in the form of a first-degree polynomial. The following expression is obtained:

$$\begin{gathered}\rho_p(U_i,\rho_0,V_0,p,v,\theta)=-18737.058 - 357.766\ U_i + 171.590\ \rho_0 - 9.504V_0 + \\ +3.41\cdot10^{-7}p - 19414.676\ v-0.097\ \theta.\end{gathered} \tag{6.9}$$

The global error of regression (6.9) reaches the value $\varepsilon_g = 0.00242$, and the maximum error increases to the value $\varepsilon_{max} = 0.29087924$, compared to the regression on the system reduced by variables, (6.8). The coefficient of determination is better for regression (6.9) than that of regression (6.8), $R^2 = 0.874$.

Also, for the density of pellets at the exit from the formation process, the second degree polynomial can be calculated in the version of the reduced variable set. Its expression is:

$$\begin{gathered}\rho_p(U_i,\rho_0,V_0,p)= -17391.251+245.906\ U_i + 210.165\ \rho_0-3.877\ V_0+ \\ +0.000018P - 1.668\ U_i\rho_0 + 0.041\ U_iV_0 + 0.0000002025078\ U_iP + 0.01008\ \rho_0V_0 - \\ +0.0000001335\ \rho_0P + 0.000000068\ V_0p - 3.573U_i^2 - 0.550131\ \rho_0^2-0.000107\ V_0^2.\end{gathered} \tag{6.10}$$

The term in the second power of pressure is negative, but multiplied by a negligible factor, 10^{-15}. The quadratic regression (6.10), has the following performances: $\varepsilon_g=0.002244$, $\varepsilon_{max}=0.26145721$, $R^2=0.892$.

B) Polynomial regression for the energy consumed

For the consumed energy, the reduced first degree polynomial regression has only two arguments, according to the correlation selection (table 6.5.), being given by the relation:

$$E_c(p)=2.65+0.00000001251\,p+0.000000001123\,p_e \tag{6.11}$$

with relation between F_{max} and the maximum pressure given by $p=\frac{4F_{max}}{\pi\emptyset^2}$ şi with the force of extracting the pellet F_e and the extraction pressure p_e. The precision of this relation is given by the values: $\varepsilon_g=0.017$, $\varepsilon_{max}=0.6439$, $R^2=0.414$. The formula of the regression polynomial of the second degree, is practically also of the first degree, because the coefficient of the term of the second degree in the compression pressure is below the negligible limit of10^{-15}:

$$E_c(p)=8.37037037-0.000000041403088\,p \tag{6.12}$$

The performance of this relation is measured by values: $\varepsilon_g=0.014$, $\varepsilon_{max}=0.6854725$, $R^2=0.575$. Considering linear regression dependent on all six experimentally varied arguments does not improve accuracy: $\varepsilon_g=0.016$, $\varepsilon_{max}=0.6101372$, $R^2=0.503$. Slightly better performances are obtained by considering the polynomial regression of the second degree, for the whole set of variables.: $\varepsilon_g=0{,}011$, $\varepsilon_{max}=0.56400485$, $R^2=0.742$.

C) Polynomial regression for the length of the pellets

The linear regression for the pellet length parameter at the output of the compression process, for the reduced set of variables, is given by the expression:

$$L(U_i,\rho_0,V_0,p)=-3.664-0.000002978\,U_i+0.013\,\rho_0+10226.285\,V_0-10^{-11}p. \tag{6.13}$$

with the precision performances estimated by values: $\varepsilon_g=0.003028$, $\varepsilon_{max}=0.30457$, $R^2=0.831$.

The first order linear regression for the length of the pellets at the exit of the forming process, for the whole set of variables considered, is given by the equation:

$$\begin{aligned}L(U_i,\rho_0,V_0,p,v,\theta)&=0.726+0.0134\,U_i-0.006042\,\rho_0+0.000334\,V_0-\\&10^{-11}\,p+0.778\,v+0.00001007\,\theta.\end{aligned} \tag{6.14}$$

The performances of this model are described by the following characteristics: $\varepsilon_g=0.00283$, $\varepsilon_{max}=0.28575897$, $R^2=0.852$.

