THE GRANUPURE PROCESS. NEW TECHNOLOGY FOR ENERGY EFFICIENT SEPARATION OF MELTS BY COMBINATION OF GRANULATION AND WASH COLUMN PURIFICATION

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3 Objectives

Melt crystallization is able to meet increasing requirements to the quality (purity) of products with moderate energy consumption owing to a high selectivity, low temperature and small heat of melting. These advantages are clearly exhibited on a laboratory scale, but in large tonnage production a high potential of melt crystallization cannot often be realized. In layer crystallization, because of low values of specific interfacial area, a high rate of crystallization is needed to be set. This results in a decrease in the efficiency of purification, an increase in the number of stages and the corresponding rise in the energy consumption. In suspension crystallization, conversely, problems arise because of too large interfacial area and are associated with the difficulty of the phase separation.

The objective of the present work is to develop a “hybrid” technology of crystallization purification which minimizes the drawbacks of layer and suspension crystallization. The method of solution is to apply a novel purification methodology consisting in the production of polycrystalline granules of a given size and optimum surface and in their further purification using different mechanisms of the impurity removal. The objectives resulting for the individual partners of the project from this general task will be outlined in the following section.

Task 1: Production and characterization of granules
One fundamental part of the Granupure Project was to produce pastilles. The aim was to optimise the pore structure of the granules formed on the cooling belt in relation to the ability to purify the granules using following purification steps. The narrow size distribution of the granules produced on SPS units and their almost spherical shape seems to be perfect especially for wash column technology. The relatively large specific surface area compared to a solid layer increases the effectiveness of the surface related purification steps.

The characterization of the pastilles were done by basic investigations regarding the purification of dropformed pastilles and the influence of the parameters during pastille production on the purification effect.

Task 2: Purification of granules

In order to perform this task the investigations had to be executed in two separate sets of experiments. The first one dealt with the influence of the pastillation parameters (impurity concentration of the melt, pastillation temperature, roughness of the pastillation surface) on the properties of the resulting pastilles (size, shape, internal crystalline structure). The second one on the other hand investigated the influence of these pastille properties on the post purification potential of the granules. To assess the purification potential of pastilles UB had to perform experiments in laboratory scale equipment. The investigated processes were crystallization, sweating, rinsing, and diffusion washing since the overall purification by a wash column process is achieved by a combination of all these steps.

The main objective of TNO in the Granupure project was to investigate the technical feasibility of the purification of granules for the following configuration: cooling belt (for the production of granules) - STR (Stirred Tank Reactor, for pre-purification of granules) and HWC (Hydraulic Wash Column, for the final purification of granules).

The work of UR had two main objectives, namely to provide a pre-purification step of solid pastilles made of a crude organic product and to concentrate the impurities of a solid residue stream from a purification process in order to recycle a small amount of the residue itself to the main process.

Task 3: Operation an modeling of wash columns

The mechanisms of purification which are put into effect in the processes of the washing and sweating of granules, including those with the use of physical methods of mass transfer intensification, had to be studied by KI. The purification experiments of the caprolactam, biphenyl and naphthalene granules varying in size from 2.5 × 0.5 to 4 × 4 mm from a number of different impurities were carried out in laboratory apparatuses. It is shown that mass transfer during washing in apparatuses of vessel and column types is limited by internal diffusion of impurities in the granule pores. The rate of purification in washing processes is determined by the magnitude of impurity diffusion coefficient, the size of granules and their residence time in apparatuses.

The research was focused on the operation and design of the Wash Column. For the purpose of the Gravity Wash Column (GWC) small scale experiments were required to obtain data of the purification rate as well as the hydrodynamic behaviour of pastilles countercurrently settling in a gently stirred wash column. Finally, in a pilot plant the cooling belt will be integrated with the GWC to obtain a first assessment of the Granupure process. Also the technical feasibility of the following configuration were investigated: cooling belt (for the production of pastilles) – STR (Stirred Tank Reactor, for pre-purification of granules) and HWC (Hydraulic Wash Column, for the final purification of granules).

Task 4: Integrated pilot plant tests

The final aim of the project was the development and testing of an integrated pilot plant in which the combination of a granulation unit and a wash column technique and /or a dry sweating unit is applied to an industrial important chemical.

