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# Instrumented Compaction Experiments under Varied Climatic Conditions

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**Abstract**

**From empirical observations in industrial practice it is known that compaction behaviour and quality of uniaxial pressed ceramic compacts may be significantly influenced by climatic conditions during granulate storage and processing. At the same time, the type of basic material, added binders and lubricants can play a major role in most cases. In the present study, an attempt was made to reach a better understanding of the interactions between climatic conditions and compaction behaviour of silicon nitride granulates with different binder/lubricant systems by using instrumented compaction experiments. In addition to the continuous measurement of the pressure-density development and the strength of the compacts, parameters are measured which characterize the friction conditions, the total compaction energy and their single parts, the elastic redeformation in axial and radial directions. From the calculation of shear and compressive stresses, conclusions can be drawn regarding acting failure mechanisms and predictions of the expected density distribution can be given. For the examined silicon nitride granulates a deleterious influence of moisture on the whole pressing process and the defect rate of the compacts after storage of granulates at a humidity of more than 50 % before compaction was verified. For this material the changes in compaction behaviour are not reversible after storage at lower humidity.**

*Keywords: Instrumented compaction, friction, silicon nitride, granulate, climatic conditions*

## I. Introduction

The pressing of powders or granulates is the most common and economical method for forming ceramic components. Thereby the properties of the "system granulate" with all its components such as basic material and types and concentrations of added binders and lubricants play one of the most decisive roles for the quality of the ceramic product. The problem is that these properties can be affected and changed by several undefined or uncontrolled external influences such as climatic conditions. The temperature, for example, may change the deformation properties of the binder drastically depending on the glass transition temperature of the binder[1]. Yet the moisture of granulates and their changes with relative humidity is as important as the temperature. For the pressing of silicate ceramic granulates, it is known that, up to an optimum value, higher moisture contents affect the total compression behaviour in a favourable way[2]. In contrast to this, the authors have various references, which result from cooperation with industrial partners in the field of granulate development, indicating that in advanced ceramics the relations are far more complicated. Some characteristic comments may illustrate the frequent situation:

- All parameters for conditioning, granulation and compaction were kept constant, but from one day to the next granulates cannot be compacted.
- Suddenly, we have problems with the occurrence of macroscopic defects such as end-capping defects or laminations in the green body.
- Granulates must be sprayed during winter time, then we have no problems during compaction.
- Granulates must be stored for some days before compaction with the container being left open.
- Sometimes it must be accepted that there is no suitable "press weather".

From industrial practice, stringent advice on the serious effects of seasons on the manufacturing process based on uniaxial pressing and on the quality of materials has also been published[3].

The intention of the present study was to attempt to get over the predominant empiricism in this field and to reach a better understanding of the interactions between climatic conditions and compaction behaviour for the case of silicon nitride with varied binder/lubricant systems.

## II. Experimental Procedure

Suitable measuring equipment is the most important requirement for a precise investigation of the processes during compaction. This means that not only compaction response diagrams must be recorded but it must also be possible to conduct a survey on the friction conditions, the elastic spring-back, the compaction energies and the distribution of the stresses from the measurements. For

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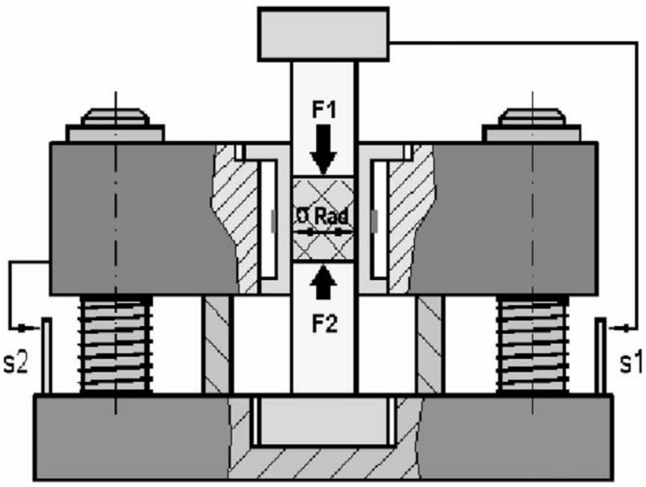
these experimental investigations a self-made instrumented pressing tool according to the schematic view in fig. 1 was used[4, 5].




$F_1$, $F_2$...Forces at the upper resp. lower punch; $s_1$ ...Way of the upper punch; $s_2$... Way of the die (optional); $\sigma_{Rad}$... Radial stress

**Fig. 1:** Schematic view of the instrumented compacting tool.

Primary measured parameters are the forces at the upper and the lower punches, the radial stress on the die wall and the distance covered by the top punch during compaction. Die and bottom punch are fixed. The instrumented compacting tool is installed in a material testing machine and allows, depending on the diameter of the cylindrical die, measurements up to a pressure of 600 MPa. For the investigation of possible influences of the die, different instrumented inserts made of hardened steel, CPM-steels or carbide with different roughness are available.

