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PREREQUISITES FOR COMMERCIALIZATION OF TUBE GRANULATOR FOR FLY ASH

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TIIVISTELMÄ

Voimalaitoksissa syntyy suuria määriä lentotuhkaa, josta iso osa päätyy jätteenä kaatopaikoille. Lentotuhkan kaatopaikkasijoitus on voimalaitoksille kallista mm. kiristyneen jäteverotuksen takia. Lentotuhkaa voidaan kuitenkin käyttää hyödyksi eri tarkoituksissa. Tuhkan hyödynnettävyyttä voidaan parantaa rakeistamalla, jolloin poistuu myös tuhkan pölyävyysongelma.

Työssä käsitellään uuden rakeistusmenetelmän, putkirakeistuksen, kaupallistamiseen liittyviä edellytyksiä. Putkirakeistusmenetelmä hyödyntää vettä, lentotuhkan sisältämää kalsiumoksidia sekä savukaasujen sisältämää hiilidioksidia ja lämpöenergiaa. Työssä selvitetään putkirakeistusjärjestelmään vaadittavat apulaitteet itse rakeistimen lisäksi, putkirakeistusprosessin mitoituksen lähtökohdat ja rakeistusprosessin liittäminen osaksi jatkuvaa voimalaitosprosessia. Lisäksi tarkastellaan putkirakeistuksen taloudellisia etuja käyttäjän näkökulmasta.

Jatkuva putkirakeistusprosessi tarvitsee toimiakseen apulaitteiden osalta tuhkajärjestelmän, vedensyöttöjärjestelmän ja savukaasujärjestelmän. Järjestelmän kytkeminen osaksi voimalaitosprosessia riippuu hyvin pitkälti kyseessä olevasta voimalaitoksesta, mutta lähtökohtaisesti tuhka tulisi saada rakeistimelle mahdollisimman tuoreena ja savukaasut tulisi ottaa savukaasupuhaltimen painepuolelta. Prosessin mitoitusta voidaan tarkastella mm. rakeistimen täyttöasteen ja viipymäajan perusteella tai rakeiden kuivumisen kannalta. Optimaalisten mitoitusparametrien selvittäminen vaatii kuitenkin testiajoja rakeistimella.

ABSTRACT

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Prerequisites for commercialization of tube granulator for fly ash

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A large amount of fly ash is produced in power plants and a big fraction of it ends up as waste to landfills. Disposal of fly ash to landfills is expensive for power plants due to for example waste taxation. However fly ash can utilized in different applications. Possibility of utilizing fly ash can be increased by granulation which also removes the dustiness problems of ash.

This Thesis deals with the prerequisites for commercialization of a new granulation technique, tube granulation. Tube granulation technique utilizes water, calcium oxide in fly ash plus carbon dioxide and heat from flue gas. This Thesis determines the necessary auxiliary equipment for tube granulation, approaches for process dimensioning and implementation of the granulation process into a continuous power plant process. In addition, the economic benefits of tube granulation are examined from the user's perspective.

A continuous tube granulation process requires the following auxiliary systems to function: ash system, water feed system and flue gas system. Implementation of tube granulation system into a power plant process depends on the specific power plant but a general principle is that fly ash should be obtained to the granulator as fresh as possible and flue gas should be taken from the pressure side of a flue gas fan. Dimensioning of the process can be examined for example in terms of degree of filling and residence time in the granulator or in terms of granule drying. Determining the optimal dimensioning parameters requires pilot tests with the granulator.

ACKNOWLEDGEMENTS

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SYMBOL AND ABBREVIATION LIST

Roman alpabet

a	distance	[m]
A	Hamaker's constant	[J]
d, D	diameter	[m]
D	molar mass ratio M_V/M_G	[-]
F	force	[N]
k	volume ratio of voids to solid	[-]
K	Coulomb's constant	[Nm^2C^{-2}]
m	mass	[kg]
q	number of atoms per unit volume	[m^{-3}]
Q	electric charge	[C]
S	liquid saturation	[%]
t	surface withdrawal parameter	[-]
T	temperature	[°C]
X	moisture content	[-]
y	volume ratio of liquid to solid	[-]
Y	humidity	[-]

Greek alphabet

λ	van der Waals constant	[Jm^6]
ϵ	voidage	[-]
ψ	equilibrium relative humidity	[-]

Subscripts

0	inlet
db	dry basis
E	outlet (exit)
G	gas

GS   adiabatic saturation

HS   hygroscopic saturation

min   minimum

P   outlet (exit)

S   saturation

V   vapor

Abbreviations

BFB   Bubbling fluidized bed

CFB   Circulating fluidized bed

COD   Chemical oxygen demand

DCS   Distributed control system

ESP   Electrostatic precipitator

ID   Induced draft

MMM   Ministry of Agriculture and Forestry (Maa- ja metsätalousministeriö)

PC   Pulverized coal

TOC   Total organic carbon

VAT   Value-Added Tax

1 INTRODUCTION

A large amount of ash is produced in combustion of fuels in heat and power plants. In many cases ash ends up as waste in landfills although it could possibly be utilized in different applications. Tightening of environmental regulations and waste taxation has increased the interest in ash utilization since disposal of ash has become expensive for the producers.

Ash from heat and power plants has several possible applications where it can be utilized. Fly ash has better utilization possibilities than bottom ash and therefore the focus in this Thesis is in fly ash. Some of the most important applications for fly ash include fertilizer use, cement and concrete production and earth construction but other applications exist as well. Suitability of fly ash for different applications depends on its physical and chemical properties. Use of fly ash is controlled by laws and regulations that specify certain requirements for ash properties in different uses. Utilization of fly ash and regarding regulations are discussed in chapter 3.

Utilization possibilities of fly ash can be improved by stabilizing it. Dry and dusty ash is also problematic to handle and store and stabilizing removes these problems. Stabilization can be done by either granulation, self-hardening or pelletizing, of which granulation is the most effective mechanism. Granulation is a process that is used to create larger agglomerates, or granules, from particulate matter. The theory of granulation is discussed in chapter 4. Currently used granulation techniques for fly ash are plate granulation and drum granulation that are based on layered granulation. These techniques are briefly presented in chapter 5.

The literature part of this Thesis begins with explaining what the ash-forming compounds in solid fuels are, how ash is formed during combustion and why ash should be utilized instead of disposing it to landfills, in chapter 2.

This Thesis introduces a new technique for granulation of fly ash, so called tube granulation. The main process of tube granulation has been invented by Mr. Hannu Kuopanportti. A tube granulator is a compact and simple granulation device that can be

implemented directly into a power plant process. Tube granulation is a continuous process that takes advantage of the high carbon dioxide content and heat in flue gas which is obtained from the power plant process. The granulation process involves chemical reactions between calcium that is found in the fly ash, water and carbon dioxide to form strong granules efficiently. (Kuopanportti 2015a.) A more detailed description of the tube granulation process is in chapter 6.

This Thesis has been done for a technology company in Finland as a part of a development project regarding fly ash granulation. The company shall later be referred simply as "Company". The Company's objective is to develop the tube granulation process and commercialize it to be sold to heat and power plants. Before commercializing, pilot tests need to be completed with a pilot granulator that has already been manufactured. However the complete tube granulation system, including auxiliary equipment, has not yet been designed and the granulation capacity of the existing pilot granulator has not been defined.

The aim of this Thesis is to figure out the basis for tube granulator process dimensioning, to define the necessary auxiliary equipment for the process and to design how the tube granulator can be implemented into a power plant process. This information works as a base for pilot testing of the tube granulation process that will be carried out at a power plant. Defining the equipment for tube granulation plus process and equipment dimensioning approaches are discussed in chapter 6. Detailed dimensioning information and calculations are excluded from this Thesis intentionally. Chapter 7 discusses the options for integrating tube granulation into a power plant process. In addition, the profitability of the tube granulator from the user's perspective is analyzed in chapter 8.

While coal ash is also discussed in this Thesis, the focus is in wood and peat ashes as these are the highest potential ashes for utilization, considering the location of the Company.

2   FUNDAMENTALS OF ASH UTILIZATION

Ash is produced at heat and power plants as an inorganic residual from combustion and the amount of ash produced is highly dependent on the used fuel. After formation, ash is collected at different locations in the power plant process. Bottom ash is removed from the furnace, a fraction of fly ash is collected from the heat transfer surfaces and most part of fly ash is collected from a dust separator device, such as an electrostatic precipitator. Collected ash is typically transported directly to a storage silo or container. Bottom ash and fly ash are usually stored separately. (Ojala 2010, 4, 7.)

Heat and power plants in Finland produce annually approximately 350 000 tons of ash from co-combustion of wood and peat, 150 000 − 200 000 tons of pure wood ash and 50 000 tons of pure peat ash. In the year 2006 about 1 million tons of coal ash was produced in Finland. (Korpijärvi et al. 2009, 16; Motiva Oy 2009.) Worldwide the annual amount of coal ash produced is approximately 600 million tons. In Finland about 50 − 80 % of the coal ash and around 30 % of the peat and wood ashes has been somehow utilized. Worldwide the average utilization rate of ashes is approximately 16 %. (Ahmaruzzaman 2009, 328; Korpijärvi et al. 2009, 22.)

2.1   Ash-forming compounds in fuels

In addition to organic, convertible matter, fuels also contain inorganic mineral matter which forms ash in combustion. The mineral matter in fuels can be called ash also before it has undergone any transformations during combustion. Different fuels contain different types of mineral matter with varying composition and association. The minerals in fuel can be found in different forms: as free ions, salts, organically bound or hard minerals. Hard minerals can be divided into two groups: included or excluded minerals. (Cieplik et al. 2011, 198-199.) Lind (1999, 19) presents an example of different types of mineral matter in a fuel particle as seen in Figure 1.
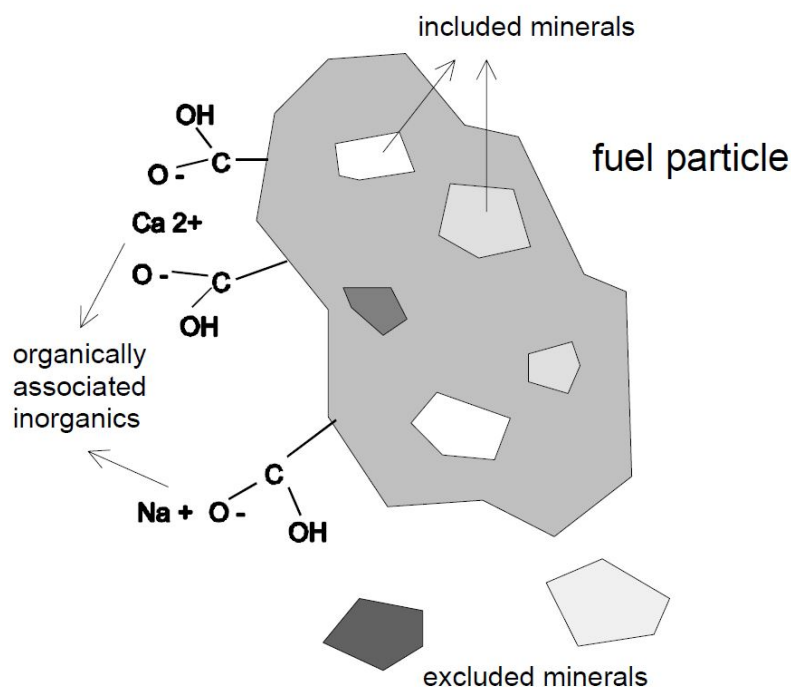
Figure 1. Included and excluded minerals as well as organically associated inorganics in a fuel particle (Lind 1999, 19).

Included minerals are the necessary inorganics for plants to grow. They are present in biomass and also in coal which is of biomass origin but has undergone the processes of conversion to peat and coal. Excluded minerals in fuels come from collecting and handling the fuel. For biomass this is due to contamination with soil during harvest or handling and for coal due to mining or handling. (Cieplik et al. 2011, 200.) In younger fuels, such as wood, most of the ash-forming compounds is found as organically bound and as salts while in high-rank coals which are very old fuels the major part of ash-forming compounds is as included minerals. Peat and younger coals are in the middle of the two extremes as they contain ash-forming compounds both as organically bound and as included minerals. (Lind 1999, 17, 20; Raiko et al. 2002, 270-272.)

Younger fuels typically contain less minerals and more volatile compounds than older fuels (Cieplik et al. 2011, 199). This means that woody biomass contains less ash-forming compounds than for example peat and coal. According to Lind (1999, 20) biomass fuels usually contain less than 1 % but up to 10 % or more of ash-forming compounds. In coals the amount is usually 5 - 15 % but can even be as high as 50 %.

Alakangas (2000, 152, 154) has listed typical ash contents in various fuels stating values of 0,5 - 3 % for wood-based biomass, 4,5 - 5,1 % for peat and 14 % for coal. Given values are as weight percentage in dry matter.

The elemental composition of ashes is usually given as oxides of certain elements disregarding whether they actually are as oxides or not (Raiko et al. 2002, 270). Table 1 presents typical elemental compositions of ashes for different solid fuels including coals, peats and wood-based biomass, as oxides of the main components. It has to be noted that different authors give different values for typical compositions and therefore the values presented should be considered only as indicative.

Table 1. Ash contents and ash compositions as oxides of main components for various solid fuels. Values are indicative. (Alakangas 2000, 39, 88, 91; Raiko et al. 2002, 271.)

Fuel	ash %	SiO_2	Al_2O_3	Fe_2O_3	P_2O_5	CaO	MgO	Na_2O	K_2O	SO_3	TiO_2	other[*]
Coals												
Lignite	5,2	9,8	1,6	11,3		51,2		0,1	0,1	1,3		24,6
	22,0	42,6	9,7	1,3		8,9		6,2	0,9	15,6		14,8
	4,9	12,6	8,0	14,0		16,9	4,8	9,3	0,2	23,7		10,5
Bituminous	7,5	43,2	21,5	13,7		6,9	3,0	2,8	0,5	0,8		7,6
Anthracite	7,4	46,6	23,6	8,1		7,0	1,2	0,1	0,5	6,0		6,9
Peats*	1,6/ 4-6	31,8/ 40-75	13,1/ 1-16	11,0/ 4-7	-/ 2-4	21,1/ 1,5-12	6/ 0,5-2,5	1,4	2,0/ 0,1-0,5		-/ 0,5-1	13,6
	16,8	20,0	5,2	70,0	2,2	4,7	0,7	0,5	0,7			- **
	19,6	57,0	13,0	17,0	1,6	4,4	1,4	2,3	2,0			1,3
Woods												
Birch***	0,3	0,9/ 3,8			3,5/ 7,7	45,8/ 57,8	11,6/ 7,7	8,7/ 7,7	15,1/ 11,5	2,6/ 3,8		11,8
Pine***	0,2	3,5/ 4,6		-/ 5,5	2,7/ 1,0	41,8/ 42,0	16,1/ 16,0	3,1/ 3,0	15,3/ 15,2	4,5		13
Spruce***	0,3	1,0		-/ 8,5	2,7/ 1,0	36,8/ 36,7	9,8/ 10,0	3,2	29,6	4,3/ 4,2		12,6
Barks												
Birch***	1,6	3,0		1,0	3,0/ 3,5	60,3	5,9	0,7	4,1	-/ 4,8		22
Pine***	1,8	14,5		3,8/ 5,0	2,7/ 2,6	40,0	5,1/ 2,6	2,1/ 2,0	3,4/ 3,3	-/ 3,7		28,4
Spruce***	3,4	21,7		1,8	2,7/ 2,6	50,5	4,2	2,8	3,5	-/ 1,6		12,8

* calculated as remainder
** remainder negative
*** different values obtained from Raiko et al. (2002) / Alakangas (2000). Information in rows without this mark is taken from Raiko et al. (2002).