4 Technical description

This section of the Publishable Final Report covers a detailed description of the techniques applied for the investigations and the chosen procedures on a task-per-task basis.

Task 1: Production and characterization of granules
The SPS granulation unit consists of a pastillator called ROTOFORMER and a cooling belt. The ROTOFORMER separates the melt into drops with uniform size and places them on the conveying steel belt. The bottom side of the steel belt is cooled in standard applications with sprayed water for cooling the pastilles below the melting point. The cooling time of the pastilles can be varied by the cooling length and the belt speed. The size and the shape of the pastilles will be generated by the ROTOFORMER- configuration in specific ranges depending of the melt properties. The wide range of possible belt sizes in width and length allows to reach nearly every customer-required capacity.
The cooling belt combines a high cooling rate per unit of cooling surface area with an excellent way to produce uniform, equal sized pastilles. The pastilles, typically “half spheres” and tuneable in the size range 1-10 mm, should facilitate a high throughput and stable operation of the GWC.

Task 2: Purification of granules
In order to perform the work described in Section 3 of this report UB first of all had to design and build laboratory scale equipment. The first piece of equipment built was a pastillation cell in order to simulate the granulation unit used by SPS. The pastillation cell allows the production of pastilles by a dropformation process onto a cooled surface. It enables the controlled setting and variation of the initial composition and temperature of the melt, the drop-off height, the roughness and temperature of the pastillation surface and of the ambient conditions namely the air temperature and humidity. For the production of the pastilles caprolactam was used as the main component containing water as the model impurity. The production parameters were examined at the following levels:

Initial temperature of the melt: 70°C
Drop-off height: 6 mm
Initial water concentration of the melt: 0.5%wt.; 1.0%wt.
Temperature of pastillation surface: 0°C; 10°C; 20°C; 30°C
Roughness of pastillation surface: rolled stainless steel; sandblasted stainless steel

The ambient conditions were kept constant throughout the experiments.

After pastille production their size, shape, and internal crystalline structure were assessed using an image analysis system.

For the investigation of the influence of pastille properties on their purification potential pastilles produced under different conditions underwent crystallization, sweating, rinsing, and diffusion washing experiments. The crystallization experiments were performed in a fluidized bed plant simulating the flow conditions prevailing in a wash column. Based on pre-tests, the process parameters during the crystallization experiments were set as follows:

Shape of the pastilles: pastille-like; egg-like
Impurity concentration of the melt: 3.0%wt.

The sweating, rinsing and diffusion washing experiments on the other hand were executed in jacketed beakers, using the parameter settings listed below:

Sweating temperature: 64°C
Sweating time: 30min; 60min; 90min
Rinsing temperature: 70°C
Impurity concentration of the rinsing melt: 0.3%wt.
Rinsing time: 5s; 10s; 15s

Diffusion washing temperature: 59.9°C...60.7°C
Impurity concentration of the diffusion washing melt: 2.7%wt.
Diffusion washing time: 30min

After each of these experiments the impurity concentration of the pastilles was measured in order to assess the purification effect.

The principle of the dry-sweating operation is to heat the solid to be purified by a hot gas stream to induce its partial melting. The effect of the loss of mass of the solid due to its melting is the reduction of the solid impurities since they are distributed preferentially in the generated melt. An effective dry-sweating apparatus requires: a high gas-solid heat transport rate and the detachment of the generated melt from the solid.

The developed technique is based on the use of a rotating equipment where solid particles, smaller than few centimetres in size, are heated up by a hot gas for a period of time in the range 10-30 minutes. the generated melt is suddenly separated from the solid particles due to the applied centrifugal force. The process allows a good purification of the organic solid and a strong concentration of the impurities in the molten phase residue stream. The ratio between the concentrations of the impurities in the residue and in the purified solid is up to 30.

The water impurity was detected by a Karl-Fisher titration instrument, whereas the content of the organic impurities was evaluated by means of a spectrophotometer.

