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Bench Mark Prebaked Anode Production with Russian Raw Materials

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Expansion of the Russian aluminium smelting is foreseen by using high amperage Rusal prebaked pot technology. For a high productivity of such modern smelters – thanks high amperage but also high current efficiency, but also for low energy consumption – the performance of the prebaked anodes is decisive.

Bench mark anode quality is therefore critical for low metal production cost. The intrinsic quality of anodes depends on the raw materials, mainly on the coke properties, and on the production equipment and their operating parameter conditions.

Based on a review of the domestic available green cokes, a prediction of the corresponding calcined coke blend and of the prebaked anode properties has been performed. Provided that a state of the art carbon plant installation is available, including the rodding shop for the butts quality, the bench mark quality of the prebaked anodes made with a representative coke blend has been defined. This forecast of the best achievable anode quality will be the basis for technical assessments of the performance of existing and future Russian carbon plants.

Keywords: green coke, calcined coke, butts, carbon plant, prebaked anode quality.

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Эталонные обожженные аноды из российских сырьевых материалов

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Планируется выполнить расширение российской алюминиевой промышленности за счет технологии электролизеров РУСАЛа с большой силой тока и обожженными анодами. Для высокой производительности таких современных заводов за счет большой силы тока, высокого выхода по току, а также низкого расхода энергии показатели обожженных анодов играют решающую роль.

Определение эталона качества анодов является критическим для обеспечения низкой стоимости производства металла. Качество самих анодов зависит от сырьевых материалов, в основном от свойств кокса, а также от технологического оборудования и его рабочих параметров.

На основании обзора имеющихся отечественных зеленых коксов был сделан прогноз свойств соответствующих шихт прокаленного кокса и обожженных анодов. При условии наличия современных цехов производства анодов, включая анодомонтажное отделение для качества огарков, был определен эталон качества обожженных анодов, изготовленных из представительной коксовой шихты. Этот прогноз наилучшего качества анодов станет основой для технической оценки показателей существующих и будущих российских цехов производства анодов.

Ключевые слова: зеленый кокс, прокаленный кокс, огарки, цех производства анодов, качество обожженных анодов.

Introduction

The changes in the availability of crude oils of different natures and the evolution of the refining industry towards higher conversion degree of their plants have substantial effect on the quality of the petroleum cokes processed by the aluminium smelters. The recent trend towards prebaked rather than Söderberg cell technology also affects the performance of the cokes and of the carbon anodes in the electrolysis process. The above mentioned issues were observed in the western world in the 1980s especially in smelters using coke from merchant calciners as they blend green cokes from several different refineries importing heavy/sour crudes rich in Sulfur and Vanadium. China experiences the same, as in the last decade the aluminium industry has greatly expanded and large amount of middle-east heavy crude oils are imported. The previous low S/V coke has been gradually replaced by high S/V coke that is a challenge for smooth use in the smelters.

In Russia the refiners mainly use oil supplied by the pipelines of AK Transneft, in which the west Siberian light oils are mixed with the heavier grades of the Ural area [1]. In the ex-USRR territory (Azerbaijan, Turkmenistan, Kazakhstan and Ukraine) most of the crude oils show S content below 0.5 % and some of the low S green cokes produced there find their way to the Russian smelters. In Russia the oil fields deliver either medium S crude oils, 0.5 % to 2.5 % S like for the Western Siberia and part of the Ural Povolzh'e fields, or high S (> 2.5 %) crude oils in most of the Ural Povolzh'e area.

New delayed coking units are built and projected, but large size/high amperage aluminium smelters are also in construction. This may negatively impact the anode performance and the smelter cost if the needed adaptations of the entire process are not performed and the operations not optimized for the high S/V anodes used in high amperage pots.

The resulting coke quality available to the Russian smelters today and in the future is addressed in this paper. Recommendations are given for a safe usage of the medium to high S/V cokes in the prebaked carbon plants and in the pots.

Green cokes : availability and quality

A good review of the Russian coke market was given in 2009 by Buzunov et al. [2]. The current green coke demand has remained practically unchanged at about 1.4 Mio tpy covering the need of a total aluminium production of 3.5 Mio tpy in Russia. The refinery groups with delayed coker units are, in descending order of tonnage, Rosneft, Lukoil, Bashneft and Sibneft. Excess green coke produced in Azerbaijan (Socar Baku) and in Kazakhstan (CCL Oil at Pavlodar) is imported by the Russian smelters. There are no real merchant calciners, while in US Gulf most of the calcined coke is produced by such independent players. In Russia the refineries having their own calcination unit are the exception (a total of 200'000 tpy calcined coke capacity at Sibneft Omsk and Lukoil Volgograd), here again a situation that is quite different from Europe where the calcination step is integrated in the refineries producing green coke.

