CURRENT PROCESS DEVELOPMENTS IN PRODUCTION OF NATURAL UO_2 GREEN PELLETS

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ABSTRACT

Nuclear Fuel Complex (NFC) is responsible for manufacturing natural UO_2 fuel for the operating and forthcoming PHWRs in India. In recent years, the following two important modifications have been made in the production line leading to improvement in productivity:

- modifications in the press-tooling, granulator and in the powder handling systems in pre-compaction press facilitating reduction of fine fraction (-100 mesh) of UO_2 powder and simultaneous processing of the virgin powder with the fine fraction

- use of an alternative and effective *Admixed Powder Lubricant* in pelletization for overcoming problems faced with zinc-stearate

The paper discusses the above process developments and highlights the benefits obtained.

1. INTRODUCTION

The natural uranium fuel fabrication plant of NFC produces high density (96-98%TD), nuclear grade uranium oxide (UO_2) fuel pellets required for all the PHWRs operating in the country. These pellets are made from UO_2 powder prepared through ammonium-di-uranate (ADU) precipitate route. The pellet fabrication is accomplished through process steps viz. pre-compaction and dry granulation, admixing of powder lubricant with the granules, compaction of green pellets, high temperature (1700°C) sintering of the green pellets followed by wet grinding of the sintered pellets to produce uniform diameter ground pellets. These pellets are visually inspected for physical integrity before they are encapsulated in zirconium alloy fuel tubes.

During the last couple of years, the plant had successfully introduced a number of developments/improvements in the pelletization viz. introduction of the *Admixed Lubricant Process Route* in place of liquid die-wall lubricant route, adoption of high performance press-tooling in final compaction, incorporation of advanced pellet design in the production of double-dish chamfered pellets etc.[1]. Through these developments, the plant achieved a significant progressive improvement in the process-recovery (Fig-1).

Currently, developmental effort was taken up in the hydraulic press used for pre-compaction and granulation of UO_2 powder. Modifications carried out helped to reduce the generation of fine fraction (-100 mesh), to avoid manual interventions during automatic operation and need of frequent replacement of granulator mesh, steel dies and punches, etc. Such modifications have helped in improving the productivity in the process and overall process recovery.

Another area needing developmental effort was to establish a suitable alternative lubricant for replacing zinc-stearate used as the admixed lubricant in the compaction of UO_2 pellets. Zinc-stearate led to many operational problems in sintering including boat stuck-up, mainly due to condensation of zinc in particulate form over the interior parts of the sintering furnace.

During de-waxing in the pre-heat section of the main furnace, the metallic zinc evolved out of the lubricant gets condensed below its volatization temperature of about 920°C. The clean muffle (Fig-2) gets gradually deposited with the zinc particulate. This particulate mass gets sintered and transforms into a hard deposit to cause disturbance/hindrance to the boat movement (Fig-3) leading to boat jamming. In extreme cases, molybdenum components worth lakhs of rupees get damaged during boat jamming (Fig-4). Consequently planned or unplanned shut down of the continuous furnace becomes necessary for cleaning the deposit. Additionally, failure of thermocouples, frequent clogging of the gas burn-off port, accumulation of zinc-oxide dust near the charge entry and the fume hood, etc. are common. At times, the growing zinc deposit gets dislodged and falls over the pellets resting on the top layer of the boat. During sintering, craters are formed on the top surface of the UO_2 pellets (Fig-5), which contribute to increased rejections.

The plant identified a promising organic *Admixed Powder Lubricant* bearing no metal component. A comprehensive suitability study was carried out using the coarser grade as well as finer grade of the lubricant. The finer grade of the organic lubricant added with the UO_2 granules in same %wt basis as that of zinc-stearate performed very effectively in all respects in mass scale production of UO_2 green pellets.

The paper discusses the developments/modifications in detail in the following sections:

2. PROCESS-DEVELOPMENT IN PRE-COMPACTION AND GRANULATION IN THE HYDRAULIC PRESS

This development has essentially the following characteristics and the modified arrangement in handling of UO_2 powder in the press is shown in (Fig-6):

2.1. Incorporation of Tungsten Carbide Die-Sleeve in place of Steel Die-Sleeve:

Die-sleeve made of cold die steel (CDS) wears out very fast in pre-compaction operation because pressing is done without using any die-wall lubricant. This brings about many process-draw backs such as continuous addition of iron contamination in the granules, increasing powder spillage from the enlarged die and need of frequent tool-replacement associating loss of production and spread of radioactive contamination in the surrounding. As an effective solution, Tungsten carbide die-sleeves were developed indigenously and adopted in the process.

