Improved Aluminium Recovery at Recycling Plants by integrated Slag Refining

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Introduction

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Raw materials dedicated to the recycling of aluminium have to contain it in a non-oxidized but metallic form. The transformation of such oxides into metal is technically and economically not feasible. The major recycling target is to recover the metallic aluminium content of scraps and residues with the highest possible yield combined with the lowest effort.

During processing and use of aluminium products a broad range of scraps, intermediates and residues are generated. A special interest is given to turnings and drosses, which build approximately 45 % of the raw materials mix to a refining plant [1]. In a close connection the by-product "slag" has to be considered, as this material has to be melted, using salt as oxide collector and atmosphere barrier. Figure 1 shows the raw materials mix in 1997 of Germany's refining operations. Table 1 summarizes the estimated amount of turnings, drosses and slags.





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Table 1: Total amount (t/y) of turnings, dross and slag in Germany 1997(* estimates)

wrought alloy turnings	27 000
cast alloy turnings	50-60 000
turnings from semis	130-170 000
turnings from final processing	50-100 000
turnings (total)	ca. 260-360 000
primary dross	15 000
remelters dross	30,000
dross from foundries	35 000
refiners dross (converter)	20 000
refiners dross (hearth furnace)	ca. 10 000
dross (total)	ca. 110 000 *
slag from tilt type rotary furnaces	20-30 000 *
slag from rotary drum furnaces	ca. 200 000
slag (total)	ca. 230 000

One target of this investigation is to optimize the salt composition in order to obtain higher aluminium yields in the furnace operation. As a second target a concept will be developed to integrate the slag treatment with the melting process in order to recover the lost aluminium content already at the refiners plant. A third target is the reduction of salt purchased from external sources.

2 Established recycling processes for aluminium turnings, drosses and slags

2.1 Conditioning of aluminium turnings

During mechanical processing of aluminium turnings from cast and forged parts are generated and collected typically classified by alloy composition. Quite often an emulsion of oil and water is used and contaminates this type of aluminium scrap.

A <u>wet conditioning</u> process is based on a countercurrent flow washing step, using centrifuges. The water/oil content is reduced to each 2 %, the removed emulsion is separated using demulgators. The final drying takes place in rotary drum coolers, followed by an iron separation using magnetic fields. By sieving the turnings are split into a > 1 mm coarse fraction and a < 1 mm fine fraction. The latter has to be compacted prior to the melting process [2, 3].

A <u>thermal conditioning</u> process is conducted in indirect heated discontinuously operated rotary drums with a well controlled oxygen deficit in the oven atmosphere. The de-oiled and dried turnings are magnetically split and seived as mentioned above. The off-gases are subsequently burned at temperatures > 800 °C and quenched to 250 °C with water to avoid PCDF-formation [4, 5].

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2.2 Conditioning of aluminium dross

Dross is a by-product of all aluminium melting processes. Depending on scrap type, alloy treatment and melting equipment, the amount, metal content and properties of dross vary in a wide range. Dross is a mixture of aluminium, aluminium-oxide and oxides of alloying elements, additionally halogenide-, carbide- or nitride-compounds of these metals are regularly encountered. Typically dross contains a large amount of metallic aluminium. This is mechanically fixed in a cellular structure of aluminium-oxide, which has a tension strength strong enough to keep aluminium 20 times of its own weight bonded. As dross contains also finely dispersed gas bubbles (foam structure), it easily separates from the aluminium melt due to the reduced density [6, 7].

The following process parameters have a strong influence on volume, morphology and metal content of dross:

- surface/volume-ratio of the scrap,
- type (organic, inorganic) and amount of impurities,
- melting conditions and melting equipment,
- melt composition (especially Mg-, Na-, Li-, Ca-content [8]),
- refining procedure,
- way of dross removal (temperature, tools),
- cooling parameters of the removed dross.