The green cokes potentially available to the Russian smelters are divided in three classes as a function of their Sulfur contents (Table 1). The low Sulfur cokes (LS) are produced by the refineries of Sibneft Omsk, Lukoil Volgograd, Rosneft Angarsk and Komsomolsk. Medium Sulfur cokes (MS) are produced at the Lukoil Perm refinery and are imported from the Socar Pavlodar refinery (Kazakhstan). Rosneft in Novo-Kuibyshevsk and Bashneft in Ufa produce high S green cokes (HS) containing very high V impurities. The weighed overall blend of the different cokes is close to 50 % LS and 25 % each of MS and HS green cokes.

The Russian classification has some analogy with those prevailing in USA [3] and in China [4], (Table 2). The major difference being that the HS Russian cokes are still lower in S than the

Table 1. Green coke properties

Property	Unit	Russian green cokes			Worldwide range of blended coke
		LS	MS	HS	
S	%	1.5	3.0	4.5	2-4
V	ppm	150	400	900	200-400
Ni	ppm	100	100	200	100-200
Fe	ppm	200	200	200	100-300
Si	ppm	150	150	250	100-200
Na	ppm	100	150	150	50-150
Ca	ppm	40	40	50	50-200
VCM	%	10	10	10	9-11
HGI	-	85	85	50	70-90

Table 2. Comparison of the green coke classifications adopted in Russia, USA and China

Russia	S (%) V (ppm)	LS 1.5 150	MS 3 400	HS 4.5 900	
USA	S (%) V (ppm)	A 0.7-1.8 50-240	C 2.1-3.8 130-460	D 3.2-5.4 220-470	NT 5.0-6.5 500-600
China	S (%)	2B 0.8-1.5	3B 2-3	4A 3-5	4B 5-7

Non -Traditional (NT) cokes used in USA [5] or than the category 4 B introduced recently as a 4th Chinese anode grade category. However the V content of the HS Russian cokes largely exceed the one processed as NT coke by the merchant US Gulf calciners at a typical level of 20 % in their blend.

Attention has to be paid on the high level of isotropy (high coefficient of thermal expansion CTE also of the anodes) of the HS green cokes, as shown by the lower level of Hardgrove Grindability Index (HGI) which reflects the hardness and inelasticity of the material, an aspect that might create too high a thermal shock brittleness of the corresponding anodes. As this issue is not a topic for Söderberg anodes, due to the slow baking rate of the carbon anodes, the Russian smelters are probably not very familiar with this anode cracking risk.

In respect of the deterioration of the purity aspect of green cokes that will be available in Russia, the same trends as observed in USA [6] and in China are expected to occur. A gradual increase of the S and V concentration in the green coke blend calcined by the Russian smelters has to be anticipated. Therefore the gradual disappearance of LS cokes (like the one of Rosneft Komsomolsk), the start-up of delayed cokers [7] processing heavier residuals (Rosneft Achinsk and Krasnodar, Bashneft Ufimsky) but also the Rusal project of Boguchansky and Taishet, with a total green coke demand exceeding 700'000 tpy of green coke, will affect the availability of the conventional raw materials used for the prebaked anodes in the Irkutsk, Sayansk and Krasnoyarsk prebaked smelters.

The weighed overall blend of the different cokes available in the first half of this decade was close to 50 % LS and 25 % each of MS and HS green cokes .Tentatively we anticipate that at the end of this decade, when all the projected prebaked smelters (1.5 Mio tpy Al capacity) will be in operation, the representative blend of green coke will be 40 % of MS cokes and 30 % each of LS and HS cokes.

Calcined coke quality forecast

The properties of the calcined “Russian Coke Blend“ are given in the Table 3, along with the range of calcined cokes from blended green cokes provided by merchant calciners based in USA, India and China.