Task 3: Operation and modeling of wash columns

The GWC (gravity wash column) is a cylindrical column with a slowly rotating stirrer and knife and relatively simple internals in order to control the axial dispersion. The column itself operates adiabatically. The pastilles that leave the column at the bottom are pumped through a melting circuit. The reflux at the bottom of the column is controlled by adjusting the product flow valve and the level of the crystal bed by the rotating knife.

The types of equipment are so called proven technology and the rules for scaling up are relatively straightforward.

TNO measured and evaluated the purification which occurred when impure caprolactam granules were brought into contact with a melt in a STR as a function of experimental parameters like the residence time, the composition of the granules and melt and the temperature. The STR was operated batch-wise as well as continuously.

In the second place TNO investigated the operation of a HWC for caprolactam granules and for an inert test system based on HMA (Hot Melt Adhesive) granules. The obtained results were compared to the operation of a HWC with caprolactam crystals.

Additional experiments regarding the effect of ultrasound of moderate power levels below the breakage threshold of granules showed little influence on mass transfer in small pores. Simultaneous effect of ultrasonic and thermal fluctuations stimulates recrystallization processes in the granules - wash melt system and appreciably increases the rate of purification (by a factor of 2 compared with that in usual washing in an apparatus with a stirrer).

Simple sweating of granules under conditions of free draining-out of liquid fractions at an acceptable product yield of 70 - 80 % gives a purification effect of only 1.5 - 2 because of insufficient separation of phases. The efficiency of purification during sweating can be enhanced by forced removal of liquid fractions under centrifugal forces or vacuum. The use of vacuum made it possible to combine sweating and distillation in a single apparatus with their simultaneous occurrence. The distillation sweating process developed in KI occurs in the region of the three-phase solid - melt - vapour equilibrium at a pressure close to the triple point pressure of a major component. In this case distillation plays an auxiliary role and is used for intense evaporation of a sweating liquid, which is impurity-enriched, and for removal of the vapour from the granule pores into a condenser (see Fig. 4.1).

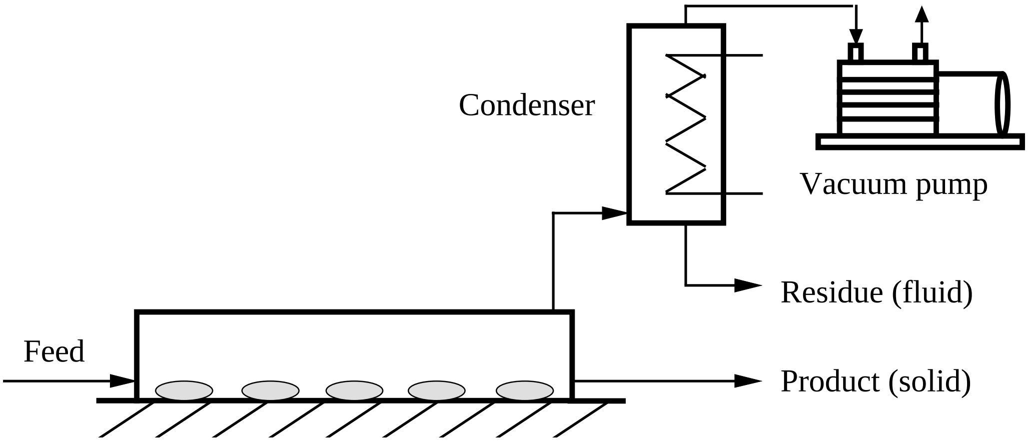

Figure 4.1: Schematic diagram of a combined process of distillation sweating.

Task 4: Integrated pilot plant

The integrated pilot plant consists of a Cooling Belt, a STR and a HWC. After creating pastilles optimised in size and shape with the ROTOFORMER / Cooler unit, the pastilles were fed to the STR for pre- purification and HWC for the final purification.

The purification of caprolactam granules in an integrated pilot was studied. The energy consumption for this configuration was evaluated and the scale-up criteria for the individual unit operations were defined. The attained purification for caprolactam belonging to the Granupure process option tested was compared to the purification measured in an earlier project for a suspension-based melt crystallisation process.

5 Results and conclusions

The present section will give a detailed overview over the results and the conclusions drawn based on the experimental findings.