A direct comparison and evaluation of the various dross conditioning processes [9, 10] is difficult, as the raw material source is different. Generally modern processes are based on a fast quenching step in order to avoid an oxidation of the incorporated aluminium metal droplets. It is followed by a mechanical benefication step which removes as much oxide as possible. Alternatively a direct melting of the > 1 mm fraction is state of the art [11]. The by-product dross-dust has to be dumped or processed separately in a slag treatment plant, the < 1 mm fraction (dross residue) can be sold to the steel industry for desoxidation purposes.

Table 2 summarizes size, aluminium content and proportion for the four products of the mechanical treatment process. It can be seen that all values vary widely depending on the history, especially the $\leq 1 \text{ mm}$ dross-residue (table 2).

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Table 2:	Products of the "c	d" consolidation	of dross [12] (*estimates)
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	particle size	origin	Al-content [% Al]	proportion [%]	next step
lump fraction	lumps	hand sorting	70-90	up to 20	melting
coarse fraction	<100 mm	sieve	50-70	30-80	salt melting
dross residue	< 1 mm	sieve	20-50	10-40	desox. in steel, landfill
dross dust	< 1 mm	filter	5-20	5-20	landfill, slag recycler

2.3 Rotary drum furnace process

Still up to now the rotary drum furnace is the most common unit to melt high impurity scraps. It consists of a cylindrical steel shell, which is horizontally mounted on wheels and lined with ceramic bricks. The ceramic lining is typically interrupted by mixing paddles in order to maintain an improved mixing and furnace operation [13].

A liquid salt consisting of sodium- and potassium-chloride is used to collect oxides and to avoid oxidation by the atmosphere. CaF_2 is added to improve the coagulation of aluminium droplets. The lining is continuously cleaned by the slag due to the rotating movement of the kiln. The productivity depends on the raw material mix, salt quality/quantity and the operation mode. Using oxygenburners, optimized furnace sealing and an automatic control of the atmosphere, the thermal yield can be moved up to 66 % [2]. A thermal energy balancing results in a calculated oxidation loss for aluminium of about 2 - 3 % of the charged metallic aluminium.

A "salt factor" is introduced to characterize/calculate the necessary amount of salt:

SF = (salt amount)/(weight of raw material mix - trial determined aluminium recovery)

The use of salt is especially necessary, if raw materials with high specific surfaces or high oxide contents are to be recycled. Depending on the technical and economic conditions a limit for the total oxide content exists (productivity, energy and salt consumption, slag treatment costs). Figure 2 shows exemplary the specific energy consumption of an aluminium rotary drum furnace as a function of the charged oxide volume. As it can be seen, the energy consumption can be reduced by choosing smaller salt factors.



Figure 2: Theoretical energy consumption of a rotary drum furnace with rising oxide contents in the Aluminium-scrap (MJ/t Al)

2.4 Smelting in a tilt type rotary furnace

A more recent melting process is based on a tiltable furnace and also operated with salt addition. But compared to the conventional rotating drum furnace the salt volume is significantly reduced (salt factor 0,4 up to 0,8). This results in a non liquid but pasty slag, impossible to tap or cast. The oven has to be tilted entirely to be emptied through the charging hole. This type of furnace is especially designed for heavy oxidized materials, eg. dross. Oxygen burners mounted close to the off-gas opening forces the flame to turn back. This leads to a high thermal efficiency and less specific energy consumption [14, 15, 16].

Table 3 [17] compares the two salt-smelting-techniques. The conventional rotating drum furnace (RDF) recovers at least 90 % of the aluminium content if charged with a standard raw materials mix. A hypothetical salt factor reduction form 1.8 to 1.0 would reduce the specific energy consumption to about 90 % of the reference operation, a further reduction is technically not possible (tapping problems). A further optimization can be realized with the tiltable rotary furnace (TRF), which allows even a 100 % dross charge.