The total equipment is installed in a laboratory in which humidity and temperature can be adjusted in a wide range. The conditions comprise a range from 15 %-80 % for the relative humidity and 16 °C-28 °C for the temperature in the laboratory.

From the primary measured parameters, a system of specific compaction parameters can be derived (fig. 2). This allows an estimation of the processes during compaction, an evaluation of the efficacy of organic additives and a prediction of the quality of the compact mainly with respect to the causes of the development of typical macroscopic pressing defects such as end-capping and laminations.

| *Parameters of the compact* | | *Elastic relaxation* | |
|---|---|---|---|
| $\rho_1$ | Density at pressure | $\Delta_{Tot}$ | Total axial relaxation |
| $\rho_G$ | Geometrical Density | $\Delta_i$ | Part inside the die |
| $\sigma_{Dia}$ | Diametral compressive strength | $\Delta_{ej}$ | Part at and after ejection |
| | | $\Delta_d$ | Radial relaxation |
| *Specific friction parameters* | | *Compaction energies* | |
| $\mu_w$ | Wall friction coefficient | $A_1$ | Theoretical energy |
| $\mu_p$ | Powder friction coefficient | $A_2$ | Total compaction energy |
| $\eta$ | Radial stress coefficient | $A_3$ | Relaxation energy |
| $F_2/F_1$ | Force transmission quotient | $A_4$ | Friction energy |
| $F_{ej}$ | Ejection force | $A_5$ | Consumed energy |

**Fig. 2:** System of specific compaction parameters.

In each case eight samples are examined, the specific parameters are averaged. The sample with the best correlation to the average is taken as the basis for the dynamic curves, for example fig. 10: "Loading and unloading curves after different storage conditions (PVA-type)".

In addition to these parameters, distributions of compressive and shear stresses as well as density can be calculated according to a model by Thompson[6], modified by results of own investigations[7]. From typical compaction curves, analyses are also conducted to determine the occurrence of stick-slip-mechanisms during compaction, the mobility of the lubricant dependent on the pressure, the plastic and elastic parts of deformation during compaction of the granulates and later during measurement of the green strength.

The starting material was a commercially available silicon nitride powder with an average particle size of 0.9 µm and 11 % sintering aids. Two types of binders, one from the group of polyvinyl alcohols and one from the acrylic emulsion group, were added in equal concentrations. A ready-for-use emulsion of a salt of a fatty acid acted as the lubricant. The compaction behaviour of granulates was examined immediately after the spray granulation process. Afterwards the granulates were stored in a laboratory atmosphere of 15 %, 30 %, 50 %, and 75 % relative humidity at a constant temperature of 22 °C until a constant moisture was reached and then they were pressed again. In each case the standard pressure was 200 MPa.

## III. Results and Discussion

### *(1) Influence of Humidity on the Compaction Behaviour*

#### (a) *Change of Moisture*

The change of moisture in granulates by absorption resp. desorption occurred in accordance with results published by Whitman[8]. The conclusion that a granulate with an added polyacrylate as the binder component shows a lower affinity for a water absorption than the polyvinyl alcohol is verified by the measurements shown in fig. 3.

| Storage at rel. humidity | Moisture content in the granulates, % | |
|---|---|---|
| | Polyacrylate | Polyvinyl alcohol |
| As-sprayed | 0,44 | 0,51 |
| 15 % | 0,40 | 0,46 |
| 30 % | 0,48 | 0,58 |
| 50 % | 0,52 | 0,72 |
| 75 % | 0,71 | 1,02 |

**Fig. 3:** Change of granulate moisture on storage under varied climatic conditions.

It seems to be of special interest for the compaction behaviour that both types of granulates exhibit a shift to higher moistures already after a storage in a relative humidity of only 30 %.

#### (b) *Properties of the Green Compact*

Continuous compaction diagrams were recorded up to a pressure of 200 MPa. The resulting densities at the endpoint show that increasing moisture always leads to a higher compressibility of granulate when a polyvinyl alcohol is added, as can be seen in fig. 4.