Task 1: Production and characterization of granules

The partners agreed to caprolactam as the test product. This fulfils the request to use an industrially important chemical. The impurity compounds were a certain amount of water and dye sudan red. The dye was very helpful to get an optical qualitative indicator during the purification steps. The investigation concerning the pastille size and shape with respect to the optimization of the efficiency of the purification steps led to a “standard-size“ pastille. SPS provided all partners with pastilles in the required size, shape and capacity.

The analysis of the pastilles' size and shape represented by their height-to-diameter ratio revealed a neglectable influence of the initial composition of the melt and the roughness of the pastillation surface. The pastillation temperature, however, influences this parameter perceivably. The results show, that the largest height-to-diameter ratios can be obtained at a temperature of 10°C which can be explained by the counter acting influence of increasing crystal growth and increasing viscosity of the melt with lower temperatures. The internal crystalline structure of the pastilles shows the same behavior in such a way that the pastillation temperature has the dominant impact on it leading to finer crystalline structures for higher temperatures. An increased initial impurity concentration on the other hand leads to finer structures because of the reduced difference between the equilibrium composition and the temperature of the surface. The roughness of the pastillation surface reduces the effect of both, the pastillation temperature and the initial composition of the melt which is attributed to the hindered heat transfer for the case of the rougher surface resulting from the reduced contact area between the peaks of the surface profile and the melt.

Task 2: Purification of granules

Crystallization: The results of the crystallization experiments clearly show that the purification of the pastilles improves with increasing weight and height-to-diameter ratio of the pastilles. This effect can be explained by the increased relative velocity between the particles and the melt on the one hand and the more uniform flow around the egg-like shaped, symmetrical pastilles on the other.

Sweating: The sweating effect of pastilles revealed the typical behaviour as it is well known for single crystals produced by a suspension crystallization process. With increasing sweating time the purification effect improves logarithmically. From the results it can be further deduced that the internal crystalline structure of the pastilles is extremely important to the sweating effect. That is, the coarser the pastilles and the larger the pores the better the separation because the importance of capillary effects holding back the remolten impurities in the pores decreases with increasing pore diameter.

Rinsing: Generally speaking, the result of the rinsing experiments is very similar to that of the sweating runs. Here again the structure of the pastilles is of considerable importance to the purification result. However, the scatter of data is so large that clear differences between the results for different pastillation temperatures cannot be detected. The initial impurity concentration and the roughness of the pastillation surface on the other hand yield clearly distinguishable differences of the purification result leading to the conclusion that the amount of impurities located on the surface of the pastilles is the determining factor for the rinsing result which can be explained by the short residence times. Consequently, a dense structure, i.e. small spherical crystals or large crystals that are homogeneously oriented lead to the best separation because major parts of the impurity are not incorporated into the crystal structure but remain on the pastilles' surface.

Diffusion washing: In case of the diffusion washing experiments the experimental data again scattered to such an extent that a clear dependence of the purification effect as a function of the pastillation temperature could not be observed. However, when averaging over all four temperatures for every combination of surface type and initial impurity concentration distinct behaviours result which show the same tendencies as the ones found for the sweating experiments. This leads to the conclusion that also for the diffusion washing the internal crystalline structure of the pastilles is the factor which has the strongest impact on the purification result.

The following table (Tab. 5.1) qualitatively summarizes the results of the experiments of UB stating the process parameter settings which promise the best purification result:

Purification Process	Initial water concentration	Surface roughness	Pastillation temperature	Purification time
Crystallization	No influence	No influence	no influence	high
Sweating	Low	High	low	high
Rinsing	High	Low	no influence	high
Diffusion Washing	Low	High	low	high

Table 5.1: .... Parameter settings for optimal purification of pastilles

Experimental runs were carried out by using rotating apparatus at bench and pilot scale. The treatment was applied to pastilles of ε-caprolactam specifically prepared by SPS and on crude products from industrial plants. In particular two industrial crude products were examined:

ε-caprolactam (CPL) and benzil-dimethyl-ketal (BDK). Both the examined systems exhibit an eutectic point. The crude CPL contained water and organic compounds as impurities, whereas the crude BDK had only organic impurities. Nitrogen and air were used as hot gas in the CPL and BDK runs, respectively.