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	rotating c	lrum furnace	tilttype rot	ary furnace
	excess salt addition	optimized salt addition	scrap mix (normal)	only dross
gas burner	cc	old air	οχι	/gen
salt factor	1,8	1,0	0	,5
scrap input [t]	11,8	13,0	6,97	6,0
salt input [t]	3,2	2,0	0,52	1,5
total input [t]		15	7	,5
Al output [t]	9,9	11,0	5,92	3,3
Al yield (kg metal/scrap input) [%]		85	85	55
slag output [t]	5,0	4,0	1,57	4,2
oxide content in slag [%]	36	49	(57
energy consumption [MJ/t] (per total input)	2600	2600	1700	1700
energy consumption [MJ/t Al] (per scrap input)	3300	3000	1830	2100
energy consumption [MJ/t Al] (per Al output)	3900	3550	2150	3800
tap to tap time [h]	ca. 6	4-8	ca. 3	2-3

Table 3: Comparison of conventional rotating drum (RDF) and tiltable rotary furnace (TRF) operation

2.5 Slag treatment/salt recovery

Closely linked to the consolidation and metallurgical processes for aluminium scraps and residues is the treatment of the "by-product" slag. In the past years the specific slag volume could be reduced from 400 - 700 kg/t to 300 - 500 kg/t [17]. The oxide input as well as oxidized aluminium from the melting procedure is collected by the salt forming a physical salt-oxide-mixture. Unavoidable are small aluminium droplets, too small to coagulate, which reduce the melting yield. Table 4 summarizes the proportion of these three phases in RDF and TRF processes.

Table 4: Composition of	slags from RDI	- and TRF-furnace	operations (*estimates)
	rotary dr (liquid	um furnace salt melt)	tilt type rotary furnace (pasty salt cake)
	typical	min/max	min/max
Aluminium metal [%]	8	5/20	5/15*
oxides, CaF ₂ [%]	37	20/40	50/60*
NaCl, KCl [%]	55	45/75	25/45*

Due to the water soluble chloride-phase, landfilling of the slag is ecologically no solution considering the risk of ground water contamination and gas-formation (H₂, CH₄, NH₃, PH₃, H₂S). Besides this, also the recovery of the melting salt is economically feasible. The salt, typically a NaCl-KClmixture, shows a minimum melting point of 664 °C at 44 % NaCl/56 % KCl (equimolarity) [18, 19].

Typically some percent of CaF_2 are added to improve coagulation of aluminium droplets. The salt due to its low surface tension creeps between the oxide film and the molten metal using cracks formed by thermal expansion or Na-gas-formation. As the solubility is almost zero the oxides form a liquid-solid-suspension with the salt, the viscosity rises. The viscosity itself is strongly depending on volume size and type of oxides dispersed in the salt. Figure 3 shows the thixotropic behaviour, forced by flat and thin γ -Al₂O₃ dispersoids.



Figure 3: Rheologic behaviour of an oxide-salt mixture with oxide-morphology in a mechanical stirred crucible [20]

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- 3 Experimental results of salt melting tests with high oxidized raw materials
- 3.1 Equipment

An electrically heated crucible was charged with NaCl/KCl-salt after the temperature exceeded 100 °C. After melting, a thermo-couple was placed 3 cm below the surface, a mechanical stirrer was mounted in a depth of 4-5 cm to keep the turbulence in the upper crucible section. Figure 4 shows a scheme of the experimental arrangement.



Figure 4: Scheme of the experimental equipment

- l-motor; 2-stirrer; 3-crucible; 4-lining; 5-aluminium; 6-salt
- 7- heater; $8-Al_2O_3$ -protection tube with thermocouple;

9- temperature measurement

3.2 Melting behaviour of fine aluminium turnings

The very fine turnings-fraction shows a tapped density of $0,78 \text{ g/cm}^2$, a particle size distribution, as presented in table 5 and a mean oxygen content of approximately 1,45 %.

 Table 5:
 Particle size distribution of a turnings-fraction (< 2 mm)</th>

fraction (mm)	>4	>2-4	>0,8-2	0,25-0,8	< 0,25
proportion (%)	0,05	0,07	45,6	45,5	8,8

Based on these data a specific surface area of about 1 m^2/kg can be assumed (calculation as a 0,8 mm aluminium foil).