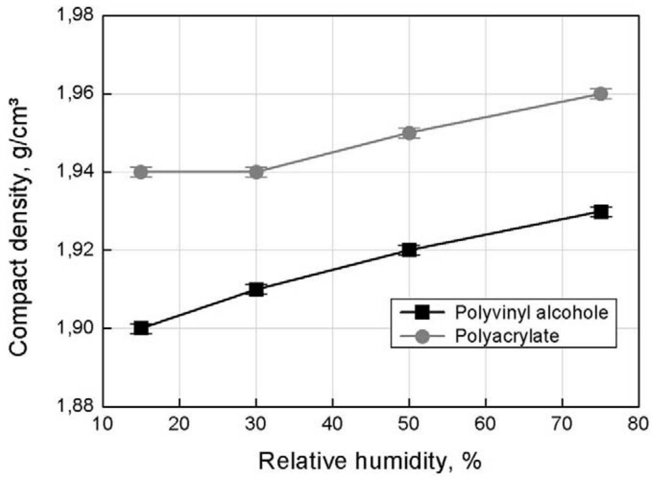



**Fig. 4:** Dependence of the density on storage conditions.

The initial value of the density obtained with pressing in the as-sprayed condition was 1.90 g/cm³ for this type. The acrylic-bonded granulates exhibit no changes compared to the basic density of compacts after storage at 15 % and 30 % relative humidity. Only at higher humidity can an increase of the density be noted.

Green strength was measured with the diametral compression test. In this test the compact is loaded on the cylindrical surface up to fracture. From the appearance of the fracture, conclusions can be drawn about the existence of latent macroscopic defects in the compact. In comparison to the basic strength of compacts pressed from the as-sprayed granulates (in both cases $\sigma_{Dia}$=1.33 N/mm²), each absorption of moisture leads to decreasing strength (fig. 5). Obviously, the binding force of the additives is negatively influenced by the moisture.

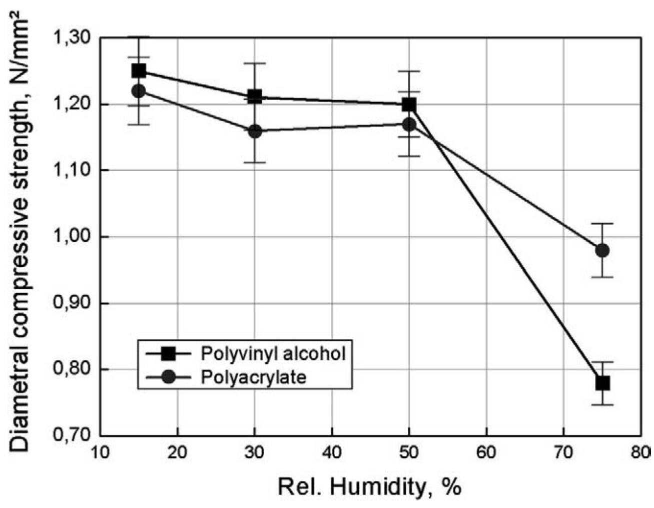



**Fig. 5:** Dependence of the green strength on storage conditions.

The strength of compacts pressed from granulates stored at humidity higher than 50 % is dramatically lower than that of those stored under drier conditions. Moreover, the fracture patterns revealed no straight plane fractures, as caused by exclusively acting tensile stresses, but showed distinct end-capping and lamination defects instead. Such granulates must be characterized as unsuitable for the compaction process.

(c) ***Frictional Parameters***

Serious changes also occur with respect to the friction conditions depending on the moisture of granulates. No inhomogeneities could be observed in the curves of loading, pressure holding, unloading and ejection for original granulates and those which were stored below a humidity of 50 %. Higher humidity in the environment of granulates causes characteristic stick-slip-mechanisms during compaction, which were frequently accompanied by characteristic noises. Typical examples for granulates containing polyvinyl alcohol are given in fig. 6.