The linear regression of the second degree in the reduced set of variables is characterized by the following values: $\varepsilon_g=0.00283$, $\varepsilon_{max}=0.28575897$, $R^2=0.852$. For the linear regression of the second degree in the complete set of variables, the following precision estimates are obtained: $\varepsilon_g=0.002851$, $\varepsilon_{max}=0.32654358$, $R^2=0.85$.

D) Polynomial regression for pellet moisture

The linear regression of the pellet moisture has the expression:

$$U_p(U_i,\rho_0,V_0)=231.414+4.416\,U_i-1.961\,\rho_0+1.11\,V_0. \tag{6.15}$$

and precision assessment: $\varepsilon_g=0.002549$, $\varepsilon_{max}=0.16885892$, $R^2=0.95$. The precision of the quadratic regression for the moisture of pellets, in the version of the reduced set of parameters, has a poor accuracy assessment and cannot be taken into account.

The linear regression on the complete set of variables has the expression:

$$\begin{aligned}U_p(U_i,\rho_0,V_0,p,v,\theta)&=232.954+4.416\,U_i-1.962\,\rho_0+0.111\,V_0-\\&-0.0000000017\,P+259.31\,v-0.02\,\theta\end{aligned} \tag{6.16}$$

Evaluation of the precision of relation (6.16) is: $\varepsilon_g=0.001804$, $\varepsilon_{max}=0.12492327$, $R^2=0.975$.

Second order quadratic regression is characterized by even better precision: $\varepsilon_g=0.001588$, $\varepsilon_{max}=0.11079441$, $R^2=0.981$.

E) Polynomial regression for pellet volume

The linear regression of the volume of the pellets at the exit of the formation process, for the reduced set of variables, has the expression:

$$V(U_i,\rho_0,V_0,p)= - 0.0002986 - 0.00000000023\ U_i + 0.000001\ \rho_0 + 8.333\ V_0. \tag{6.17}$$

Precision performance of relation (6.17) are given by $\varepsilon_g=0.002957$, $\varepsilon_{max}=0.3042955$, $R^2=0.838$. For the second degree regression, with the reduced set of variables, the accuracy assessment is given by the following values: $\varepsilon_g=0.00277$, $\varepsilon_{max}=0.28586421$, $R^2=0.857$.

The linear regression of the first degree with the complete set of variables, for the volume of the pellets at the exit from the process has the expression:

$$\begin{gathered}V(U_i,\rho_0,V_0,p,v,\theta)=0.000059 +0.000001\ U_i -0.0000004917\rho_0+0.00000002719V_0+\\ +0.00006352\ v+0.000000000988\ \theta.\end{gathered} \tag{6.18}$$

The performance of regression (6.18) are given by: $\varepsilon_g=0.002774$, $\varepsilon_{max}=0.32612782$, $R^2=0.857$. Second order linear regression with complete set of variables has even better accuracy performance: $\varepsilon_g=0.001957$, $\varepsilon_{max}=0.28689592$, $R^2=0.929$.

The precision characteristics for all the investigated interpolation relations for each output parameter as a function of input and control parameters are concentrated in Table 6.6.

Table 6.6. Precision estimators of the dependency laws of the output parameters (qualitative) on the input and control parameters

Parameter	Degree of regression	Versions	ε_g	ε_{max}	R^2
ρ_p	1	Reduced	0.002538	0.27598956	0.862
		Complete	0.002420	0.29087924	0.874
	2	Reduced	0.002244	0.26145721	0.892
		Complete	0.001703	0.26425000	0.938
E_c	1	Reduced	0.017000	0.64390000	0.831
		Complete	0.016000	0.61013720	0.503
	2	Reduced	0.014000	0.68547250	0.575
		Complete	0.011000	0.56400485	0.742
L	1	Reduced	0.003028	0.30457000	0.831
		Complete	0.002830	0.28575897	0.852
	2	Reduced	0.002830	0.28575897	0.852
		Complete	0.002851	0.32654358	0.850
U_p	1	Reduced	0.002549	0.16885892	0.950
		Complete	0.001804	0.12492327	0.975
	2	Reduced	-	-	-
		Complete	0.001588	0.11079441	0.981
V	1	Reduced	0.002957	0.3042955	0.838
		Complete	0.002774	0.32612782	0.857
	2	Reduced	0.002770	0.28586421	0.857
		Complete	0.001957	0.28689592	0.929