2.2   Ash formation in combustion

Combustion of fuel can be divided into four phases that are drying, release of volatile gases (pyrolysis), volatiles burning and char combustion (Raiko et al. 2002, 60-61). These phases happen in series but they can also overlap especially when the fuel particles are large (Raiko et al. 2002, 186). This is the case for example in fluidized bed combustion where the fuel particle size is typically rather large (Lind 1999, 28).

Ash formation in combustion happens through multiple different mechanisms and it is affected by fuel properties and used combustion technique. Part of the ash-forming compounds is released into flue gases already during the pyrolysis but most part is released during char combustion. (Lind 1999, 24; Raiko et al. 2002, 273.)

During combustion some part of the ash-forming compounds volatilize into flue gases. The fraction of volatilized compounds is noted to be higher with increasing combustion temperatures and highly reducing conditions. The fraction of volatilization also depends on the age of the fuel and it can be from a few percent of the total ash with old coals to up to 10 % with younger coals and even higher with biomass. This can be explained with the fact that older fuels contain more included and excluded minerals which do not volatilize easily. On the other hand ash-forming compounds that are organically bound and as salts, which are the major ash-forming compounds in younger fuels, are readily volatilized during combustion. (Lind 1999, 24-25.)

After volatilization the reduced compounds move away from the char particle. As soon as they reach oxidizing conditions while moving through the char boundary layer, the compounds are oxidized. The resulting oxides have much lower equilibrium vapor pressures than the original reduced compounds which lead to highly supersaturated vapors. These vapors nucleate in the char boundary layer forming new particles which grow in size by coagulation, condensation and agglomeration. Ash particles formed by nucleation and the subsequent growth mechanisms usually have a diameter of less than 1 μm. Particles of this size in the ash size distribution are called the fine particle mode. (Lind 1999, 25.)

Most of the ash-forming compounds remain in the char after volatilization and form residual ash particles that are larger (diameter > 1 μm) than the fine particles formed by nucleation. There are several mechanisms affecting how residual ash particles are formed. One of these mechanisms is melting and coalescing of minerals inside or on the surface of the char particle. (Lind 1999, 26-28; Raiko et al 2002, 273.) Ash melting behavior is affected by the combustion temperature. In PC (pulverized coal) boilers where the ash is formed in the temperature range of 1000 to 1500 °C, ash melts and attains a spherical shape. In fluidized bed boilers the combustion temperature is much lower and ash is formed at temperatures from 800 to 900 °C. At this temperature the ash doesn't melt and it keeps the original shape of the mineral matter which leads to coarse, rough edged, ash. (Basu 2006, 166.)

If the fuel particles fragment to a high degree, it is possible that one fragment contains only one mineral particle which forms a stand alone ash particle. For fuels that don't fragment much, all the minerals in a fuel particle may coalesce to form one or several ash particles. Minerals that are excluded go through chemical reactions and oxidation and they can also fragment due to rapid temperature increase. Ash forming compounds that are organically bound coalesce on the char surface. These compounds form beads that again coalesce with each other, forming residual ash particles that are released into flue gases during char burn-out. (Lind 1999, 26-28; Raiko et al 2002, 273.) In addition, in fluidized bed combustion the collisions of bed material particles can cause them to break and some fragments can be small enough to be captured in the flue gases as fly ash particles that are in the size range of residual ash particles (Lind 1999, 43).

After initial formation, the ash particles may still go through some transformation. Firstly, ash particles may collide with other ash, fuel or bed material particles in the flue gases or in the bed of a fluidized bed boiler, resulting in particle size growth or decrease. Collisions of ash particles in the flue gases can cause particle growth but this is only significant in the fine particle mode where particle concentrations are high. In fluidized bed combustion the collisions of ash, fuel and bed material particles in the bed play a more important role. Collisions in the dense bed may cause either particle size growth by agglomeration or decrease by fragmentation. Secondly, it is possible that the

volatilized compounds condense on the surface of ash particles or chemically react with them. (Lind 1999, 27-29.) The volatilized compounds can also chemically react with the bed material particles (Lind 1999, 37).

In fluidized bed boilers the bed material particles that grow too large to be fluidized are removed from the furnace as bottom ash. The bed material particles can grow by the above-mentioned collisions of ash and bed particles as ash particles layer on the bed particle's surface, or by agglomeration with other bed particles. Attrition caused by the collisions can also result in the ash particles being detached from the bed particle's surface. (Lind 1999, 36-37.)

Figure 2 presents a schematic figure of the ash formation mechanisms during CFB (circulating fluidized bed) combustion of solid wood-based biomass as determined by Lind (1999, 67) in her ash formation experiments.
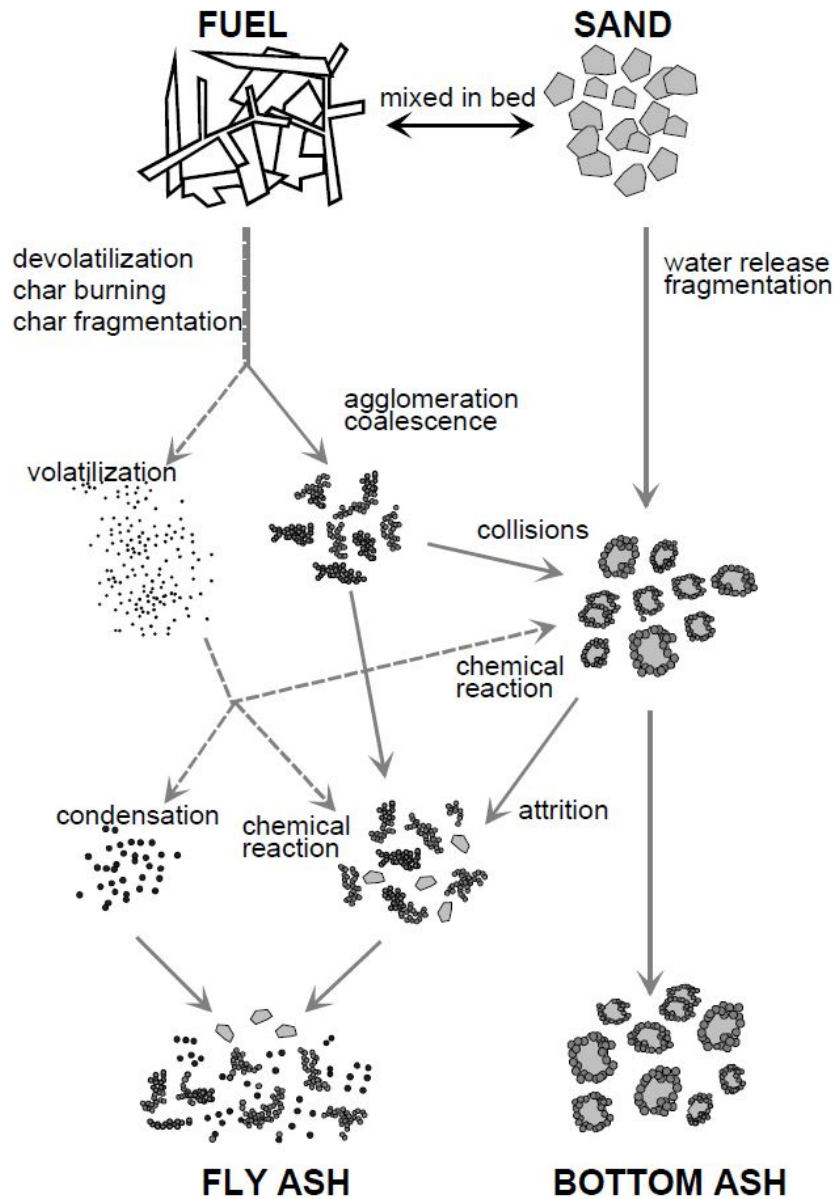
Figure 2. A simplified example of ash formation mechanisms during CFB combustion of solid wood-based biomass (Lind 1999, 67).

In fluidized bed boilers about 40 – 80 % of fuel ash leaves the boiler as fly ash. In BFB (bubbling fluidized bed) boilers the fluidizing air velocity is reasonably low and only the finest ash particles leave the boilers as fly ash and the mean diameter of fly ash particles is 3,0 – 5,0 μm. In PC boilers typically over 80 % of fuel ash leaves as fly ash. The mean diameter of the fly ash particles in PC boilers is 5,0 – 8,5 μm. (Basu 2006, 166.)

2.3 Basis for ash utilization

Fly ash doesn't contribute to energy production and from that perspective it is seen as a by-product and as waste. However the composition and properties of fly ash can make its utilization beneficial in several applications and therefore ash can be seen as a resource and an important component of the fuel. The nutrient content, geotechnical properties and physicochemical characteristics of fly ash make it suitable for use in different applications. (Ahmaruzzaman 2009, 328.)

From an environmental perspective fly ash is a serious problem unless it can be utilized (Ahmaruzzaman 2009, 328). Disposing of the large amount of ashes produced in combustion plants is a great burden to landfill sites and reserves a lot of space that could be used for other purposes (Ahmaruzzaman 2009, 331; Korpijärvi et al. 2009, 11). Utilizing ash also helps in the efforts of reducing the use of non-renewable resources (Kiviniemi et al. 2012, 6).

The most natural cycle for ash from burning wood is to return it in back to forests. When forests are cut the logging residues are often harvested and used as fuel. The logging residues contain lots of nutrients that are therefore lost from the forest ecosystem. At the same time the acid-buffering capacity of the soil is lowered. In order to maintain the nutrient and acid-buffering balance in the ecosystem, it is wise to return the wood ash to the forest land where the biomass was obtained. (Skogsstyrelsen 2007.) Due to the goal of increasing the use of renewable energy, logging residue usage in energy production in Finland is expected to rise significantly in the future. This will increase the loss of nutrients in forests which makes recycling of ash even more important. (Ojala 2010, 1.)

Disposal of ashes is expensive due to waste taxes and landfill site fees. By utilizing fly ash these costs can be minimized and even some profit can be made from selling the ash as a product. (Ahmaruzzaman 2009, 331.) Possible ways for utilization of ash and related regulations are discussed in chapter 3.

3   UTILIZATION OF GRANULATED ASH

Ash from burning wood and peat can be utilized in several applications such as fertilizer for forests and fields, soil enrichment, road building, landfill structures, additive in asphalt and cement and also in dumping. However utilization of ash often causes some costs and ash quality may not meet certain laws and regulations that restrict the utilization of ash. Therefore it is in many cases cheaper to transport the ash to a landfill unutilized or place it in a power or heat plant's own banking site. (Ojala 2010, 1.)

It is possible to improve ash quality and properties by simple actions such as choosing the appropriate storing method and keeping different types of ashes separated (Ojala 2010, 4). If water is added to ash when it's removed at the plant it is easier to handle because it won't dust but if ash starts to harden by itself it will be harder to make it homogenous again (Isännäinen et al. 2006, 7). The most usual and important way of separating different ash types is to collect and store bottom ash and fly ash separately at a plant (Ojala 2010, 7). Fly ash has better utilization possibilities than bottom because fly ash has less unburnt content and it is homogenous. For example bottom ash from fluidized bed boilers isn't suitable as a fertilizer since it mostly contains bed material. (Ramboll Finland Oy 2012, 1; Isännäinen et al. 2006, 7.)

Fly ash quality can be further improved by collecting and storing ash from the electrostatic precipitator's (ESP) electric fields separately if such a dust reduction system is used. The ESP's first electric field collects the largest ash particles while the smallest particles are collected in the last electric fields. This method of separating different particle sizes is called fractionating. The majority of fly ash is separated in the ESP's first electric field but it is possible to optimize fractionating of ash by tuning the operation of the ESP. Small ash particles contain higher concentrations of reactive and soluble matter and heavy metals than large particles which means that the fly ash collected from an ESP's first electric field is of better quality and could be stored separately. (Isännäinen et al. 2006, 8; Korpijärvi et al. 2009, 20-21.)

Loose ash from combustion plants is very dusty and it causes several problems in handling, storing, transportation and spreading. Dusty ash causes health hazards when it

is inhaled or in contact with skin or mucous membrane. It also easily penetrates sealed areas of handling equipment where it causes erosion and hot ash may even start a fire. In order to remove these problems loose ash must be stabilized before it can be utilized. (Väätäinen et al. 2000, 12-14.) Ash can be pretreated by self-hardening, granulating or pelletizing (Ojala 2010, 8; Huotari 2012, 10). Current granulating technologies are discussed in chapter 5 and the theory behind granulating is explained in chapter 4.

3.1   Applications

Granulated ash can be used in many different applications. Possible applications for ash are determined by the properties of ash that vary with different fuels, boiler types, ash separation systems and ash storing methods. Granulated ash can be utilized for example as a fertilizer, in earth construction, in concrete production and in waste water treatment.

Ash can be used in several applications in earth construction. Ash can be used in different forms - as pure ash, compacted ash, mixed with other secondary products or as a binder. (Kiviniemi et al. 2012, 3.) Ash mixtures are used to combine the desired properties of ash and other secondary components and to minimize the negative properties of both materials. Commonly used secondary components with ash are fiber clay and tailings, such as sand from enrichment of calcite. (Kiviniemi et al. 2012, 18; Ramboll Finland Oy 2012, 8.)

Dry stored and strengthening fly ash can be used in different stabilization techniques that are stabilizing soft soil, stabilizing road and field pavements, stabilizing low-grade soil and sediment and stabilizing contaminated soil. In these applications fly ash can be used alone or mixed with commercial binders or other by-products. (Ramboll Finland Oy 2012, 8.)

Bottom ash from combustion plants can be utilized to replace sand in earth construction but this is not further discussed here since the focus in this Thesis is in fly ash (Ramboll Finland Oy 2012, 7).

3.1.1   Forest fertilizer

Fertilizers are used in forests to enrich the soil with nutrients of which concentration in the soil is too low for the need of trees. The goal of fertilizing is to increase the growth speed of trees or to ensure a healthy nutrient balance in the soil for a long period of time. (Huotari 2012, 41.)

Ash from burning wood and peat can be used as a forest fertilizer since it contains nutrients needed by trees (Ojala 2010, 1). Ash from burning biomass from fields or animal manure can also be used as a fertilizer or a raw material in fertilizer production (Huotari 2012, 9). Wood based ash can fertilize the ground and increase the growth of trees for at least 20 years while the fertilizing effect of artificial fertilizers starts to weaken in 10-15 years after fertilization (Ojala 2010, 2; Ramboll Finland Oy 2012, 2). Ash fertilizer is most effective in forests where the soil is rich with nitrogen and lacks phosphorus and potassium. Typically this is in peatland forests and former peat production lands that are afforested. (Huotari 2012, 41-42.)

Ash from burning wood only is the most suitable fertilizer as it has all the required nutrients in appropriate ratios for trees excluding nitrogen which it doesn't have at all. Wood ash can therefore be distributed in forests without adding any extra nutrients in it. In Finland pure wood ash is not generated much because wood is usually burned in combination with peat. (Huotari 2012, 6.)

Wood ash composition varies depending on the wood species and the part of the wood used and typically contains 0,2 - 3 % phosphorus (P), 0,5 - 10 % potassium (K), 5 - 40 % calcium (Ca) and less than 0,1 % borium (B). The most important nutrients for trees in wood ash are phosphorus, potassium and trace elements. Compared to wood ash, peat ash has lower amount of nutrients especially potassium, calcium, magnesium and borium. Peat ash is also more coarse and its granulation and fertilizing properties are not as good as in wood ash. On the other hand peat ash usually has lower content of heavy metals than wood ash. (Huotari 2012, 6; Ojala 2010, 5.)

Spreading ash fertilizer can be done either by land or air. Spreading by land requires a solid soil to carry heavy machinery and therefore especially in peatland forests it must be done during winter time when the ground is frozen. Ground spreading also requires an intermediate felling with preparation of vehicle paths to be done. Spreading by air is done with a helicopter and it can be done at any time of the year and it doesn't require any silvicultural measures. Air spreading always requires granulated ash but ground spreading could also be done with loose ash. (Huotari 2012, 43-44.)