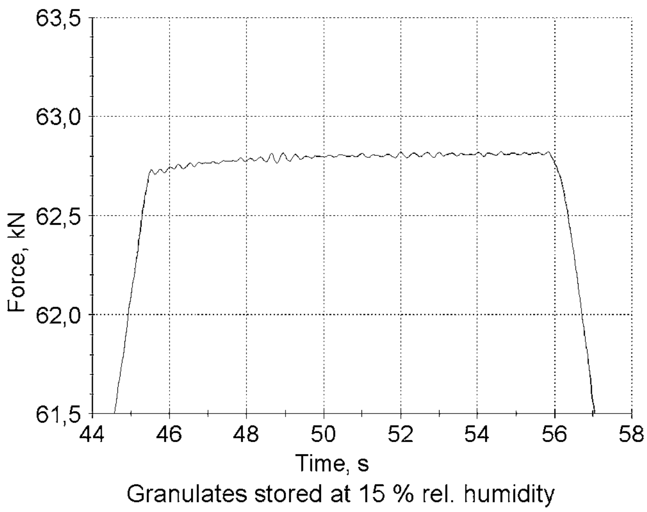



**Fig. 6a:** Forces at the upper punch during pressure holding time, granulates stored at 15 % relative humidity.

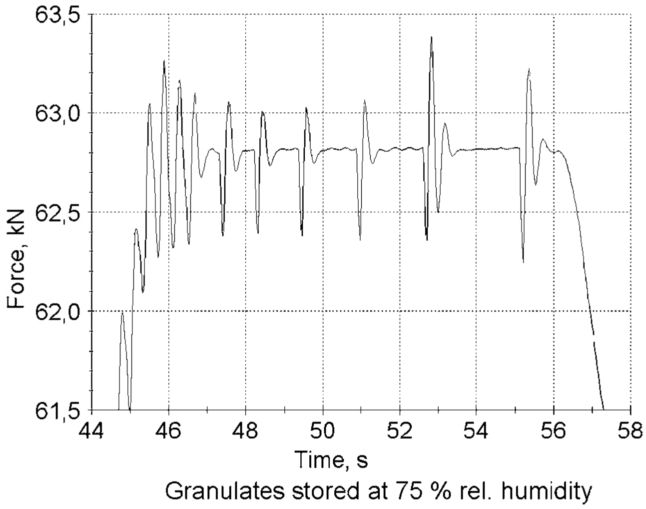



**Fig. 6b:** Forces at the upper punch during pressure holding time, granulates stored at 75 % relative humidity.

Granulates with polyacrylate behave in the same way after identical storage. It is necessary to avoid these mechanisms in respect of the surface quality of the compacts and the wear of the die.

A view of the changes of the axial force transmission quotients at different storage conditions shows clearly that the efficacy of the lubricant is affected by higher moistures in the granulates (fig. 7). Apparently, build-up of a uniform stable lubricant layer is not possible in such cases.

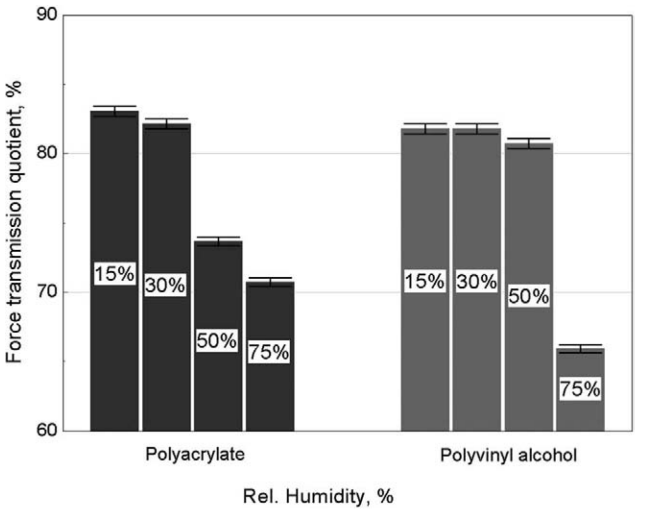



**Fig. 7:** Influence of storage conditions on the axial force transmission.

Yet, the degree of the influence is clearly dependent on the binder type. While, with the polyvinyl alcohol, the force transmission quotient after the storage of granulates at 50 % relative humidity is diminished to a very limited extent, the quotient decreases considerably on compaction of the polyacrylate type. As a consequence, storage at higher humidity leads to still higher losses of pressure transfer to the lower punch for both binder versions.