The typical results obtained from bench-scale experiments are reported in Tab. 5.2 and Tab. 5.3.

PRODUCT	**SHAPE**	**RUN-TIME** [min]	**$\Delta M/M$** [%]	**$k_{d\,H_2O}$**	**$k_{d\,ORG}$**
CPL	Pastilles 2x4 mm	20	15	0.05	0.30
BDK	Particles 1÷4 mm	30	16		0.16
BDK	Particles 1÷4 mm	20	16		0.10

Table 5.2: ....Dry-sweating runs on crude products

In Tab. 5.2 $\Delta M/M$ is the molten fraction of the original solid and k_d is the distribution coefficient of the impurity, that is the ratio between the estimated content of the impurity in the purified solid and in the original sample.

The purification of very crude products was also investigated. In such a case specific analyses were made near the laboratories of the industrial suppliers. The results are reported in Table 5.3 .

PRODUCT	**RUN-TIME** [min]	**$\Delta M/M$** [%]	**FEED**	**PRODUCT**	**RESIDUE**
			(Permanganate Number)		
CPL, very crude	10	24	600	8000	
			(Purity grade by gaschromatography).		
BDK, crude	20	14	97.3	99.97	
BDK, very crude	10	50	70.0	97.5	85.0

Table 5.3: .... Dry-sweating runs on industrial crude and very crude products

From the obtained results it can be observed that a very good purification is obtained in all the cases. The molten fraction is smaller than 20% of the solid in case of crude product. When very crude products are purified a higher molten fraction is required. The smaller recovery factor obtained in this latter case is justified by the low value of the product to be processed.

Results similar to those ones above reported were obtained by using the apparatus at pilot scale.

In conclusion the developed dry-sweating technique appear very promising to purify organic products. In particular, when very crude products, that is residue streams, are treated more than 50% of the original product is purified and can be recycled to the main purification process, and the impurities are very concentrated in the new residue stream. Therefore the technique can be considered an additional step of a purification section, which strongly increase the recovery factor of the overall purification process.

On basis of the results obtained by TNO it can be concluded that the pre-purification of granules in a melt using a STR is promising as a step preceding the (final) purification of granules in a HWC, which offers the following advantages:

- a STR is a simple (and relatively cheap) device, which offers an easy method for preparation of the feed suspension of the HWC (which should be done anyway);
- a substantial part (60-85 %) of the impurities can already be removed from the granules in the STR for residence times ≥ 45 minutes. Pre-purification in the STR reduces the required residence time in the wash column and lowers the separation duty of the wash column.

From the research on operation of the HWC it was concluded that the operation becomes easier in the sequence: caprolactam granules → caprolactam crystals → HMA granules. The extremely good results obtained with the ideally-shaped HMA granules reveal that operation of the HWC with granules can be very easy. The relatively difficult operation of the HWC for caprolactam granules is likely to be caused by the relatively flat shape of the caprolactam granules and/or the formation of fines due to breakage of the granules and/or unwanted crystallization due to under-cooling of the melt during preparation of the feed suspension in the STR.

Task 3: Operation and modelling of wash columns

In the combined process the rate of the impurity removal from granules increases by two orders of magnitude compared with that in the diffusion washing process. A rapid removal of sweating liquid favours the retaining of the shape of granules and prevents their breakage. Along with a high rate, the combined process has higher purification efficiency than that of individual processes of sweating and conventional distillation. For instance, during the distillation sweating of the caprolactam, naphthalene and biphenyl granules of 4×4 mm, the concentration of a number of impurities was lowered by a factor of 10 for 5 - 20 minutes at a yield of no less than 70 - 80 %.

The distinctive feature of the combined process is the possibility of an effective removal of not only impurities that are more volatile but also impurities that are several times less volatile than a major component. For instance, in the biphenyl - naphthalene system with relative volatility coefficient $\alpha_o = 5$, both the purification of the biphenyl granules from naphthalene impurity in amounts of 4 wt % and conversely were equally successful.

In KI theoretical fundamentals of distillation sweating have been developed which allow main operating parameters of the process, the achieved degree of purification, product yield and the required energy consumption to be calculated. The possibility of solving the problem of scale-up in multi-layer placement of granules on the heating surface is shown.