3.1.2   Concrete production

Fly ash from combusting coal can be used in concrete production to partially replace cement. Partial replacement of cement in concrete can improve the properties of concrete and reduce its unit cost. (Ahmaruzzaman 2010, 348.) Coal ash has good properties as a binder and it has been used in concrete production for a long time. Nowadays ash from multi-fuel combustion can be used as well but a major portion of the fuel mix has to be coal. (Ramboll Finland Oy 2012, 7.) Small particle size and higher roughness favors the use of fluidized bed boiler fly ash over PC boiler fly ash in concrete and cement production (Basu 2006, 166).

Suitability of solely wood based ashes in concrete production has been researched in Finland. The findings are that concrete including wood based ash requires significantly more water than conventional concrete or coal ash based concrete but the concrete including wood ash has better compression strength than conventional concrete. (Ramboll Finland Oy 2012, 7.)

3.1.3   Earth construction

Ash can replace natural stone in earth construction. Ash can be utilized in road, street, field and bench construction, as a binder or filling material in dredged bottom sediments and soft soil stabilizing. Ash has also been used for example in surface structures in

landfill site closing and in sound barriers. (Ramboll Finland Oy 2012, 7.) Use of fly ash in soil stabilization, road construction and pavement construction can be really cost effective especially if the construction site is close to a power plant (Ahmaruzzaman 2010, 353-354).

Massive fly ash structures are lamellar structures that consist of only fly ash or fly ash stabilized with a reinforcing binder. Usability of fly ash in different structures depends on the technical properties of fly ash. Massive fly ash structures can be used in road, street and field structures from embankment fills to lower part of road base. Using massive fly ash structures instead of conventional structural materials has the following advantages: better thermal insulation capacity, higher loadbearing capacity, lightness of the structure, saving natural resources and total thickness of the structure can be lower. (Kiviniemi et al. 2012, 22.)

3.1.4 Waste water treatment

Research has shown that ash from biomass can be used in waste water treatment to remove phosphorus, lead, cobalt, cadmium, copper and zinc. Granulated coal ash can also be used to remove arsenic or boron from waste water. (Ramboll Finland Oy 2012, 8; Polowczyk et al. 2013, 100; Polowczyk et al. 2007, 40.) Tests made at the University of Oulu also show that granulated ash from biomass can be used to reduce chemical oxygen demand (COD) and total organic carbon (TOC) values of waste water. The reduction rates can be can be over 99,5 % of COD and over 80 % of TOC. (Ramboll Finland Oy 2012, 8.)

Research made at the University of Oulu includes adsorption tests with granulated ash where the phosphate removal efficiency of granulated ash was examined. The research shows that over 97 % phosphate reductions can be easily achieved with granulated ash without adding any secondary materials in it. Ash from two different combustion plants was used and both ashes were from a fuel mix of 75 % peat and 25 % wood. (Karvonen et al. 2012, 37, 77-78.)

Ash can be used in phosphate removal also by dissolving the aluminum and iron ions in ash. Aluminum and iron ions can act as flocculation agents in waste water treatment to remove phosphate from water. This method however is most efficient with fine ash and therefore granulated ash can't be used as it is. (Karvonen et al. 2012, 92.)

3.2 Regulations for ash utilization

In Finland the use of ash as a product or material in different applications is controlled by several laws and regulations. The Finnish legislation also encourages the utilization of ash by making it more profitable than dumping ash to landfill sites.

The Finnish law covering taxation of waste (Law 1126/2010) has defined fly ash from combustion plants as taxable waste since the beginning of year 2011. The goal of this law is to encourage utilization of waste that is usable but often ends up unutilized to landfill sites. (Ramboll Finland Oy 2012, 1.) In Finland the legislation regarding landfill sites (Government Decree 331/2013) also forbids disposal of waste that contains organic matter more than 10 % expressed as total organic carbon (TOC) or loss on ignition (LOI) to landfill sites for non-hazardous waste. However this does not apply to bottom and fly ashes from energy production if the dissolved organic carbon content is less than 800 mg in a kilogram determined with a liquid-solids ratio of 10 liters per one kilogram of dry matter.

3.2.1 Fertilizer use

When ash is used as a forest fertilizer in Finland the combined phosphorus (P) and potassium (K) content has to be at least 2 % and calcium (Ca) content at least 6 %. In other than forest use the neutralizing ability (Ca) of ash has to be at least 10 %. It is allowed to add inorganic fertilizer products to granulated ash in order to improve its usability and to fulfill minimum requirements. If boron (B) is added to ash it cannot be used at groundwater areas or conservation areas. (MMM decree 24/11.)

The maximum amount of harmful metals in fertilizers is regulated by law in Finland (Table 2 and Table 3).

Table 2. Maximum allowed concentrations of harmful metals in ash used as a forest fertilizer in mg/kg dry material (MMM decree 24/11).

Element	Limit value (mg/kg$_{db}$)	Maximum load (g/ha/a)
Arsenic (As)	40	Maximum 160 g/ha over 60 years period
Mercury (Hg)	1,0	
Cadmium (Cd)	25	Maximum 100 g/ha over 60 years period
Chromium (Cr)	300	
Copper (Cu)	700	
Lead (Pb)	150	
Nickel (Ni)	150	
Zinc (Zn)	4500[1]	

[1] Exceeding the limit value of zinc in ash used as a forest fertilizer is allowed only in peatland forests when the lack of zinc has been proved by either soil, leaf or needle analysis. The limit value in this case is 6000 mg Zn/kg dry material.

Table 3. Maximum allowed concentrations of harmful metals in ash used as a field fertilizer or as its raw material in mg/kg dry material (MMM decree 24/11, Ramboll Finland Oy 2012, 10).

Element	Limit value (mg/kg$_{db}$)
Arsenic (As)	25
Mercury (Hg)	1,0
Cadmium (Cd)	2,5
Chromium (Cr)	300
Copper (Cu)	600[1]
Lead (Pb)	100
Nickel (Ni)	100
Zinc (Zn)	1500[1]

[1] Exceeding the limit value of zinc and copper is allowed when the lack of these elements has been proved by a soil analysis.

3.2.2 Concrete production

In concrete production the European standard EN 450 Fly ash for concrete part 1 & 2 set the limits for fly ash quality that can be used. The standard EN 450-1 describes that in co-combustion the minimum percentage, by dry mass, of coal has to be at least 60 %

with the exception that if the co-combustion material is only from green wood the percentage of coal shall not be less than 50 %. Green wood is described as "wood originating from trees, bushes and shrubs that is created when processing wood as cross-cut ends, planings, saw dust and shavings used in the form of dust, chips and pellets". The percentage of ash from co-combustion materials has to be 30 % or less by dry mass in order to use the ash in concrete production.

Detailed chemical, physical and other requirements for fly ash in concrete production use are described in the standard EN 450-1. The scheme for evaluation of conformity of fly ash is described in part 2 of the standard, EN 450-2.

3.2.3 Earth construction

In Finland the use of ash products in earth construction is regulated by the Government Decree 403/2009, so called MARA-decree, which applies to utilization of waste in earth construction. The decree sets the limits for total amount of harmful materials and for solubility. If the limit values in the ash are not exceeded it can be utilized in earth construction by notification procedure. Additional permitting processes and suitability studies are required if the limit values are exceeded. (Ramboll Finland Oy 2012, 10.)

Table 4 lists some of the limit values for fly ash described in the decree 403/2009. This table does not include all the elements listed in the decree.

Table 4. Some of the limit values for concentration of harmful materials and their solubility (Government Decree 403/2009).

Element	Concentration (mg/kg$_{db}$)	Solubility (L/S = 10 l/kg) Covered structure / Paved structure
Arsenic (As)	50	0,5 / 1,5
Barium (Ba)	3000	20 / 60
Cadmium (Cd)	15	0,04 / 0,04
Chromium (Cr)	400	0,5 / 3,0
Copper (Cu)	400	2,0 / 6,0
Lead (Pb)	300	0,5 / 1,5
Zinc (Zn)	2000	4,0 / 12

4   GRANULATION OF FLY ASH

Different authors have different definitions for the scope of processes included in the term granulation. Also the same processes of particle size enlargement are described with different terms in different industries. Sherrington & Oliver (1981, 3) defines granulation as a process where particles as a powder or powder/binder mixture are built up to clusters, or granules. Granulation is divided to several processes that produce granular material. The processes are following: compaction by compression or extrusion, agglomeration by agitation, drop formation by globulation and heat bonding by nodulization or sintering. (Sherrington & Oliver 1981, 2-4.) The focus of this Thesis is clearly in granulation by agitation and therefore mainly this process is discussed. From now on the process of forming granules by agitation is described simply as agglomeration and the word granulation is used as a general heading for this process.

4.1   Fundamentals of particle size enlargement

The growth of granules is explained with two different mechanisms. First is collision and successful adherence of primary particles and second is collision and attachment of particles around a nucleus forming a layer. Both mechanisms lead to discrete granule formation. (Sherrington & Oliver 1981, 7.)

Particle size enlargement is based on cohesive forces at particle contact points. The cohesive forces hold the granules together. The type and magnitude of the cohesive forces along with particle size distribution and the number of bonding contact points per particle determine the strength of the granules.

4.1.1   Particle-particle attractive forces

Particle-particle attractive forces are forces that hold the particles together when they come into close contact. These forces can be divided to

1. Intermolecular attractive forces

2. Electrostatic forces
3. Liquid bridge modes
4. Solid bridges.

Intermolecular attractive forces

Intermolecular forces are active only up to a distance of 0,1 μm and any roughness in molecular surface will diminish the forces rapidly. The main intermolecular forces that affect real particulate systems where molecular surface isn't smooth are van der Waals dispersive forces. Van der Waals forces are effective at distances less than 0,1 μm. To calculate the van der Waals forces between particles of different size the following equation can be used. (Sherrington & Oliver 1981, 7-8.)

$$F_{vdW} = \frac{\pi^2 q_0^2 \lambda}{12a^2}\left(\frac{d_{p1}d_{p2}}{d_{p1}+d_{p2}}\right)$$
(1)

where F_{vdW} van der Waals forces [N]

             q_0 number of atoms per unit volume [$1/m^3$]

             λ van der Waals constant [Jm^6]

             a separation distance [m]

             d_p particle diameter [m]

Van der Waals constant λ in equation (1) can be obtained from equation

$$\pi^2 q_0^2 \lambda = A$$
(2)

where A Hamaker's constant [J]

Hamaker's constant in equation (2) is quoted to range from $(0,01 - 200) \times 10^{-19}$ Joules while the most commonly accepted figure is 10×10^{-19} Joules (Sherrington & Oliver 1981, 9).

Electrostatic forces

Electrostatic forces have almost always a role in the attraction between particles. Electrostatic attractive forces are usually a result from friction between particles but rubbing is not necessarily needed to produce the forces as mere contact can cause the effect. The magnitude of electrostatic forces in particle binding is rather small compared to other binding mechanisms but they help in particle growth by holding or orientating particles in a contact region for a period of time before other binding mechanisms start to dominate. For smooth particle surfaces electrostatic forces can be greater than van der Waals forces. For relatively large and dry particles electrostatic forces have a decisive role in initial adhesion of the particles. (Sherrington & Oliver 1981, 11.) In the application of granulation of fly ash this should be taken into account.

The maximum electrostatic force of attraction F_{elec} when two oppositely charged particles come near each other can be calculated with the following equation.

$$F_{elec} = -K \frac{Q_1 Q_2}{d_p^2} \left(1 - \frac{2a}{d_p} \right) \tag{3}$$

where F_{elec} electrostatic force of attraction [N]

   K Coulomb's constant $= 8{,}9876 \times 10^9$ Nm^{2}C^{-2}

   Q electric charge [C]

When the separation distance of particles is much smaller than particle diameter, $a \ll d_p$, the following equation can be used.

$$F_{elec} = -K \frac{Q_1 Q_2}{d_p^2} \tag{4}$$

Liquid bridge modes

The presence of liquid can assist particle growth by producing cohesive forces with the following mechanisms:

1. Adsorbed liquid layers on particles
2. Mobile liquid bridges
3. Viscous or adhesive binders.

In case only adsorbed liquid layer films are present on the surface of the particles the following two phenomena contribute to the attractive forces between two particles:

1. Surface is smoother which increases the available particle-particle contact area
2. The effective distance between particles is decreased which increases the role of van der Waals or electrostatic forces.

The cohesive forces obtained from adsorbed liquid layers are weak compared to other liquid bridge mechanisms where more liquid is present. The cohesive strength of the agglomerates increases as sufficient amount of liquid is added to produce liquid bridges. The different saturation states in a randomly packed agglomerate can be presented as shown in Figure 3. (Sherrington & Oliver 1981, 11-12.)
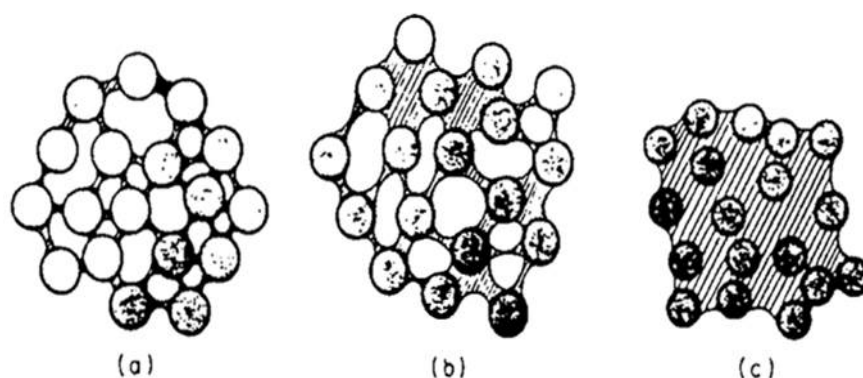
Figure 3. Saturation states of liquid in a randomly packed agglomerate: a) pendular bridging state; b) funicular state; c) capillary state (Kuopanportti 2015a).

The amount of saturation in different states in Figure 3 can be described with liquid saturation S. Liquid saturation is the pore volume occupied by liquid divided by the total pore volume available in the agglomerate. (Sherrington & Oliver 1981, 12.)

In pendular state the agglomerate is held together by bridge necks formed by liquid. In pendular bridging the cohesive force is a result of the effect of surface tension and the negative suction pressure caused by the curvature of the liquid meniscus at the air-liquid interface. (Sherrington & Oliver 1981, 12-13.) In theory the cohesive force should increase as saturation decreases because the suction pressure within the liquid rises while the surface curvature increases. However in practice the tensile strength has been noted to remain approximately constant or decrease when liquid saturation is decreased. This can be explained with the fact that in reality the particles are never smooth which causes a gap between them. Saturation state in the agglomerate is considered pendular until the liquid bridges start to coalesce which happens when enough liquid is present. (Capes 1980, 28-30.)

In the case of capillary state the whole pore space is saturated and there are no single bridges between the particles. The only force holding the granule together in the capillary state is caused by the capillary pressure at the granule boundary gas-liquid interface. It has been noticed that the range of liquid saturation where only capillary pressure is present starts from $S > 80$ %. (Sherrington & Oliver 1981, 16-17.)

The transitional stage between pendular and capillary states is called the funicular state where liquid saturation is 25 % $< S < 80$ %. Various degrees of saturation inside the granule can be noticed in this state. Both pendular bridges and completely filled voids exist and the total cohesive strength is affected by both the forces of pendular bridges and the capillary suction pressure. When S is reaching 80 % practically all voids are filled with liquid and the contribution of pendular bridges to the cohesive strength rapidly decreases to zero. (Sherrington & Oliver 1981, 17-18.)

According to Capes (1980, 32) the tensile strength of an agglomerate is highest in the case of capillary state and lowest in pendular state. The tensile strength of pendular state

is approximately one third of the tensile strength of capillary state. The strength of funicular state depends on saturation S and is between the tensile strength of pendular and capillary states.