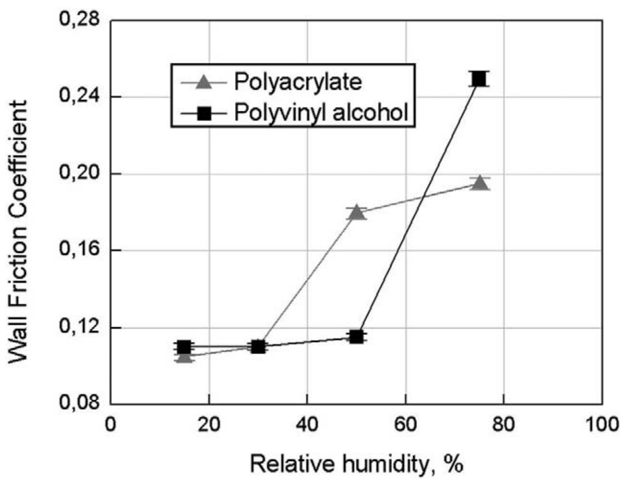



**Fig. 8a:** Wall friction coefficients at varying storage conditions.

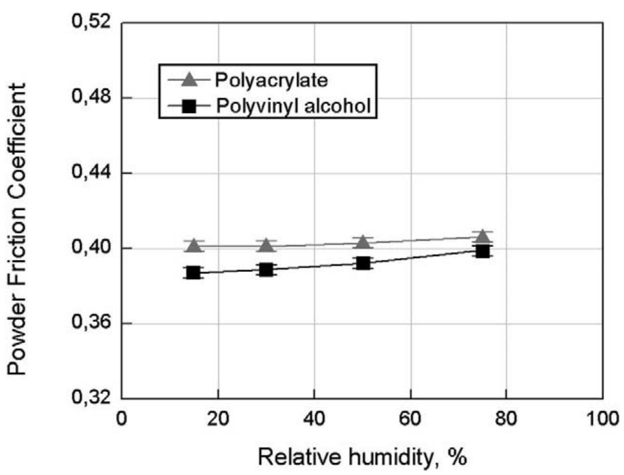



**Fig. 8b:** Powder friction coefficients at varying storage conditions.

The results gained for the wall friction coefficient (fig. 8) should give enough reason to reconsider our knowledge on the influence of moisture. The information obtained from the compaction of silicate ceramics, in which higher moisture is in principle beneficial for the pressing process and the properties of the compact cannot be uncritically transferred to advanced ceramics. After storage in up to 50 % humidity, the basic wall friction coefficient of the as-sprayed type with polyvinyl alcohol remains almost unchanged. Interestingly, the combination of the acrylic binder with the lubricant proved to be more sensitive again and shows a steep rise of friction in the region of 50 % humidity.

The dependence of the powder friction coefficient on the moisture is not as distinct as with silicate ceramics (fig. 8). The possibilities for changing the powder friction for advanced ceramics are very limited. An exception has proven to be a specific modification of the powder surface, for instance by silanisation[9].

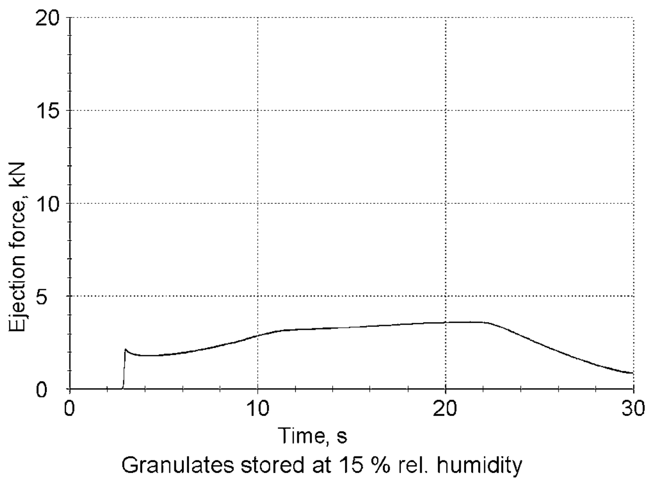



**Fig. 9a:** Courses of the ejection forces in static and sliding friction regions (PVA-type), granulates stored at 15 % relative humidity.

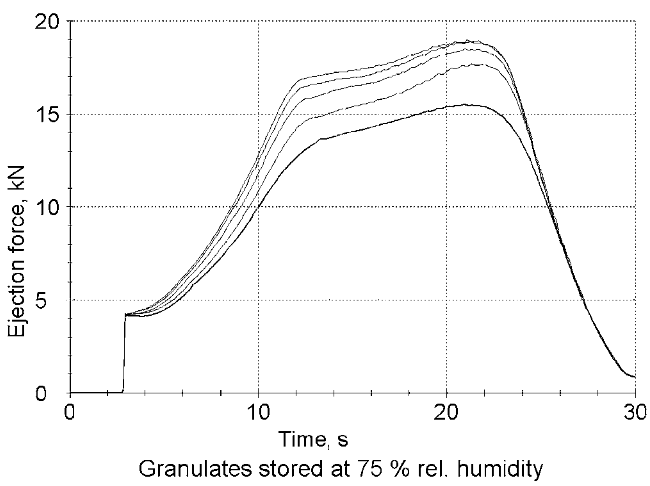



**Fig. 9b:** Courses of the ejection forces in static and sliding friction regions (PVA-type), granulates stored at 75 % relative humidity.

An affirmation of all previous results is given by the course of the dynamically measured ejection forces (fig. 9). These reduplicate for the static friction and reach a four-times-higher amount in the sliding region when the granulates stored at 15 % and the ones stored at 75 % relative humidity are compared.

These characteristic changes of ejection forces are independent of the binder type and take place for acrylic and polyvinyl alcohol in the same way. After a dry storage of granulates the results are reproducible at a low force level. Wet granulates require high forces and show, from one experiment to another, the phenomenon of increasing forces in the sliding region. This is caused by an abraded powder layer at the die wall until an equilibrium in the size of the gap between punch and die wall is reached.