A simple mass transfer model, based on the structure of the pastilles, was combined with a hydrodynamic settling model with axial dispersion in order to quantify experimental results. This model was used to estimate the parameter sensitivity of the GWC operation.

In a labscale GWC operating under total reflux the relevant model parameters were experimentally determined. In this GWC also the operating bottlenecks could be identified. These tests were executed with 1.8 mm caprolactam pastilles containing 1 wt% of water. The purification rate constant, $k_p=2.5\text{x}10^{-3}$ s^{-1}, the non-removable impurity fraction, $x_{eq}=0.35$ and the axial dispersion coefficient, $D_{ax}=10^{-5}$ m^2 /s. The measured production capacity was 0.22 ton/m^2/hr which was limited by the submersion rate of the pastilles at the top of the column

It was found that the purification for the combination STR-HWC was significantly better than the purification measured for the STR alone, which proves that additional purification occurs in the HWC. More than 99 percent of the model impurity Sudan Red was removed when the combination STR-HWC was applied. In that case the HWC was operating under total reflux. The thus-obtained purification efficiency for the combination STR-HWC is comparable to single step suspension-based melt crystallization processes (and better than that for single-step layer based melt crystallization processes).

It was concluded from the integrated pilot plant tests with the combination Cooling Belt - a (batch-wise operated) STR and a HWC that this Granupure process option is (experimentally) difficult to operate for caprolactam. The underlying reasons are: (1) the formation of fines due to breakage of granules and/or unwanted crystallization and (2) the relatively flat shape of caprolactam granules. It is expected that less practical problems will occur when the STR is operated continuously, which is the preferred way of operation in an industrial process but not feasible for the pilot plant tests at the studied scale. The relative flatness of the caprolactam granules is among other things caused by the relatively low viscosity of the melt. This leads to the expectation that the potential for application of the Granupure process is better for compounds with a viscosity of the melt higher than that for caprolactam (which is about 15 mPa.s). Despite of all practical difficulties it was proven experimentally that operation of the Granupure process for caprolactam with a Cooling Belt, a STR and a HWC is possible. TNO analysed the energy consumption for the combination STR-HWC and defined the scale up criteria for the following three cases:

1. ‘Direct’ purification: 99 wt % caprolactam granules as feed; product 99.95 wt %; yield ≥ 80%;
2. ‘Pre-purification’: 99 wt % caprolactam granules as feed; product 99.8 wt %; yield ≥ 90%;
3. ‘Final purification’: 99.8 wt % caprolactam granules as feed; product 99.95 wt %; yield ≥ 90%;

It was concluded that ‘Final purification’ is the preferred application of a HWC, but application of the combination STR-HWC for ‘Direct’ purification and ‘Pre-purification’ purposes seems to be feasible for the typical caprolactam feed streams found in industry.

In summary, the conclusions that can be drawn from TNO’s activities in the Granupure project are:

- The essential condition for successful application of the Granupure process, which is that the physical processes during granulation result in an inner structure of the granules which enables purification, is proven by the experiments,
- Operation of the Granupure process for the purification of caprolactam using the combination STR-HWC is rather difficult, but technically possible. It is expected that the advantages, i.e. the chance on successful application, of the Granupure process increase with increasing viscosity of the melt.

Task 4: Integrated pilot plant

The two pastillation units were assembled in the laboratories of the corresponding partners for the integrated pilot plant tests. In the last period of the GRANUPURE-Project the Integrated-Pilot-Plant was running and gave the data for scaling up to industrial sizes. The results showed, that combinations of the SPS granulation unit with the HWC, the GWC and also with the dry-sweating unit are not only possible but also useful.

Scale up rules both on the micro-scale of the pastille as on the macro-scale of the GWC were used to built the integrated pilot plant. This pilot plant was tested and it was shown that the specific throughput increased to 0.7 ton/m^2/hr, which is 70% of the settling capacity.