Solid bridges

Solid bridges are produced when the liquid used in granule formation changes phase from liquid to solid. In many applications the forming of granules with the help of liquid is only an intermediate step in the process and the final properties of the product are determined in the solid phase. The strength of the final product can be increased when liquid is changed to solid phase. The phase change from liquid to solid often happens when temperature or pressure is increased. Solids bridging can happen by the mechanisms listed below. (Sherrington & Oliver 1981, 18.)

1. Crystallization of dissolved material
2. Hardening binders
3. Particle melting or localized fusion welding
4. Particle deformation followed by sintering
5. Chemical reaction

Solid bridges formed by these mechanisms can have the tensile strength equal to the tensile strength of the constituent particles, or even greater in some cases.

4.1.2 Moisture content for granulation

The moisture content of a granulating mass has a significant influence in the end product of the process and most continuous granulation processes are primarily controlled by regulating the amount of liquid added to the process. Moisture content can be described as a volume ratio of liquid to solid in the process. (Sherrington & Oliver 1981, 21-22.)

The moisture content in wet granulation is generally considered to be such that the voids in the agglomerate are just saturated (Capes 1980, 55). Experiments on sand grains granulation show that granule size growth can occur with moisture contents of 90 - 110 % of that required to saturate the voids in the packed sand grains. The results also show that with low moisture contents the granules are only closely packed while with higher moisture contents the granules are less highly packed. 85 - 93 % of the granule voids were found to be saturated in these experiments. (Sherrington & Oliver 1981, 22.)

It has been widely observed that moisture content in the granulating mass has a significant effect on the growth of granules. Higher moisture contents lead to increased surface moisture which results in a higher chance of particles sticking to each other when they collide. (Sherrington & Oliver 1981, 22-23.) However the necessary moisture content for granulation can be lower than that of required to completely saturate the voids. Accurate values of required moisture content are very hard to predict since there are several factors that affect the value, some of which are explained below. (Capes 1980, 56.)

If the components in granulation include water soluble material the amount of solution needs to be taken into account in the total liquid content which will increase the amount of liquid in the process. When soluble components are involved the temperature of the process influences the amount of solution which means that the required moisture content is temperature dependent. One example where the moisture-temperature relationships are important is fertilizer granulation. Other factors that affect the required moisture content include the wetting properties of the liquid/solid system, agitation conditions and mixing time, liquid and solid properties and the fraction of voids actually filled with liquid as the voids may contain trapped gas. (Capes 1980, 56; Sherrington & Oliver 1981, 22-23.)

It has also been noticed that the required moisture content is relatively less for small granules (1 or 2 mm diameter) than for larger granules. For small granules the required moisture content is much less than that to saturate the voids completely while for larger granules the required moisture content approaches the theoretical demand necessary to

completely saturate the voids. (Capes 1980, 58.) Sherrington & Oliver (1981, 22) present a simple model, shown in Figure 4, to understand this phenomenon.
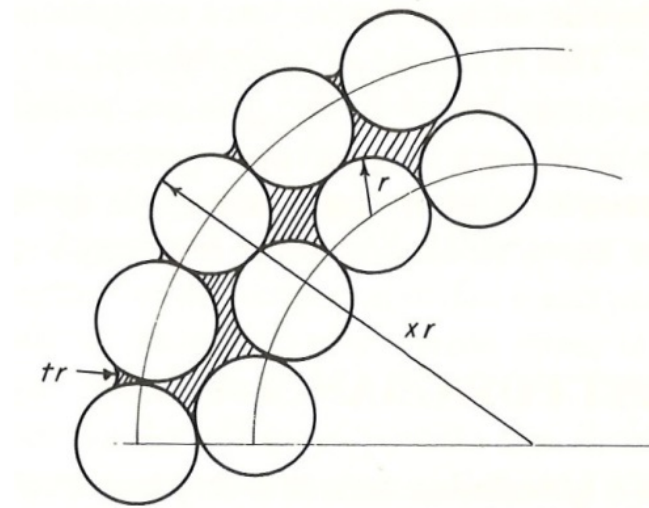

Figure 4. A magnified view of the granule surface: r = grain radius; xr = granule radius; tr = depth of withdrawal of liquid from granule surface (Sherrington & Oliver 1981, 22).

In Figure 4 the voids inside the granule are saturated but the surface is considered as dry. Liquid is withdrawn into the interstices between the particles on the surface in order to develop negative capillary pressure. This happens with both small and large granules. As smaller granules have higher surface area relative to volume than large granules, their moisture content is also lower. By using the geometrical considerations in Figure 4 the following equation can be written. (Capes 1980, 58; Sherrington & Oliver 1981, 23.)

$$y = k \left(1 - \frac{3td}{D} \right)$$

(5)

where
y volume ratio of liquid to solid [-]
k volume ratio of voids to solid [-]
t surface withdrawal parameter [-]
d feed particle diameter [m]
D granule diameter [m]

Equation (5) can be used to examine the relationship between required moisture content and granule size. However, accurate calculations for required moisture content cannot be made since values of k and t have to be determined experimentally for a specific agglomeration system. (Capes 1980, 58-59.)

4.2   Granule formation and growth

After the fundamentals of agglomeration, the physical properties of the binding liquid and the solid feed material, the granulator geometry and the process conditions are well understood, it is possible to make predictions for granule formation and growth in granulation processes. These factors determine the granule growth rate and the properties and form of the final product. (Sherrington & Oliver 1981, 24.)

The two granulation techniques that have been studied most are the rotating drum and the inclined pan granulators. The literature of granule growth processes has focused in the contribution of binding liquid content in the above-mentioned techniques. Sherrington & Oliver (1981, 24) state that three basic mechanisms for granule growth have been generally accepted using either a narrow or a wide particle size distribution. The mechanisms are:

1. nucleation of primary particles by random coalescence,
2. a transition region dependent on either:
   a. coalescence in a preferential mode or;
   b. crushing and layering
3. a 'ball' growth region.

Mechanisms (1), (2)(a) and (3) describe the growth with wide size distributions and (1) and (2)(b) with narrow size distributions. (Sherrington & Oliver 1981, 24-25.)

Granule growth mechanisms (1-3) above are observed to happen in numerical order. Experiments show that with both narrow and wide size distributions the growth process starts with rapid random coalescence (1). This is then followed by a transition region (2)

which is dependent on the particle size distribution. With a wide size distribution the growth continues by coalescence in the transition region. In this region the mechanism is size dependent and here the growth rate reaches its maximum. With a narrow size distribution, experiments show that as the granule size grows the intensity of the collisions increases leading to breakage of the colliding granules and even complete crushing of smaller granules. The crushed fragments and fines are then picked up by larger granules and the growth continues by layering. In mechanisms 1-2 the particle-particle bonds are stronger than the shear-generated torque caused by the collisions which makes coalescence possible. When the granule size reaches a critical point the torque becomes stronger than the particle-particle bond strength and balling becomes the dominant granule growth mechanism (3). (Sherrington & Oliver 1981, 25-26.)

The product granules are stronger when the particle size distribution is wide than when it is narrow. This is explained with a lower amount of point contacts in a narrow size distribution compared to a wide size distribution. With a wide size distribution the surface of the granule has been noticed to have excess moisture which is a result of the packing structure of the granule. As the granule rolls its structure becomes more packed and liquid is pushed from inside the granule to the external surface. The excess liquid on the surface allows some plasticity and deformation which makes further coalescence easier. The particle rearrangement behavior with a narrow size distribution is quite the opposite. As the granule is subjected to shearing or tearing forces while it rolls, dilatation occurs and the volume of the granule expands. This leads to liquid moving into the granule and its external surface may appear dry. Coalescence is less likely to happen and the granule will break down more easily. (Sherrington & Oliver 1981, 27.)

The granule growth behavior mentioned above applies well to granulation processes where the granules are formed by rolling, e.g. in rotating drum granulators or inclined pan granulators. However, different mechanisms can be dominant in other types of granulators. For example, in a fluidized or spouted bed granulator where the feed material is dry and binding liquid is sprayed on to or into the granulation material, layering or 'onion-skinning' has been noticed to be the dominant mechanism when using an aqueous salt solution as a binding liquid. (Sherrington & Oliver 1981, 28.)

4.3   Packing of particulate material

It is needed to understand the packing behavior of particulate material to estimate the voidage in the granules. As mentioned in chapter 4.1.2 the required binding liquid amount for granulation is determined from the voidage of packed material.

For an ideal case of smooth monosize particles the packing can be either regular or random. There are six possible arrays for regular packing of particles which are shown in Figure 5 and listed in Table 5 with their corresponding voidages. (Sherrington & Oliver 1981, 33.)
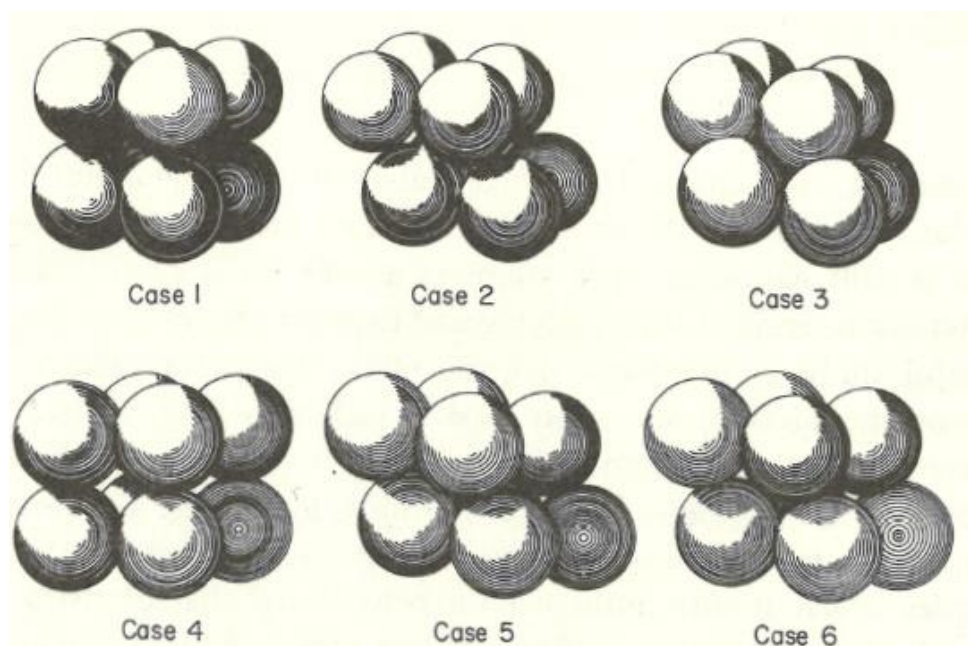

Figure 5. Geometric arrangements for smooth monosize particles (Sherrington & Oliver 1981, 34).

Table 5. Geometric arrangements in Figure 5 and the corresponding voidages (Sherrington & Oliver 1981, 34).

Case	Geometric arrangement	Voidage
1	Cubic	0,4764
2	Orthorhombic	0,3954
3	Rhombohedral (pyramidal)	0,2595
4	Orthorhombic	0,3954
5	Tetragonal-spheroidal	0,3019
6	Rhombohedral (hexagonal)	0,2595

When the particles are poured into a container and allowed to reach an arrangement by itself the packing is called random. Studies show that in random packing there is a network of somewhat regular packing regions that are basically of orthorhombic and tetragonal-spheroidal type. (Sherrington & Oliver 1981, 33-34.) Random packing can be used to describe the packing behavior in the case of handling fly ash where ash is stored in a silo or a container and it is not compressed in any way.

For random packing the particle assembly can be described with loose and dense random packing. The voidage of the random assembly is usually between the characteristic values for loose and dense packing presented in Table 6. It has also been shown that the voidage ϵ in random packing is nearly always in the range of $0,3624 < \epsilon < 0,4$. (Sherrington & Oliver 1981, 33-34.)

Table 6. Voidage of loose and dense random packing (Sherrington & Oliver 1981, 33).

	Voidage
Loose random packing	0,3990
Dense random packing	0,3634

4.4   Drying of particulate material

The tube granulator described in chapter 6 could be thought as sort of a cylindrical dryer since drying is one of its objectives. Therefore this chapter mostly focuses on drying in

a drum dryer which is the most similar drying process to what happens in the tube granulator.

When a hot gas is used to evaporate moisture from the material that is being dried, the evaporated moisture is "captured" by the gas and its humidity rises. Humidity Y is defined as the mass ratio of moisture vapor to dry gas according to equation (6). (Keey 1992, 117.)

$$Y = \frac{m_V}{m_G} \tag{6}$$

where m_V mass of moisture vapor [kg]

            m_G mass of dry gas [kg]

As the hot unsaturated gas flows through or over the wet material, heat from the gas is used to evaporate moisture from the material. This leads to a decrease in the temperature and an increase in the humidity of the gas. Evaporation continues until the gas is saturated and its humidity cannot be increased any further. If no heat is lost to the surroundings this is called adiabatic saturation which sets one limit to the drying capacity of a dryer. This limit is also called the cool limit. (Keey 1992, 123-124.)

When drying in a rotary drum dryer where the wet solids and gas flow through the dryer at the same time, the mass balance is

$$\frac{\dot{F}_0}{1 + X_0}(X_0 - X_P) = \pm \dot{G}(Y_{G0} - Y_{GE}) \tag{7}$$

where $\dot{F}_0$ mass flow of solids [kg/s]

            X_0 inlet moisture content of solids [-]

            X_P outlet moisture content of solids [-]

            $\dot{G}$ mass flow of gas (dry basis) [kg/s]

            Y_{G0} inlet gas humidity [-]

            Y_{GE} outlet gas humidity [-]

In equation (7) the positive sign is used when the materials flow is countercurrent and the negative sign in cocurrent flow. (Keey 1992, 258-259.) A rotary drum dryer with cocurrent flow of materials is presented in Figure 6.
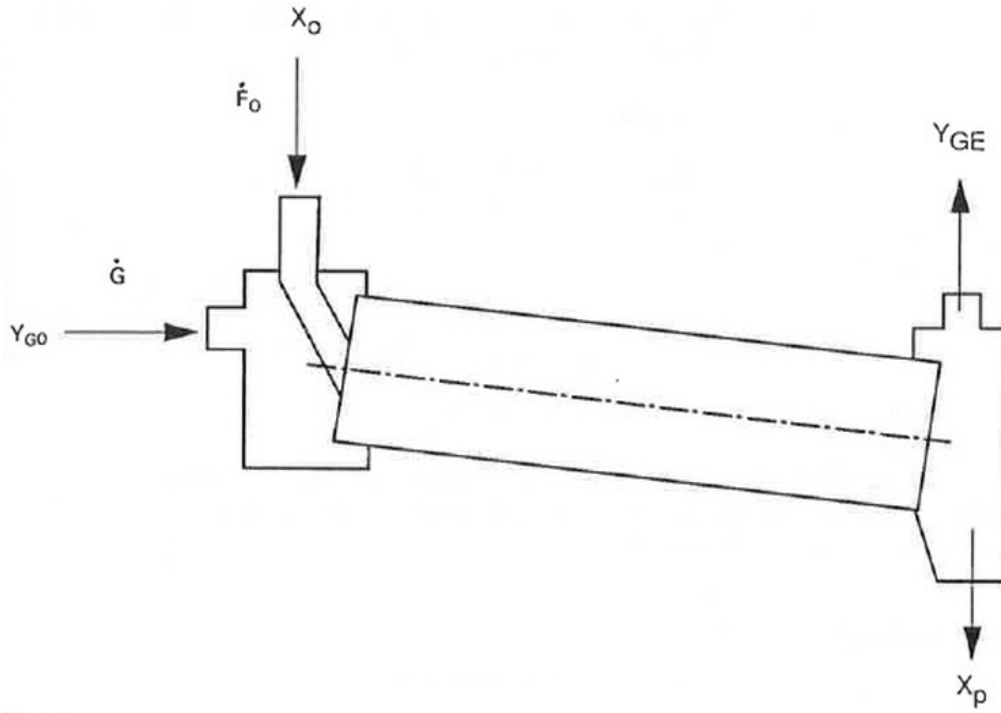

Figure 6. Rotary drum dryer with cocurrent flow (Keey 1992, 259).