Figure 6.12. presents graphically the dependence of pellets density on the moisture of the raw material, in the version of linear and quadratic regression, with a small number of arguments and with the complete set of arguments. A theoretical optimum of the input moisture of the pelletized material is observed, which is around 11%.

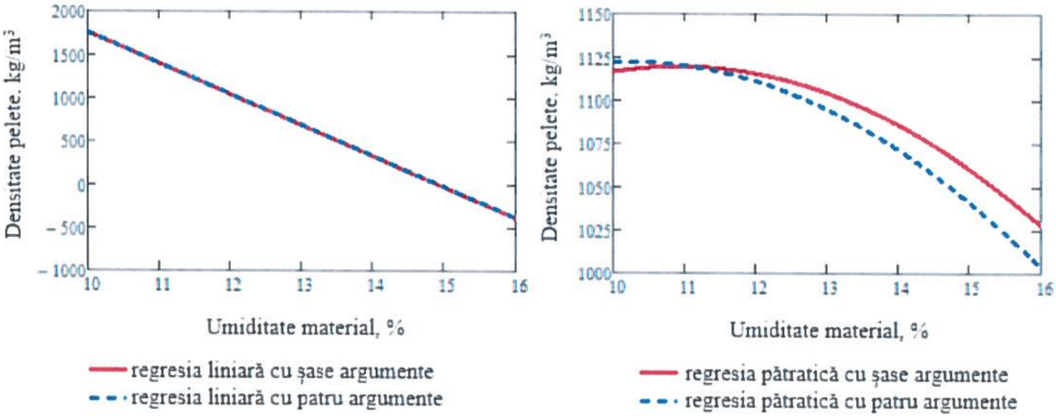


Fig. 6.12. Dependence of pellet density on raw material moisture, in different regression versions, for the constant values: ρ_0=140kg/m³, V_0=0,00001837 m³, p = 300 MPa, v = 0,002m/s, θ=80°C

The energy consumed during the pelleting process depends most strongly on the compression pressure and the pressure required to extract the pellet from the die. However, moisture also significantly influences the energy consumed. Figure 6.13 shows the variation of the consumed energy with the working pressure (implicitly with the compressive force and the extraction force), for four values of the moisture of the raw material. **Thus, an optimal operating point is observed from the point of view of the working process, corresponding to the value of 270 MPa for which the energy consumption is constant regardless of the moisture of the raw material.**

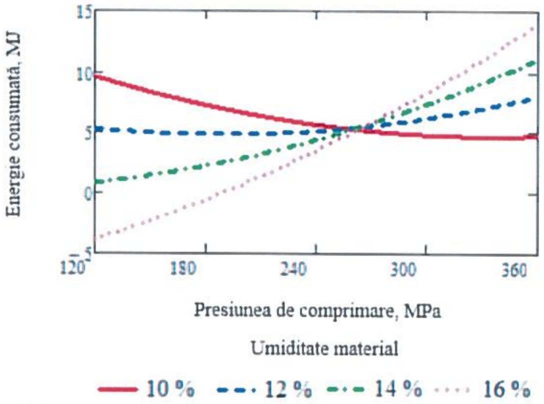


Fig. 6.13. Dependence of the energy consumed of the maximum pressure applied, for various values of misture, the other variables being kept constant: V_0=0,00001837 m³, p = 300 MPa, v = 0,002 m/s, θ = 80 °C

Also, the energy consumption E_c, can be optimized using one of the non-linear regression versions to obtain the multi-parameter model of its variation.