The properties are predicted by using well established correlations on the impact of impurities present in the coke [8, 9] and of its relevant green physical properties like the VCM and HGI, on rotary kiln calcined coke [10]. For the calcination degree a moderate real density target is chosen to avoid significant desulfurization of the high S green cokes present in the blend as shown in Fig. 1. This aspect

Table 3. Calcined coke properties (*30 % LS / 40 % MS / 30 % HS)

Property	Unit	Russian coke Blend*	Worldwide range of blended coke
S	%	2.7	2.0-3.5
V	ppm	500	200-400
Ni	ppm	160	100-200
Fe	ppm	250	100-300
Si	ppm	200	100-200
Na	ppm	150	50-150
Ca	ppm	50	50-200
Bulk density 2-1 mm	Kg/dm ³	0.83	0.78-0.83
Real density	Kg/dm ³	2.06	2.04-2.08
Sp. El. Resistance	μΩm	470	450-520
CO ₂ reactivity	%	12	5-12
Air reactivity 525 °C	%/min	1.0	0.3-0.6

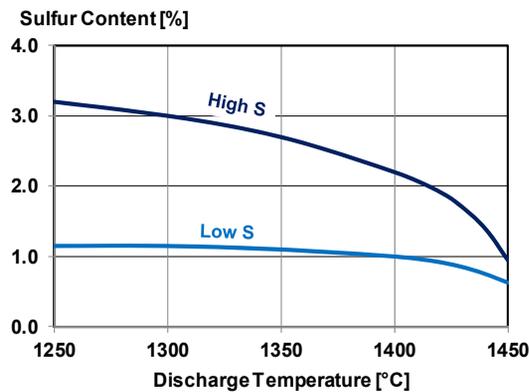


Fig. 1. S Loss for HS and LS cokes during calcination

is treated later in Chapter 5 dealing with optimum processing conditions for producing bench mark anodes.

Concerning the porosity, illustrated by the tapped bulk density figures, the Russian coke blend shows a level on the high side of the worldwide range of blended coke as a higher isotropy related to the usage of HS green coke is anticipated. For the other properties, the Russian calcined coke blend appears to be within the range of blended coke currently used in North America, India and the Middle East except for the V content that is higher and for the Na content that is on the high side of the range. The CO₂ and air reactivities are on the high side and well above the worldwide ranges of blended cokes. While the catalytic effect of V on the air reactivity cannot be eliminated, some improvements could be achieved for the Na content, another strong catalyst, by deeper desalting of the crude oil in refineries [11].

The impact of the poor purity, increased S content compared to LS cokes and drawbacks of the calcined coke properties on the prebaked anode properties are addressed in the next chapter.

Prebaked anode quality forecast

Prediction of the anode properties can be done by using the extended pilot plant studies made by several researchers [12–14] but also by evaluating results of full size anodes resulting from a worldwide survey of smelters using blended cokes with different levels of impurities and physical characteristics [15]. Table 4 shows the anode properties of representative smelters using blended cokes as a basis.

The dispersion of the mean and 2σ values given in the worldwide range with blended coke is a combined result of the range of the coke properties and of the anode plant peculiarities (equipment, processing conditions and control). As modern carbon plants are planned to be installed for the future Russian prebaked smelters, it may be anticipated that the best anode quality is targeted for the anode plant of Taishet designed to deliver its blocks to four smelters (Boguchansky, Taishet, Irkutsk and Krasnoyarsk). The bench mark anode quality with the Russian coke blend is therefore listed in Table 4.

The apparent density of these bench mark Russian coke anodes being on the high side of the worldwide range, all mechanical properties are in the upper side of the given ranges. The CTE and the elasticity modulus being high, the anode process control shall be impeccable to achieve consistent anode production, as shown by the lowest possible variabilities of the properties like the resistivity and flexural strength. Crack free anodes are mandatory for avoiding thermal shock issues [16], and this is shown by the low 2σ values of both mentioned properties.