If the inlet and outlet moisture contents of solids are specified and the solids are nonhygroscopic then the minimum gas flow in terms of adiabatic saturation can be determined with the equation below (Keey 1992, 259).

$$\dot{G}_{min} = \frac{\pm \dot{F}_0 (X_0 - X_P)}{(1 + X_0)(Y_{GS} - Y_{G0})} \qquad (8)$$

where $\dot{G}_{min}$ minimum mass flow of gas [kg/s]

Y_{GS} adiabatic saturation humidity [-]

Also in equation (8) the positive sign is used when the materials flow is countercurrent and the negative sign in cocurrent flow. In countercurrent flow the minimum gas flow obtained from equation (8) is an operational limit in drying. If lower gas flow is used

then dew will condense on the wet feed material. Adiabatic saturation humidity Y_{GS} in equation (8) represents the case when the solids are nonhygroscopic. In case the solids are hygroscopic then the hygroscopic saturation humidity Y_{HS} should be used instead. When the solids are hygroscopic the required minimum gas flow will be higher. (Keey 1992, 259.)

The adiabatic saturation humidity Y_{GS} refers to the absolute humidity of the gas at the adiabatic saturation temperature T_{GS}. The following equation presents the humid enthalpy change:

$$\frac{Y_{GS} - Y_G}{T_{GS} - T_G} = \frac{\bar{C}_{PY}}{H_L - H_V} = -\frac{\bar{C}_{PY}}{\Delta H_{VS}} \tag{9}$$

where $\bar{C}_{PY}$ humid heat [kJ/(kgK)]

               H_L enthalpy of liquid [kJ/kg]

               H_V enthalpy of vapor [kJ/kg]

               ΔH_{VS} latent heat of vaporization [kJ/kg]

In equation (9) the latent heat of vaporization ΔH_{VS} and the enthalpies of liquid and vapor are at the adiabatic saturation temperature. The adiabatic saturation temperature can be solved from equation (9) by iteration. The humid heat $\bar{C}_{PY}$ in equation (9) is expressed by the following equation. (Keey 1992, 125-126.)

$$\bar{C}_{PY} = \bar{C}_{PG} + \bar{C}_{PV} Y_G \tag{10}$$

where $\bar{C}_{PG}$ specific heat of air [kJ/(kgK)]

               $\bar{C}_{PV}$ specific heat of water vapor [kJ/(kgK)]

The adiabatic saturation temperature and the corresponding adiabatic saturation humidity can be solved from equation (9) by iteration (Keey 1992, 126).

As mentioned above, if the solids are hygroscopic meaning that moisture is bound to the solids, then the hygroscopic saturation humidity should be used in equation (8). In this

case the enthalpy of binding (or heat of wetting) should be taken into account. Now the cool limit is not at the adiabatic saturation humidity but at the relative humidity corresponding to equilibrium with the moisture in the solids instead. The limiting humidity in hygroscopic saturation can be estimated with the equation below. (Keey 1992, 127-128.)

$$Y_{HS} = \frac{\psi D Y_S}{D + (1 - \psi) Y_S} \tag{11}$$

where ψ equilibrium relative humidity (water activity) [-]

D molar mass ratio M_V/M_G [-]

Y_S saturation humidity ($\psi = 1$) at the limiting temperature [-]

Taking the heat of wetting into consideration, an enthalpy balance for the limiting conditions for adiabatic hygroscopic saturation can be expressed. The expression of humid enthalpy change in equation (9) becomes as follows in the case of hygroscopic saturation. (Keey 1992, 128.)

$$\frac{Y_{HS} - Y_G}{T_{HS} - T_G} = \frac{\bar{C}_{PY}}{H_L - \Delta H_W - H_V} = -\left(\frac{\bar{C}_{PY}}{\Delta H_V + \Delta H_W}\right)_{HS} \tag{12}$$

where T_{HS} hygroscopic saturation temperature [°C]

ΔH_W heat of wetting [kJ/kg]

Equation (12) can be used to solve the hygroscopic saturation humidity by iteration assuming that the heat of wetting is known. A value obtained from equation (11) can be used as a first estimate. Hygroscopic saturation humidity can also be obtained by using Figure 7 to determine the hygroscopic saturation temperature and looking up the corresponding tabular value for humidity. Figure 7 gives the hygroscopic saturation temperature as a function of the adiabatic saturation temperature T_{GS} and relative equilibrium humidity ψ. (Keey 1992, 129.)
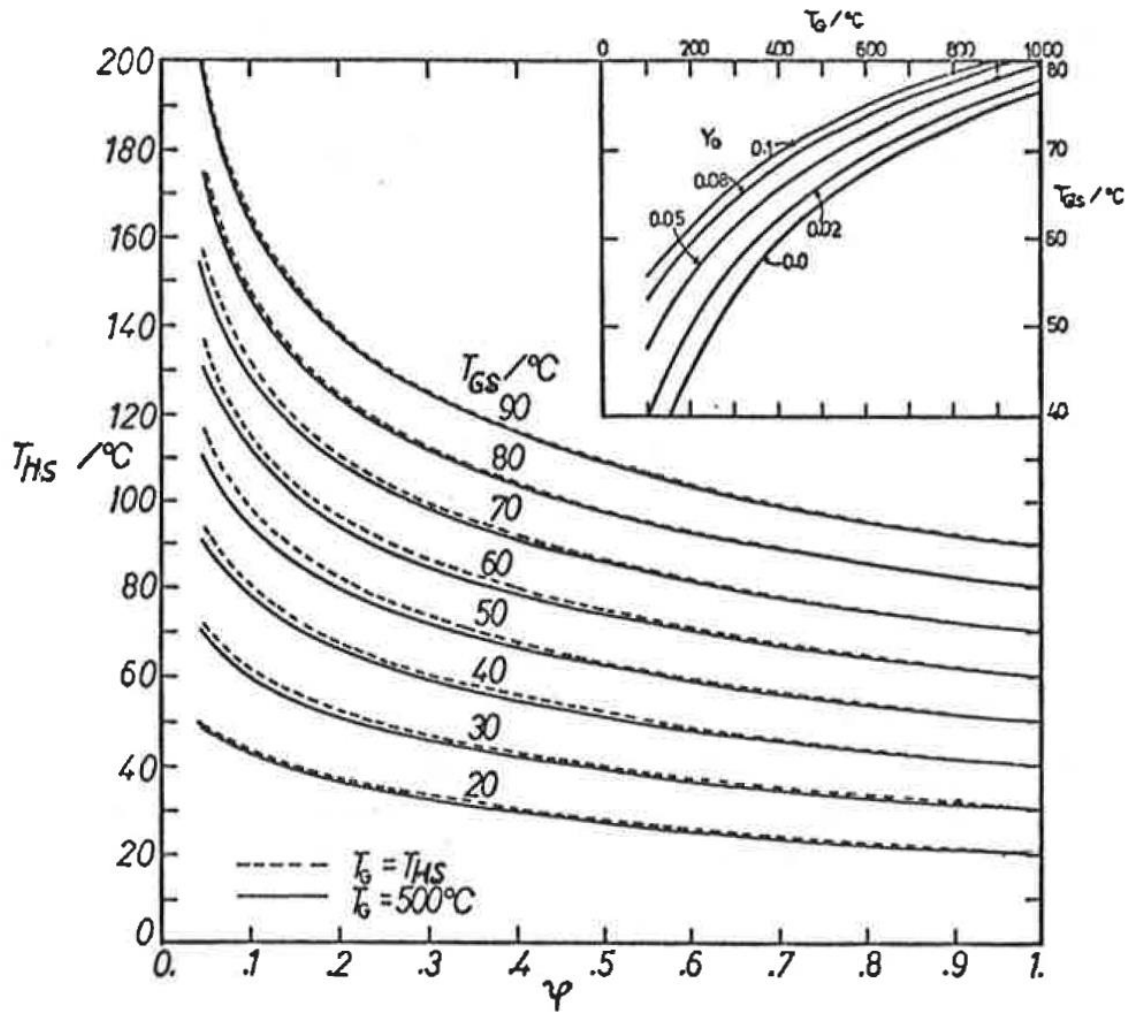
Figure 7. Hygroscopic adiabatic saturation temperature for water vapor in air as a function of adiabatic saturation temperature and equilibrium relative humidity (Keey 1992, 130).

5   CURRENT GRANULATING TECHNOLOGIES

As mentioned before, dusty ash has to be stabilized in order to handle and transport it efficiently and safely. Granulating is one of the techniques for stabilizing ash and it is the most effective stabilizing method used at the moment (Huotari 2012, 10). Granulating technologies currently in use are plate granulator and drum granulator. Other stabilizing methods in use are self-hardening and pelletizing. Common to all stabilizing methods is adding water in ash to start the chemical hardening. (Isännäinen et al. 2006, 9-10.) Granulating technologies are discussed individually later in this chapter. Self-hardening and pelletizing are briefly explained below to get an idea of alternatives for granulating.

Self-hardening is the simplest of stabilizing methods. Self-hardening process consists of moistening the ash and waiting for it to dry and harden naturally. Ash is mixed with water in a screw moistener or a vertical-shaft mixer where the target moisture content of ash is approximately 30 - 40 %. Then it is moved to pile at the mill area where it is let to harden for some weeks depending on the ambient temperature. Finally the hardened ash is screened and the largest pieces are crushed during loading for transportation. Self-hardening reduces the dustiness of ash significantly but it doesn't remove the problem completely. (Ramboll Finland Oy 2012, 16; Isännäinen et al. 2006, 9-10.) The end moisture content in self-hardening is approximately 25 % (Korpilahti 2003, 10).

Pelletizing of ash can be done with a roller mill or a matrix-moulding press. Both techniques are based on pressing ash to get compact material. Rolling produces either a thin sheet of ash or strings of ash when grooved rolls are used. After rolling the pressed ash is cut to pellet-like pieces or left as it is hoping that it will crumble enough in the following processing and handling. (Korpilahti 2003, 15-17.)

In matrix-moulding press moisturized ash is pressed through an aperture plate. The quality of the ash pellet produced is good but the technology has some problems. Erosive and corrosive ash is pressed with great force so it causes the aperture plate and other parts to wear fast. Ash also hardens like cement which causes blockages in the aperture plate. (Korpilahti 2003, 17; Isännäinen et al. 2006, 11.)

Granulating has several advantages over other stabilizing methods. For example hardening of ash is a lot faster in granulating than in self-hardening and the product is less dusty and has lower moisture content. (Huotari 2012, 10.) Granulating is the only stabilizing method that produces ash granules that are nearly equal in size (Korpilahti 2003, 6). It is also possible to improve the nutrient content of ash in granulation by adding other substances, such as industrial fertilizers or waste material, to the process (Huotari 2012, 10).

5.1   Plate granulator

In plate granulating the ash is first moisturized with water and then fed to a rotating plate. The plate is in inclined position and it has a blade that rotates in the opposite direction with the plate. Ash rolls on the plate and forms granules that eventually roll over the edge of the plate and drop onto a conveyor. A screening system can be added to the process after the plate to remove the smallest and largest granules. (Ramboll Finland Oy 2012, 16.) Particles that are separated as too large in the screening can be crushed and added to the accepted size granules (Korpilahti 2003, 13). Figure 8 shows an example of a plate granulator.

Figure 8. Plate granulator (left) at Hakoselkä granulation plant (right) (Ramboll Finland Oy 2012, 18).

Figure 9 shows an example of a plate granulating process. Here we can see that the granules that are too small or too large can also be recirculated in the process.
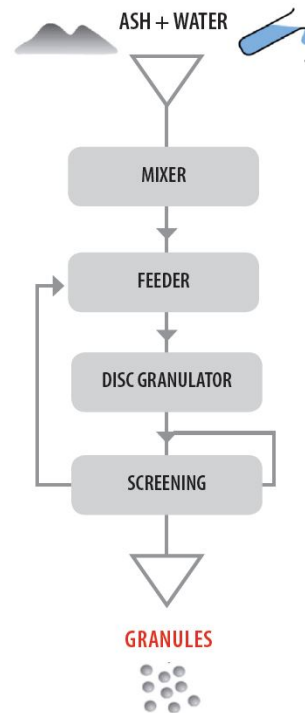
Figure 9. Plate granulating process used by Tecwill Granulators Oy (Tecwill Granulators 2013).

After the granulating process the granules are stored and further hardening occurs. After approximately two weeks the granules are hard enough and ready to be utilized. (Ramboll Finland Oy 2012, 16.)

5.2   Drum granulator

The basic principle of forming granules by rolling moisturized ash on a rotating surface is same in a drum granulator and a plate granulator. A drum granulator is a large rotating cylinder where ash rolls on its inner surface. (Korpilahti 2003, 5-6.) Ash is moisturized to approximately 20 % water content in a mixer before it enters the drum (Pusenius 2012, 15). Like the plate granulator, the drum granulator is also in an inclined position so the ash granules eventually pour out from the end of the cylinder and are dropped onto a conveyor (Ramboll Finland Oy 2012, 17).

Figure 10. Ash granules from a drum granulator in test use (Isännäinen et al. 2006, 10).

At FA Forest Oy's granulating plant in Viitasaari, Finland the ash granules from the granulator drum are moved into an intermediate storage where they harden. After the granules have hardened enough they are screened and the granules that are too large are crushed and returned back into granulation process. The granulation process of FA Forest's drum granulator plant is shown in Figure 11. (Pusenius 2012, 16.)
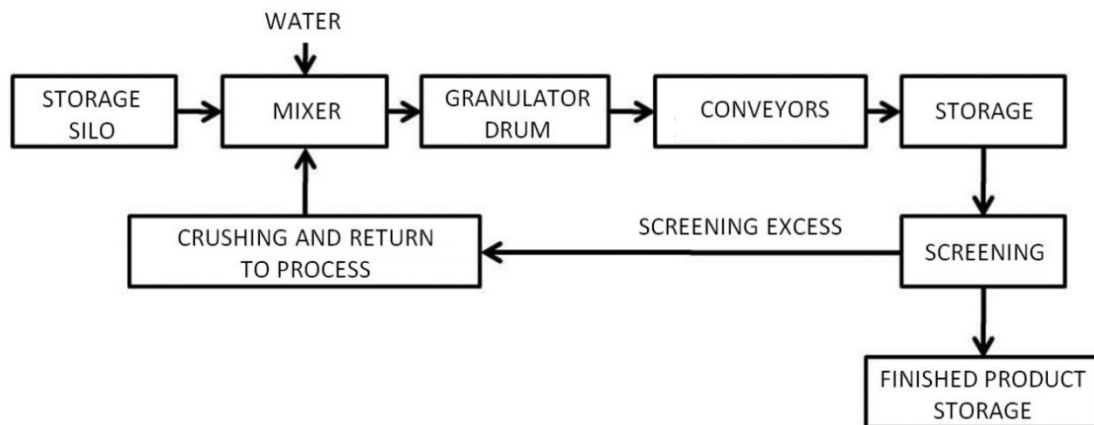
WATER

```
┌──────────┐      ┌─────────┐      ┌──────────────┐      ┌──────────────┐      ┌─────────┐
│ STORAGE  │ ──→  │  MIXER  │ ──→  │  GRANULATOR  │ ──→  │  CONVEYORS   │ ──→  │ STORAGE │
│  SILO    │      │         │      │    DRUM      │      │              │      │         │
└──────────┘      └─────────┘      └──────────────┘      └──────────────┘      └─────────┘
```

Figure 11. An example of a drum granulation process used at FA Forest's Viitasaari plant (Pusenius 2012, 16) (translated).