The energy consumption on the pellet mass unit (E_{cmp}) function is formed, by combining three qualitative functions from the set of five that are available, so that the obtained model is dimensional correct:

$$E_{cmp}\left(U_i,\rho_0,V_0,p,v,\theta\right)=\frac{E_c\left(U_i,\rho_0,V_0,p,v,\theta\right)}{\rho_p\left(U_i,\rho_0,V_0,p,v,\theta\right)\cdot V\left(U_i,\rho_0,V_0,p,v,\theta\right)},\ (\mathrm{J/kg}) \qquad (6.19)$$

Figure 6.14. presents the variation of the energy consumed depending on the compression pressure for 5 moistures of the material, obtaining an optimum point located around the pressure of 250 MPa, for which the energy consumed is constant regardless of moisture.

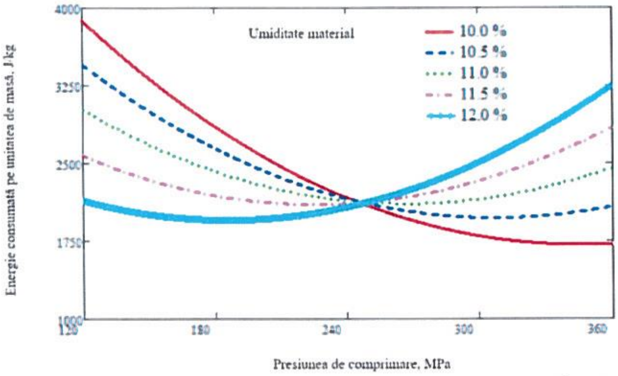


Fig. 6.14. Variation of energy consumption specific to the mass unit E_{cmp} of pellets, with the compression pressure for five values of the moisture content of the material

6.4. Conclusions

The two dependency laws tested and calibrated on the experimental data (model 1 and model 2 presented in chapter 4) are linear depending on the density of the raw material. Therefore, the density of the pellets depends linearly on the density and volume of the raw material introduced into the pelleting process. The density dependence of the pellets is non-linear with respect to the relative pressure and therefore with the maximum pressing force of the material in the die. The density dependence of the pellets is also nonlinear in relation to the moisture of the raw material introduced into the process, as well as in relation to the temperature of the die.

The statistical interpolation study shows that good accuracies are possible using reduced sets of variables, but also complete sets. The analysis of the polynomial regression study highlights that the quality (output) parameters vary monotonically with respect to the input and control parameters, within the limits of the experimental field of work. Precision performances can guide the researchers and the users of the results towards choosing a suitable interpolation version in order to improve the working regimes.

After conducting the experimental research on the stand and validating the two mathematical models proposed in Chapter 4, the data obtained showed a strong correlation between the experimental data and the theoretical estimates, $R = 0.837$ for model 1 and $R = 0.711$ for model 2.

For the data obtained on the ring die pelleting machine, there was an even stronger correlation between the experimental data and the theoretically estimated data, $R = 0.911$ for model 1 and $R = 0.847$ for model 2.

CHAPTER 7

GENERAL CONCLUSIONS. CONTRIBUTIONS. RECOMMENDATIONS AND PERSPECTIVES

7.1. Conclusions on the theoretical and experimental researches

As a result of the theoretical and experimental research carried out within the present doctoral thesis "**Researches on optimizing the process of biomass densification**", the following general conclusions can be drawn:

1. the densification of biomass depends on the complex interaction between the material properties and the parameters characteristic to the equipment used for densification.