2.2. Incorporation of Perforated SS Sheet (Mesh Size 5) in place of SS Wire Mesh Sieve (Mesh Size 8) in the Oscillatory Granulator :

The sieve with the smaller opening size allowed rubbing of the pre-compact cake over an extended duration leading to generation of more fines and lesser granules. Moreover, the frequent damage of the sieve was associated with generation of oversize granules beyond acceptable limit and frequent replacement of sieve after each 1.5 to 2MT granule production. The incorporation of a perforated SS sheet of larger opening size in the place of wire mesh sieve has greatly contributed towards increased granule-output and has avoided the above operational problems. A life equivalent to about 20 times of the life of wire-sieve has been realized.

2.3. Automatic Recycle of the Separated Fines:

Earlier in the process, the fine fraction separated from the granulated powder in the vibro-siever was collected in a cylindrical container and this container was manually replaced and handled like the input virgin powder container on campaign basis. This required additional human efforts and caused activity spread up in the process. A simple innovative concept was realized in practice by replacing the fines-collecting container with a conical container and conveying the fines automatically to the press hopper by pneumatic suction.

2.4. Benefits from the Process Development:

- Increase of productivity: All the above developments directly or indirectly have contributed in improving the equipment productivity. The production capacity of the press has been enhanced by about 50%.

- Improvement in process-recovery: The overall process recovery in production of finished UO_2 pellets from powder is further increased by about 3%. The pellets produced from such granules always have smooth and shiny sintered surface (Fig-7). Processing of the fines as a separate batch caused significant rejections earlier due to low sintered density, bulging and cracks in sintered pellets. The spillages of powder are also absolutely controlled.

Additionally, the modifications in the press have helped in extreme containment of radioactive dust in granule production.

3. EXPLORING AND ADOPTING ORGANIC BASED POWDER LUBRICANT IN PLACE OF ZINC-STEARATE POWDER LUBRICANT

The organic lubricant was adopted in mass scale production of UO_2 fuel pellets after analysis of its physical and chemical characteristics and analysis of the critical observations in pressing and sintering trials.

3.1. Main Characteristics of the Organic Lubricant

- The purity of the lubricant is far superior to zinc-stearate (Table-1). The critical impurity elements and the impurity normally seen in zinc-stearate are compared.
- While analyzing the particle size distribution of the dry powder in laser diffraction, it was observed that both the coarser grade and the finer grade of the organic lubricant are little coarser than zinc-stearate (Fig-8).
- As observed in Thermo-Gravimetric-Analysis (TGA) in hydrogen atmosphere (Fig-9), the organic lubricant burns-out completely only at around 400°C where as zinc-stearate, even when heated up to 900°C, volatilizes by about 90% indicating that it needs further higher temperature for complete volatilization.

3.2. Trial-Observations:

- The finer grade organic lubricant is equally effective as zinc-stearate in compaction of UO_2 pellets without requiring any additional die-wall lubricant.
- The organic lubricant is found to produce higher sintered density in pellets than that obtained using zinc-stearate lubricant. The relative sintered density of pellets made with varying percentage of lubricants is shown in Fig-10.
- The coarser grade leads to tiny surface cracks in sintered pellets (Fig-11) where as the finer grade produces a defect-free smooth sintered surface. Greater than 95% acceptability is achieved on visual inspection of ground pellets.

3.3. Adoption in Regular Production:

The performance of the organic was confirmed initially in discrete trials, followed by continuous production in hundreds of kilograms and finally in continuous mass scale production of sintered UO_2 pellets in tonnage scale. Subsequently, the admixed organic lubricant was successfully adopted in the regular production of natural UO_2 pellets.

3.4. Benefits from Organic Lubricant:

- Unlike Zinc-stearate, the organic lubricant burns out cleanly and completely without formation of any deposit or shoot inside or outside the furnace. Thus the operational problems faced with zinc-stearate in the sintering furnace has effectively been solved by adopting this organic based powder lubricant.
- The lubricant helps to produce higher sintered density, thus the rejects or re-sintering due to low density are minimized in production.