A similar procedure was developed to design an industrial purification plant using the pastille size and the column size as the main design variables. This design procedure was tested on the purification of 100 kt/yr of caprolactam pastilles. The main results are an optimum pastille size, d_p=3.3 mm, and a GWC with a diameter, d_{col}=2.95 m and a height H=6 m. The energy consumption of the system exists of cooling energy for the cooling belt, $Q_{cooling\ belt}$=-600 kW_{th}, melting energy for the GWC, Q_{melter}=550 kW_{th} and stirring power for the GWC, P_{stir}=40 kW_{el}.

The main overall conclusion is that the combination of a cooling belt with a GWC provides an interesting option for ultra-purification of organic bulk products. An advantage of the process lay out is apart from the low energy demand the relatively simple process equipment.

5 Exploitation plans and anticipated benefits

The exploitation plans of and the benefits anticipated by the individual partners are in the focus of the subsequent sections.

SPS

SPS is interested to find new applications for the steel belt technology. The granulation is a great demand of the chemical industry. The possibility to use the granulation technology also for purification steps increases the field of working. The potential market for the GRANUPURE process is the market where up to now the melt crystallisation is established. Together with the partners we are in contact with potential users from the chemical industry. SPS will also inform own customers who are dealing with bulk chemicals not only in granulation but also in purification.

UB

The investigations conducted by UB basically have a fundamental character. For this reason a direct exploitation of the results as well as an achievement of benefits by UB itself is not possible. However, UB will make its results available to the project partners and any third party thus enabling the specific optimization of both processes, the production and purification of polycrystalline particles. Part of the results obtained during the project will be published in a PhD thesis.

UR

The developed technique can be predominately applied to the following process:

- Purification of the residue stream from the purification section of a caprolactam plant, typically the evaporation section
- Recovery of crude BDK from the final residue of the BDK production plant.

Accordingly, the dry-sweating technique is proposed for application in the industrial plant for the production of ε-caprolactam and benzyl-dimethyl-ketal.

In addition the dry-sweating process is proposed for R&D studies finalised to the purification of an organic eutectic system.

TUD

TUD will advocate the application of the Cooling Belt/GWC combination by publications and conference contributions. The research will be continued till the end of 2000. The technical results supported by a scientific background will be published in a PhD dissertation.

The pilot plant facilities at TUD with a typical production capacity of 2 kg/hr are available for tests for the process industry. These tests will be executed in close co-operation with SPS.

TNO

TNO and SPS will promote the Granupure process by means of publications/presentations and their marketing contacts with potential end-users in the chemical industry. TNO and SPS can carry out technically feasibility studies for interested end-users with an integrated pilot plant consisting of a cooling belt, a STR and a HWC. The next stage after a successful technical feasibility study is development and testing of a (transportable) demonstration unit on semi-industrial scale. The final stage in the RTD trajectory, which follows after the tests with the demonstration unit, is design, construction and operation of an industrial installation. The first industrial installation is very important because of the acquirement of the status of “proven technology”. The anticipated benefits for the Granupure process are: lower costs and/or improved product quality and/or energy saving compared to conventional/competing technology. Financial support could be helpful/required for execution of the described exploitation trajectory.

KI

The process of distillation sweating can be used on an industrial scale in the step of additional (preliminary) purification combined with granulation and washing in a counter-current column. The area of industrial application is the purification of substances that have the triple point pressure $P_{t.p}$ higher than 1 mm Hg. A fairly large number of organic compounds have such a saturated vapour pressure (anthraquinone - $P_{t.p} = 100$ mm Hg, anthracene - 40, benzene - 36, dimethylterephthalate - 10, acetic and salicylic acids - 10, p-dichlorobenzene - 9, naphthalene and phthalic anhydride - 8, p-xylene - 4, phenol - $P_{t.p} \approx 1$ mm Hg).

A high selectivity of distillation sweating allows a good purification to be achieved with a lesser fraction of the substance undergoing phase changes and accordingly lesser energy consumption than those in conventional processes (see Tab. 6.1).

No	Purification process	α_{eff}	Q [kJ/kg]
1	Simple sweating	3 - 3.5	220
2	Sublimation	3.5 - 4	650
3	Conventional distillation	4 - 5	540
4	Dynamic layer crystallization	5 - 6	450
5	Distillation sweating	9 - 11	130

Table 6.1: Effective separation factors α_{eff} and energy consumption Q on phase changes in different processes.