2. the biomass pelleting machines perform the compaction by forcing the passage of the material through the orifices of a special die.
3. in order for the *material* to pass through the orifices of the die and form pellets, it must have the following properties:
    - adequate flow and cohesion capacity;
    - particle size (if the material is too fine, it results in a high flow, but a low cohesion; if the material is too coarse, it has a good cohesion, but a low flow, it can even cause the equipment to block);
    - superficial adhesion forces (important for the agglomeration of the material and for the resistance in time of pellets);
    - adhesiveness (given by the chemical composition of the biomass, very important being the lignin content of the raw material);
    - hardness (the existence of too hard particles in the mass of material creates difficulties in agglomeration);
    - particle distribution (a sufficient quantity of fine particles is required to bind the larger particles together);
    - adequate moisture depending on the nature of the material (lower values for woody biomass and higher for agricultural biomass);
    - mechanical resistance to compression.
4. in order to ensure a smooth process and an adequate quality of the pellets obtained, there are a number of *command and control parameters* that must be taken into account.:
    - compression force (a compression force large enough to force the material to pass through the orifices of the die must be ensured);
    - working temperature during the process (it is necessary to develop a temperature sufficiently high to gelatinize the starch and activate lignin, which leads to the binding of pellets, but not as high so that the ignition phenomenon does not occur during compression.);
    - pelleting speed or retention time (a speed too high causes the material to not have sufficient time to form bonds and thus produce inferior quality products, which are destroyed shortly after obtaining);
    - wear of used equipment elements (high wear of the elements makes the process more difficult, leads to poor quality and even process failure).
5. the mathematical models developed for the compaction process help to present the behavior of the particles during the compaction process and can help to optimize the parameters to obtain good quality products. Several types of equations that express the relationship between the compaction parameters of raw materials are available in the literature.
6. taking into account the parameters that influence the pelleting process, a mathematical model that expresses the final density of pellets as determined by the final pressure applied during the process, the initial material moisture and the initial density of the material was proposed, but also a mathematical model obtained. by dimensional analysis, using Π theorem, which expresses the density of pellets as determined by pressure, heat, the initial density of the material, the pelleting speed of and the initial volume of the material.
7. the experimental researches conducted on a single pellet stand are used to simulate the pelleting conditions in a press, allowing a detailed analysis of the factors that influence the quality of the pellets obtained, offering the possibility to carry out a large number of experiments, easily varying the input and control parameters.
8. in the thesis, the experiments were conducted using two dies with a single channel, with pelleting orifices of 8 mm, respectively 10 mm. The material used was fir tree sawdust with particle sizes <2 mm.

9. the input and control parameters, chosen according to the experimental plan, were: the humidity of the material *U* (10%; 13%; 16%), temperature during the process θ (70 °C; 80°C; 90 °C), compaction force F_{max} (10 kN; 20 kN; 30 kN) and pelleting speed *v* (1.3 mm/s; 2.1 mm/s; 2.8 mm/s). For each of the input and control parameters, three different values were considered (resulting in a total of 243 experiments for each used die).
10. the output parameters, those that express the quality of the pellets and the efficiency of the pelleting process are expressed by:
    - pellet length at the exit of the process;
    - pellet volume at the exit of the process;
    - pellet density at the exit of the process;
    - pellet moisture at the exit of the process;
    - the durability of pellets over time;
    - the energy consumed to obtain the pellets.
11. as a result of the experimental research and the analysis of the experimental data obtained, a series of correlations were made between the parameters of the working process which give the first reliable information about the intensity of the connection between the different variables that describe the process of compressing the biomass material. The results of the correlations were highlighted and the consequences of the values of the components of the correlation matrix on the intensity of the correlation between correlations were explained.
12. there were strong connections between the density and the relative pressure of compaction, and of both with the raw material moisture.
13. it was noted that a high raw material moisture (16%) leads to a high moisture of the pellets, the loss in moisture through the pelleting process being on average 2.94% for the 8 mm die and 3.06% for the 10 mm one, which causes most of the pellets to disintegrate. The same behavior has the volumetric mass of the raw material, where its growth leads to obtaining pellets that disintegrate more easily.
14. the energy consumed E_c depends moderately on the compaction force F_{max} and insignificantly on the other input and control parameters. On average, E_c had higher values for the 10 mm die for U_i = 10% and 16% (by 5.6%) and smaller U_i = 13% (by 11,05%).
15. the dependencies of most qualitative parameters are related to the input parameters (the moisture of the material) and less to the control parameters. The dependence on the compression speed and the temperature of the die were low for all parameters.
16. after conducting the experimental research on the stand and validating the two mathematical models, a strong correlation between the experimental data and the theoretical estimates was obtained, $R = 0.837$ for model 1 and $R = 0.711$ for model 2.
17. the interaction between the parameters of the pelleting process was studied using the method of statistical modeling, more precisely the polynomial regressions. The correlations between the output parameters (those that indicate the quality of the pelletizing process) and the input and control parameters, show that the five output parameters are in strong relation with only a part of the input and control parameters. Pellet length (L_p), pellet density (ρ_p), pellet moisture (U_p) and pellet volume (V_p) strongly depend on the volumetric mass of the material (ρ_0), material moisture (U_i), material volume (V_0) and weaker than the maximum compression force (F_{max}), except for U_p that does not show any relation with F_{max}, according to the correlation calculation.
18. a theoretical optimum of the moisture inlet of the pelleted material was observed, with values around 11%.
19. an optimum operating point was observed from the point of view of the working process, corresponding to the value of 270 MPa for which the energy consumption is constant regardless of the moisture of the raw material.