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The dry turnings scrap charge, after preheating to 400 °C, was inserted close to the stirrer shaft into the vortex and immediatly immersed by the melt. A heat exchange coefficient α between melt and scrap can be defined using the temperature drop after charging.

$$\alpha = \frac{m \cdot c_p \cdot \Delta T_1}{F \cdot t \cdot \Delta T_2}$$

- m mass of scrap added [kg]
- F specific surface area $[m^2/kg]$
- $c_p \quad \ \ thermal \ capacity \ salt \ [J/kg \cdot K]$
- ΔT_1 salt temperature drop at charging [K]
- ΔT_2 Al temperature increase [K]
- time to reach minimum of salt temperature [s]



Figure 5 presents the heat transfer depending on the turbulence conditions and on the amount of scrap added. It shows that the rotating speed of the stirrer, i.e. mixing condition, has a significant effect on the time between charging and melting. The local turbulence is strongly depending on the viscosity of the melt, which is again a function of oxide content and stirring condition (thixotropy).



Figure 5: Heat exchange coefficient in stirred salt melts depending on rotating speed of the stirrer (portion: 10 g per charging step)

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Aluminium dross was split by sieving into a powder and a coarse fraction. Both fractions again were each split into 6 subfractions. The particle size distribution of the coarse fraction is shown in figure 6, the volume > 40 mm was additionally separated by hand into high and low metallic contents.



Figure 6: Particle size distribution of aluminium dross

From figure 7 a strong dependence between particle size and metal yield can be observed. Particles less than $75 - 125 \mu m (\log d_p < 1)$ form the bottom line of any metal recovery.



Figure 7: Metal recovery from coarse dross and dross powder with rising particle size

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It has to be stated that, even with an addition of 2 % CaF_2 , the metal yield of dross dust is always unsufficiently low. Additional trials were conducted with the fine fraction of the aluminium dross, in order to further investigate the interrelation between particle size and CaF_2 -addition on the metal yield.

As figure 8 indicates a further increase of CaF_2 to 3 % did not influence significantly the metal yield and the processing of dross powder < 1 mm is most likely uneconomic.





3.4 Effect of the oxide-content in slag on metal recovery

Industrial supplied slag from RDF-operation was analysed in respect of the contents of the major components. Table 6 shows the results of the chemical analysis of recycled salt and slag after scrap/dross treatment. The values indicate an untypically high salt factor.

 Table 6: Composition of salt and slag from industrial RDF-operation

	Al met. [%]	Al ₂ O ₃ [%]	CaF2 [%]	NaCl [%]	KCl [%]	
recycling salt	-	0,43	0,13	69,3	27,4	
slag	7,5	28,0	1,6	62	2,9	

This slag was used to investigate the effect of the oxide content on the metal recovery yield. The slag was diluted with recycling salt 2, 2,5 and 3 times of the slag weight. The slag-salt-mixture was melted and stirred for 1 hour in order to allow the dispersed aluminium droplets to coagulate. From figure 9 it can be seen that an increasing oxide content in the slag up to 15 % helps coagulation of the droplets into larger diameters, which allows settling from the slag.

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Above 15 % the increasing viscosity hinders coagulation and further settling. Industrial values with a slag of 28 % Al_2O_3 fit quite well in the drawn relationship.

3.5 Salt refining in molten condition

From the lab scale tests described above, it could be seen that the maximum metal yields can be achieved at oxide contents of 10 - 15 % in the salt. This is due to a countercurrent effect of increasing friction between the suspended oxides and the oxide film and increasing viscosity. Conventional rotary drum furnaces are operating with 25 - 30 % oxides in the tapped slag. In order to develop an integrated smelting-slag refining concept, extended tests were conducted to remove oxides immediately from the molten process slag.

First trials on filtration of oxide-containing NaCl/KCl slags showed quickly strong difficulties in holding back the fine oxides, as they are part of a quite wide particle size distribution from < 1 to $<< 1000 \mu m$. Using cake filtration techniques, soon a very dense oxide layer is formed on top of the felt and, due to the small density difference, the liquid salt can not penetrate it sufficiently.