6 TUBE GRANULATION

A tube granulator is a new technique for granulation of fly ash. The principle process of tube granulation is invented by Kuopanportti (2015a). The tube granulation process includes rather simple and compact equipment. Granulation is done in a continuous process directly at the power plant, integrated into the power plant process. Similar to other granulation techniques, such as drum granulation and plate granulation, the formation of granules in the tube granulator is based on layered granulation.

The most significant difference between the tube granulation process and other granulation processes is that flue gas is introduced in the tube granulation process. The process utilizes the heat and high content of carbon dioxide in flue gases. A simplified example of the tube granulation process diagram is presented in Figure 12.
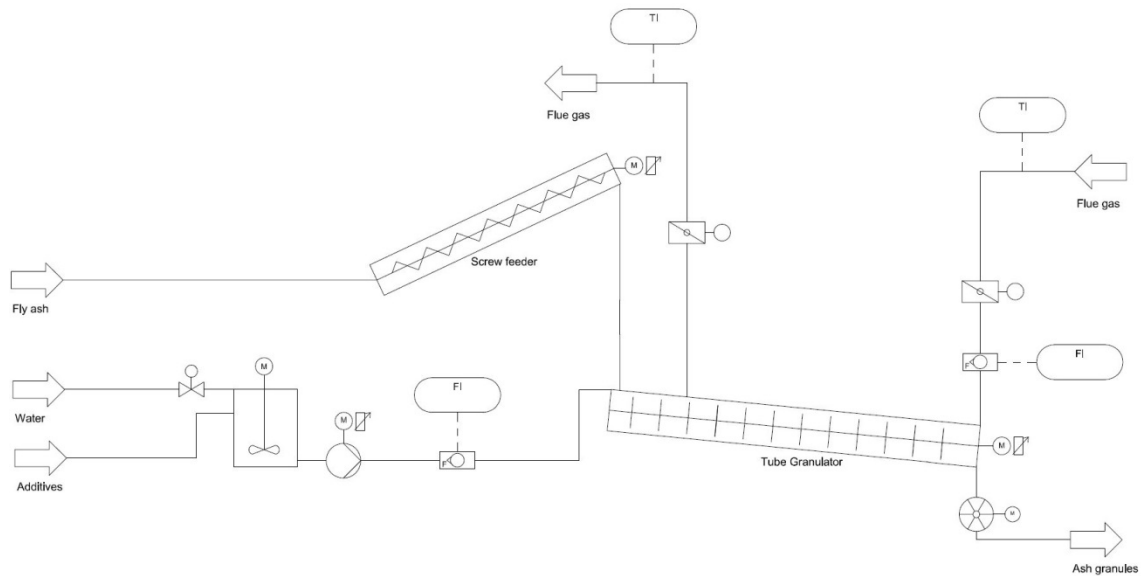

Figure 12. A simplified process diagram of an example execution of the tube granulation process.

6.1 Process

The tube granulation process is carried out in a cylindrical reactor/mixer. The process includes fly ash and water which are fed from the other end of the granulator and flue

gas which is fed from the other end of the granulator. Inside the granulator is a shaft with blades which is used to create intensive mixing of the feed substances and to create a driving force that makes the ash flow inside the cylinder. (Kuopanportti 2015a.)

The process can be divided into three parts that happen consecutively inside the granulator. The process consists of a moistening section, a reaction section and a drying section. These sections describe the state of the process inside the granulator and there are no physical boundaries between the sections. The main process of tube granulation is shown in Figure 13.
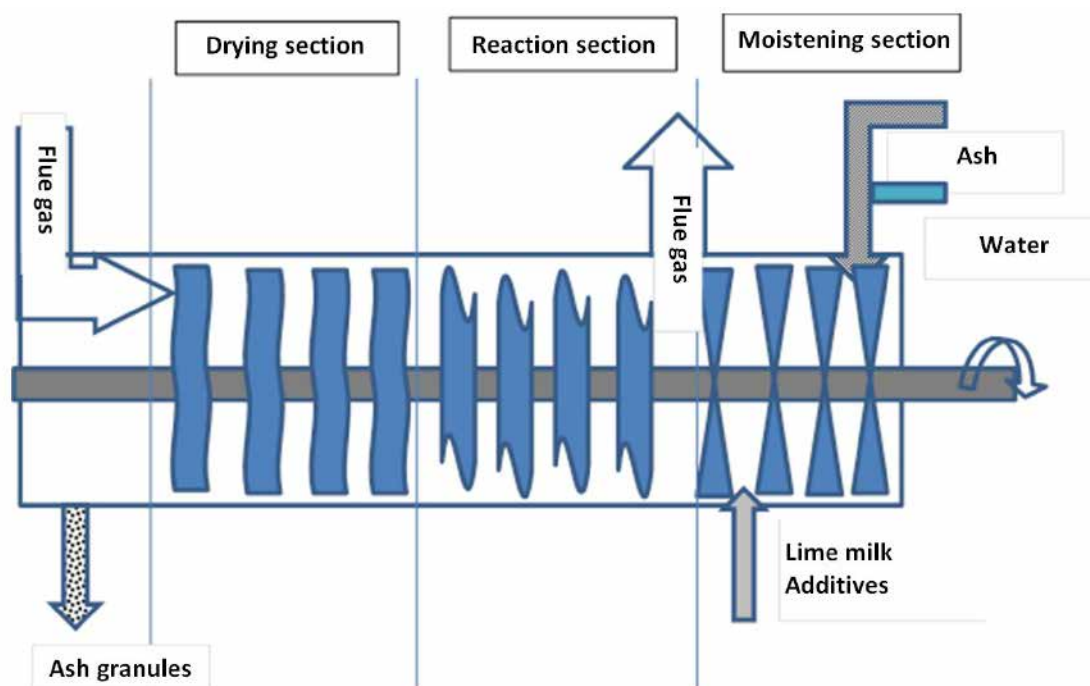

Figure 13. Tube granulation process principle (Kuopanportti 2015a).

Fly ash enters the granulator into the moistening section where water is added to moisten the ash. Water forms liquid bridges between the ash particles which binds the particles together. The rotating blades inside the granulator intensify the mixing of water and ash particles which accelerates the formation of liquid bridges. Moist agglomerates collide into each other and they stick together, forming bigger granules.

Fly ash contains a high amount of calcium oxide (CaO) which reacts with the added water. Calcium oxide and water form calcium hydroxide (Ca(OH)$_2$) according to the following chemical reaction:

$$CaO + H_2O \Rightarrow Ca(OH)_2 \tag{13}$$

Intensive mixing inside the granulator causes carbon dioxide (CO$_2$) from the flue gases to dissolve in water effectively. Carbon dioxide reacts with the calcium hydroxide and calcium carbonate (CaCO$_3$) is formed in the liquid bridges between the particles and on the surface of the agglomerates. Calcium carbonate is formed according to the following chemical reaction:

$$Ca(OH)_2 + CO_2 \Rightarrow CaCO_3 + H_2O \tag{14}$$
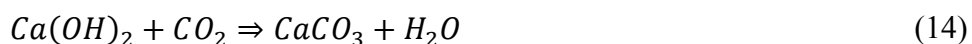
The formation of calcium carbonate is fast in the beginning of the reaction as long as there is carbon dioxide dissolved in the water. The use of flue gas instead of air increases the effectiveness of this reaction as flue gas contains a very high amount of carbon dioxide compared to air. The chemical reactions described in equations (13) and (14) occur in the reaction section of the granulator where the water is in liquid form. In practice these reactions begin as soon as the ash and water come into contact as flue gas, fly ash and water are always present simultaneously in the reactor tube.

The chemical reaction between calcium oxide and water requires the calcium oxide to dissolve in water. Mixing accelerates the dissolving but it still requires some time for a high amount of calcium oxide to dissolve. Although fly ash usually contains a relatively high amount of calcium oxide, the rate of dissolving may not be fast enough for the chemical reactions to be as effective as needed. Therefore additional calcium can be added in liquid form, for example as lime milk, in the moistening section of the granulator. This might be needed in case peat is used as the main fuel in the boiler as peat ash has lower calcium oxide content than wood ash.

In the drying section of the granulator the heat from hot flue gases evaporates water from the granules. As the granules dry the liquid bridges containing calcium carbonate become solid bridges. This leads to increased strength in the end product.

The quality of the end product can be increased by feeding additives into the granulator (Kuopanportti 2015a). For example, the nutrient content of the fly ash may not meet the minimum requirements for forest fertilizer use so additional nutrients can be added to fulfill these requirements.

There are several process conditions that can be changed to improve the strength and to reduce the solubility of the end product granules. The following changes have a positive effect on the quality (Kuopanportti 2015a):

- increase the amount of additional calcium
- increase the reaction temperature
- increase the amount of carbon dioxide
- increase the residence time of the process.

What also affects the effectiveness of the process and the quality of the granules is how fresh, or how shortly after being separated from the flue gases, the fly ash is obtained. With higher ash temperatures less energy in the drying process is required and the chemical reactions happen faster. Also the self-hardening of ash begins as soon as it comes into contact with the moisture in air. As a result of self-hardening the amount of active calcium oxide in ash decreases and the effectiveness of the granulation process suffers. Therefore the fly ash shouldn't be stored for long after it has been separated from the flue gases. (Kuopanportti 2015b.)

6.2 Equipment

The tube granulator itself is a rather simple device but for a complete continuous granulation system, some auxiliary equipment is also needed. The complete tube granulation system consists of the tube granulator, water and additives feed system, ash

feeding and removal system and flue gas system. Following is a description of the necessary equipment.

6.2.1 Tube granulator

The structure of the tube granulator is simple. The granulator is a cylinder which has connections for flue gas inlet and outlet, fly ash inlet and outlet for the end product granules. There is also a hatch on the top of the cylinder for inspection purposes. Water and additives can be injected into the same inlet as fly ash. Inside the cylinder is a shaft that has blades attached to it. The shaft is rotated by a motor connected to the cylinder end. A frequency converter connected to the motor is used to control the rotation speed of the shaft.

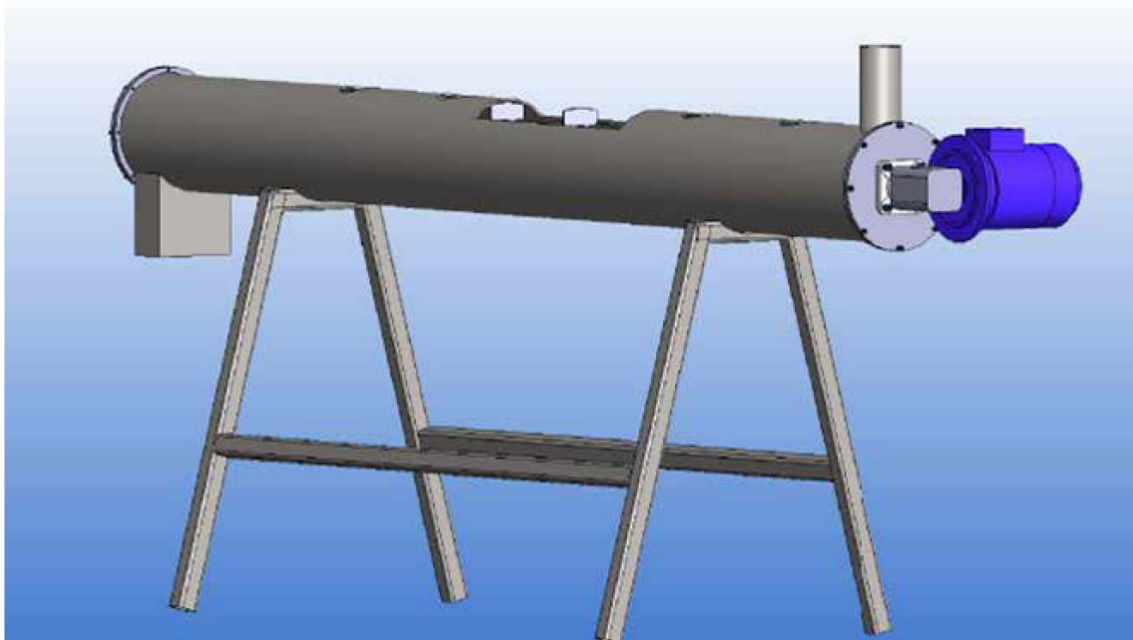A drawing of an example tube granulator is presented in Figure 14.

Figure 14. Drawing of an example tube granulator.

The shaft can be drawn out of the granulator for maintenance and cleaning by opening the end plate of the cylinder. In the pilot granulator the blades are attached to the shaft

in a way that enables adjusting the angle and vertical position of the blades or replacing them if needed. This makes it possible to optimize the position and angle of the blades during the test runs.

The granulator will be placed in an inclined position so that the granule outlet point is at the lower end of the granulator. This helps the granules to flow through the granulator where the counter-current flue gas flow creates an opposing force. Ensuring the ash flow through the granulator could be done by increasing the shaft rotation speed high enough but this would also increase the force towards the granules in collisions with the mixing blades. By placing the granulator in an inclined position, the shaft rotation speed doesn't have to be increased too much when flue gas flow is increased.

Hot flue gas runs through the granulator so all the connections and the hatch has to be gas tight. Depending on the flue gas connection points, there may also be negative pressure inside the granulator and gas tightness is important to prevent air inleakage. It should also be remembered in the granulator design that fly ash can easily get into the bearings of the shaft and damage them unless properly designed. The granulator has to be insulated for safety reasons and to minimize heat losses.

6.2.2 Auxiliary equipment

The tube granulation process requires a set of auxiliary equipment for different purposes. The combination of the equipment may vary case by case as there are different ways to carry out some of the necessary functions. The following is a listing of the basic auxiliary equipment involved in the tube granulation process. An example of the auxiliary equipment assembly can also be seen in Figure 12.

To be able to transport the dry ash from a fly ash silo or a container to the granulator, a feeding device is needed. The simplest solution for this is to use a motor-driven screw feeder. In order to control the mass flow a frequency converter is needed for the motor. To minimize the risk of flue gases getting out of the process through the screw feeder

there can be a rotary feeder between the screw feeder and the granulator. The motor of the rotary feeder doesn't necessarily have to be equipped with a frequency converter as it can be designed to operate at higher capacity than the screw feeder. In case it is decided not to use an existing fly ash silo for storage, then a container for the dry ash is also necessary. Having a separate ash container has some benefits compared to a fly ash silo which is explained in chapter 7.2. The other end of the granulator, where the granules come out, should be equipped with a rotary feeder. Similar to the inlet, this is to minimize the risk of flue gases exiting the system from the wrong place. The suction at the flue gas outlet combined with this rotary feeder should be enough to ensure that flue gas doesn't leak from the system.

Flue gas ducts need to be installed that connect the granulator into the flue gas system of the power plant. To minimize heat losses and for safety reasons, the flue gas ducts need to be insulated. Control dampers are needed in both the inlet and outlet ducts in order to control the flue gas flow into the granulator and to shut off the gas flow when the granulator is not in use. In case the sealing must be absolutely gas tight then on/off dampers are also needed.

The water used in the granulation process is taken from the power plant's water system so piping is needed for the connection. Water is injected into the granulator through feed nozzles. The water pressure of the mill water system may be sufficient in which case only a control valve and a flow meter are required to adjust the water flow. In case a higher water pressure is needed, a separate motor-driven pump equipped with a frequency converter is needed. Before the pump there is a stop valve in the water pipe. The amount of feed nozzles depends on the size of the granulator but two nozzles should be enough for most cases. The placement of the feed nozzles is so that water is sprayed in the same direction as ash entering the granulator, i.e. downwards. This way the whole cross-sectional area of the inlet duct can be easily covered with the spray and also the water spray doesn't throw ash on the duct walls.

If some additives are used in the process then a chemical feeding system is necessary. The chemical feeding equipment depend on what type of additive is used and whether it

is injected directly into the granulator or mixed with water. If the additive is in liquid form and it is injected directly then a chemical tank, a chemical pump and a feed nozzle is required. Also a frequency converter for the pump is needed to control the mass flow. If the additive is mixed with water then a mixing tank is necessary and only one feed line is needed. This kind of equipment assembly also requires a pump equipped with a frequency converter to control the pressure and mass flow from the mixing tank.