### (d) *Elastic Relaxation*

An explanation of the causes for pressing failures can also be derived from the analyses of the proportions of the elastic relaxation during unloading as well as during and after ejection. In fig. 10, typical loading-unloading-graphs with characteristic differences between the compaction of dry und wet granulates are shown.

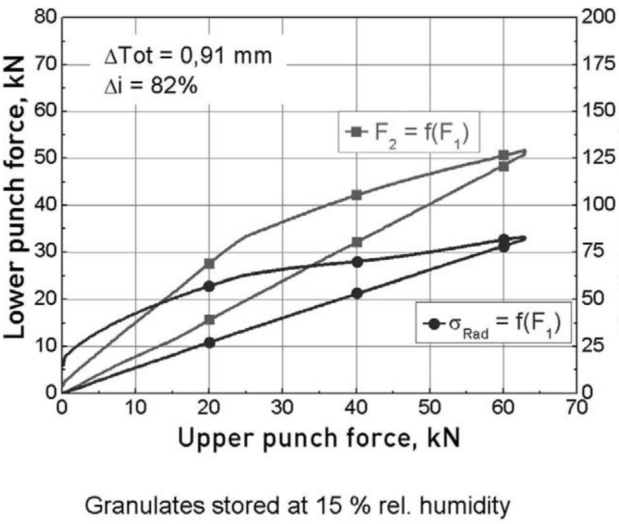



**Fig. 10a:** Loading and unloading curves after different storage conditions (PVA-type), granulates stored at 15 % relative humidity.

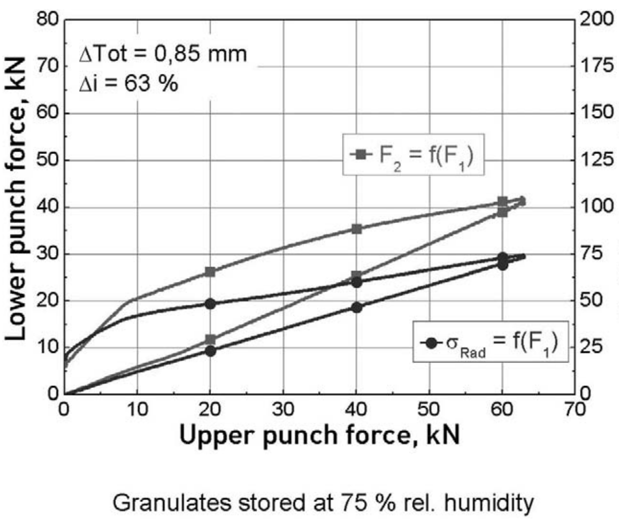



**Fig. 10b:** Loading and unloading curves after different storage conditions (PVA-type), granulates stored at 75 % relative humidity.

The original as well as granulates stored under dry conditions exhibit a higher amount of total axial relaxation. In both cases, a clear and early breakaway of the compact from the die wall at unloading is significant (fig. 10). For the wet granulates, this breakaway happens later and to a lesser extent. Consequently, the rate of axial relaxation, which takes place already inside the die, decreases if the moisture content in granulates is on a higher level. This means that the green body still contains more elastic stresses when it reaches the top edge of the die. The additional possibility of radial expansion then leads to the observed end-capping-defects and laminations if compacts are pressed from granulates previously stored at higher humidity. Naturally, the radial expansion is larger in all such cases.

### (e) *Compaction Energies*

Changing moistures influence the deformability of granulates, too. From the system of energies can be derived that with higher moistures, the elastic deformations increase up to the specified pressure. The force-way-graph proceeds considerably steeper in comparison to the dry granulate, which results in lower total compaction energy (fig. 11). Therefore, the region of "over-pressing" is reached at lower pressure. To exceed this pressure limit is