A second series investigated a simple gravity segregation process. Only a small portion of cleaned salt was obtained and the oxide-salt-bottom-phase showed a much too high salt content.

In a third test series, an IME-optimized centrifugal rotor was immersed in the slag. After two minutes at a rotating speed of 400 rpm the rotor was pulled up and accelerated to a higher rotating

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speed. The liquid salt slag was centrifugally extracted in order to receive a dry filter cake. The cake was removed and the rotor immersed again. Figure 10 presents the main part of this experimental equipment – the rotor head.



Figure 10: Scheme of the centrifugal rotor head. 1. rotor bottom, 2. area of suspension, 3. segregated oxide, 4. rotor top, 5. suspension inlet, 6. salt outlet

The separation results are shown in figures 11 - 13. Up to CaF₂-content of 4 %, a constant oxide content of nearly 45 % and a CaF₂-content of < 3 % in the cake could be obtained. The CaF₂-content depends more or less on the salt matrix. It was interesting to see and in contradiction with the common understanding (10 % solubility in the ternary diagram NaCl-KCl-CaF₂ at 800 °C), that, at a very low level of 3,8 %, CaF₂ cannot be dissolved. It is separated in the solid state from the slag and found in the oxide phase of the rotor. A systematic investigation of the NaCl-KCl,CaF₂-System is necessary and part of a current investigation at IME.



Figure 11: CaF₂- and oxide contents in the solid residue of the centrifugal rotor head with increasing CaF₂-additions to the salt

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With increasing rotating speed the centrifugal acceleration is improved, a separation factor $g = \omega^2 r/g$ can be used to classify the fluid flow system. The positive effect is shown in figure 12, as the oxide content can even exceed the 50 % level, the salt matrix is recovered with higher yields.





Probably the most exciting result is shown in figure 13. If $2 \% \text{CaF}_2$ is added to the salt, the metallic aluminium content in the solid residue of the rotor, which would be the product delivered to external slag refiners, can be reduced from > 5 % to much less than 0.5 %.



Figure 14: Integration of the centrifugal salt cleaning process into the standard rotary drum furnace operation

aluminium production



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oxide removal centrifuge

alt/oxide

- mix

1897

purchased recycling salt:

1440 kg

1033 kg

i.e. - 28%

411 kg

0.2 %

2381 kg

2385 ko

77 ka

4.1 Concept for rotary drum furnaces

Rotary drum fumaces are especially useful if scraps and residues with increased oxide and/or impurity levels have to be recycled. The results presented about the melting behaviour of fine turnings, dross powder, dross dust in conjunction with the influence of oxide contents force to develop a concept, applicable to "high oxide" processes. In respect to metal recovery yield, it is recommended to control the oxide level in the salt to 10 - 15 % and avoid significant higher values. But presently rotating drum furnace operations cannot run economically with such low oxide levels (= high salt/scrap ratio) due to the high slag volume, external slag treatment cost, increased energy consumption as more salt has to be melted, and a reduced productivity. The idea is to implement an inline slag-cleaning/oxide removal process step in order to recover molten recycling salt and the dispersed aluminium metal already at the recycling plant. It is recommended to use a separate heated ladle with a centrifugal rotor head to treat the tapped slag immediately after scrap melting. A high concentrated oxide/salt mix with very low aluminium inclusions will be the product given to salt recyclers, liquid salt can be separated and returned to the next drum melting sequence and segregated aluminium can be added to the main stream. A certain amount of salt will be kept in the ladle to dilute the next slag tapping to the optimum level of 15 % oxides.