Table 4. Properties of prebaked anodes

Property	Unit	Worldwide range with blended coke		Bench mark with Russian blend	
		\bar{X}	2σ	\bar{X}	2σ
Baked apparent density	Kg/dm ³	1.55-1.60	0.02-0.04	1.60	0.02
Sp. Electrical resistance	$\mu\Omega\text{m}$	52-56	3-6	53	3
Flexural strength	MPa	10-14	2-4	14	2
Compressive strength	MPa	40-55	8-15	55	8
Elasticity modulus	GPa	4.0-5.5	1.0-2.0	5.5	1.0
CTE	10^{-6} K^{-1}	3.08-4.4	0.3-0.6	4.4	0.3
Thermal conductivity	W/mK	3.5-4.5	0.5-1.0	4.2	0.5
Real density	Kg/dm ³	2.07-2.09	0.01-0.02	2.07	0.01
S	%	1.8-3.2	0.1-0.4	2.5	0.1
V	ppm	180-350	10-40	450	20
Fe	ppm	200-400	100-200	350	100
Si	ppm	100-200	50-100	200	50
Na	ppm	150-300	100-200	250	100
Ca	ppm	50-200	50-100	50	50
Air permeability	nPm	0.5-1.5	0.5-1.0	0.5	0.5
CO ₂ reactivity Residue	%				
Dust	%	90-96	1-4	95	2
Dust	%	1-3	1-3	1	1
Air reactivity Residue	%				
Dust	%	70-90	5-10	75	8
Dust	%	1-5	1-4	2	2

A moderate and consistent baking degree is anticipated to cope with the desulfurization issue of the HS coke present in the blend and to avoid underbaked anodes with dusting propensity in the cells. The Na contamination from the butts recycling is minimal as the difference between the anode and coke Na values is only 100 ppm (~ 400 ppm in butts). As a consequence, the CO₂ reactivity is excellent and the air reactivity residue still lies within the typical range of worldwide anodes made with blended cokes despite the handicap of much higher V concentration.

To achieve these ambitious figures all steps of manufacturing from the green coke blending to the butts recycling need to be professionally performed.

The long optimization chain for producing bench mark anodes

The experiences made on the usage of high S and of isotropic cokes have been reported in many papers, already some decades ago [17–19] or more recently [20]. Usage of 20 to 40 % of very high S/V cokes by blending them to relatively low S/V conventional anode grade cokes have been experimented successfully. However their drawbacks can be summarized as follows:

- extra fine size porosity due to desulfurization during calcination with increased SO₂ emission and lower yield;
- higher hardness resulting in a drop of the ball mill fines capacity, wear of crushers and higher iron contamination in the dry aggregate;
- higher resiliency of the coke, lower shrinkage and higher loss during baking resulting in higher green to baked anode density differences [17];
- higher net carbon consumption, lower current efficiency and higher energy consumption in pots sensitive to air burn;
- higher V content in metal and higher SO₂ concentration in the cell off-gas [18].

Optimizations of the entire process are thus needed to avoid the traps in the coke and anode production. Special care should be given to select equipment and installation in the production.

Delayed Coking in the Refinery

Very high S/V residual coking being considered as a feedstock giving intrinsically a low value fuel grade coke, the trap would be to design and run the unit as a fuel coker with very low recycle and short cycle time. This would result in a more impure and high VCM green coke production as the coke production is minimized. The aluminium industry should consider paying a reasonable price for such NT coke, a price being higher than for fuel grade coke. Otherwise the refinery has no motivation to design and run an anode grade coker and ignores the importance of a good control of the quality and consistency of the green coke.

Rotary Kiln Calcining

Most of the small rotary kilns (100'000 tpy) installed in the Russian smelters suffer from poor run length and their maintenance is problematic. Improvement of the design and quality of the lining are needed and a capacity raise to a state of the art production of 300'000 tpy per unit.

The blending of the green cokes directly after the discharge in a blending bed covered stockyard (see Fig. 2) with longitudinal Chevron stacking and frontal reclaiming by harrows guarantees a homogenous

kiln feed in term of purity, sizing, VCM and moisture. Compared to the current system used in Russia, consisting of piling of the green cokes separately in bays and reclaiming them intermittently by using the crane grab, a much better stability of the calcining process is guaranteed. This has a positive impact on the yield and the calcined coke quality and consistency. Hence this is a must for the stability of all downstream processes.

A big issue addressed in the literature is the selection of the calcining degree target for reaching the best level of coke properties [21], like for the air reactivity or the electrical resistance, but more importantly the best achievable anode performance [22]. The issue is to minimize the negative effects of the coke denitrification and desulfurization (Fig. 3), as observed on the apparent density (Fig. 4), especially on HS cokes present in the coke blend [23 – 25].

Eventually a moderate calcination severity appears to be the best compromise for avoiding the creation of extra micro-porosity, which also means poorer oxidation resistance of the coke and of the other mechanical issues related to the coke behavior during forming and baking.

For a good control of the calcination degree a modern and automatic system adapting the burner power to a selected finishing temperature level is mandatory. A routine quality control including the measurements of the specific electrical resistance and the tapped bulk density among other things has to be implemented as well.