- By using organic lubricant, the furnace emission is become environment-friendly, generation of hazardous zinc-oxide waste is eliminated. Cost saving is significant by not installing dust collectors any time.

- By eliminating zinc-stearate, incidence of boat stuck-up and consumption of molybdenum components worth lakhs of rupees are likely to be minimized.

4. DISCUSSION

4.1. Since the fine fraction is extremely reduced and it is processed in mixed condition along with the virgin powder, the granules are always softer in nature. The green pellet undergoes coherent shrinkage in sintering resulting in smooth and shiny sintered surface (Fig-7). The granules made using the fine fraction alone as a separate batch are normally denser and harder in nature requiring10-12% lesser die-fill in final compaction. The compaction pressure is not likely to destroy the denser granules resulting in some large pores, because of which such pellets might develop defects like crack, low density and bulging in sintering [2].

4.2. Since the organic admixed lubricant burns out completely at only 400°C (Fig-9) without leaving any residue in the furnace, such lubricant is most sought-after lubricant in P/M industries and in fuel pellet fabrication.

4.3. As required, the admixed lubricant/binder should get expelled completely from the UO_2 pellet before the densification process is initiated. Because the organic lubricant gets volatilized at about 400°C, it does not interfere at all in the sintering process. Hence, higher sintered density is observed in case of the organic admixed lubricant. The dip in sintered density in case of zinc stearate is higher with higher quantity addition (Fig-10). Complete volatilization of zinc-stearate is likely to take place much above 900°C. Hence, zinc vapor may get trapped in the pores during densification, which intervenes in pore-shrinkage resulting in lower sintered density.

4.4. The coarser grade organic lubricant, even with smaller quantity results in uniformly distributed tiny surface cracks (Fig-11) probably because the coarser particles even in lesser quantity-addition intervene inter-particle bonding [3].

4.5. Zinc-oxide vapor is health hazardous and hence zinc-stearate poses environmental hazard. Therefore, the development of the organic lubricant in compaction of UO_2 pellets is of immense worth.

4.6. The replacement of zinc-stearate by the organic lubricant will be highly useful for advanced sintering technology such as low temperature oxidative sintering. Such organic lubricant is also advantageous for a walking beam type sintering furnace where any solid deposit as in the case of zinc-stearate may affect mechanical movement of the beams.

5. CONCLUSION

5.1. The process development carried out in pre-compaction and granulation in the hydraulic press has been highly advantageous in improving equipment productivity, pellet quality and containment of radio active dust.

5.2. The process development involving exploration and adoption of an organic based admixed lubricant in place of the zinc-stearate is a unique development in compaction of UO_2 pellets. Being 100% organic in nature, technically it is an advanced lubricant associated with innumerable advantages such as lower de-waxing temperature, residue-less burnout, environment friendly emissions, improvement of sintered density of pellets, etc. Such lubricant is likely to facilitate low temperature oxidative sintering and use of walking beam sintering furnace.

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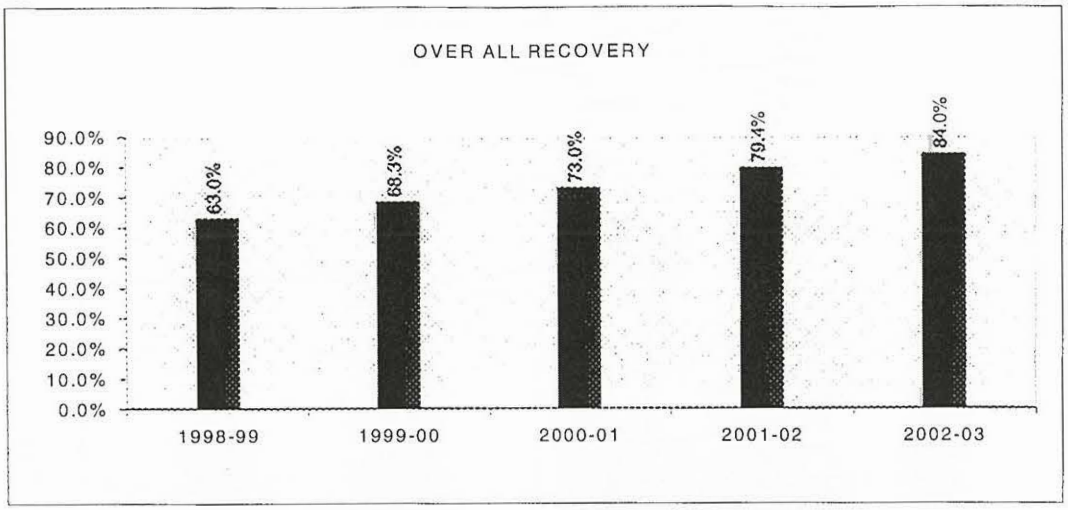