A continuous tube granulation process is integrated into the power plant's distributed control system (DCS) and the process is monitored and controlled with the help of certain measurements. The type and amount of different measuring instruments depends on how extensively the process conditions need to be known. The basic instruments include a flow meter for water and additives and a surface level measurement for dry ash storage. In the flue gas side useful information can be obtained with temperature and flow measurements. The surface level measurement in fly ash silo or ash container is important so that the ash storage is not drained empty. The screw feeder rotational speed is controlled by the surface level measurement in order to maintain the surface level within specified limits. Temperature measurements in the flue gas side give information about the heat transfer in the process. There might be a temperature measurement in the power plant's flue gas system after the flue gas fan, or ID (induced draft) fan, in which case an additional temperature measurement is only needed in the flue gas outlet duct from the granulator. A volumetric flow measurement in the flue gas inlet duct is used to control the flue gas flow by using the control dampers.

6.3   Process and equipment dimensioning

The desired ash handling capacity sets the base for dimensioning of the granulation process. As the tube granulation process is a new invention there are no straightforward guidelines for the process dimensioning. This chapter introduces some approaches for the dimensioning.

The tube granulation process includes multiple stages that need to be examined separately. One must consider that sufficient amount of ash particles is fed for efficient

agglomeration by layering, that the residence time in the granulator is long enough for the reactions to happen and that the desired degree of drying is achieved. When the ash handling capacity of the granulator is calculated with these different approaches, different capacities are obtained from each method. Therefore the dimensioning has to be done by either choosing which approach is to be used or by estimating the optimal design parameters from the different approaches.

The first step in the process dimensioning is to determine the moisture content of the ash-water mixture. This can be calculated when the voidage in the packing of ash particles and the required degree of saturation of these voids are known. For simplification the ash particles can be assumed smooth and monosize. This gives us an approximation for the moisture content and the optimal moisture content can then be determined empirically by adjusting the water flow into the granulator. The voidage ϵ of smooth monosize particles in random packing is between 0,3624 and 0,4 as noted in chapter 4.3. As presented in chapter 4.1.2 moisture contents of 90 - 110 % of that required to saturate the voids have been noticed to be optimal for granulation. However the key idea in tube granulation is to obtain strong solid bridges between particles by rapid formation of calcium carbonate and subsequent drying and therefore a lower moisture content than suggested can be used (Kuopanportti 2015b). Capillary state of saturation is not necessarily needed and the saturation S could even be less than 80 %.

One approach for the granulator dimensioning is by choosing a degree of filling and a residence time for the particles inside the granulator. This approach is based on two facts that affect the granulation efficiency. The first is that there should be enough ash particles inside the granulator for a high rate of collisions of particles. The more collisions of particles happen the higher is the chance that particles stick together and form bigger agglomerates.

The other fact is that the chemical reactions that happen in the process require a residence time. The residence time also has an impact on the drying of the granules. The dissolution of carbon dioxide through the liquid layer is relatively fast at the tube granulator conditions where the flue gases are hot and the feed materials are well mixed.

Also the subsequent formation of calcium carbonate is very fast in these conditions. What requires the most time is the formation of liquid calcium hydroxide from solid calcium oxide and liquid water. (Kuopanportti 2015b.) The required residence time in the granulator is shortened if enough lime milk is added in the process. The residence time can be controlled by adjusting the angle of the mixing blades, changing the shaft rotation speed, adjusting the inclination of the granulator and changing the flue gas flow rate. The optimal combination of these variables has to be found empirically as no earlier references for this kind of a process are available.

One method to search the optimal variables mentioned above is to initially choose an angle of inclination of the granulator and then change the other parameters. The rotation speed of the shaft can be chosen so that it creates as much driving force to the ash flow as possible without directing too much shearing stress which would result in broken granules. Some of the bigger granules are allowed to break into smaller granules as long as the granule size distribution stays in the desired range. When the optimal rotation speed is found the residence time can be controlled by the flue gas flow. Flue gas flow should be as high as possible in order to maximize the available carbon dioxide and the drying of the granules. As the flue gas flow is counter-current to ash flow it creates an opposing force on the ash flow and tries to push back the ash particles. Therefore the flue gas flow can be increased only as long as the ash flows freely through the granulator.

Finally the ash handling capacity can be calculated by using the residence time and choosing a degree of filling. A higher degree of filling means higher capacity and improved granulation because of more collisions of particles while a lower degree of filling means dryer and stronger granules.

Another approach for the dimensioning is to set a target moisture content for the product granules and examine the process in terms of drying. Adiabatic saturation humidity can be used to define the required minimum flue gas flow in the granulator and furthermore the maximum ash handling capacity of the granulator. The adiabatic

saturation humidity can be solved by iteration using equation (9) in the following form (Keey 1992, 126):

$$\hat{T}_{GS}^{(k+1)} = T_G - (Y_{GS}^{(k)} - Y_G)\frac{\Delta H_{VS}^{(k)}}{\overline{C_{PY}}} \tag{15}$$

which gives an estimate to T_{GS} from a previous value. The adiabatic saturation humidity Y_{GS} is a tabular value that depends on T_{GS}. The iteration scheme for the adiabatic saturation humidity can be presented as below (Keey 1992, 126).

$$Y_{GS}^{(k+1)} = Y_{GS}\left(\hat{T}_{GS}^{(k+1)}\right) \tag{16}$$

Keey (1992, 126) suggests that the adiabatic saturation temperature will be a bit higher than the dew point temperature of the gas at the inlet and therefore the following equation can be used to obtain the first value of T_{GS} for iteration in equation (15).

$$T_{GS}^{(1)} = T_D(Y_G) + \delta T \tag{17}$$

where T_D dew point temperature [°C]

              δT small temperature increase, e.g. 2 °C

When the humidity of the inlet flue gas is known the dew point temperature can be obtained by using Dalton's Law of Partial Pressures. After obtaining the adiabatic saturation humidity by iteration of equations (15) and (16), and deciding the desired product granule moisture content, equation (8) can be used to calculate the minimum flue gas flow. The minimum flue gas flow is obtained as mass flow of dry gas per mass flow of moistened ash.

The flue gas mass flow in the granulator can be calculated by specifying the granulator dimensions and choosing a flue gas velocity. The granulator cross-sectional area and the flue gas velocity define the volumetric flow. The mass flow is obtained by using the flue gas density at the given pressure and temperature. Pressure can be assumed to be close to 1 atm when the gas enters the granulator and the temperature after flue gas fan

can be used. Now the moist solids material flow can be obtained by using the minimum flue gas flow per feed material flow as obtained before. The ash handling capacity of the granulator is the fraction of dry ash mass flow in the feed material flow.

7 IMPLEMENTATION OF TUBE GRANULATOR INTO POWER PLANT PROCESS

The placement of the tube granulation equipment and connection points to the process have to be designed case by case as power plant processes and layouts are different. If the tube granulator is to be installed as a part of a new power plant delivery then the implementation can be optimized. However it has to be remembered that the tube granulation process is not a main process in the power plant and therefore it has to be adapted according to the plant design. When installed into an existing power plant the placement of the tube granulation equipment may involve some challenges. The available space for the equipment at existing power plants requires some optimization in order to find a solution with reasonable investment costs and good conditions for the process. Figure 15 presents one possible solution for tube granulator implementation into a power plant process.
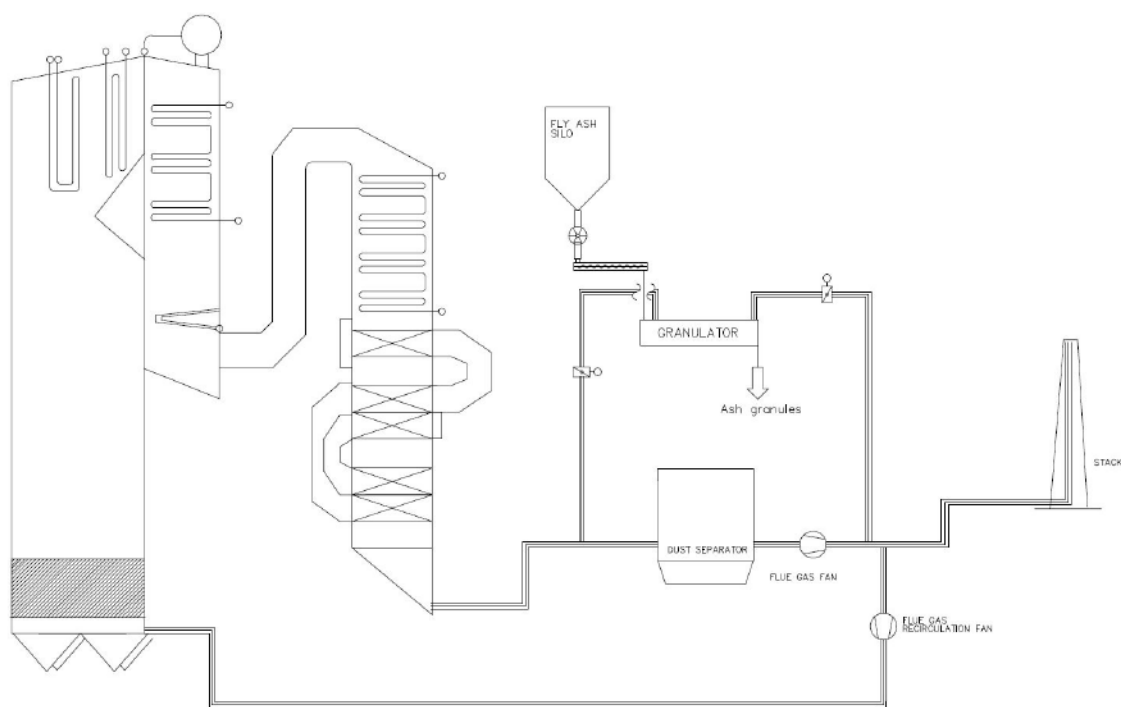

Figure 15. Example of tube granulator implementation into a power plant process.

There are several things that have to be taken into account when planning the implementation of a tube granulator. Connections of the flue gas, ash, water and additives systems into a power plant process are discussed in the following chapters.

7.1 Flue gas connections

There are a few possible solutions for the flue gas connection points that mostly depend on the power plant design. If a separate fan for the tube granulator inlet flue gas is not used then a sufficient pressure difference between the inlet and outlet connection points is needed. A pressure difference is required to create a high enough gas flow through the granulator. To achieve the pressure difference, there must be a flue gas fan between the inlet and outlet connection point. The flue gas must be taken from the pressure side of the fan and returned to the suction side of the fan. In a power plant process which includes flue gas recirculation the flue gas inlet connection can be at the pressure side of the recirculation gas fan to obtain the biggest pressure difference. Figure 15 shows an example connection where the flue gas is taken after the flue gas fan and is returned to the flue gas path before a dust separator.

In case sufficient pressure difference cannot be achieved between the inlet and outlet connection points, a separate fan for the granulation process flue gas is needed. This might be the case if the flue gas inlet connection cannot be installed to the pressure side of a flue gas fan or if the flue gas duct from the connection point to the granulator becomes unreasonably long. Especially at existing power plants the layout may require the granulation equipment to be placed far away from the flue gas fan and installing a separate fan can be cheaper than to make very long ducts.

The flue gas outlet connection point should be placed before the flue gas fan in the flue gas flow path. For maximal pressure difference it would be best to place the connection point as close as possible to the flue gas fan as there are pressure losses in the flue gas path. Another good choice is to make the connection before a dust separator, for example an electrostatic precipitator (ESP) or a cyclone. The reason for this is that in some situations there can be some ash entrained in the granulator outlet flue gas flow.

The fly ash entering the granulator is very fine and it is possible that some of the smallest particles get through the moistening section without touching the water spray. Also there might be a malfunction in the water spraying system which can cause a large amount of ash to get entrained in the exiting gas flow before feeding of ash is stopped. Another possible situation where ash can get back into the flue gas duct is when the flue gas flow in the granulator rises so high that it reaches the fluidization velocity for the moistened ash agglomerates.

If entrained ash gets into the flue gas duct and there is no dust separator before the flue gas fan and the stack, the dust emission levels will increase and the flue gas fan blades may be damaged due to erosion. Placing the flue gas outlet connection point before the dust separator minimizes those risks. Therefore it can be suggested to do so as long as the plant layout allows it and sufficient pressure difference is achieved.

7.2   Ash connections

The tube granulation process is continuous and the ash flow into the granulator must be controllable in order to adjust the water flow accordingly. This means that there must be enough ash available to the feeding device so that the flow rate can be properly controlled.

The fly ash collected from the dust separators is usually at first collected in an ash hopper and then transported pneumatically to the fly ash silo. The pneumatic transmitters send a batch of ash periodically. Also while a cyclone gathers fly ash into the ash hopper quite continuously, an ESP collects the fly ash on collecting plates from which the ash is dropped into the ash hoppers periodically by rapping devices. This means that the tube granulator can't be placed directly after the dust separator and a buffer storage is needed.

At existing power plants the fly ash silo can be used as a buffer storage. In this case the ash feeding device can be installed directly to the silo. Figure 15 presents such a case where the screw feeder for the tube granulator is connected to the fly ash silo which is

equipped with a rotary feeder. However the fly ash silo which is meant for storing ash is unnecessarily large as a buffer storage because the idea is to granulate the ash as soon as possible after it has been collected from the flue gases. By adding a smaller container as a buffer storage the storage size can be optimized. This also gives more freedom in the placement of the equipment as otherwise the granulator has to be close to the fly ash silo or additional ash conveying lines have to be added. At power plants where the fly ash silo is outdoors cold weather can be a problem as well if the fly ash silo is used since the temperature of the fly ash in the granulation process should be as high as possible. Basically if the optimal place for the granulator is close to the fly ash silo it is simplest to use the existing silo but otherwise a separate buffer storage can be a better solution.

In case a small buffer storage is used the ash conveying system has to be designed so that the whole granulation process can be bypassed and ash can be conveyed to a bigger silo. This applies to both existing and new plants and is important so that the power plant process can be operated normally in case the granulator is out of operation because of maintenance or a malfunction. At existing plants that already have ash conveying systems from the ash hoppers to the fly ash silo the connection to the buffer storage should be made in the conveying line with a possibility to switch the route to the fly ash silo. In case a totally new ash handling system is being designed one solution would be that the ash conveying line from the hoppers leads to the buffer storage and a second conveyor between the buffer storage and the fly ash silo is made.

7.3   Water and additives connections

Water for the granulation process can be taken from the power plant's raw water network although the process conditions can be improved if warmer water is available. This means that the water connection for the granulator can be made in the nearest raw water or process water line based on whichever can be used. The water can be injected into the granulator from its own feed line in which case the additives would be injected from another feed line if they are used. Another possible solution is to mix the water and additives streams into a single feed line.

When separate feed lines are used for water and additives, both the streams can be precisely controlled by a control valve in the water feed line and a pump equipped with frequency converter in the additive feed line. In this case the water feed line is directly connected to the power plant's water network and the additives line to a chemical tank. The downside of using separate feed nozzles is that the uniformity of mixing of water and additives is harder to achieve.

In case the water and additives are mixed in a mixing tank, both streams are primarily connected to the tank after which they are combined into a single stream. Now the amount of water and additives going to the mixing tank must be precisely controlled. The uniformity of the solution is achieved by efficient mixing.

8 ECONOMIC BENEFITS

This chapter presents some economic benefits that are obtainable by the user of the tube granulator. Savings and profits achievable depend on what would be done with the ash without the tube granulation process and how can the ash be utilized after granulating. Savings can be obtained from taxes, landfill site fees and transportation costs. If the granulated ash can be sold as a product then the power plant will get profits.

A large power plant with two boilers and a combined fuel power of approximately 580 MW is taken as an example in the economic calculations. About 50 000 t of fly ash is collected annually at the plant. The calculations are based on Finnish legislation and cost information.