Fig. 2. Circular blending bed with stacker and reclaimer

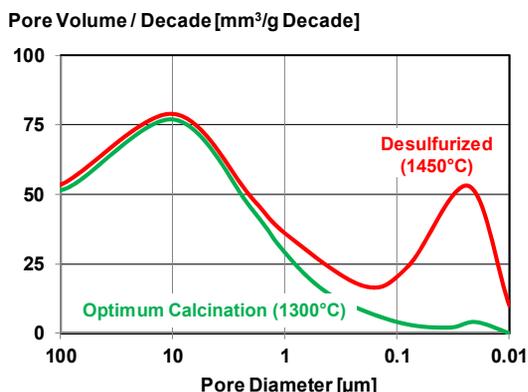


Fig. 3. Microporosity generated by over calcining of HS coke

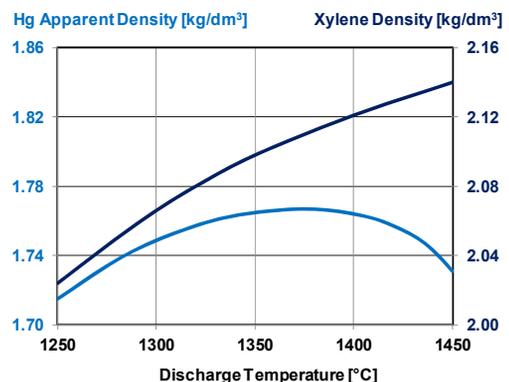


Fig. 4. Decrease of the apparent density at too a high calcining degree

Paste Plant

The cleaning of the butts is unfortunately often beyond the responsibility of the paste plant. However, having in mind that coarse butts is a dense raw material (denser than coarse coke), processing of butts in crushers and sieving machines that maximize the amount of coarse butts (20 – 4 mm) will provide a good basis for high density and low permeability anodes [26]. Minimizing the amount of coarse porous coke is advantageous while feeding the ball mill with the overflow of the finest coke fraction guarantees a fines production rate in line with the nominal expectation, despite the relative hardness of the blended coke.

As there are some drawbacks in the usage of the Russian blend cokes related to the thermal shock and anode burning aspects, it is important to optimize the processing conditions of the state of the art preheating/mixing/forming equipment. It is the goal to avoid any crack formation and inhomogeneities in the anode block at an elevated apparent density that guarantees a low air permeability to compensate the inferior air reactivity of the anode. This means usage of high softening point pitches promoting a low baking loss but also the availability of efficient preheater, powerful continuous mixer and vacuum forming machine. A paste cooler is a must as it allows mixing optimizing separate from forming. At last an in-plant routine quality control for the fines (Blaine tester) with a control model coping with block density fluctuations complete the picture for producing first class green blocks with zero scrap delivered to the bake furnaces.

Baking Furnace

The overall effect of the baking conditions on the anode properties was well illustrated by Fischer et al. [27]. The baking dilemma for high S coke anodes was already addressed in the paper [22] and more recently detailed industrial experiences on the minimization of desulfurization during baking were reported. [28].

Figures 5 and 6 show that an excessive anode baking temperature above 1150 °C results in lower anode weight combined with a deterioration of its air reactivity residue, without noticeable decrease of the anode electrical resistivity but a significant increase of the thermal conductivity.

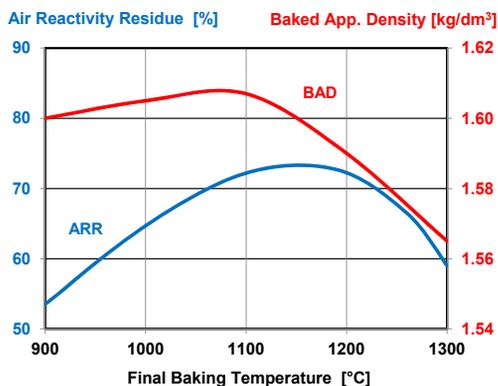


Fig. 5. Air reactivity / apparent density vs. anode baking temperature

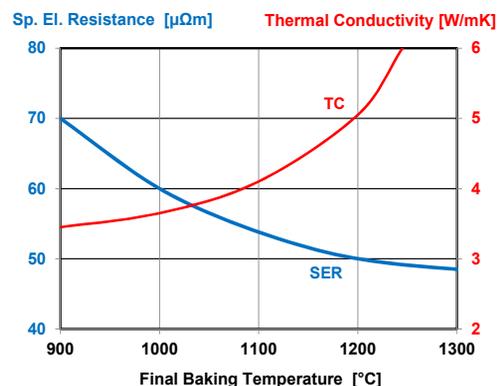


Fig. 6. Sp. el. resistance / therm. Conductivity vs. anode baking temperature

The loss of anode weight through denitrification and desulfurization can reach more than 20 kg for a 1 ton anode block. The higher thermal conductivity increases the anode top temperature that can double the air burn rate, especially if the anode is not well covered. The anode cycle time may then have to be reduced to avoid lower pot performance in terms of metal output.