Fig-1: Progressive Improvement in Recovery

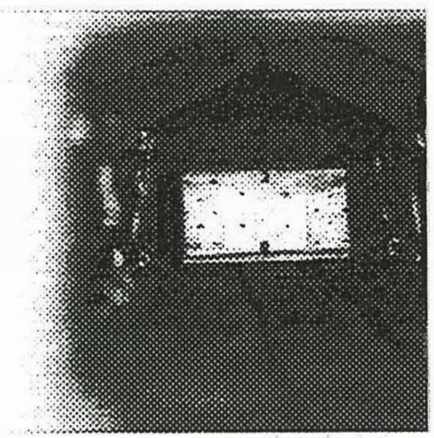

Fig-2: Clean Muffle

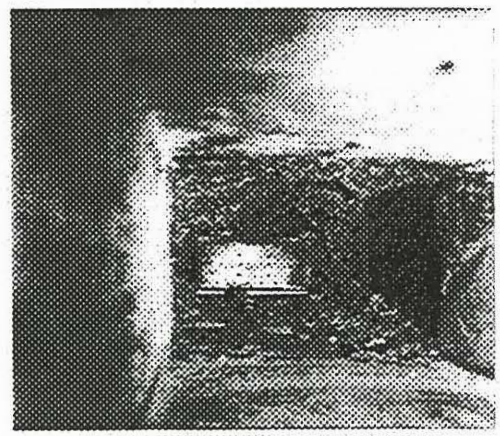

Fig-3: Hard Zinc Deposit around Muffle

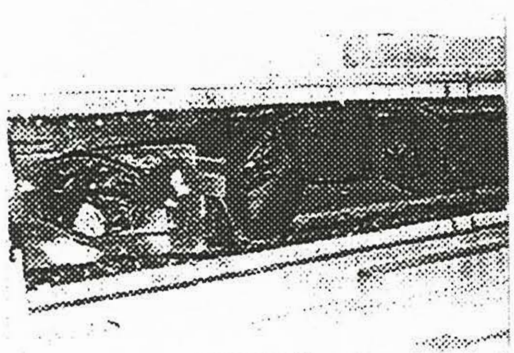

Fig-4: Boat Jamming & Damage of Moly Items

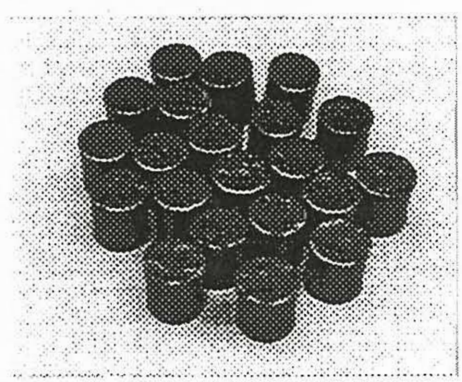