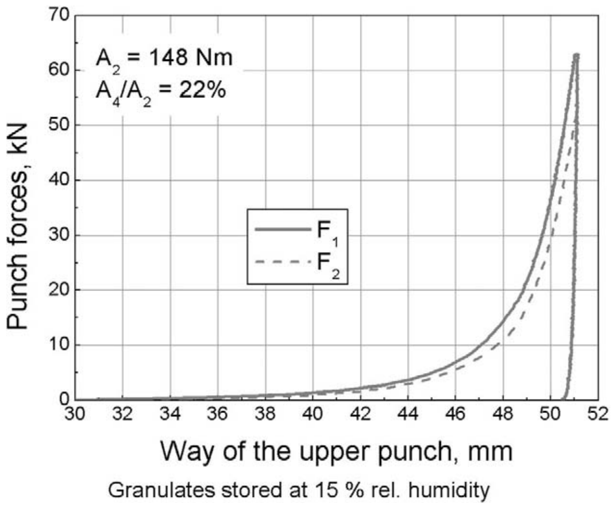



**Fig. 11a:** Characteristic force-way-graphs (PVA-type), granulates stored at 15 % relative humidity.

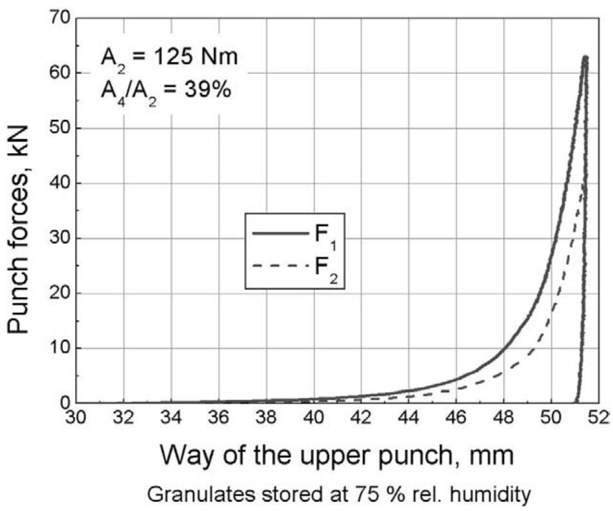



**Fig. 11b:** Characteristic force-way-graphs (PVA-type), granulates stored at 75 % relative humidity.

one of the most important reasons for the development of compaction failures.

As it could be expected from the friction coefficients, the part of energy losses also becomes larger with higher moisture of granulates. Again, granulate with the acrylic binder is more affected by this mechanism.

### (f) *Distribution of Pressure and Shear Stresses*

The calculated stress distributions support all previously made statements on the influence of moisture changes. The pressing of granulates with increasing moisture leads to a doubling of shear stresses for granulates with polyacrylate as the binder and somewhat more even when a polyvinyl alcohol is added (fig. 12).

| Type of Granulate (Binder and storage) | Max. axial pressure gradient, MPa | Max. radial pressure gradient, MPa | Max. shear stress MPa |
|---|---|---|---|
| PVA, 15% rel. humidity | 72 ± 0,4 | 39 ± 0,3 | 12 ± 0,1 |
| PVA, 75% rel. humidity | 134 ± 0,7 | 75 ± 0,5 | 26 ± 0,2 |
| PAc, 15% rel. humidity | 67 ± 0,4 | 35 ± 0,2 | 11 ± 0,1 |
| PAc, 75% rel. humidity | 116 ± 0,6 | 64 ± 0,4 | 22 ± 0,2 |

**Fig. 12:** Distribution of stresses in compacts pressed of different granulates.

The binders are not able to compensate for these stresses and defects will develop, most of which appear in the form of end-capping. In the same way, the differences in the axial distribution of pressure stresses increase, resulting in larger density gradients. A graphic example for compacts pressed from silicon nitride granulate with PVA-binder, stored under different climatic conditions is given in fig. 13.

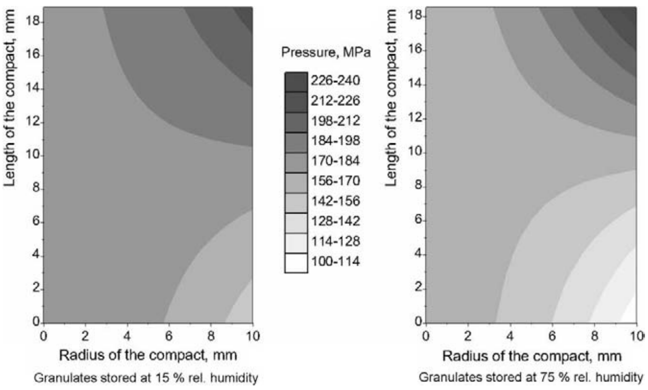



**Fig. 13:** Distribution of pressure in compacts pressed of different stored granulates.

### *(2) Influence of Cycled Climatic Conditions*

A simple example can illustrate the problems that may arise if climatic influences act on granulates in an uncontrolled manner.

In fig. 14, the results of instrumented compaction experiments with two parts of a granulate charge are summarized. These parts originated from the same barrel. Therefore, the type and concentration of organic additives as well as the granulation parameters were identical.