Figure 14 shows a very simplified mass flow balance of a standard dross treatment process (black numbers) and an improved process (italic numbers).

dross input

rotary drum furnace

2000 ka

1080 kg

1157 kg

1.0. + 7%

oxygen: 21 kg

60 %

2,0 %

8.0 %

salt factor

lab metal vield

Al droplet loss

Al oxidat. loss

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slag by oxidation and as only 1080 kg (e. g. 90 % yield related to a laboratory test) will be recovered to a demand of 1440 kg salt, typically bought from a recycling plant. As always aluminium is lost m associated costs. The new separation equipment would recover approx. 80 kg molten Al and 410 kg 4,1 % dispersed aluminium metal. Even the latter is given away today for external recovery with test had shown a maximum melting recovery of 60 %, a salt factor of 1,8 is practicable. This leads Assuming that only 2000 kg dross will be charged to a drum furnace, whereby a previous melting treated externally. molten recycling salt. This means 1900 kg salt/oxide mix (= 75 % of present volume) have to be from droplets as a metal bullion. Approx. 2400 kg of slag will be tapped with about 36 % oxide and

The advantages are

- increased aluminium yield (+ 7 %)
- reduced slag treatment costs (- 25 %)
- reduced salt purchased (- 30 %)
- reduced melting energy consumption (- 8 to 10 %)

for the new process step and costs for consumption materials (lining, etc.) Additional costs will be thermal energy needed to keep the slag liquid, capital- and personnel costs

4.2 Concept for hearth furnace dross

to recover the metallic aluminium content straight from the hot dross is presented below day the dross is cooled, collected and given to external refining operations (see 2.2, 4.1). A concept At remelter-plants dross is continuously produced in melting- or warming-up hearth-furnaces. To-

processing are produced (wrought alloy). supplied by indirect gas/oil-burners or even electrically by submerged electrodes [21]. Figure 15 quite compact form and operates with high throughput and flexibility. The energy input may be Much higher Al-recovery-yields can be expected. The centrifuge-vessel can be constructed in a system (separate chamber), the CaF2-content is controlled and adjusted. The liquid aluminium is much lower than in RDF-furnaces (see 3.4). Excess oxide is continuously removed by a centrifugal special care has to be taken regarding the salt addition, if Al-billets for a subsequent rolling/forging presents a conceptional flow chart of the above mentioned process (inline treatment of hot dross). A forced to separate from the oxide-network, coagulates and segregates due to the higher density The high aluminium containing dross is hot charged to a salt melt with optimized oxide content

rized in Table 7, based on the input of 1000 kg dross A preliminary comparison between the actual and a process-integrated dross treatment is summa-

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Figure 15: Integrated centrifugal dross treatment in molten salt with a continuous oxide removal resulting in increased metal yields

Table 7: Process data and expected product volumes for dross treatment processes (conventional

and proposed proce	ss-integrated salt melting)	
	conventional dross treatment	hot charging/centrifugal salt
	process	melting process
	(related to 1 t hot dross)	(related to 1 t hot dross)
aluminium recovered [kg]	approx. 480	approx. 670
aluminium yield (related to	69	96
hot dross content) [%]	-	
salt addition [kg]	approx. 170	approx. 300
dross residue (losses) [kg]	approx. 410	none
slag (to recycling plant) [kg]	approx. 350	approx. 650
energy consumption [MJ/t]	5000	1000

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Advantages are expected in

- heat energy use of the hot dross
- reduction of aluminium oxidation compared to a cooling/mechanical treatment-/warming-upprocess
- reduced aluminium losses
- reduced by-product volume (slag and dross residue)

5 Summary

Extensive experiments show that a reduced oxide content in KCl-NaCl-CaF₂-slags leads to an increased aluminium recovery in recycling processes. A centrifuge-based equipment was developed to continuously remove oxide from such melts. This can be process-integrated in conventional rotary drum furnace plants, in order to recover aluminium from slag and to recycle salt directly in the plant. A mass balance is presented for such a concept. A series of economic advantages can be achieved.

On the other hand also for remelting plants the process integration of the centrifuge should be feasible. Hot dross from hearth furnaces can be treated immediately and the incorporated aluminium is recovered. Using an optimized Al_2O_3 level in the dross treatment vessel, the maximum metal yield can be achieved.

In both cases a salt-oxide mixture (50 : 50) is produced, much lower in volume at lower recycling costs as compared to the state of the art process.

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