The biphenyl - naphthalene system. The energy consumption is given per 1 kg of product for a tenfold purification.

To carry out the process, vacuum apparatuses used in industry for sublimation or drying can be employed. Additional savings in energy consumption are possible if the combined purification process in the region of the three-phase equilibrium is applied directly in the granulation step.

7 Photograph, diagram or figure to illustrate potential applications of the project

The following two photographs show the integrated pilot plants set-up at TUD and TNO:

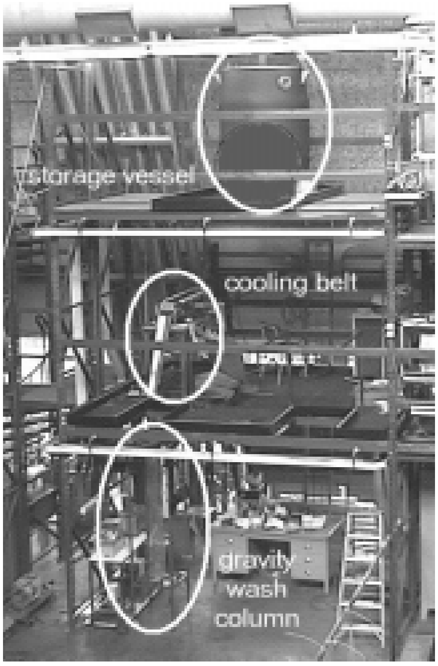

Figure 7.1:

An overview of the TUD integrated pilot plant is given in the figure showing the 200 liter storage vessel, the 0.2x1 m^2 cooling belt and the 0.075x1.5 m^2 GWC.

From the temperature controlled vessel, located at the top floor the melt is pumped to the feeding block located at the start of the cooling belt. The size and number of pastilles that are formed are controlled by the stroke and the frequency of the membrane feed pump and the number of holes in the feeding block. The rotation speed of the cooling belt is adjusted to the feed system in order to obtain a uniformly covered belt. At the end of the belt a scraper takes of the pastilles. The pastilles are “launched” by the high angle gutter into the pilot GWC situated on the ground floor.

The GWC consists of three glass tubes. This lay out is necessary to ensure that the wash column operates adiabatically.

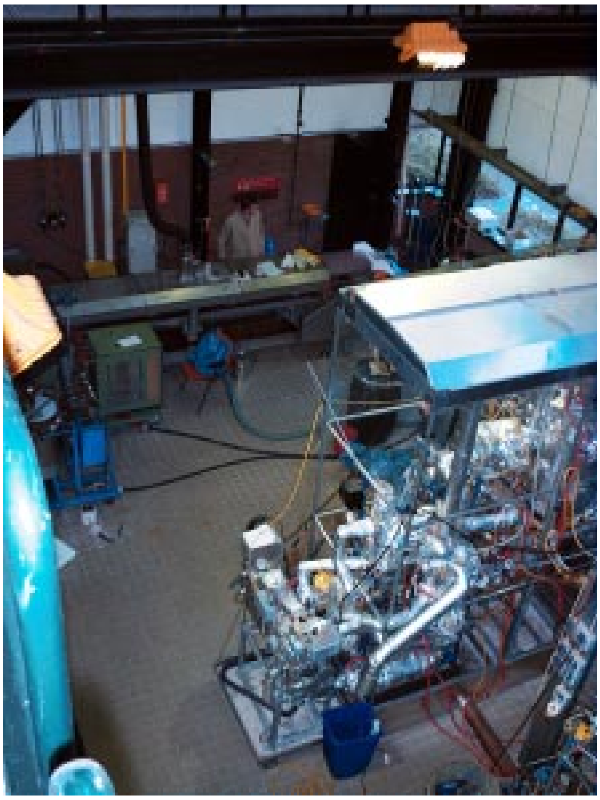

Figure 7.2:

Overview of the integrated pilot plant for technical feasibility studies for the Granupure process at TNO premises in Apeldoorn. The cooling belt can be seen at the background and the HWC pilot plant (including the STR) is situated at the left hand front side.