20. for the energy consumed per unit mass, an optimum point was obtained around the pressure of 250 MPa, for which the energy consumed is constant regardless of moisture.
21. for the data obtained on the ring die pelleting machine, there was an even stronger correlation between the experimental data and the theoretically estimated data, $R = 0.911$ for model $\rho_p(\rho_0, p, U_i) = a\rho_0 \left(\frac{p}{p_0}\right)^b U_i^c$ and $R = 0{,}847$ for model $\rho_p(\rho_0, p, U_i) = \rho_0 \left(\frac{Q}{\rho_0 \cdot v^2 \cdot V_0}\right)^a \left(\frac{p}{p_0}\right)^b U_i^c$.

7.2. Personal contributions

As a result of the theoretical and experimental researches conducted within the present thesis, a series of personal contributions of the author are revealed, from which we can mention:

1. performing a synthetic analysis of the specialized literature regarding the current state of theoretical and experimental research in the field of biomass compaction, in general, and pelletization, in particular, by studying a number of more than 300 specialized papers in the country and abroad, out of which 132 can be found in the bibliography of the present doctoral thesis, the others being given in the semester doctoral reports.
2. development of two mathematical algorithms regarding the density of biomass pellets depending on the process input and control parameters.
3. development of an experimental single pellet stand in order to conduct the experimental researches of biomass densification.
4. development of an experimental methodology for pelleting biomass using the experimental stand.
5. physico-chemical characterization of the biomass used in the pelleting process by determining the moisture content, ash, volatile matter, lower calorific power.
6. conducting the experimental researches and processing the experimental data;
7. validation of mathematical models regarding the biomass densification, both on the experimental stand and on a ring die pelleting machine.
8. performing the statistical modeling and optimizing the biomass pelleting process.
9. giving a set of conclusions and recommendations that may prove useful to the specialists in the field of biofuels, contributing to the deepening of knowledge in the field of biomass densification.
10. the results obtained from the studies and researches carried out within the present doctoral thesis were valorized by the elaboration and publication of a number of 14 scientific papers (2 ISI, 4 ISI Proceedings, 8 IDB, 5 communications) in specialized journals, in volumes of some national and international conferences, by presenting 5 of them in national and international scientific events in the form of oral or poster presentation, as author and co-author.
11. submission of a national patent application with the title "*Kit for optimizing the working process of ring die pelleting equipment*", registered with the State Office for Inventions and Trademarks with the number A00592 / 20.08.2018.

7.3. Recommendations and perspectives

The researches could continue by:

1. improvement of the stand for biomass densification in the laboratory by diversifying the dies used and by modifying the heating mode of the dies;
2. theoretical and experimental researches regarding the biomass densification by conducting detailed tests to determine the mechanical durability of the pellets obtained;
3. experimental researches on biomass densification using agricultural biomass, for the verification of mathematical models for this type of biomass.