Fig-5: Formation of Craters on Pellets due to Zinc-deposit from Muffle

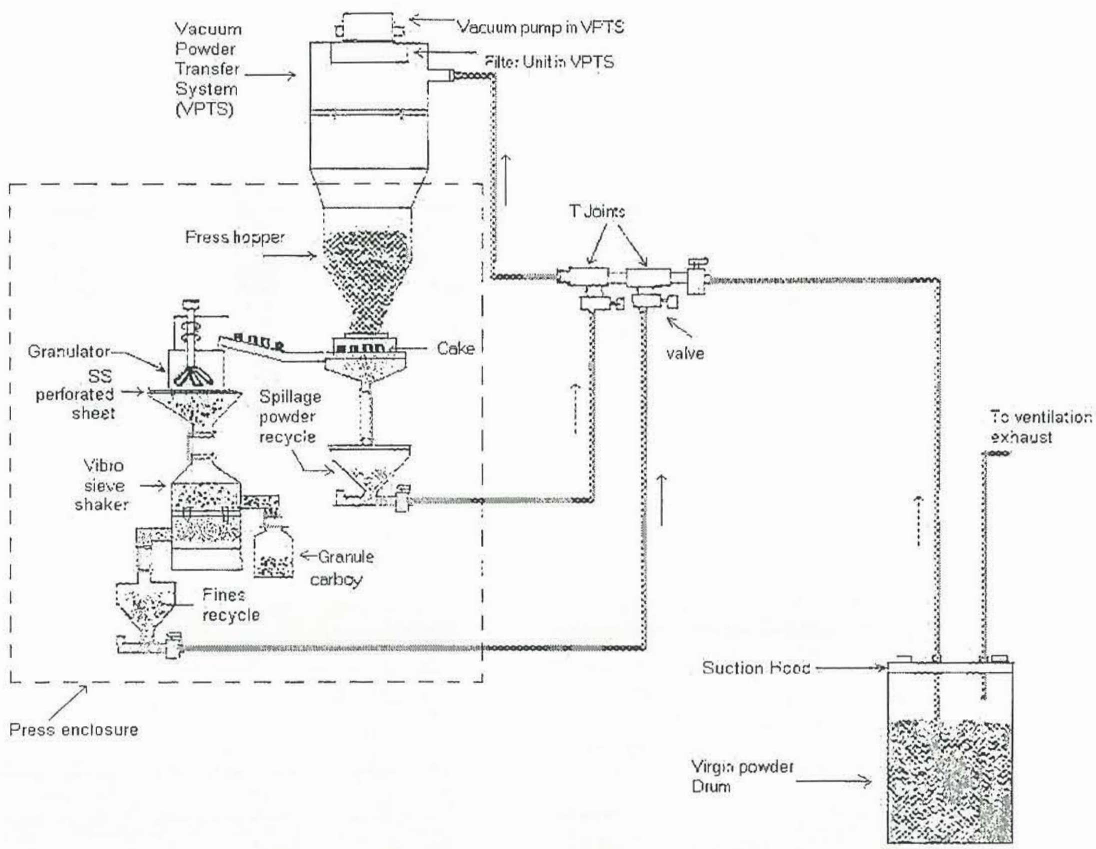

Fig-6: Modified arrangement in Hydraulic Pre-Compaction Press

Fig-7: Smooth & Shiny Sintered Surface in pellets

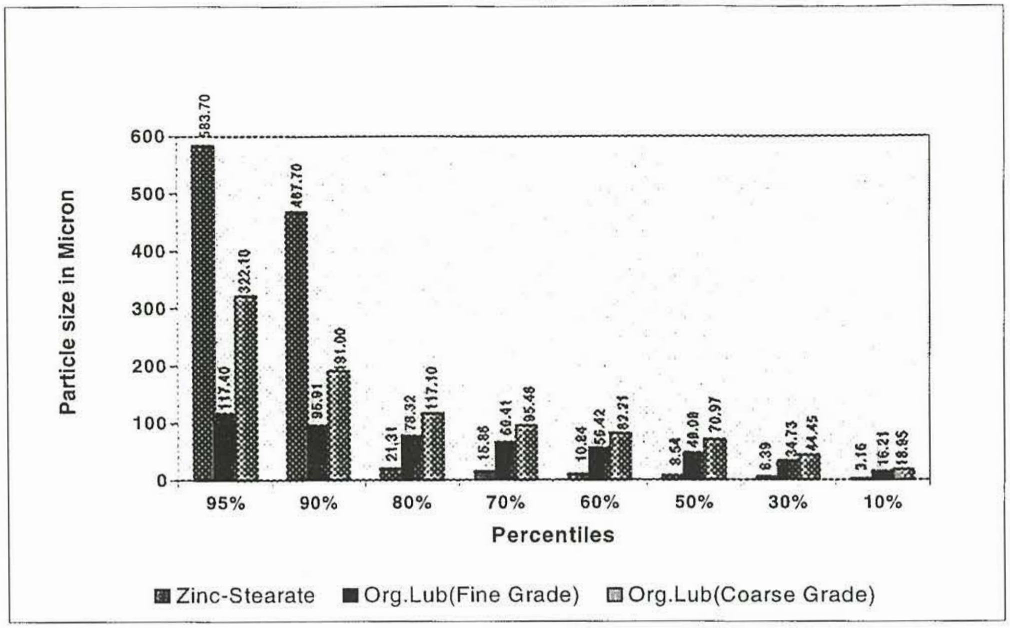

Fig-8: Particle size Distribution of Lubricants

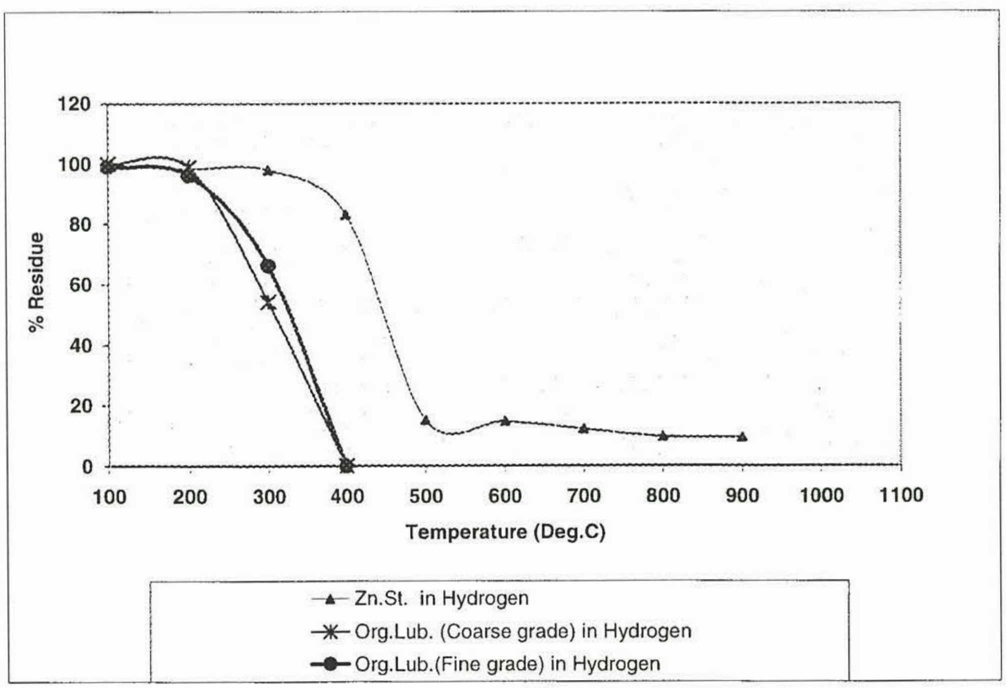