By far the biggest savings are obtained when granulated ash is utilized somehow compared to a situation where the ash ends up as waste. As mentioned in chapter 3.2 fly ash is defined as waste unless utilized and it is taxable according to The Finnish law covering taxation of waste (Law 1126/2010). The taxation has been increased in a revision of the above-mentioned law in 2014 (Law 1072/2014) when it was raised from 50 €/t to 55 €/t and a new revision for the law has already been accepted which has raised the tax to 70 €/t in 2016 (Law 1401/2015). For the example power plant the waste tax in 2016 would be equal to

$$50\,000\,\frac{t}{a} \cdot 70\,\frac{€}{t} = 3\,500\,000\,\frac{€}{a} \tag{18}$$

The amount of waste tax calculated in equation (18) is calculated for dry ash ($X = 0$). The actual amount of waste ash is higher since dry and dusty ash has to be moistened before it can be placed in a landfill.

In addition to the waste tax, landfill site fees need to be paid as well. The fees can be around 65 - 90 €/t without Value-Added Tax (VAT) (Oulun Jätehuolto 2015, Vestia 2015). Granulating fly ash improves the possibility of utilizing it which means that the waste tax in equation (18) and the landfill site fees could be avoided.

Savings are also earned in transportation costs. Savings are earned through bigger transport capacities for granulated than dusty ash and need for less transportation steps as the ash is granulated directly at the boiler plant. Dusty ash has to be transported by tank trucks or in closed containers where the maximum capacity is 30 t while granulated ash can be transported by full articulated vehicles where the maximum capacity is 40 t (Ramboll Finland Oy 2012, 27). Savings are also earned in the case where ash is granulated instead of simply moistening and self-hardening it for easier transportation. The target moisture content of ash granules from the tube granulation process is around 10 - 15 % while self-hardened ash is at approximately 25 % moisture content, as mentioned in chapter 5. Therefore less water and more ash is being transported when the ash is granulated instead of self-hardening.

According to available data from year 2010 the road traffic costs in Finland are following (Tervonen & Ristikartano 2010, Ramboll Finland Oy 2012, 27):

- Articulated vehicle's fuel costs 19,48 c/km and other operating costs 22,45 c/km without taxes
- Fuel taxes 20 c/km as they are about half of the fuel price
- Articulated vehicle's capital costs 10,32 €/h
- Driver's hourly costs 24,14 €/h
- Overhead costs and margin approximately 10 %

From the values above, total cost per distance is 0,62 €/km and total cost per time is 34,46 €/h. For given distance, driving speed, loading time and unloading time, the unit costs for 30 t and 40 t transport sizes can be calculated. A distance of 50 km shall be used with an average driving speed of 60 km/h. Loading and unloading of ash is estimated to take 0,5 h in total. For a round-trip the transport costs would be as follows.

$$1,1\left(2 \cdot 50\ km \cdot 0,62\frac{€}{km} + \left(0,5\ h + 2 \cdot \frac{50\ km}{60\frac{km}{h}}\right)34,46\frac{€}{h}\right) \tag{19}$$

$$= 150,3\ €$$

The unit cost for transportation can be calculated by dividing the transportation costs from equation (19) with transport distance and capacity. Unit cost for a 30 t transport is calculated below.

$$\frac{150{,}3\ €}{30\ t \cdot 50\ km} = 0{,}10\ \frac{€}{tkm} \tag{20}$$

In equation (20) the distance isn't multiplied by two because it is assumed that the truck returns empty. For a 40 t transport the unit cost would be 0,075 €/tkm.

Assuming that the ash would normally be transported to a granulation plant but is now granulated at the power plant, one transport step less is needed. For the example power plant the annual transportation costs to a granulation plant at 50 km distance would be following.

$$50\ 000\ t \cdot 50\ km \cdot 0{,}1\ \frac{€}{tkm} = 250\ 000\ € \tag{21}$$

The unit cost for 30 t transport size is used in equation (21) since the ash is transported as dry to the granulation plant.

In case the ash would normally be moistened and self-hardened before transporting it, for example to be utilized or to a landfill site, savings are obtained through less water being transported. Both granulated and self-hardened ash can be transported by full articulated vehicle transports and the unit cost for 40 t transports is used. For the example power plant the transportation costs for a 50 km distance would be as follows.

Self-hardened ash at 25 % moisture content:

$$\frac{50\ 000\ t}{0{,}75} \cdot 50\ km \cdot 0{,}075\ \frac{€}{tkm} = 250\ 000\ € \tag{22}$$

Granulated ash at 10 % moisture content:

$$\frac{50\,000\ t}{0,90} \cdot 50\ km \cdot 0,075\ \frac{€}{tkm} = 208\,333\ €$$ (23)

Annual savings for transporting granulated ash instead of self-hardened ash can be obtained from the difference of costs from equations (22) and (23) which in this case is 41 667 €. Transportation costs at different distances from 10 km to 90 km are presented in Figure 16.
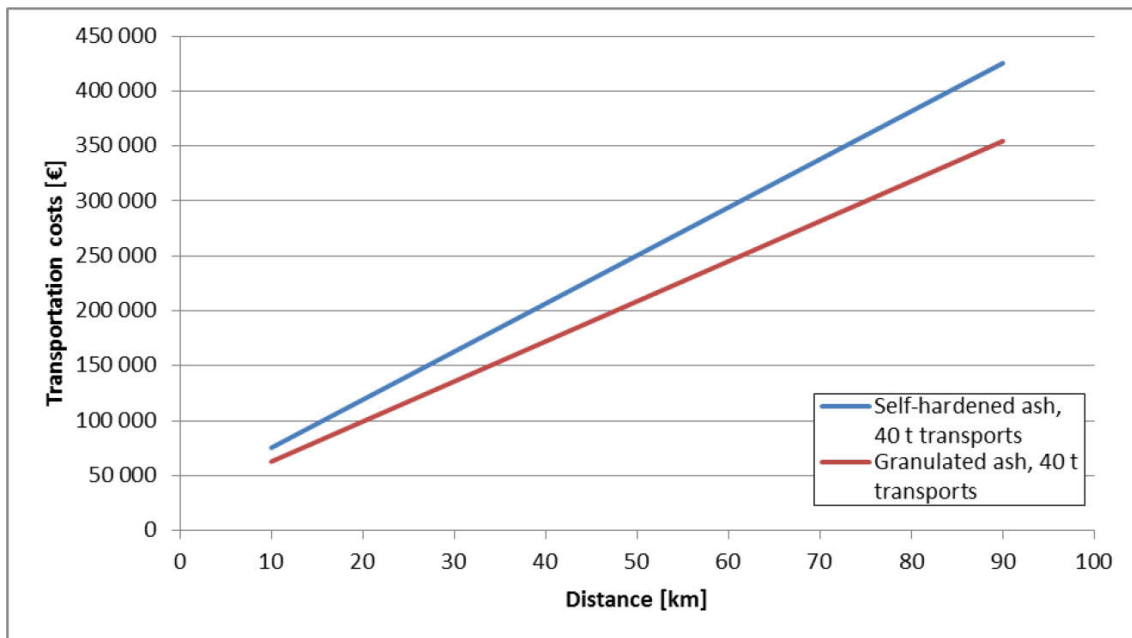

Figure 16. Transportation costs for self-hardened ash at 25 % moisture content and granulated ash at 10 % moisture content.

When the ash is granulated at the power plant it is also possible to obtain sales profits from the granulated product. Normally the ash would be transported to a granulation plant which would get the added value from processing the ash. By selling the ash granules the product liability is at the power plant which includes some expenses due to for example necessary permits and quality analyses (Ojala 2010, 10-12). However the sales profits could be significant. For example, in case the ash granules are sold as forest fertilizer the sales price can be 70 - 80 €/t according to Soikkeli (2015). That is the purchase price of Metsänhoitoyhdistys Keski-Savo which sells forest fertilizing for

forest owners. The price is for ash fertilizer as delivered to the site where it is used. Possible sales profits for the example power plant are calculated below.

Assuming that the average transport distance is 50 km from the power plant to the forest and the ash granule moisture content X is 10 %, the transportation costs would be 208 333 € as obtained from equation (23). With a 70 €/t sales price the annual profits would be:

$$\frac{50\ 000\ t}{0{,}90} \cdot 70\ \frac{€}{t} - 208\ 333\ € = 3\ 680\ 556\ € \tag{24}$$

This calculation is based on the assumption that all the ash produced can be utilized as ash fertilizer. However it is unrealistic to assume that such a high amount of ash can be sold as a forest fertilizer at all times since the demand may not be that high, or at least the transport distance can be much higher. Assuming that a portion of the total ash produced is sold as a forest fertilizer and the rest is defined as waste a simple estimation for profits or costs can be made. Table 7 below presents profitability calculations using 50 000 t annual dry ash, taking into account the percentage of ash utilized, the waste taxes, landfill site fees and sales profits. Transportation and costs are not included in these calculations and the granulated ash is at 10 % moisture content.

Table 7. Profitability in terms of ash utilization percentage. (kt = 1000 tons, k€ = 1000 euros)

Total dry ash 50 000 t							
Ash utilized from total ash		0 %	20 %	40 %	60 %	80 %	100 %
Waste ash (X = 10 %)	kt	56	44	33	22	11	0
Utilized ash (X = 10 %)	kt	0	11	22	33	44	56
Waste tax (70 €/t)	k€	3 889	3 111	2 333	1 556	778	0
Landfill site fees (75 €/t)	k€	4 167	3 333	2 500	1 667	833	0
Ash fertilizer sales (70 €/t)	k€	0	778	1 556	2 333	3 111	3 889
Total profit	k€	- 8 056	- 5 667	- 3 278	- 889	1 500	3 889

Considering the assumptions made above, it can be seen that over 60 % of the granulated ash would have to be utilized as forest fertilizer in order to obtain a positive total profit. It can also be noticed that the differences in total profit, or costs, are significant when the utilization percentage increases. Keeping in mind that 50 000 t fly ash annually is a relatively high amount, it is reasonable to examine lower amounts as

well. Figure 17 includes total profit calculated as in Table 7 with three different fly ash amounts.
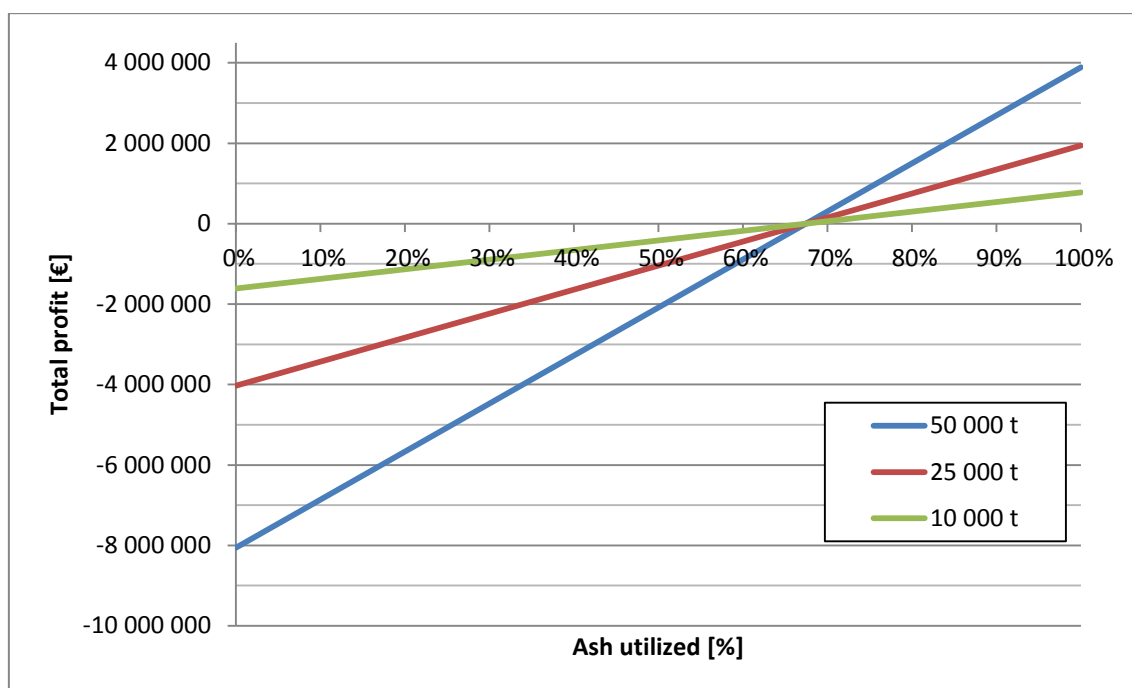

Figure 17. Total profit for 50 000 t, 25 000 t and 10 000 t annual dry ash amounts. Ash utilized percentage tells the portion of ash sold as forest fertilizer while the remaining portion is waste.

9 CONCLUSIONS

Currently a large fraction of the fly ash produced in heat and power plants ends up as waste while there could be a significant potential to utilize it in different applications. Utilization of fly ash is reasonable for both economic and environmental reasons since disposal of ash as waste is expensive, utilizing it reduces waste streams from industry and it can be used to replace use of non-renewable materials.

Dry and dusty fly ash has to be stabilized for efficient handling, storing and utilizing. The most effective stabilizing method is granulation. Currently used granulation techniques for fly ash are plate granulator and drum granulator. A new technique for granulation called tube granulation was presented in this Thesis. Formation of ash in combustion and theory of granulation, granule packing and drying were discussed as a background for tube granulation.

The tube granulation process takes advantage of the heat and high content of carbon dioxide in flue gas. Based on the literature it can be expected that granulation in the tube granulator is far more efficient than in conventional granulators since higher temperature and carbon dioxide accelerate the process. One of the objectives of this Thesis was to design the auxiliary equipment that is necessary in the complete tube granulation system. An example auxiliary equipment assembly was defined, however some parts of the system can be executed with different equipment and the optimal equipment assembly may vary in different cases. For example, a screw feeder and rotary feeders can be in the ash system, control dampers are needed in the flue gas system and possibly pumps and feed nozzles are required in the water feed system.

As the tube granulation process is a new invention there were no earlier experiences that could be used as a reference in the dimensioning. For this reason the dimensioning had to be approached from different perspectives. The examined approaches were based on a sufficient degree of filling in order to ensure a high rate of collisions of the ash particles, a required residence time for the chemical reactions to happen and achieving a desired degree of drying. By examining the process dimensioning from these

perspectives, it was noted that process values obtained from each approach differ from each other considerably. An exact dimensioning method could not be defined and the design values have to be chosen from a combination of these different approaches. These values can be used as a base for dimensioning the test equipment but it is necessary to do empirical testing in order to find out the optimal design parameters.

Implementation of tube granulation into a continuous power plant process was examined considering the connections of water and additives, flue gas and ash. Flue gas can be taken directly from the flue gas ducting of the main power plant process. If possible, connection points should be chosen so that the inlet connection is after a flue gas fan and the outlet connection is before the fan. This way the pressure difference over the flue gas fan can be taken advantage of and an additional fan may not be needed for the granulation process. Ash to the tube granulator can be taken from a fly ash silo or a separately installed storage tank. A feeding device such as screw feeder must be installed between the ash storage and the granulator. Water can be taken directly from the power plant's water network. Precise connection points depend on the power plant layout and they have to be designed case by case.

Economic benefits of granulating and utilizing fly ash were roughly examined from the ash producer's perspective. Example calculations were made by examining the savings earned from avoiding waste taxes and landfill site fees, differences in transportation costs and possible profits gained from selling the granulated product. It can be seen that the waste taxes and landfill site fees cause great costs for the producer of the ash and by far the biggest benefit in utilizing ash comes from avoiding these costs. It can be profitable to granulate the ash even if only a part of it can be utilized, especially if the amount of ash produced is high. Also transportation costs for granulated ash are lower than for dry or self-hardened ash since granulated ash can be transported by full articulated vehicles and less water is being transported than in self-hardened ash.

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