An examination of the moisture of both granulates immediately before the beginning of the compaction showed nearly the same contents, also. In short, the experiments were conducted with identical granulates according to conventional understanding. Yet, the system of specific compaction parameters showed surprising results. This concerns not so much the compact density, but its strength in a serious manner. The numerical value was not solely influenced, but the break patterns after the measurement of the strength were also affected drastically. For compacts pressed from part 1 of the granulate, in fig. 15 a straight break can clearly be seen. For compacts from part 2, a typical end-capping-defect combined with distinct laminations is visible. This granulate must be considered as not pressable.

| PROPERTY | | PART 1 | PART 2 |
|---|---|---|---|
| Moisture of granulate | [%] | 0,55 | 0,56 |
| Compact properties | $\rho$ [g/cm³] | 1,90 ± 0,001 | 1,92 ± 0,001 |
| | $\sigma_{Diam.}$ [N/mm²] | 1,33 ± 0,055 | (0,98 ± 0,040) end-capping |
| Friction | $F_2/F_1$ [%] | 82 ± 0,4 | 61 ± 0,3 |
| | $\mu_W$ | 0,110 ± 0,0009 | 0,278 ± 0,0021 |
| | $\mu_P$ | 0,390 ± 0,0013 | 0,391 ± 0,0013 |
| | $F_{Ej}$ [kN] | 2,2 ± 0,08 / 3,9 ± 0,15 | 5,5 ± 0,21 / 19,8 ± 0,7 |
| Energy | $A_2$ [Nm] | 147 ± 2,0 | 144 ± 2,0 |
| | $A_4$ [Nm] | 19 ± 0,5 | 44 ± 1,2 |
| Relaxation | $\Delta$Tot [mm] | 0,94 ± 0,018 | 0,86 ± 0,017 |
| | $\Delta$i [%] | 83 ± 3,3 | 60 ± 2,4 |
| Stress distribution | $\Delta\sigma_{ax}$ [MPa] | 70 ± 0,4 | 152 ± 0,8 |
| | $\tau_{max}$ [MPa] | 11 ± 0,1 | 32 ± 0,3 |

**Fig. 14:** Results of measurements of two parts of one and the same granulate.

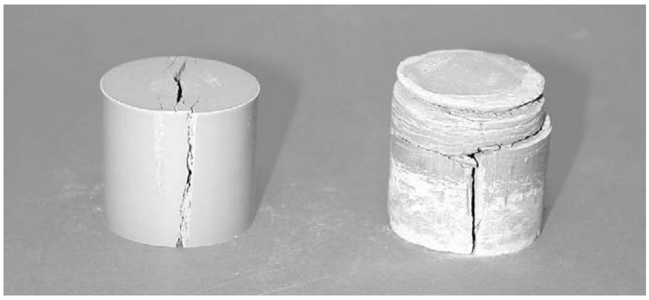


**Fig. 15:** Appearance of the compacts after measurement of diametral compressive strength (Granulates part 1 at the left, part 2 at the right).

All other parameters in fig. 14 also demonstrate that something must have happened with part 2 of the granulate. For example, the axial force transmission was diminished at 20 %, the wall friction coefficient rose, and especially the ejection forces increased drastically. In the same way, the results for the part of energy loss and the distribution of pressure moved in an unfavorable direction. Directly relevant to pressing defects are the calculated high shear stresses and the decreased fraction of elastic relaxation inside the die.

The explanation for the compaction riddle is very simple and will become clear with a view of the procedure described in fig. 16.

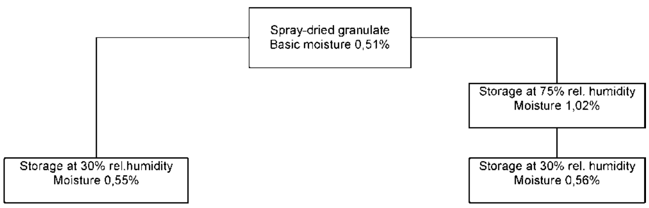



**Fig. 16:** Solution to the compaction riddle.

The basic spray-dried granulate was divided into two parts. The first one was hermetically sealed off from climatic influences before compaction. The second one was stored in an open container in the laboratory for 2 days and absorbed water until an equilibrium moisture of 1.02 % was reached. After the subsequent storage of the granulate under 30 % humidity the basic moisture nearly was reached again.

## IV. Conclusions

The achieved results show that the compaction behaviour and the quality of uniaxial pressed compacts are significantly influenced by climatic conditions during granulate storage and processing. Furthermore, it was demonstrated that not every fact known about the role of moisture during compaction of granulates from silicate ceramics can be applied to the group of advanced ceramics. For the examined silicon nitride granulates with the described binder/lubricant combinations, after their storage at a humidity of more than 50 %, a deleterious influence of moistures was verified. The experiments have shown that the absorption and desorption are indeed reversible processes for that case, but not the compaction behaviour. The influence of cyclic changing climatic conditions must be avoided under all circumstances.

Generally speaking, greater effort is needed for the most important base materials of advanced ceramics to gain a better understanding of the complicated correlations and interactions between the binder and lubricant properties of different types under varying humidity, temperatures and pressures. The determination of a system of specific pressing parameters with an instrumented compacting tool is a suitable way to achieve such progress and to overcome the widespread empiricism.

## Acknowledgements

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