Fig-9: Thermogravimetric curves of the Lubricants

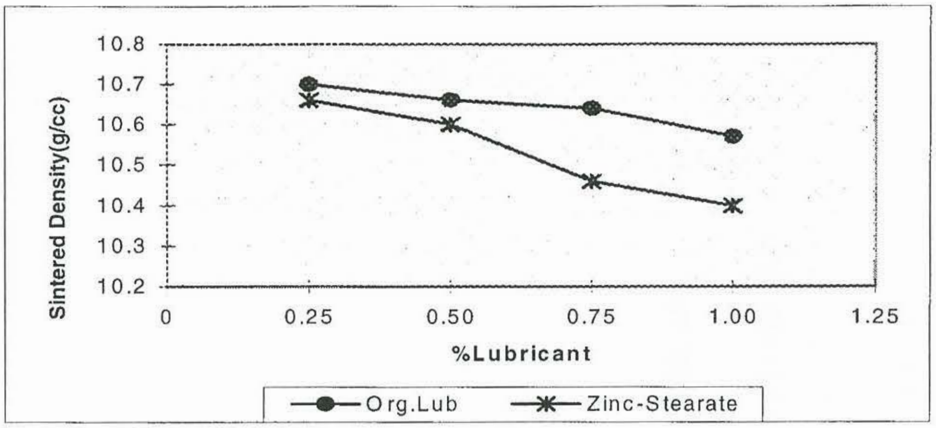

Fig-10: Sintered Density Vs % Lubricant

Fig-11: Tiny Surface-Cracks in case of Org.Lub.Coarse-Grade

Table-1: Comparison for Purity

Impurity Elements	In Zinc-Stearate (ppm)	In Org. Lubricant (ppm)
B	1	<1
Ca	<2000	<25
Cd	1	<1
Fe	60	<25
Mg	<100	<25
Mn	40	<10
Na	<4000	<50
Zn	10-11%	-