Hence a compromise should be aimed between low electrical resistivity of the block and minimum air burn without dusting issues. Furthermore minimal deviations related to the fire control and to the temperature distribution within the pit should be targeted. Emphasis during the design engineering phase of the baking furnace should be given to the pit and flue design but also to the selection of a relatively long nominal cycle time that is favorable for the pit temperature distribution. A metal capacity creep of 15 % of the smelter through higher current intensity shall also be considered in the calculation of the pit dimensions (longer anodes!) and in the number of fires and pits eventually needed. A representative routine anode quality control including real density or crystallite size testing together with a sound interpretation of their levels and variations, taking into account the one found on the coke [29], will allow an optimization of the baking step. The effect of the butts cleanliness on the reactivity dust values shall also take into account the degree of calcination of the coke blend [29].

Anode in the Cells

Thermal cracking of dense and high CTE anodes being the first potential concern with anodes made of Russian coke blend, the cell designer shall be cautious with the magneto-hydro-dynamic conditions, thus avoiding too strong bath movements. The bath chemistry in the operations shall be selected for moderate superheat. The dissolution of the crust formed on the cold anode block just after setting shall not be too rapid so the thermal stresses in the anodes remain moderate. Pot start-up conditions shall integrate these aspects; bad experiences have been made during this phase with unacceptable cracking rate of the blocks.

All publications made on the effect of high S and V anodes demonstrate that air burn is critical especially in shallow cavity pots and in high current density cells where the thermal equilibrium is easily disrupted so that the anode cover thickness is minimized to maintain a good ledge protection and a high current efficiency. It is recommended to select a combination of cell design and operation where the anodes remain extremely well protected [30], as the extent of air burn decreases exponentially with the thickness of the protective layer. The detailed and accurate simulation of the cell thermal state (also the dynamic one!), appears to be of prime importance, especially with the HS air reactive anodes.

Crushed anode cover sizing and alumina content are important for low permeability of the anode cover in service [31, 32]. An anode setting cycle that is favorable to limit the risk of poorly protected anode top just after the anode change is also helpful [33]. Last but not least in the practices and methods of covering, the timing of adding the anode cover and of the final dressing of the anodes [34], should be considered by the operators and the management.

Anode Butts

Although the HS anodes appear to be less sensitive to the catalytic effect of the Na related to butts recycling [19], it has to be emphasized that an impeccable cleaning of the butts through several stages in the rodding area is badly needed. The air reactivity figure without butts addition would be just fair

as the V content is high and the integrity of the baking furnace has to remain at the highest standard for avoiding poor baking conditions with risks of desulfurization together with dusting related to poor bake furnace state [35].

Recycling of baghouse fines from butts crushing and from bake furnace should be avoided. Bath impregnated butts (swimmer mainly) should be discarded as well. The crushing step of butts in the paste plant was described earlier so it is just mentioned here that crushers producing a lot of fines (like hammer or impact crushers) should not be used.

Conclusions

With the restart of idle capacity in existing Russian smelters and the completion of new large smelter projects by the end of this decade, the availability and quality of the green cokes for prebaked anodes become a major issue. Developing a cooperation in petroleum coke supplies between refiners and smelters, as started by Rosneft/Rusal [36], is surely one answer to the above mentioned issue. The experience and knowledge on the impact of HS cokes in calciners, anode plants and anode use in the smelters are decisive for a good performance in the cells.

The selection of powerful equipment and state of the art control system in the carbon side are of prime importance for guaranteeing the consistency of the products in the long chain of fabrication. Optimization of the processing parameters, taking into account the available quality of coke blend that might deteriorate, will allow the production of bench mark anodes. In the same line the complete integration of the impact of the anodes on the electrolysis process, which is facing new challenges [37], will provide a good basis for the lowest possible aluminium production cost.

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